

Determination of particle size distributions from acoustic wave propagation measurements

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Abstract

The wave equations for the interior and exterior of the particles are ensemble averaged and combined with an analysis of Allegra and Hawley (J. Acoust. Soc. Am. **51**, 1545 (1972)) for the interaction of a single particle with the incident wave to determine the phase speed and attenuation at low volume fractions. The theory is shown to compare very well with the measured attenuation. The inverse problem, i.e. the problem of determining particle size distribution given the attenuation as a function of frequency, is examined using regularization techniques that have been successful for bubbly liquids. It is shown that, unlike the bubbly liquids, the inverse problem can be solved satisfactorily only in some specific cases that depend on the nature of particles and the size and frequency range.

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1 Introduction

Determining the particle size distribution of a solid-liquid mixture is of great practical interest. It has been suggested in the literature that this distribution may be determined by measuring the attenuation of a sound wave propagating through the mixture as a function of the frequency of the wave. The main premise is that the attenuation caused by a particle as a function of frequency depends on its size and therefore the attenuation measurements can be inverted to determine the particle size distribution — at least when the total volume fraction of the solids is small enough so that the particle interactions and detailed microstructure of the slurry play an insignificant role in determining the acoustic response of the slurry. Indeed, this general principle has been exploited successfully to determine the size distribution of bubbles in bubbly liquids^{1–3}. Commercial “particle sizers” based on acoustic response are in the process of being developed/marketed for characterizing solid-liquid mixtures⁴. The main objective of this paper is to investigate under what circumstances such a problem can be solved for solid-liquid systems. It will be shown that the success of the acoustic method for determining detailed particle size distribution is limited to very specific cases.

The problem of determining the acoustic response of a slurry given its particle size distribution is referred to as the forward problem. When the total volume fraction of the particles is small, the problem is relatively easy since then one only needs to understand the interaction between a single particle and the incident sound wave. This has been examined by a number of investigators in the past with notable contributions from Epstein and Carhart⁵ and Allegra and Hawley⁶ who considered suspensions of particles as well as drops. The latter investigators also reported experimental results verifying the theory for relatively small particles for which the acoustic wavelength is large compared with the particle radius. The theory developed by these investigators is quite general and accounts for attenuation by thermal, viscous, and scattering effects as described in more detail in Sections 2-3. The case of monodisperse non-dilute suspensions has been examined by Varadan *et al*⁷ who used an effective medium approximation to account for particle interactions. Their analysis was concerned only with the attenuation due to scattering. In section 2 we present the theory for the forward problem with the main aim of reviewing the important physical effects causing the attenuation. The derivation for the attenuation proceeds along different lines than that followed by Epstein and Carhart or Allegra and Halwey in the way the one particle analysis is used to predict the attenuation of the suspension. These investigators calculated the energy dissipation per one wavelength to estimate attenuation while we use the method of ensemble averages to determine both the phase speed and attenuation of waves. The method of ensemble averages will be more convenient for developing a suitable expression for attenuation in non-dilute suspensions, if desired, using either an appropriate effective-medium approximation or direct numerical simulations.

In Section 3 we present new experimental data for nearly monodisperse polystyrene particles whose radii are comparable to the wavelength and validate the theory described in Section 2 over a non-dimensional frequency range much broader than examined by previous investigators. We also

summarize in that section the different physical mechanisms that cause attenuation in suspensions. The attenuation as a function of frequency is shown to undergo several peaks owing to the resonances in shape oscillations in agreement with the theory prediction. It also gives some indication of the range of frequency and attenuation measurable with our acoustic device.

In Section 4 we consider the inverse problem, i.e., the problem of determining the particle size distribution given the total attenuation as a function of frequency and the physical properties of the particles and the suspending liquid. At small particle volume fractions, this amounts to solving a linear integral equation for the unknown size distribution. This is an ill-posed problem: small changes in the attenuation data can cause large changes in the size distribution. Thus, for example, several very different particle distributions could give rise to essentially the same attenuation-frequency curve. This, of course, is a rather well known difficulty in most inverse problems which involve solving a Fredholm integral equation of the first kind with a smooth kernel. Techniques have been developed to “regularize” the problem. We use the well known Tikhonov regularization technique⁸, which replaces the ill-posed original problem with another well-posed problem involving an integral equation of the second kind while minimizing the fluctuations in the particle size distribution. Minimizing of the fluctuations is rationalized on the grounds that in most practical situations the particle size distribution is smooth. This regularization technique has been shown to work very well for the inverse problem in bubbly liquids².

We apply the above technique to the suspensions of polystyrene and glass particles. We find that the technique works well for the polystyrene particles but not for all glass particles. We also find that for polystyrene particles the technique works only when the attenuation is given over an appropriate frequency range — a frequency range that is too narrow or too broad may give erroneous estimates of the distribution. An inverse technique based on linear programming also failed to produce the correct particle size distribution when not a large enough frequency range is covered by the supplied attenuation data. This suggests that the prospects for determining the detailed particle size distribution from acoustic measurements are somewhat limited. (In situations where more might be known about the nature of particle size distribution, e.g. unimodal with a Gaussian or log-normal distribution, one might be able to determine the size distribution through appropriate curve fitting as has been done, for example, by McClements and Coupland⁹ but this is not addressed here.)

The reasons for why the size distributions for some particle suspensions are not recovered by the inverse technique, while so far no limitations were reported for bubbles can be given in terms of differing resonance nature of these suspensions. In the case of bubbles in most typical applications, the resonance occurs at frequencies for which the wavelength is relatively large compared with the bubble radius. This resonance is due to volume oscillations; the shape dependent resonances are unimportant and, as a consequence, there is effectively one resonance frequency for each bubble size. Thus, the peaks in the attenuation-frequency curve give a reasonable indication of the bubble

sizes, already at wavelengths that are relatively large. The situation with the particles is different as their resonance behavior is governed by shape oscillations. These resonances occur only at high frequencies where the wavelength becomes comparable with the particle size. In order to have attenuation-frequency curves that show as much variation with particle size those of bubbles, one has to provide data up to high frequencies. The reason, then, why we find difficulty in solving the inverse problem for glass particles is that there are no significant resonance peaks in that case.

