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ON WAVE PROPAGATION IN RANDOM PARTICULATE COMPOSITES

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Abstract—A new method is presented for analysis of wave propagation in random particulate viscoelastic composites. The method incorporates both the scattering effect and viscoelastic losses as well as the Kramers–Kronig relationships valid for any casual linear system. Explicit expressions for the attenuation and dispersion are given and compared with available experimental data.

1. INTRODUCTION

The recent appearance of five papers[1–5] devoted solely to the frequency-dependent response of disordered composites clearly indicates the increasing interest in this topic. The work of Gaunaurd and Überall[1] made use of the method due to Ament[6] whereas Kligman, Madigosky, and Barlow[2] employed a slightly different approach due to Chaban[7]. The essential procedure in both of these works is based on equating the scattered field in the composite at hand to that due to a sphere made of the effective homogeneous material. A different approach to the problem was taken by Junger [3]. His method can be viewed as a dynamic analog of the mixture rule of Reuss, well-known in the static analysis of composites. Two other papers, by Kinra *et al.*[4] and by Kinra and Anand[5], reported experiments with an epoxy-glass particulate composite. While similar studies were published previously (see[8–9]), this work appears to be the first sound experimental investigation of harmonic wave propagation in particulate composites.

The method, systematically presented in this paper, is an extension of the approach briefly outlined by Beltzer[10] for the case of elastic porous media. The results obtained in [10] were shown to agree with those of Junger[3]. Section 2 of the present paper deals with deducing of the basic equations for the general case of a viscoelastic particulate composite. The approach is based on combining the scattering analysis with the Kramers–Kronig (K–K) relationships. In Section 3, we consider a viscoelastic matrix with the linear law of attenuation. Explicit results for dispersion and attenuation in the composite are given and compared with experiments.

2. BASIC EQUATIONS

The theories presented in [1, 2, 6, 7] take equivalency of the scattering in the composite and that in an "effective sphere" to be the single criterion for identification of the effective parameters. Both attenuation and dispersion are deduced solely from the scattering analysis. It seems more logical, however, that in addition to scattering, some general properties of the linear systems should be taken into account in formulating the effective medium. The appropriate method is given herein.

Let us consider harmonic wave propagation (with frequency ω) in an isotropic medium defined by its viscoelastic compliance $J(\omega) = J_r(\omega) + iJ_i(\omega)$ and density ρ. The Kramers–Kronig relationships, valid for any casual linear system, and the associated links imposed between $J_r(\omega)$ and $J_i(\omega)$ were derived and rederived many times (see, e.g. Ben-Menachem and Singh[11] and O'Donnell *et al.*[12]). In particular, they yield the following result

$$J_i(\omega) = -\frac{2\omega}{\pi} P \int_0^\infty \frac{J_r(\omega_1)}{\omega_1^2 - \omega^2} d\omega_1 \quad (1)$$

where P denotes the Cauchy principal value. Let us introduce the complex wave number, $K(\omega)$ defined as

$$K(\omega) = K_r(\omega) + iK_i(\omega) = \omega[\rho J(\omega)]^{1/2} \tag{2}$$

$$K_r(\omega) = \omega/c(\omega);\ K_i(\omega) = \alpha(\omega) \tag{3}$$

where $c(\omega)$ is the dispersive phase velocity and $\alpha(\omega)$ is the attenuation. In the almost forgotten paper of Futterman[13], it was shown that the K–K relationships can be also formulated in terms of the refraction index, $n(\omega)$, given by

$$n(\omega) = n_r(\omega) + in_i(\omega) = K(\omega)/(\omega/c_0) \tag{4}$$

where c_0 is the nondispersive limit of the phase velocity occurring at low frequencies. We shall need only one of his equations similar to eqn (1)

$$n_i(\omega) = -\frac{2\omega}{\pi} P \int_0^\infty \frac{n_r(\omega_1)}{\omega_1^2 - \omega^2}\,\mathrm{d}\omega_1; \quad (n_i(\infty) = 0). \tag{5}$$

Recently O'Donnell et al.[12], making use of the technique due to Bode[14], pointed out that for intervals at which both $J_r(\omega)$ and $J_i(\omega)$ are slowly varying functions (no sharp resonances), eqn (1) yields the following approximate relationship:†

$$J_i(\omega) \simeq -\pi\omega(\mathrm{d}J_r(\omega)/\mathrm{d}\omega)/2 \tag{6}$$

In view of the similarity between eqns (1) and (5), it follows that

$$n_i(\omega) \simeq -\pi\omega(\mathrm{d}n_r(\omega)/\mathrm{d}\omega)/2. \tag{7}$$

