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RESONANT SCATTERING OF ELASTIC WAVES BY A RANDOM DISTRIBUTION OF INCLUSIONS†

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Abstract—Moon and Mow considered the propogation of a plane longitudinal wave in a random particulate composite consisting of a homogeneous distribution of rigid spheres in welded contact with an elastic matrix. The main assumptions of their model are the inclusions are heavy and rigid, the wavelengths are large and the suspension is dilute. One of the conclusions of their analysis is that there exists a cut-off frequency which is associated with the translational resonance of the individual particle in its elastic surroundings. The purpose of the present experimental work is to demonstrate that in spite of the fact that their model rests on a number of rather restrictive assumptions, it predicts the resonance frequency with a reasonable accuracy. Therefore the model is good, well outside the range of its underlying assumptions.

1. INTRODUCTION

In a recent series of papers, the propogation of ultrasonic waves in particulate composites has been studied experimentally by Kinra *et al.*[1–5]. The primary quantity of interest was the phase velocity of longitudinal and shear waves in the composite $\langle C_1 \rangle$ and $\langle C_2 \rangle$, respectively. In [1], the attention was confined to the low-frequency regime. In [2], all three frequency regimes of interest were studied: low, intermediate and high. Consistent with the predictions of Moon and Mow[6], the wave propogation was found to occur along two separate branches, namely, the acoustical branch and the optical branch. The two branches were found to be separated by a range of frequencies over which neither $\langle C_1 \rangle$ nor $\langle C_2 \rangle$ could be measured: this range was labeled the "forbidden zone." However, one of the key assumptions of [6]—that the inclusions should be very much heavier than the matrix—was not satisfied in [2]. Therefore it is not at all surprising that a *quantitative* comparison, between the results of Moon and Mow and the experiments of [2], was found to be rather poor. This problem was rectified in [3] where lead—rather than glass—inclusions were used; only $\langle C_1 \rangle$ was measured. As predicted by Moon and Mow, the longitudinal phase velocity was found to increase rapidly with frequency *in the vicinity* of the predicted cut-off frequency. However, this increase is *continuous* and, therefore, it is not possible to deduce precisely the cut-off frequency, n_c, from the $\langle C_1 \rangle$ data. This provided the motivation for the present work. Here, the emphasis is on the *amplitude* of an ultrasonic wave received through a specimen: expectedly, as $n \rightarrow n_c$, the amplitude goes through a sharp minimum. Thus in contrast to the phase velocity data, n_c can be determined fairly precisely from the amplitude data. The measured n_c was found to be in excellent *quantitative* agreement with the predictions of Moon and Mow. In the next section we briefly outline the theory of Moon and Mow.

2. THEORY OF MOON AND MOW

Consider a single *rigid* sphere in an *elastic* matrix. If the particle is linearly displaced from its equilibrium position (no rotation) and let go, it will undergo damped

† This paper is dedicated to Protessor Vijay Kumar Stokes.

translational vibrations. The damping here is not due to the conversion of elastic energy into heat (viscous damping), but rather due to the radiation of mechanical energy away from the sphere in the form of elastic waves (radiation damping). The equation of motion for the sphere was derived by Pao and Mow[7], and its transient response was studied by Mow[8]. Later Moon and Mow[6] showed that this equation of motion has the form of a damped oscillator with memory. By taking this equation of motion as a starting point, they constructed a model for the aggregate behavior of a random particulate composite under the following key assumptions.

1. The spheres are dispersed in a statistically random and homogeneous manner: i.e. in the aggregate, the composite behaves like an isotropic homogeneous medium.
2. The inclusions are *heavy*: i.e. $\rho'/\rho \gg 1$, where ρ is the density, and ( ) and ( )′ denote the matrix and the inclusion material, respectively.
3. The inclusions are *rigid*.
4. The volume fraction of inclusions $\bar{C} \ll 1.0$, the so-called dilute suspension. This assumption allows the authors to ignore multiple interaction effects between the particles.
5. The wavelengths are large. Let λ_1 and λ_2 be, respectively, the wavelength of the longitudinal and the shear disturbances in the matrix, a be the radius of the sphere, then λ_1 and $\lambda_2 \gg a$. In other words, $k_1 a$ and $k_2 a \ll 1$, where the wave number $k = 2\pi/\lambda$.