2 The forward problem

The wave equations for both the interior and exterior of particles have been derived by Epstein and Carhart⁶. They were interested in the attenuation of sound waves in fog and therefore their analysis was concerned with drops instead of particles. The stress tensor for a viscous fluid used by them for the interior of the drops was subsequently replaced by Allegra and Hawley⁵ by that of an elastic solid to determine the attenuation of sound waves in a solid-liquid suspension. In this section we shall ensemble average a resulting wave equation to obtain the effective wavenumber of the suspension at arbitrary volume fraction, the real and imaginary parts of which give the wave speed and attenuation. Thus, the attenuation is not calculated by means of an energy dissipation argument^{5,6}, but directly from averaging the relevant wave equation. The result contains certain coefficients that remain to be evaluated for a given microstructure. In the present study, since we are primarily concerned with determining the size distribution, we shall evaluate the coefficients in the limit of small volume fractions. In a separate study, where we shall present experimental results for non-dilute suspensions, we shall extend the theory to treat non-dilute suspensions.

2.1 Theory

Epstein and Carhart⁶ first linearized the conservation equations for mass, momentum, and energy. The pressure and internal energy were then eliminated by introducing the linearized equations of state to yield equations in terms of density, velocity, and temperature. Next, the time dependence of all quantities were expressed by the factor $\exp(-i\omega t)$ — which is henceforth suppressed — and the velocity was expressed as

$$\mathbf{v} = -\nabla\Phi + \nabla \times \mathbf{A},$$

with $\nabla \cdot \mathbf{A} = 0$. With this form of \mathbf{v} it is possible to eliminate the temperature and density from the governing equations to yield a fourth-order partial differential equation for Φ and a second order equation in \mathbf{A} . The former in turn can be split into two second order wave equations upon a substitution $\Phi = \phi_c + \phi_T$ to finally yield three wave equations:

$$(\nabla^2 + k_c^2)\phi_c = 0, \tag{1}$$

$$(\nabla^2 + k_T^2)\phi_T = 0, \tag{2}$$

$$(\nabla^2 + k_s^2)\mathbf{A} = \mathbf{0}. \quad (3)$$

The wavenumbers in the above equations are given by

$$k_c = \omega/c + \frac{\iota}{2} \left[\left(\frac{4}{3}\mu + \kappa \right) / \rho + (\gamma - 1)\sigma \right] \omega^2/c^3, \quad (4)$$

$$k_T = (1 + \iota)(\omega/2\sigma)^{1/2}, \quad (5)$$

$$k_s = (1 + \iota)(\omega\rho/2\mu)^{1/2}. \quad (6)$$

Here, c is the phase speed in pure liquid, ρ is the density, κ and μ are, respectively, the compressional and dynamic viscosity, $\gamma = C_p/C_v$ is the ratio of specific heats at constant pressure and volume, τ is the thermal conductivity, and $\sigma = \tau/\rho C_p$ is the thermal diffusivity.

Inside the particles similar equations hold with the dynamic and compressional viscosity replaced by $\tilde{\mu}/(-\iota\omega)$ and $(\tilde{\lambda} + 2\tilde{\mu}/3)/(-\iota\omega)$, where $\tilde{\mu}$ and $\tilde{\lambda}$ are the Lamé constants. Henceforth a tilde refers to the interior of particles.

Eqn. (1) and its counterpart inside the particles describe the sound wave propagation through the suspension. Note that the wavenumber has an imaginary part; sound waves in pure fluid are attenuated by viscous and thermal energy dissipation¹⁰; the term inside the square brackets in (4) is commonly referred to as the ‘diffusivity of sound’. The total attenuation coefficient in both liquid and in the solid particle will henceforth be treated as additional physical properties. The other two wave equations describe waves that arise from thermal conduction and finite viscosity: we note that the modulus of k_T is inversely proportional to the thermal penetration depth $\sqrt{\sigma/\omega}$ and that of k_s to the viscous penetration depth $\sqrt{\mu/\rho\omega}$. The thermal (ϕ_T) and shear (\mathbf{A}) waves have generally very high attenuation and are unimportant in acoustic applications.

We now proceed to ensemble average the wave equation (1) to find an expression for the effective wavenumber of a wave propagating through a solid-liquid suspension. Introducing an indicator function g defined to be unity inside the particles and 0 outside, the ensemble averaged value of ϕ_c is

$$\langle \phi_c \rangle = \langle g\tilde{\phi}_c + (1 - g)\phi_c \rangle.$$

To obtain a wave equation for $\langle \phi_c \rangle$ we first take the gradient of the above equation to yield

$$\nabla \langle \phi_c \rangle = \langle g\nabla\tilde{\phi}_c + (1 - g)\nabla\phi_c \rangle + \langle (\nabla g)(\tilde{\phi}_c - \phi_c) \rangle. \quad (7)$$

As argued by Sangani¹¹, upon assuming that the particles’ spatial distribution is homogeneous on a macroscale, the last term in (7), being a vector, can only depend on quantities such as $\nabla \langle \phi_c \rangle$ and $\nabla \nabla^2 \langle \phi_c \rangle$. Anticipating that $\langle \phi_c \rangle$ will satisfy a wave equation we express the last term on the right-hand side of the above equation in terms of $\nabla \langle \phi_c \rangle$, i.e., we write

$$\langle (\nabla g)(\tilde{\phi}_c - \phi_c) \rangle = \lambda_1 \nabla \langle \phi_c \rangle,$$

where λ_1 depends on the parameters such as the volume fraction, k_c , and \tilde{k}_c . The divergence of (7) is given by

$$\begin{aligned}\nabla^2\langle\phi_c\rangle &= \langle g\nabla^2\tilde{\phi}_c + (1-g)\nabla^2\phi_c \rangle + \langle (\nabla g) \cdot (\nabla\tilde{\phi}_c - \nabla\phi_c) \rangle + \lambda_1\nabla^2\langle\phi_c\rangle \\ &= -k_c^2\langle\phi_c\rangle - (\tilde{k}_c^2 - k_c^2)\langle g\tilde{\phi}_c \rangle + \langle (\nabla g) \cdot (\nabla\tilde{\phi}_c - \nabla\phi_c) \rangle + \lambda_1\nabla^2\langle\phi_c\rangle.\end{aligned}\quad (8)$$

Writing

$$\langle (\nabla g) \cdot (\nabla\tilde{\phi}_c - \nabla\phi_c) \rangle = \lambda_2\tilde{k}_c^2\langle\phi_c\rangle; \quad \langle g\tilde{\phi}_c \rangle = \lambda_3\langle\phi_c\rangle$$

we find that $\langle\phi_c\rangle$ satisfies a wave equation

$$\left(\nabla^2 + k_{\text{eff}}^2\right)\langle\phi_c\rangle = 0 \quad (9)$$

with the effective wavenumber given by

$$k_{\text{eff}}^2 = \frac{k^2 + \lambda_3\left(\tilde{k}_c^2 - k_c^2\right) - \lambda_2\tilde{k}_c^2}{1 - \lambda_1}. \quad (10)$$

The real part of the effective wavenumber is the frequency divided by the phase speed in the mixture and the imaginary part the attenuation.