We proceed now with application of the presented results to the dynamics of random heterogeneous media. Let us consider a material consisting of a low-loss viscoelastic matrix with a completely random distribution of spherical perfectly elastic inclusions. The typical radius of the inclusions is denoted as a and the mean interinclusion separation as $S (S \gg a)$. No other information is available on the geometry of the composite. The volume fraction, ϕ, and S are related by [2]

$$\phi = (4\pi a^3/3)/S^3. \tag{8}$$

Decay of a disturbance propagating in such a medium is caused by the attenuation in a viscoelastic matrix, $\alpha_v(\omega)$, and that due to scattering by the inclusions, $\alpha_{sc}(\omega)$. According to Kuster and Toksöz[15], the effective total attenuation is given by

$$\alpha(\omega) = n_i(\omega)(\omega/c_0) = \alpha_v(\omega) + \alpha_{sc}(\omega). \tag{9}$$

The first term, $\alpha_v(\omega)$, the characteristic of the matrix, is assumed to be known, whereas the explicit expressions for $\alpha_{sc}(\omega)$ were given in the works of Waterman and Truell[16] and Yamakawa[17]. In particular, the Yamakawa result, valid for a dilute suspension, can be cast in the form

$$\alpha_{sc}(\omega) = 3\phi/(8\pi a^3)\gamma \tag{10}$$

where γ is the scattering cross section of a single inclusion. The explicit low-frequency approximation for $\alpha_{sc}(\omega)$ will be given in the next section.

Now one observes that eqns (10), (9) and (5), as well as eqns (10), (9) and (7), constitute a closed system which allows determination of the refraction index, $n(\omega)$. Hence, the complex wave number $K(\omega)$, can also be found via eqn (4) if the nondispersive

†Although no estimates of the accuracy are given in [14] it seems that eqn (6) is of an asymptotic nature.

low-frequency velocity c_0 is known. Fortunately, extensive literature exists on effective non-dispersive (static) behavior of disordered composites. Most of the results are almost identical in a dilute suspension limit (small volume fraction of inclusions, ϕ). In particular, according to [2] (see also [20])

$$c_0^2 = (\lambda_0 + 2\mu_0)/[(1-\phi)\rho_1 + \phi\rho_2)] \tag{11}$$

$$\lambda_0 + 2\mu_0 = (\lambda_1 + 2\mu_1)\left\{1 + 3\phi\left[\frac{\lambda_1 + 2/3\mu_1 - \lambda_2 - 2/3\mu_2}{3\lambda_2 + 2\mu_2 + 4\mu_1} + \frac{20}{3}\cdot\frac{\mu_1(\mu_1 - \mu_2)}{\mu_2(16\mu_1 + 6\lambda_1) + \mu_1(14\mu_1 + 9\lambda_1)}\right]\right\}^{-1} \tag{12}$$

with ρ, λ and μ denoting density and the elastic Lame' parameters, respectively and the subscripts 0, 1 and 2 standing for the effective medium, matrix, and inclusion. For moderate magnitudes of the volume fraction, $\phi \simeq 0.2$–0.4, more accurate results due to Datta[18] or Chen and Acrivos[19] should be applied.

In the following section, we will focus on experimental investigations of wave propagation in a viscoelastic particulate composite presented in [4, 5] and will compare them with the present theory.

3. EXPERIMENTS AND THEORY

Very recently Kinra *et al.*[4] and Kinra and Anand[5] reported on experimental studies of wave propagation in a disordered composite consisting of an epoxy matrix filled with glass spheres. The densities of the constituents are given by $\rho_1 = 1.180\ \text{g/cm}^3$; $\rho_2 = 2.492\ \text{g/cm}^3$ and the wave velocities by $c_{1\alpha} = 2.54 \times 10^5\ \text{cm/s}$; $c_{1\beta} = 1.16 \times 10^5\ \text{cm/s}$; $c_{2\alpha} = 5.28 \times 10^5\ \text{cm/s}$; $c_{2\beta} = 3.24 \times 10^5\ \text{cm/s}$, where the subscript α stands for P-waves and β for S-waves. The experiments showed that the inclusions can be viewed as purely elastic ones whereas the losses in the matrix can be approximated by a linear law

$$\alpha_v(\omega) = m\omega \quad (\omega \geq 0) \tag{13}$$

where m is a constant ($m = 0.456 \cdot 10^{-6}/2\pi$ sec/cm). The dispersion and attenuation were measured for frequencies in the interval 0.3 – 5 MHz. The obtained data show no explicit resonances, at least for the Rayleigh region $|Ka| < 1$.