Moon and Mow examined the propagation of time-harmonic longitudinal waves in such a material. They showed the existence of two separate branches of wave propagation—the acoustical branch and the optical branch—separated by a cut-off frequency, n_c. Using a composite consisting of glass spheres of a constant radius in an epoxy matrix, Kinra and Anand[2] measured $\langle C_1 \rangle$ vs n in both the long-wavelength and short-wavelength regimes. Following Moon and Mow they conjectured that their data falls along these two branches. We note that a glass/epoxy composite violates the second assumption above. With $\omega = 2\pi n$ and $k = C_1/C_2$, the cut-off frequency is given by [6]

$$\omega_c^2 = \omega_0^2[1 + \rho'\bar{C}/\rho(1 - \bar{C})], \tag{1}$$

where

$$\omega_0^2 = 9\rho C_1^2/a^2\rho'(2k^2 + 1). \tag{2}$$

Here ω_0 is the resonant frequency of a single particle in an unbounded medium. If the wave speeds in eqns (1) and (2) are substituted for, in terms of elastic constants E and ν, it turns out that the normalized cut-off frequency $(k_1 a)_c$ is independent of the Young's modulus, and depends only on the Poisson's ratio, ν:

$$\Omega_c^2 = (k_1 a)_c^2 = \frac{9(1 - 2\nu)}{(5 - 6\nu)}\left[\frac{\rho}{\rho'} + \frac{\bar{C}}{1 - \bar{C}}\right]. \tag{3}$$

The differential equations governing the motion of the composite are eqns (12) of Ref. [6]; both equations are of order two and have constant coefficients. Analogous to the classical spring/mass/dashpot system, Moon and Mow identified an undamped natural frequency ω_0, given by eqn (2), and a damping coefficient c, given by

$$c = 9\rho(2k^3 + 1)/2\rho'\tau_0(2k^2 + 1)^2, \tag{4}$$

where τ_0 is a characteristic time of the problem, namely, the time required by the longitudinal wave to travel the radius of the sphere: $\tau_0 \equiv a/C_1$. Carrying the analogy a bit further, we define a dimensionless damping coefficient, ξ (see any elementary text on vibrations, e.g. [9]). It turns out that $\xi = c/\omega_0$, or

$$\xi = 1.5(\rho/\rho')^{1/2}(2k^3 + 1)/(2k^2 + 1)^{3/2} \tag{5}$$

Thus the composite behaves like an underdamped, critically damped or overdamped system, whenever $\xi < 1$, $\xi = 1$ or $\xi > 1$, respectively. Equation (5) was used in the design of the present experiments. Note that $\xi \sim (\rho/\rho')$; hence the choice of lead inclusions in any epoxy matrix; $\rho/\rho' = 0.106$. Thus $\xi = 0.31 < 1$ (underdamped oscillator), and one may expect to see a sharp resonance peak at n_c.

The purpose of the present work is as follows. Clearly, the theory of Moon and Mow is based on some rather restrictive assumptions. For example, the model assumes rigid inclusions. Lead is hardly "rigid" compared to epoxy. In view of the fact that its shear modulus is small compared to its bulk modulus, it would be more appropriate to treat lead like a fluid inclusion. Further, the theory completely ignores multiple scattering effects. In our experiments, however, the rescattering effects are expected to be quite strong for all but the lowest volume fraction of inclusions ($\bar{C} = 5\%$). Finally, the theory assumes $ka \ll 1$; the experiments cover all three regimes of interest, namely, ka small, comparable and large compared to one. It would seem, therefore, that their model would have very limited applications. We will show that this is not the case. In fact, their model accurately predicts n_c across the entire range of volume fractions; namely, from a dilute suspension ($\bar{C} = 5.4\%$) to a concentrated mixture ($\bar{C} = 34\%$). Thus their theory is shown to be good, well outside the range of its underlying assumptions.

3. EXPERIMENTAL PROCEDURES

The experiment was designed around the following heuristic argument. For a fixed specimen thickness and input amplitude, let A be the amplitude of the wave received through the specimen. Then, as $n \to n_c$, A should go through a sharp minimum.