Up to this point the analysis is rigorous and without any assumption. Applying the boundary conditions of continuity of temperature, flux, velocity, and traction at the surface of the particles, and solving the resulting boundary value problem numerically, it is possible, in principle, to determine the phase speed and attenuation at arbitrary volume fraction using the above formulation. Special simplifications occur when the wavelength is large compared with and the viscous and thermal depths are small compared with the particle radius for which numerical computations using the multipole expansions developed in recent years (see, e.g., Sangani, Zhang, and Prosperetti¹²) can be readily used for determining the attenuation at arbitrary volume fractions. Alternatively, one may use a suitable effective-medium approximation to account for the particle interactions in non-dilute suspensions using the above framework. We shall pursue this further in a separate study¹³ devoted to non-dilute suspensions where we shall also present experimental data for the same. Since our interest in the present study is in determining size distributions, it is necessary to consider only the simplest case of dilute suspensions.

In dilute suspensions the particle interactions can be neglected, and the coefficients λ_{1-3} can be evaluated from the solution for ϕ_c for a single particle given by Allegra and Hawley⁵. Accordingly, the conditionally averaged $\langle\phi_c\rangle(\mathbf{x}|\mathbf{x}_1)$ given a particle centered at \mathbf{x}_1 is given by

$$\langle\phi_c\rangle(\mathbf{x}|\mathbf{x}_1) = \exp(i\mathbf{k}_c \cdot \mathbf{x}) + \exp(i\mathbf{k}_c \cdot \mathbf{x}_1) \sum_{n=0}^{\infty} \iota^n (2n+1) A_n h_n(k_c r) P_n(\mu) \quad (11)$$

where $r = |\mathbf{x} - \mathbf{x}_1|$, $\mu = \cos\theta$, θ being the angle between $\mathbf{x} - \mathbf{x}_1$ and \mathbf{k}_c , h_n is the spherical Bessel function of the third kind (corresponding to an outgoing scattered wave) and P_n is the Legendre

polynomial of degree n . The first term on the right-hand side of the above expression is the unconditionally averaged $\langle \phi_c \rangle(\mathbf{x})$ whose amplitude is taken to be unity with no loss of generality.

Inside the particle centered at \mathbf{x}_1

$$\langle \tilde{\phi}_c \rangle(\mathbf{x}|\mathbf{x}_1) = \exp(i\mathbf{k} \cdot \mathbf{x}_1) \sum_{n=0}^{\infty} \iota^n (2n+1) \tilde{A}_n j_n(k_c r) P_n(\mu), \quad (12)$$

where j_n is the spherical Bessel function of the first kind. Similar expressions are written for the conditionally averaged ϕ_T and \mathbf{A} . This results in expressions with a set of six unknowns for each mode n . Application of the aforementioned boundary conditions of continuity of velocity, traction, temperature, and flux yield six equations in six unknowns for each n . There were some minor errors in the equations given in Epstein and Carhart⁶ and Allegra and Hawley⁵; the correct equations are given in Appendix. Although it is possible to solve for the unknowns analytically in certain limiting cases, it is best to solve them numerically since we are interested in covering a wide frequency range for inverse calculations.

We now return to the calculations of the coefficients λ_{1-3} . Upon using the identity

$$\nabla g = -\mathbf{n} \delta(\mathbf{x} - \mathbf{x}_i),$$

with \mathbf{x}_i being a point on solid-liquid interface and \mathbf{n} the unit normal vector at the point, λ_1 is given by

$$\lambda_1 \nabla \langle \phi_c \rangle(\mathbf{x}) = - \int_{|\mathbf{x}-\mathbf{x}_1|=a} \mathbf{n} \left[\langle \tilde{\phi}_c \rangle(\mathbf{x}|\mathbf{x}_1) - \langle \phi_c \rangle(\mathbf{x}|\mathbf{x}_1) \right] P(\mathbf{x}_1) dA_1. \quad (13)$$

Here, $P(\mathbf{x}_1)$ is the probability density for finding a particle in the vicinity of \mathbf{x}_1 . Similarly, we have for λ_2 and λ_3

$$\lambda_2 \langle \phi_c \rangle(\mathbf{x}) = - \int_{|\mathbf{x}-\mathbf{x}_1|=a} \mathbf{n} \cdot \nabla \left[\langle \tilde{\phi}_c \rangle(\mathbf{x}|\mathbf{x}_1) - \langle \phi_c \rangle(\mathbf{x}|\mathbf{x}_1) \right] P(\mathbf{x}_1) dA_1. \quad (14)$$

and

$$\lambda_3 \langle \phi_c \rangle(\mathbf{x}) = \int g(\mathbf{x}) \langle \tilde{\phi}_c \rangle(\mathbf{x}|\mathbf{x}_1) P(\mathbf{x}_1) dV_1. \quad (15)$$

The above integrals must be evaluated keeping in mind that the integration variable is \mathbf{x}_1 . Thus, for example, in (13) and (14) we must consider all particles whose surface pass through the point \mathbf{x} . To carry out integrals we use the identity

$$e^{i\mathbf{k}_c \cdot \mathbf{x}_1} = e^{i\mathbf{k}_c \cdot \mathbf{x}} e^{i r k_c \mu} = e^{i\mathbf{k}_c \cdot \mathbf{x}} \sum_{m=0}^{\infty} \iota^m (2m+1) j_m(k_c r) P_m(\mu) \quad (16)$$

and the orthogonality of the Legendre polynomials over spherical surfaces. The resulting expressions are

$$\lambda_1 = -\frac{3\phi}{z} \sum_{n=0}^{\infty} (-1)^n [(2n+2)j_{n+1}(z) - 2nj_{n-1}(z)] [\tilde{A}_n j_n(\tilde{z}) - j_n(z) - A_n h_n(z)], \quad (17)$$

$$\lambda_2 = -\frac{3\phi}{\tilde{z}} \sum_{n=0}^{\infty} (-1)^n (2n+1) j_n(z) \left[\tilde{A}_n j'_n(\tilde{z}) - \frac{z}{\tilde{z}} j'_n(z) - A_n \frac{z}{\tilde{z}} h'_n(z) \right], \quad (18)$$

$$\lambda_3 = \frac{3\phi}{z^2 - \tilde{z}^2} \sum_{n=0}^{\infty} (-1)^n (2n+1) \tilde{A}_n [\tilde{z} j_n(z) j'_n(\tilde{z}) - z j_n(\tilde{z}) j'_n(z)]. \quad (19)$$