The law given by eqn (13) is widely employed to model viscoelastic losses. However, it is usually overlooked that eqn (13) leads to infinite value of wave velocity, unless a cut-off frequency, ϵ, is introduced. Discussions of this interesting effect can be found in the works of Futterman[13] and Knopoff and MacDonald[21]. Thus, this equation should be replaced by the Futterman law containing two experimental constants, m and ϵ

$$\begin{aligned} \alpha_v(\omega) &= m(\omega); \quad && \omega \geq \epsilon \\ \alpha_v(\omega) &= 0; && \omega < \epsilon \end{aligned} \tag{14}$$

where $\epsilon \neq 0$. Futterman indicated that due to the logarithmic dependence between the phase velocity $c(\omega)$ and ϵ, only an approximate estimate of ϵ is needed. Since ϵ is a typical frequency of the interval at which dispersion-attenuation can be neglected and is located below the region of interest, it follows that for the discussed experiments $\epsilon \simeq 10^6\ \text{sec}^{-1}$. For this magnitude of frequency, the dimensionless wave number, $K_{1\alpha}a = \omega a/c_{1\alpha}$, equals ~ 0.06. Since c_o, given by eqns (11) and (12), arises (see [2, 15]) as the limit of the effective speed when $K_{1\alpha}a \to 0$, one expects this value to yield a fair approximation for $c(\epsilon)$*

$$c(\epsilon) \simeq c_0. \tag{15}$$

The aforementioned remarks on the existence of a cut-off frequency affect the analysis of

*One, thus, neglects the slight attenuation in the interval $0 < \omega < \epsilon$ due solely to the scattering by the inclusions.

the experiments [4, 5] rather than the presented data. However, it will be shown that in theoretical considerations, this fact has radical consequences.

Next let us deduce explicit expressions for attenuation and dispersion in the composite, making use of the results of Section 2. Under the conditions of a dilute suspension and the Rayleigh frequency region, the attenuation due to scattering (see eqn 10) is given by Yamakawa[17] as follows ($K_{1\alpha}a < 1$; $K_{1\beta}a < 1$)

$$\alpha_{sc}(\omega) \simeq \Delta\omega^4 \tag{16}$$

where

$$\Delta = 3\phi a^3\{2B_0^2 + 2/3[1 + 2(c_{1\alpha}/c_{1\beta})^3]B_1^2 + 1/5[2 + 3(c_{1\alpha}/c_{1\beta})^5]B_2^2\}/(4c_{1\alpha}^4)$$
$$B_0 = 1/3(3\lambda_1 - 3\lambda_2 + 2\mu_1 - 2\mu_2)/(4\mu_1 + 3\lambda_2 + 2\mu_2)$$
$$B_1 = 1/3(1 - \rho_2/\rho_1)$$
$$B_2 = -20/3\mu_1(\mu_2 - \mu_1)/(16\mu_1\mu_2 + 6\lambda_1\mu_2 + 14\mu_1^2 + 9\lambda_1\mu_1)$$

with λ, μ standing for the Lame' parameters.

Invoking eqns (13) and (9), one finds

$$\alpha(\omega) = m\omega + \Delta\omega^4 \tag{18}$$

$$n_i(\omega) = c_0(m + \Delta\omega^3) \tag{19}$$

for the interval

$$\omega \geq \epsilon;\ \omega a/c_0 < 1.$$

Since no sharp resonances are present, the approximation given by Eq. 7 is valid to yield for the same interval

$$\mathrm{d}n_r(\omega) = -2c_0(m\omega^{-1} + \Delta\omega^2)\,\mathrm{d}\omega/\pi \tag{20}$$

$$n_r(\omega) = 1 - 2c_0[m\ \ln(\omega/\epsilon) + \Delta(\omega^3 - \epsilon^3)/3]/\pi \tag{21}$$

where $n_r(\epsilon) = 1$ was substituted on the basis of eqns (4) and (15). Thus, the low-frequency approximation for the phase velocity, $c(\omega)$, is given by

$$c(\omega) \simeq \{c_0^{-1} - 2[m\ \ln(\omega/\epsilon) + \Delta(\omega^3 - \epsilon^3)/3]/\pi\}^{-1}. \tag{22}$$

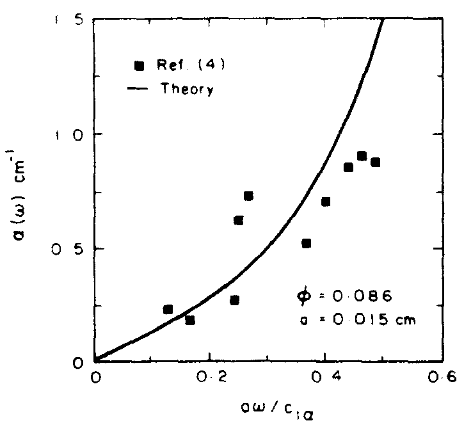

Fig. 1. Wave attenuation in a composite, $\phi = 0.086$.