An ultrasonic through-transmission direct-contact apparatus was used; it is schematically shown in Fig. 1. The Time Mark Generator/Pulse Generator/Function Generator/R.F. Amplifier combination was used to produce a toneburst (pulsed sine wave) of desired duration and center frequency. The electrical signal was applied to one of a pair of accurately matched piezoelectric transducers, and the specimens were acoust-

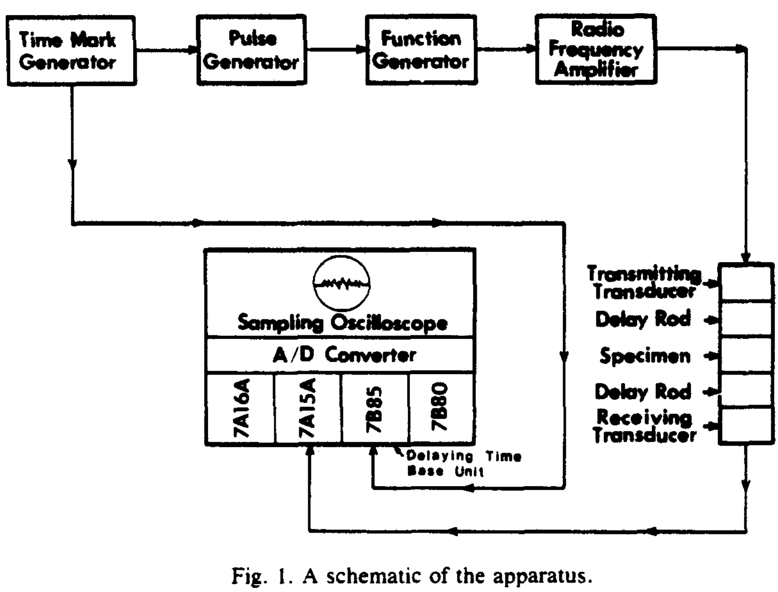


Fig. 1. A schematic of the apparatus.

ically coupled through polystyrene delay rods; a thin film of castor oil was used as a couplant. In order to minimize the random variations in the oil layer thickness, the entire assemblage was subjected to a constant axial load of about 100 N (20 lbf.). The received amplitude A was measured as a function of frequency n. Generally the toneburst was 10–20 cycles long, and the measurement was made with a reference peak near the center of the toneburst where the signal had reached steady state (i.e. away from the two ends where transients are present). This allows us to assume that consistent with the model, the composite had reached steady time-harmonic motion when the measurement was made. Since the transfer function of the measurement system (Fig. 1 without the specimens) is a smooth function of frequency, and since the objective here is to locate n_c, no attempt was made to deconvolve the received signal with respect to the transducer response. (In the early stages when this deconvolution was carried out, it was found to make no measurable difference in locating n_c).

Now n_c is an intrinsic property of the composite; it does not depend upon the overall dimensions of the composite. Therefore, in order to guard against fortuitous results, two specimens of significantly different thicknesses (W) were tested for each volume fraction ($\tilde{C}$). In each case essentially the same n_c was obtained for both thicknesses.

Specimen preparation

Lead spheres of 0.66-mm radius were suspended in an EPON-828Z epoxy matrix[10]. Now the theory assumes an elastic matrix, whereas, strictly speaking, epoxy is viscoelastic. Fortunately, over $0.3 \leq n \leq 5.0$ MHz, C_1 and C_2 were found to be frequency-independent and the attenuation, α, of the neat matrix was found to be small compared to $\langle\alpha\rangle$ for the composite; i.e. the scattering effects dominate the viscoelastic effects and it appears reasonable to assume that the matrix behaves in an elastic manner.

The diameter of the largest transducer used was 25.4 mm (1 in.). In order to eliminate the error due to the reflection of the scattered waves at the lateral boundaries, the cross-sectional dimensions of the composite were chosen to be 50.8 × 50.8 mm (2 × 2 in.). A mold with sixteen compartments was fabricated. A mixture of the resin and the hardener was thoroughly degassed in a vacuum chamber, and then poured into the mold. Subsequently, very accurately measured amounts of lead balls were sprinkled. Whenever clustering of the spheres occurred, they were dispersed by hand. Upon curing, each square was machined accurately to a thickness calculated by $d = a(4\pi/3\tilde{C})^{1/3}$. The desired number of the squares was randomly stacked, a thin layer of the matrix epoxy was applied to the mating surfaces, the assemblage was subjected to a suitable pressure, and the curing cycle was repeated. Finally, the two longitudinal faces were polished and lapped parallel to 25 μm. It is at this point that the final value was assigned to the volume fraction of inclusions, $\tilde{C}$, by accurately measuring the density of the specimen, $\langle\rho\rangle$:

$$\tilde{C} = (\langle\rho\rangle - \rho)/(\rho' - \rho). \tag{6}$$

Table 1. List of specimens

Specimen Number	$\tilde{C}$(%) Nominal	$\tilde{C}$(%) Measured	W (mm)
13	5	5.4	15.60
14	"	"	9.30
15A	15	15.9	7.62
16A	"	"	22.61
17	25	26.1	9.12
18	"	"	5.44
19	35	34.0	7.98
20	"	"	4.83
21	50	52.0	7.21
22	"	"	4.32

Table 2. Acoustic properties of the constituents

Material	C_1 (mm/μs)	C_2 (mm/μs)	ρ	ν	α (nepers/mm)
Epon 828Z	2.64	1.20	1.202	0.372	0.043 @ 1 MHz
Lead	2.21	0.86	11.3	0.411	0.026 @ 2 MHz

An important question is whether there is a rigid bond (no-slip condition) between the inclusions and the matrix as assumed in the theory. First, epoxy shrinks by about 5% as it cures; this virtually guarantees a compressive stress at the interface. Second, the individual squares (before they are assembled to form the composite) are sufficiently translucent, particularly at low $\bar{C}$, to permit a photoelastic examination. A "clover-leaf" type fringe pattern was observed around each sphere, indicating the presence of an interface stress; the first reason above was used to conclude that this stress is compressive: hence, the assumption of welded contact is justified.

A list of all the specimens used in this investigation is given in Table 1.

4. RESULTS

We first report the properties of the constituents. Several specimens with thickness ranging from 15 to 50 mm were cast from the next epoxy. C_1, C_2 and α were measured in the frequency range of $0.3 \leq n \leq 5.0$ MHz and are given in Table 2; C_1 and C_2 are accurate to ±0.3%. See [4] for a detailed description of the methods to measure C_1, C_2 and α, as well as for a systematic error analysis. For the case of lead, direct measurements cannot be carried out on the spherical particles. Three sheets of lead (5.08, 11.51 and 19.35 mm thick) made up of the same composition lead as was used to make the spheres, were obtained from the vendor[11] and tested.

The attention is now drawn to Fig. 2. Here, A_0 is the response of the measurement system, i.e. the amplitude at the receiver (in millivolts) with the specimen removed (see Fig. 1). Transducers with 0.5-MHz center-frequency were used in this measurement; this explains the peak in A_0 at 0.5 MHz. A_0 was measured at very small intervals of Δn = 0.01 MHz.

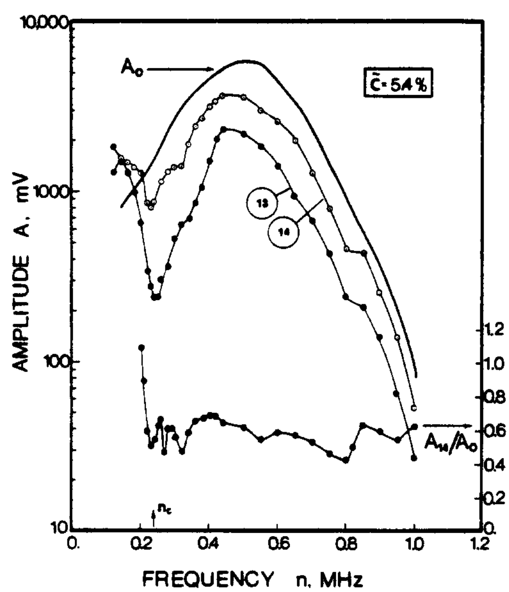


Fig. 2. The amplitude of the received signal exhibits a sharp minimum at the resonance frequency for specimens of two different thicknesses. Transducers: 0.5 MHz. A_0 is the frequency response of the measurement system. The deconvolved response, A_{14}/A_0, is also shown on a linear scale to the right.