Here, ϕ is the volume fraction of the solids, $z \equiv k_c a$ and $\tilde{z} \equiv \tilde{k}_c a$ are the non-dimensional wavenumbers, and primes denote derivatives. Expressions (17)-(19), together with the expression for the effective wavenumber (10) complete the description of a solid-liquid mixture at low volume fractions.

In the above we have derived expressions for the attenuation and wave speed by calculating the effective wavenumber directly. An alternative derivation of the attenuation coefficient is to calculate the energy dissipation per wavelength in the mixture and divide the result by the energy per wavelength. The result for the attenuation per unit length is then^{5,6}

$$\alpha = -\frac{3\phi}{2z^2 a} \sum_{n=0}^{\infty} (2n+1) \text{Re} A_n. \quad (20)$$

Although it is not obvious that the resulting expressions for the attenuation by the two methods are the same, both methods were found to give essentially the same numerical results for attenuation as a function of frequency in dilute suspensions.

The above analysis may be extended to account for the effect of finite volume fraction through a suitable effective-medium approximation. Sangani¹¹ showed that the first correction of $O(\phi^{3/2})$ to the dilute $O(\phi)$ approximation for bubbly liquids can be simply derived through an effective-medium approximation. This correction is most significant near the resonance frequency of bubbles, and to correctly capture the behavior near resonance it is important to replace the pure liquid wavenumber (k_c in the above analysis) by the effective wavenumber. Thus, in the present context, $z \equiv k_c a$ in Eqns (17)-(19) for λ_{1-3} , is replaced by $z_{\text{eff}} \equiv k_{\text{eff}} a$, while the wavenumber in pure liquid in the expression for k_{eff} , Eqn.(10), has to be retained. The latter expression is then iterated to obtain a converged solution for k_{eff} . The effective-medium approximations have been found to be quite useful in the related study of light scattering by suspensions (see, e.g., Ma, Varadan and Varadan¹⁴). For very high volume fractions the other physical properties of the so-called effective medium must also be modified. In a separate study¹³, where we shall report experimental data for dense slurries, we shall examine several different versions of effective-medium approximations in more detail.

Finally, the above analysis can be extended in a straightforward manner to account for the particle size distribution when the total volume fraction of the particles is small. Let us write the attenuation by particles of radius between a and $a + da$ as an attenuation density $\hat{\alpha}(\omega, a)$ times the volume fraction of those particles $\phi(a) da$; we shall refer to $\phi(a)$ as the volume fraction distribution. At low volume fractions these contributions can be ‘summed’ over all particle sizes to give for the total attenuation $\alpha_{\text{tot}}(\omega)$:

$$\alpha_{\text{tot}}(\omega) = \int_{a=0}^{\infty} \hat{\alpha}(\omega, a) \phi(a) da. \quad (21)$$

It is customary to express the particle size distribution in terms of its number density distribution $P(a)$. The volume fraction distribution is related to $P(a)$ by $\phi(a) = (4\pi a^3/3)P(a)$.

The effective-medium approach described earlier can also be readily extended to account for the particle size distribution. The coefficients λ_{1-3} are first determined as functions of a for an assumed value of the effective wavenumber and these are integrated after multiplying by $\phi(a)da$ to yield estimates for the average values of λ_{1-3} for the suspension. These are substituted in (10) to determine k_{eff} . If this estimate of k_{eff} is different from the the assumed value, then λ_{1-3} are estimated for the new value of k_{eff} , and the process is repeated until the assumed and estimated values of the effective wavenumbers agree with each other.

3 Discussion and comparison with experimental data

Figures 1-2 show the predictions for the attenuation and wave speed as a function of frequency f for 79 μm radius polystyrene particles at a volume fraction of 0.05. The frequency f in Hz is related to ω by $\omega = 2\pi f$. The physical properties used in computations are given in Table 1. We note that the wave speed only changes if the frequency becomes very large and that these changes coincide with strong changes in the attenuation as well. Hence we expect that the measurement of phase speed will not provide significantly new information over that obtained from the attenuation measurements alone. We shall henceforth focus on the results for attenuation.

The attenuation of sound waves in a suspension is different from that in pure liquid because of four effects. First, the attenuation of sound in pure solid is different from that in pure liquid, and hence simply the presence of the particles changes the attenuation from that of pure liquid. Second, changes in temperature are different in a solid than in a liquid, and this causes a heat flow through the surface of the particles. This heat flux is out of phase with the sound wave passage and this leads to attenuation referred to as the thermal attenuation. Third is the viscous energy dissipation caused due to the motion of boundary of the suspended particles with respect to the liquid. Finally, the fourth is the attenuation due to scattering.

Allegra and Hawley⁵ showed that when the particle size is much smaller than the wavelength and much greater than the thermal and viscous penetration depths $\sigma^{1/2}/\omega$ and $\nu^{1/2}/\omega$, the resulting viscous and thermal attenuations increase as $f^{1/2}$. On the other hand, when the penetration depths are much greater than the particles, both attenuation contributions increase as f^2 . This transition occurs at very low frequencies — about 2 Hz for 100 micron radius particles in water — and will not be considered here. Attenuation due to scattering becomes important when the non-dimensional wavenumber $z = k_c a$ becomes comparable to unity. For small but finite z the scattering losses increase as f^4 . Thus, one expects that the change in the attenuation behavior from $f^{1/2}$ at low frequencies to f^4 will provide an important indication of the particle size. These asymptotic ranges are indicated in Figure 1. We see that the transition to the f^4 behavior does not fully occur for

the particles considered here. As the frequency is increased particles undergo several resonances as described in more detail below, and this is responsible for the several peaks seen in Figure 1.