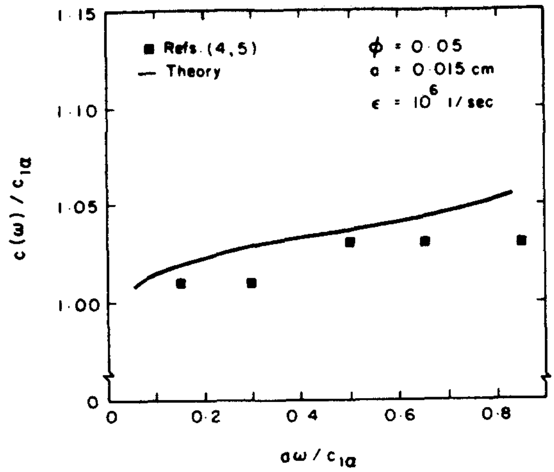

Fig. 2. Wave dispersion in a composite, $\phi = 0.05$.

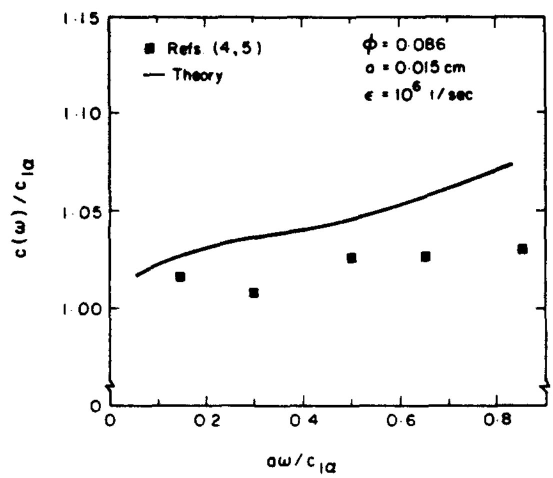

Fig. 3. Wave dispersion in a composite, $\phi = 0.086$.

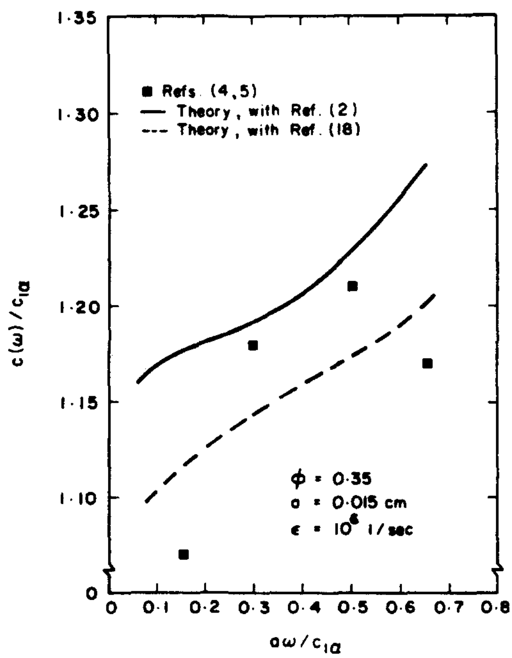

Fig. 4. Wave dispersion in a composite, $\phi = 0.35$.

As expected, the logarithmic dependence appearing in eqn (22) indicates that ignoring the cut-off frequency effect ($\epsilon = 0$) leads to the breakdown of the theory, confirming, thus, the conclusion of Ref. [21] on inconsistency of eqn (13) with the linear theory of wave propagation.

Comparisons between the experimental data (dilute mixture and the Rayleigh frequency interval) and eqns (18) and (22) are shown in Figs. 1–4. A very satisfactory agreement for attenuation, $\alpha(\omega)$, is obtained, as displayed in Fig. 1. The dispersion is slight, as Figs. 2–4 show for different values of the volume fraction, $\phi = 0.05$, 0.086 and 0.35. Computations confirmed the remark of Futterman on a slight correlation between $c(\omega)$ and ϵ. The values of $c(\omega)$ computed for $\epsilon = 10^5\,\text{sec}^{-1}$ were found to differ about 3% from those presented in Figs. 2–4, computed for $\epsilon = 10^6\,\text{sec}^{-1}$.

Figure 4 needs the following explanations. For moderate magnitudes of the volume fraction, such as $\phi = 0.35$, the use of the result of Datta[18] is more justified than the use of eqn (11), taken from[2]. In fact, as Fig. 4 shows, incorporation of the Ref. [18] result makes the comparison more favorable.

4. CONCLUSIONS

A new method was presented for analysis of disordered viscoelastic composites which yields a convenient means for the extension of static results to the dynamic case. The method consists of computation of the losses due to scattering as well as a viscoelastic losses and subsequent application of the Kramers–Kronig relationships to derive the wave speed, $c(\omega)$. The possibility of incorporating any theoretical or empirical value for the non-dispersive velocity c_0 is an advantage of the method. The method, along with the Futterman law of viscoelastic losses, provides the results matching experimental data on wave propagation in epoxy-glass particulate composites. It is believed that a good agreement between frequency-dependent data for the theory and experiments is demonstrated here for the first time.

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