Table 3. Measured and calculated values of n_c in MHz

$\bar{C}(\%)$	0.054	0.159	0.261	0.340	0.520
Calculated	0.237	0.318	0.397	0.462	0.639
Measured	0.24	0.30/0.36	0.40	0.47	0.60/0.65

The amplitude received with lead/epoxy specimens 13 and 14 is also shown in Fig. 2. For each specimen, all settings were kept the same and only the frequency was varied. Each frequency response curve was obtained in one continuous session. Furthermore, all settings were kept fixed in going from the first specimen (13) to the second (14) of the same volume fraction. The point of this precaution is that both A_{13} and A_{14} are equally affected by the *systematic* errors (subscripts refer to the specimen number). Both A_{13} and A_{14} were measured at extremely small frequency intervals of $\Delta n = 0.01$ MHz; only half the raw data could be plotted in Fig. 2. The minimum is distinctly clear in both curves. Note that a logarithmic scale has been used; on a linear scale, the minimum is considerably sharper. For specimen 13 ($W = 15.60$ mm), the same minimum voltage was observed at $n = 0.24$ and 0.25 MHz; for specimen 14 ($W = 9.30$ mm), the minimum was found at $n = 0.23$ MHz. Therefore, we claim an accuracy of ± 0.01 MHz in the measurement; i.e. $n_c = 0.24 \pm 0.01$ MHz. This is in excellent agreement with the calculated value of $n_c = 0.237$ MHz from (1); see also Table 3 and Fig. 7.

Properly, the specimen response should be deconvolved relative to (divided by) the measurement system response prior to extracting n_c. This has not been done; we briefly mention the reasons. A_{14}/A_0 is also plotted in Fig. 2 on a *linear* scale shown on the right. Near the calculated $n_c = 0.24$ MHz, there is one minimum as expected. However, there are two additional minima which have nothing to do with the physics of the problem, and owe their existence to the fact that we have divided one set of experimental data by another, and that both sets suffer from ubiquitous random errors. It is recognized that the deconvolution will shift the position of the minimum; however, an elementary calculation showed this shift to be negligibly small. We conclude that insofar as extracting n_c from the raw data is concerned, more precise (although very slightly less accurate) information can be obtained from the raw data than from the deconvolved data.

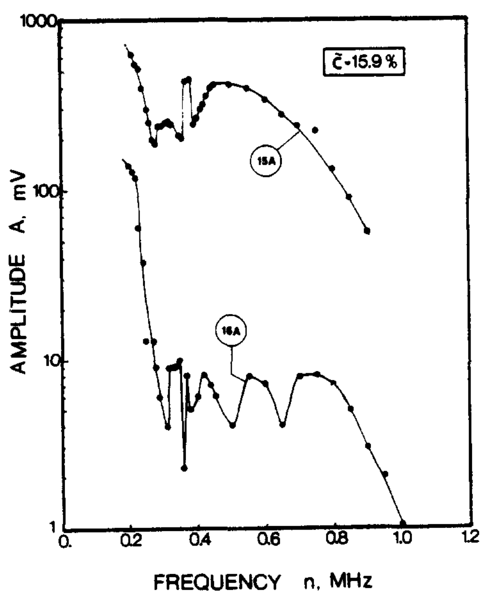


Fig. 3. Amplitude vs frequency for specimens of two different thicknesses. The signal in the range $0.29 \leq n \leq 0.36$ MHz suffered from harmonic distortion. Transducers: 0.5 MHz.

The results for specimens 15A and 16A are presented in Fig. 3 ($\bar{C}$ = 15.9%); the system response, A_0, is omitted. For reasons which are still not clear, this particular volume fraction presented some peculiar problems not encountered for $\bar{C}$ = 5.4%. Therefore the conclusions regarding n_c are not as unequivocal as was the case with 5.4% volume fraction, Fig. 2.

We first refer to the thicker specimen 16A (W = 22.61 mm). As n is increased from 0.2 MHz, A_{16} goes through a sharp and reproducible minimum at n = 0.31 MHz, which is in excellent agreement with the calculated value of n_c = 0.318; see Fig. 7 and Table 3. However, as n is further increased, the signal begins to suffer from harmonic distortion. In other words, even though the wave entering the composite was sinusoidal, the wave leaving it did not reach time-harmonic steady state during the period of observation, which is limited by spurious reflections from the boundaries of the apparatus. This objection applies to the data in the range $0.29 \le n \le 0.36$ MHz. Therefore, in this range, the amplitude cannot be trusted quantitatively. In particular, there is apparently a second sharp minimum at n = 0.36 MHz. There is a second problem associated with this part of data: the noise level was about 1 mV; the minimum at 0.36 MHz has an amplitude of 2.2 mV. Thus the signal-to-noise ratio is less than satisfactory.