Figure 3 shows the contributions to the total attenuation from each P_n mode. The $n = 0$ mode corresponds to radial (volume) oscillations of the particles, $n = 1$ to the translational oscillations, $n = 2$ to the ellipsoidal P_2 -shape deformation oscillations, and so on. The density of polystyrene particles is essentially the same as that of water and hence their translational oscillations are very small. As a consequence, the viscous attenuation is small and the small frequency behavior is governed by the thermal attenuation of the $n = 0$ mode. At higher frequencies the $n = 0$ mode begins to increase first as f^4 due to scattering losses but the contribution from the $n = 2$ mode soon becomes important as it undergoes a resonance at about 3 MHz frequency. The $n = 3$ and $n = 1$ modes undergo resonances next, and so on. We see that the $n = 0$ mode undergoes a broad maximum around 9 MHz. Although not shown here it too undergoes a resonance with a sharp downward peak at about 21 MHz. Thus, we see that the attenuation varies with frequency in a rather complicated manner at high frequencies owing to various resonances. We should note here that the behavior of this kind for polystyrene particles has also been reported by other investigators in the past. For example, Anson and Chivers¹⁶ and Ma, Varadan and Varadan¹⁴, who restricted their analysis to scattering losses only, found essentially the same behavior, and in earlier investigations^{17,18} mainly focussing on the determination of waves reflected by immersed objects, high-amplitude reflected waves were found at certain resonance frequencies.

Figure 4 shows attenuation as a function of non-dimensional wavenumber $k_c a$ for particles of radii 50 and 79 microns. We see that the curves for these two radii are essentially the same indicating that, at least for polystyrene particles, the thermal or viscous effects have negligible influence on the resonance frequency. The first resonance corresponding to $n = 2$ appears at $k_c a \simeq 1.4$.

Allegra and Hawley⁵ tested (20) extensively against their experiments and found very good agreement. However, their particles were always smaller than $1\mu\text{m}$, so that the wavelength was always much greater than the particle size. No resonance behavior was observed in their experiments. Although the above-mentioned paper by Ma, Varadan and Varadan¹⁴ presents experimental data on light scattering in the small-wavelength regime, no data on attenuation of sound waves by particles were presented. To test how well the theory works for larger particle sizes, we carried out an experiment that will be described in detail (along with more experiments on concentrated slurries) elsewhere.¹³ In this experiment the attenuation of sound waves was measured in a frequency range of 1-10 MHz in a solid-liquid mixture of polystyrene particles with 79 ± 3 micron mean radius and 1.8 micron standard deviation at 0.05 volume fraction. Monochromatic tonebursts, at incremental frequencies, were transmitted by a transducer on one side of a small vessel in which the mixture was being stirred; a second transducer received the signal and sent it to a LeCroy 9310A digital oscilloscope. The amplitude of the signal for pure water was measured, as was that for the solid-liquid

mixture. The excess attenuation was determined by

$$\alpha = -\frac{1}{d} \log \left(\frac{V_{\text{mix}}}{V_{\text{H}_2\text{O}}} \right),$$

where d is the distance between the transducers and V_{mix} and $V_{\text{H}_2\text{O}}$ the voltage amplitudes of the received signals in the mixture and pure water, respectively. The distance between the transducers was 2" at low frequencies and 1" at higher frequencies; this was necessary because the attenuation at higher frequencies was too high to produce significant noise to signal ratio in the larger vessel.

Figure 5 shows the comparison between theory and experiment. At the two gaps in the frequency domain (where the theory predicts very high peaks) the attenuation became again so large that the signal to noise ratio vanished even in the smallest vessel. Good agreement is found between experiments and the theory except near resonance frequencies where small differences are seen. There are two possible reasons for these small differences. The first is concerned with the finite volume-fraction effect. To investigate this we have also plotted in Figure 5 a result from an effective-medium approach described in the previous section. The resulting attenuation changes, but in the wrong direction. The second reason is that the particles were not exactly monodispersed. Using the method described in the previous section, a log-normal particle size distribution was introduced with a mean radius of $77\mu\text{m}$ and $1.8\mu\text{m}$ standard deviation, which lies within the manufacturers' specifications. The result for the attenuation, the dash-dotted curve in Figure 5, shows close agreement with the data. Thus, we conclude that the agreement between the theory and experiment is excellent, and that the small observed differences are due to small polydispersity of the suspension.

The attenuation behavior displayed by polystyrene particles is not generic as can be seen from Figure 6 which shows the attenuation behavior for glass particles. Since the density of the glass particles is significantly different from that of water, the glass particles execute significant translational oscillations. As a consequence, the low frequency behavior is completely governed by the viscous effects and the $n = 1$ mode. Note that the small frequency attenuation is about two orders of magnitude greater for glass particles than for the polystyrene particles. Also we see a considerably different behavior at higher frequencies. The attenuation does not seem to peak at several frequencies. Rather, for each n we see broad "hills" separated by narrow "valleys." The total attenuation does not appear to go through several resonances. The difference in the behavior for the glass and polystyrene particles at these high frequencies seems to arise mainly from the different elastic properties of the two materials.

4 The inverse problem

We now consider the inverse problem: given the total attenuation α_{tot} as a function of f we wish to determine $\phi(a)$ using (21). The straightforward method of solving the integral equations, i.e.

discretizing the integral domain into a number of elements and converting the integral equation into a system of linear equations in unknowns $\phi(a_k)$ at a selected number of points a_k in the domain, cannot be used since the resulting equations will be ill-conditioned. Figure 7 illustrates the ill-posed nature of the problem. Figure 7a shows a volume fraction distribution $\phi(a)$ of polystyrene particles in water together with a distribution that has superimposed large-amplitude noise; Figure 7b shows their corresponding attenuation versus frequency graphs. Clearly, for these two different particle size distributions the differences in attenuation are not visible (in fact, the differences are of the order of 0.1%). In other words, one can superimpose on true particle distribution another highly oscillatory distribution without affecting the total attenuation significantly.

4.1 Method

Since the true particle distribution is expected to be smooth, we must only allow solutions that are reasonably smooth. There are several ways of accomplishing this. In the present study, we shall use primarily a regularization technique due to Tikhonov⁸ which was successfully used for bubbly liquids by Duraiswami²; an alternative method is presented at the end of this Section. Accordingly, we multiply (21) with $\hat{\alpha}(f, a)df$ and integrate over the frequency range to obtain a simpler integral equation in which the right-hand side is only a function of a :

$$\int_{f_{\min}}^{f_{\max}} \int_{a_{\min}}^{a_{\max}} \hat{\alpha}(f, a)\hat{\alpha}(f, a')\phi(a')da'df = b(a) \equiv \int_{f_{\min}}^{f_{\max}} \alpha_{tot}(f)\hat{\alpha}(f, a)df, \quad (22)$$

where (a_{\min}, a_{\max}) and (f_{\min}, f_{\max}) are radius and frequency ranges. The above integral equation is now regularized as explained below by adding a small term $\epsilon(\phi - l^2\phi'')$ (where primes denote derivatives) to its left-hand side. Thus, we obtain

$$\epsilon[\phi(a) - l^2\phi''(a)] + \int_{a_{\min}}^{a_{\max}} K(a, a')\phi(a')da' = b(a), \quad (23)$$

where l is a suitably defined lengthscale and $K(a, a')$ is a kernel defined by

$$K(a, a') = \int_{f_{\min}}^{f_{\max}} \hat{\alpha}(f, a)\hat{\alpha}(f, a')df. \quad (24)$$

Eqn (23) is an integro-differential equation and needs two boundary conditions. Usual practice is to take the derivative of $\phi(a)$ to be zero at the two end points:

$$\phi'(a_{\min}) = \phi'(a_{\max}) = 0 \quad (25)$$

Note that a_{\min} and a_{\max} are not known in general *a priori*. One expects ϕ to be zero also at the two end points. Thus, the range $(a_{\min} - a_{\max})$ must be determined by trial and error so that both ϕ and its derivatives are approximately zero at the extreme values of a .

Now it can be shown that the solution of (23) subject to the boundary conditions given by (25) minimizes

$$E + \epsilon \int_{a_{\min}}^{a_{\max}} \left[\{\phi(a)\}^2 + l^2 \{\phi'(a)\}^2 \right] da, \quad (26)$$

where E is the measure of error between the actual attenuation and the computed attenuation:

$$E = \int_{f_{\min}}^{f_{\max}} \left| \int_{a_{\min}}^{a_{\max}} \hat{\alpha}(f, a) \phi(a) da - \alpha_{tot}(f) \right|^2 df. \quad (27)$$

Since both E and the second term, i.e., the integral, in (26) are nonnegative, minimization of (26) ensures that the solution of (23) will be free from large oscillations in ϕ . In other words, highly oscillatory distributions such as the one shown in Figure 7a are rendered inadmissible when (23) is solved with finite, positive ϵ in place of the original integral equation (22). Thus, we have regularized the problem of determining ϕ .

If we choose a large ϵ then we decrease the oscillations in ϕ but increase the error in $\phi(a)$ since then the equation solved is significantly different from the original integral equation. Small ϵ , on the other hand, yields unrealistic $\phi(a)$ having large oscillations when the data $\alpha(f)$ are not exact. An optimum choice of ϵ then depends on the magnitude of uncertainty/error in the attenuation–frequency data. In the calculations we shall present here the exact $\alpha_{tot}(f)$ is first determined using the forward theory for a given $\phi(a)$ and a small random noise of about 1% magnitude is added to it before the inverse calculations are made. Thus, we have an estimate of the error in the data but in general this estimate may not be known reasonably accurately. To determine the optimum ϵ , we solve (23) for several different ϵ 's and plot E versus ϵ to find a minimum in E . This, however, may lead to distributions in which $\phi(a)$ may have unphysical negative values for some a . The constraint $\phi(a) \geq 0$ for all a is satisfied *a posteriori* by setting $\phi(a) = 0$ for all a 's for which the solution of (23) gave negative values of ϕ . The computed value of E for a given ϵ is then based on $\phi(a) \geq 0$.

The integro-differential equation (23) was solved as follows. After discretizing the domain ($a_{\min} - a_{\max}$) into $N - 1$ equal segments and the frequency domain into $M - 1$ logarithmically equal segments we first evaluate the kernel $K(a_i, a_j)$ for $i, j = 1, 2, \dots, N$ (cf. (24)) using a trapezoidal rule for the integration over the frequency range. As pointed out by Duraiswami², it is essential to calculate the integral over particle radius very accurately. We assumed that $\phi(a)$ varied in a piecewise linear manner in each segment and used a 12-point Gauss-Legendre quadrature to evaluate the integral in (23). A second-order central difference formula was used to evaluate $\phi''(a)$ at all points except the end points a_{\min} and a_{\max} . The boundary conditions $\phi'(a_{\min}) = 0$ and $\phi'(a_{\max}) = 0$ were approximated using, respectively, second-order forward and backward difference formulae. Application of (23) at all the discretization points together with the boundary conditions can be expressed with the above scheme as a system of linear equations:

$$\sum_{j=1}^N A_{ij} \phi_j = b_i, \quad i = 1, 2, \dots, N, \quad (28)$$

where $\phi_j = \phi(a_j)$ and $b_i = b(a_i)$. The above set of equations was normalized by dividing all the equations with the greatest element of the kernel $K(a_i, a_j)$, K_m for all i, j times the segment length $\Delta a = (a_{\max} - a_{\min})/(N - 1)$. This set of equations was subsequently solved using a standard IMSL subroutine for linear equations.

Once ϕ_j are determined for a selected value of ϵ , we satisfy the constraint $\phi_j > 0$ by setting, as mentioned earlier, $\phi_j = 0$ for all negative ϕ_j . The error E as given by (27) was subsequently evaluated using a trapezoidal rule for integration over the frequency range. The optimum value of ϵ was determined by stepping logarithmically through several values of ϵ and plotting E versus ϵ .