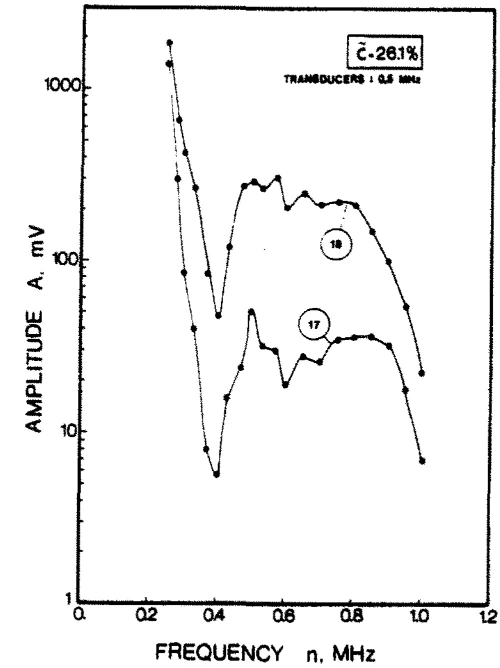
We now turn to the (thinner) specimen 15A. The first minimum occurs at 0.28 MHz (cf. calculated n_c = 0.318 MHz). Consistent with 16A, there is a second minimum at 0.36 MHz. The large jump in A (from 200 to 440 mV) in going from 0.36 to 0.37 MHz was considered very peculiar, and accordingly, checked several times.

Clearly, it is not possible to assign an unequivocal value to the measured n_c. With that proviso, the measured n_c is either 0.30 or 0.36 MHz.

The results for $\bar{C}$ = 26.1% are plotted in Figs. 4(a) and 4(b). Both 0.5- and 1.0-MHz transducers were used. A very sharp minimum was observed for both transducers and both thicknesses at n = 0.4 MHz; this is in excellent agreement with the calculated n_c = 0.397 MHz. We are unable to answer the following question. For the nominal, $\bar{C}$ = 5%, 25% and 35% (to follow), an unequivocal sharp resonance was observed. Why was a similar sharp resonance not observed for an intermediate $\bar{C}$ = 15%, Fig. 3?

The results for $\bar{C}$ = 34.0% are plotted in Figs. 5(a) and 5(b). Once again, data was collected with both 0.5- and 1.0-MHz transducers. Referring first to Fig. 5(a), a sharp minimum at 0.466 MHz was observed for both thicknesses. For specimen 19, the minimum amplitude was below 1 mV; i.e. comparable to the noise (hence not plotted). When we repeated the experiments with 1-MHz transducers, the minimum occurred at 0.5 MHz. Since n_c is very much closer to the natural frequency of the 0.5-MHz transducer than it is to that of the 1.0-MHz transducer, the measurements with the 0.5-MHz transducers are considered far more reliable; hence, n_c = 0.466 ≃ 0.47 MHz (calculated value = 0.462 MHz). If all four curves are examined, one finds a second minimum which does not occur at a fixed frequency. The average of the frequencies at which it occurs is, roughly, n = 0.7 MHz. A plausible explanation for this additional minimum will be offered later on.

Fig. 6 shows the results for the case of a concentrated suspension, $\bar{C}$ = 52%. At this high-volume fraction, most of the spheres are indirect contact with their neighbors. Therefore one would hardly expect a good comparison between the experiments and a dilute-suspension theory. Nevertheless, the experimental results are included for the sake of completeness. The theory predicts n_c = 0.639 MHz. For the thinner specimen 22, we notice a small, rather poorly defined, minimum at about 0.6 MHz (a well-defined minimum at 0.9 MHz is believed to be due to some other resonance). Unfortunately, corroborating results could not be obtained with the thicker specimen 21, because the received amplitude fell below 1 mV where it gets buried in the noise. Nevertheless, the first minimum (in the sense of increasing n) was noted at 0.65 MHz (the broken lines imply that its measured values were below 1 mV). Based on the data obtained with specimen 22, we assign the value of 0.6 MHz to n_c; however, it is quite questionable whether it indeed corresponds to the rigid body translation resonance under consideration.