A typical result ($N = 30$, $M = 112$, $f_{\min} = 0.1\text{MHz}$, $f_{\max} = 17\text{MHz}$, $a_{\min} = 15\mu\text{m}$, and $a_{\max} = 35\mu\text{m}$) for the error E in the resulting attenuation as a function of ϵ is shown in Figure 8. Note that ϵ here is the actual ϵ divided by $K_m\Delta a$. We see a clearly defined optimum value of ϵ . Computations were also made with larger M to confirm that the resulting volume fraction distribution was not affected by the further refinement in the integration over the frequency range. A remark should also be made of the choice for the length l in (23). We may regard both ϵ and l as parameters to be chosen so as to minimize the error E . Taking $l = (a_{\max} - a_{\min})/n$ we computed E by varying both ϵ and n with n varied from 1 to N . The three-dimensional plot of E versus n and ϵ showed that E was much more sensitive to the choice of ϵ than was to n . In general, the results with n close to N were slightly better than with those near $n = 1$. Based on this observation we chose $n = 30$. For larger values of N ($N > 40$) we found, that choosing $n = N$ led to more oscillatory behavior for ϕ_j . This is to be expected since choosing larger n , and, hence smaller l , permits larger values of $\phi'(a)$.

4.2 Results and Discussion

We now present results for the volume fraction distribution obtained using the above technique. We used forward theory to generate attenuation data for an assumed volume fraction distribution. Small random noise can be added to the data thus generated to mimick possible errors arising in the attenuation measurement. This is satisfactory since we are primarily interested in assessing the procedure for solving the inverse problem. If the procedure gives erroneous results even for this case, it will certainly break down in practice.

The frequency range over which experiments can be carried out depends on the technique used to transmit and receive the sound waves. Transducers are available (and used in our laboratory) to cover at least the range 0.1-15MHz. We shall choose here the same range to investigate the success and limitations of the above technique to solve the inverse problem although we shall also consider one or two cases with a larger frequency range to enquire if better estimates of $\phi(a)$ could be achieved if the attenuation data at higher frequencies were to be made available.

We consider first particle sizes that are of the same order of magnitude as the wavelength somewhere in this frequency range, which is the case for particles of about 10-100 microns radius (for

larger particles observed behavior of the attenuation is shifted to lower frequencies). A particle size distribution that is often used is a log-normal distribution, which results in volume fraction distributions such as the smooth one shown in Figure 7a. We attempt therefore to recover that distribution from the corresponding attenuation data. As in the forward problem, we shall investigate polystyrene particles and glass particles in water, as the first are almost neutrally buoyant and deformable while the latter are very rigid and much heavier than water; the physical properties used in the present calculations are listed in Table 1.

We begin with the results for polystyrene particles with a narrow size distribution in the range of 20-30 μm . The particle size range for the inverse calculations is first taken to be much greater — 5-100 μm ; the frequency range was 0.1-17MHz. Figure 9 shows that the volume fraction distribution as evaluated from the inverse technique is in very good agreement with the input distribution. The result for the size distribution can be improved further by making the particle size range smaller; the new result is shown in the same figure.

In Figure 10 we consider a more complicated, bimodal size distribution in the range of 20-45 μm with peaks at about 25 and 38 μm . The frequency range included the first two resonance peaks of the attenuation. We see once again that the inverse procedure recovers this distribution very well.

A major difficulty in solving an ill-posed problem is that small errors in the input (attenuation) data cause large changes in the solution. Of course errors are present in the experimentally obtained attenuation data. The calculations presented so far were made with no added noise. To mimic the practical situation, we added random noise of 5% standard deviation to the input data; this is about the same as the order of magnitude of the errors present in the experimental results shown in Figure 5. The resulting volume fraction distribution, shown in Figure 11, does confirm that small fluctuations in the input data only cause small deviations in the output. When the calculations were repeated with a noise of 10% standard deviation, the computed particle size distribution was found to be considerably different from the input distribution, although the main features of the size distribution were preserved by the inverse computations.

The results discussed so far suggest that the inverse problem can be solved with reasonable success. We now illustrate some limitations. The inverse method gave erroneous particle size distributions for smaller particles when the same frequency range as the above (0.1-20 MHz) was used. Of course, in order that the size of the particles be determined there must be at least one transition — from the thermal attenuation dominated regime to the scattering dominated regime which occurs roughly speaking at $k_c a = O(1)$. If the particles are very small then this transition may not occur over a fixed frequency range. But, as we shall presently see, the results are very sensitive to the frequency range chosen for computations even when this transition is included in the range.

Figure 12 shows the effect of varying f_{max} on the computed distribution. As seen in the figure

the resonance in the shape oscillations of the particles leads to a change in the slope of the curve just before the first resonance. This transition occurs just beyond the point marked by a circle in Figure 12a. We see a marked improvement in the results when f_{\max} is chosen corresponding to a point marked plus in Figure 12a over those obtained with a point corresponding to the circle which does not include the second change in slope. At this point, the size distribution for the largest particles is accurately predicted, but not yet that for the smaller particles (that have at this point still not undergone a second transition). Also shown in Figure 12b are the results when f_{\max} is chosen to coincide with the end of first peak, the point marked square in Figure 12a. This is seen to yield very accurate results for the size distribution. A similar result is shown in Figure 13 for much smaller particles with a broad size distribution. The resonance peaks of the different particle sizes overlap when the distribution is broad, as seen in Figure 13b. Nevertheless the conclusion that a second change in slope should be included in the input attenuation data is confirmed. Finally, Figure 14 shows a result for the bimodal distribution shown also in Figure 10, but now after cutting off the attenuation (input) data right after the first peak. The second peak in the attenuation in this case was mainly due to the smaller particles. The result is that the first peak in the volume fraction distribution is not at all recovered.

Clearly there is a significant error in the obtained volume fraction for the unsuccessful cases discussed so far. This is compensated by an overprediction of the volume fraction density of large particles, which give a higher attenuation at large frequencies. Hence at large frequencies the attenuation is well predicted in these cases, but not so at low frequencies where scattering-scaling does not work. To avoid the dominance of attenuation data that have a large magnitude, one may divide the data by the frequency squared and then solve the inverse problem. However, only small improvements were found by using this device; actually, the inverse-problem result shown in Figure 14b was obtained in this way (when not doing that the result is worse, but qualitatively the same).

Some insight into why the choice of f_{\max} drastically affects the results may be gained from Figure 16 which shows the three-dimensional plots for the kernel $K(a_i, a_j)$ for the same values of f_{\max} as considered in Figure 12. We see that when $f_{\max} = 10.4\text{MHz}$, corresponding to the circle in Figure 12a, the kernel has a maximum for $a_i = a_j = a_{\max}$. The kernel for smaller particles is very small and as a consequence the inverse technique could determine the larger particle size volume fraction correctly but fails for smaller particles. In contrast to this the kernel for $f_{\max} = 17.1\text{MHz}$, corresponding to the end of first peak, shows significant variations for wide range of values of a_i and a_j , and this apparently leads to a much better inverse solution. Finally, the kernel for $f_{\max} = 30.4\text{MHz}$, corresponding to the end of the third resonance peak, show a less pronounced structure.