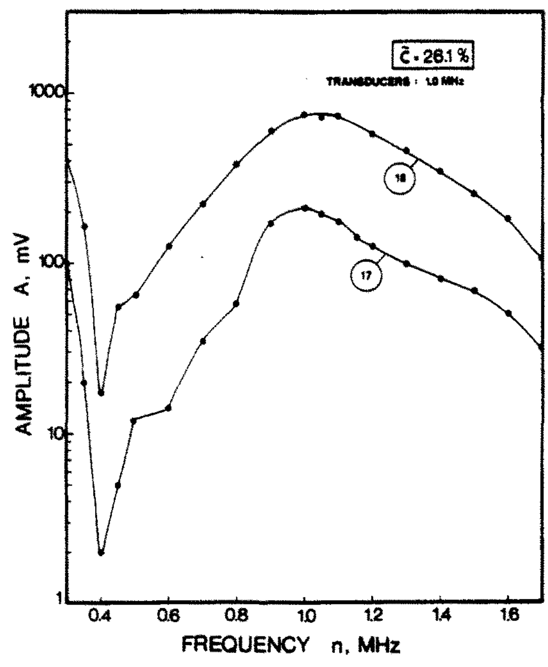


Fig. 4. (a) The amplitude exhibits a minimum at 0.4 MHz for specimens of two different thicknesses. Transducers: 0.5 MHz. (b) When 1.0-MHz transducers are used (instead of 0.5 MHz the minimum still occurs at 0.4 MHz. Therefore the measurement is independent of the transducer response.

In addition to the expected minimum associated with the resonance, other minima were observed in these experiments. A plausible explanation is offered next. By virtue of the assumption that the inclusion is rigid, the only motion accessible to the particle in the model of Moon and Mow is the rectilinear translation. Now, lead is hardly "rigid" compared to epoxy; see Table 2. For a compliant or an elastic inclusion, there are many other resonances possible. These have been studied explicitly by Flax and Uberall[12], who showed that there is a doubly-infinite series of resonances. Furthermore, these occur when $k_1 a \gtrsim 0.5$. In the frequency range $0.2 \le n \le 1.8$ MHz, we have $0.3 \le k_1 a \le 2.8$; i.e. the wave number is in the correct range for some of the lower resonance modes to get excited. This may very well be the explanation for the additional minima.

In order to bring out the dependence of the resonance frequency on the volume fraction, the theory and the experiment are compared in Fig. 7; the agreement is considered remarkably good. Here we have plotted $\Omega_c = (k_1 a)_c$. Note that $k_1 a = O(1)$ in

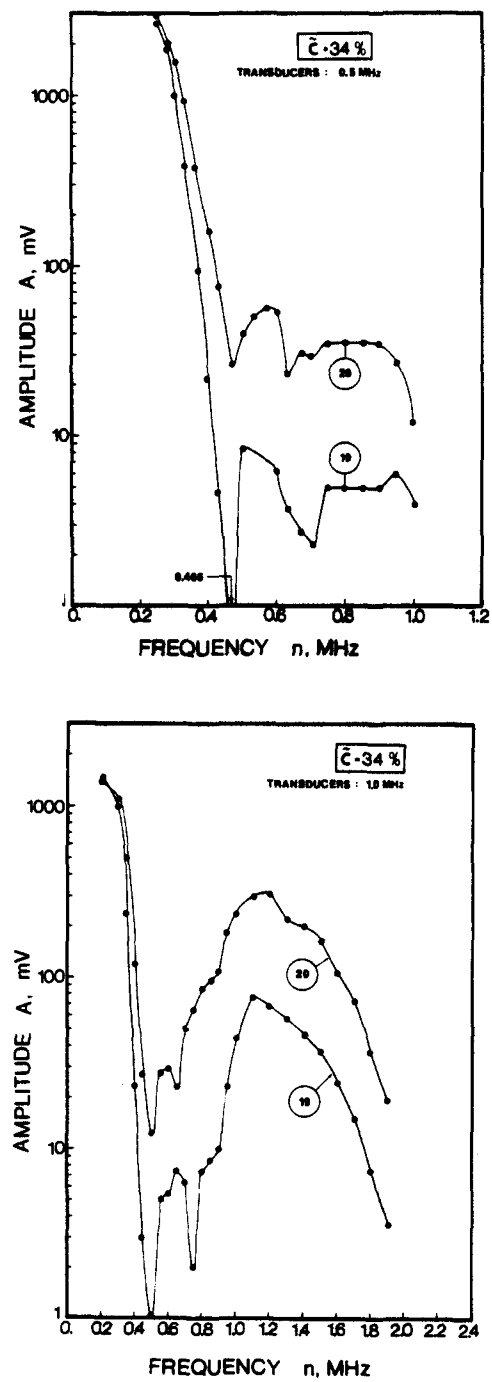


Fig. 5. (a) The minimum amplitude occurs at 0.466 MHz for both specimens. Transducers: 0.5 MHz. (b) When 1.0-MHz transducers are used (instead of 0.5 MHz), the minimum shifts to 0.5 MHz.

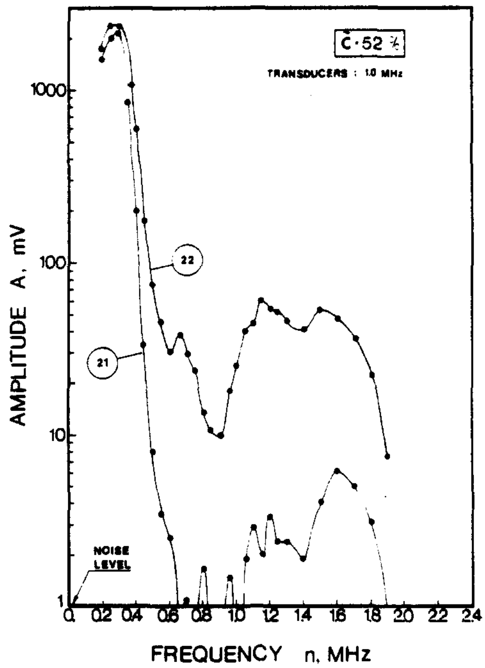


Fig. 6. Amplitude vs frequency for two specimens of different thickness.

our experiments, whereas the theory assumes a vanishingly small $k_1 a$. We are led to conclude that even though the theory of Moon and Mow is based upon a number of rather restrictive assumptions, it accurately predicts the resonance frequency. We note that lead is very much heavier than the matrix; on the other hand, lead is *not* very much stiffer than the matrix. Therefore from the viewpoint of the scattering phenomenon, it may well be that in the present instance, the inertial-mismatch effect dominates the elastic-mismatch effect; hence, the excellent comparison between the experiment and the heavy-inclusion theory.

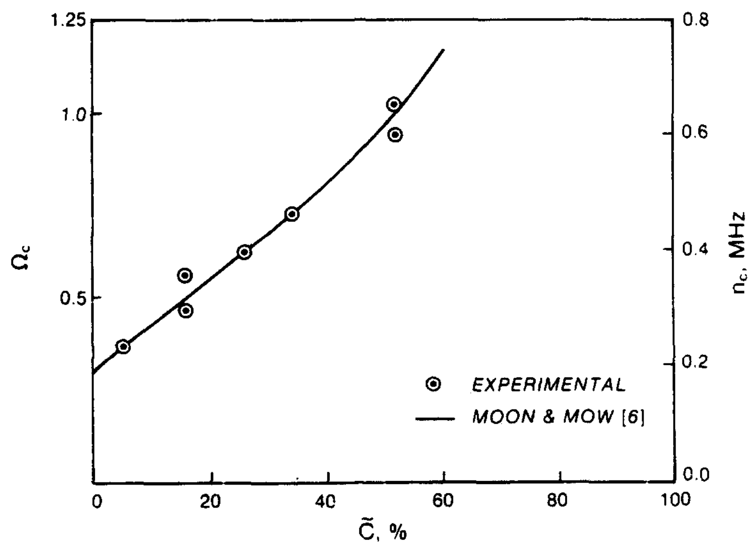


Fig. 7. The comparison of the theory[6] and the experiment was found to be excellent, even when $\tilde{C}$ is large and even though $k_1 a = O(1)$.

5. CONCLUSIONS

An approximate model for the longitudinal wave propagation behavior in a random particulate composite has been given by Moon and Mow. We have measured the resonance frequency for a lead/epoxy composite as a function of the volume fraction of inclusions. The model has been shown to predict the measured values quite accurately even when the following assumptions of the model are clearly violated: (1) The inclusions are rigid, (2) the wavelength is large compared to the size of the inclusion and (3) the volume fraction is small. Thus the model has been shown to be quite good, well outside the range of its underlying assumptions.

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