It is also instructive to examine the kernel and the results of inverse calculations for the problem of determining bubble-size distribution in bubbly liquids examined by Duraiswami². The inverse procedure works very well for bubbly liquids as can be seen from Figure 17a which shows the input

and computed bubble size distributions to be excellent agreement. The kernel for this case has smooth variations over a wide range of bubble radii as seen in Figure 17b. The attenuation as a function of frequency is shown in Figure 17c. The main reason for the success of the inverse technique for bubbly liquids seems to be that there is only one resonance frequency for bubbles of each size. As long as the frequency range is broad enough to cover the resonance frequency all the bubbles, it is possible to determine the size distribution.

Returning now to the polystyrene particles, we note that the frequency range for solving the inverse problem should contain at least one resonance peak or two transition points (or change in the slope in the attenuation-frequency curve). But one might suspect that including a large frequency range with many resonance peaks may cause a problem too as follows. The first three resonance peaks seen in the total attenuation may either correspond to the peaks for the P_2 -mode oscillations for two different size particles, or may correspond to P_2 , P_3 and P_4 modes of the same size particles (etc.). We therefore show results in Figure 15 where attenuation data have been used that include the first resonance peak, the first three peaks and a very large frequency range. We see that although all the results shown are reasonable there is a slight deterioration of the results when more than one resonance peak is included.

A next possible complicating factor is that the particle size distribution may be very wide so that it may so happen that the frequency at which the second transition for smaller particles occur may be greater than the frequency at which the second resonance of the larger particle occurs. Again, this proved to be no significant difficulty as is seen from Figure 18, where successful inverse problem solution for a very broad size distribution of polystyrene particles (5-160 μ m radius) is shown.

Another reason besides a limited frequency range why resonance peaks may not be present in the provided attenuation data is that the resonance behavior is different for different materials. Indeed, when now turning from polystyrene to glass particles, we recall that the main difference between the attenuation data for both particles is the absence of the peaks for glass particles (see Figure 6). In accordance with this, we have found that results for the inverse problem for glass particles are poor in general. Figure 19a shows an inverse problem result for glass particles that is still reasonable, but that shown for the larger particles in Figure 19b is really poor. The reason for this lies of course in the shape of the kernel $K(a_i, a_j)$, which for this latter case is shown in Figure 19c. Comparing with the corresponding kernels for polystyrene particles (Figure 16b) and for bubbles (Figure 17c), we see that this particular kernel is very flat, indicating that this problem is very hard to solve.

The results presented so far show that the success of Tikhonov regularization to solve the inverse problem is limited. Although we have discussed at length reasons why the method works well for bubbles but not for particles, it is possible that other techniques for solving the inverse problem may be more successful. For that reason we have attempted an alternative method that we shall

briefly describe here.

The alternative method of solving the inverse problem that we tried is to reformulate the inverse problem as a linear programming problem that can be solved using a standard optimization routine^{2,19}. The simplest scheme is to minimize the error

$$\int_{f_{\min}}^{f_{\max}} \left| \int_{a_{\min}}^{a_{\max}} \hat{\alpha}(f, a) \phi(a) da - \alpha(f) \right| df \quad (29)$$

instead of the square of this expression at each frequency. Constraints on the solution are used *a priori* in linear programming; here we use that $\phi(a) \geq 0$. Imposing an upper bound on the total volume fraction (maximum packing) can also be incorporated but is not essential. To reformulate this problem into a linear programming problem, we first discretize (29) and introduce new variables by

$$\sum_{j=1}^N A_{ij} \phi(a_j) - \alpha(f_i) = u_i - v_i, \quad u_i, v_i \geq 0, \quad (30)$$

with A_{ij} the discretized integral operator in (29). Now, minimizing the absolute value of (30) can be shown¹⁹ to be equivalent to minimizing

$$\sum_{i=1}^M u_i + v_i \quad (31)$$

with (30) as a constraint together with $u_i, v_i \geq 0$ ($i = 1, \dots, M$) and $a_i \geq 0$ ($i = 1, \dots, N$). Essential here is the notion that at the optimum $u_i v_i = 0$ at all i , which makes the solutions of the two minimization problems (29) and (31) identical.

Although using the above linear programming scheme is a completely different way to solve the inverse problem than Tikhonov regularization, the findings are very similar. We concluded above that Tikhonov regularization breaks down when the frequency range over which attenuation data are supplied is made too small. In Figure ** we present the linear programming results for the same problem as was used in Figure ** above and see no improvement (and, for that matter, no deterioration either). The other inverse problem that could not be solved by Tikhonov regularization was that for relatively large glass particles (****size). But again, that problem could not be solved by using linear programming either. Hence linear programming has confirmed the conclusion that the inverse problem becomes very difficult to solve in cases where there is no clear resonance behavior.

5 Conclusion

Solving the inverse problem successfully is thus restricted to non-rigid particles that cause peaks in the attenuation vs. frequency curve. In such cases, the attenuation data should include the range where the beginning of the first peak occurs (see Figure 6b) for all particle sizes that are present.

Even if the experimental data show this point, problems still occur when the size distribution is broad. Having attenuation data well in the acoustical scattering regime causes problems, too. The absence of the attenuation peaks for very rigid particles such as glass beads makes the inverse problem very difficult to solve. Hence we see that recovering volume fraction distributions for solids requires skill and would be difficult to automate.

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Appendix

In this Appendix we give the set of linear equations for unknowns that include the coefficients A_n required to calculate the attenuation from (10) and (17)-(19) or (20). These equations are derived from the boundary conditions on the surface of a test particle. In addition to the coefficients A_n, \tilde{A}_n of the solution of (1) outside and inside the particle, respectively, similar coefficients arise due to the solution of (2) and (3), denoted by B_n and C_n . (Note that (3) is an equation for the vector \mathbf{A} rather than a scalar velocity potential, but only the azimuthal component of \mathbf{A} is non-zero, hence only a scalar coefficient C_n .)

$$\begin{aligned} z_c j_n'(z_c) + A_n z_c h_n'(z_c) + B_n z_T h_n'(z_T) - C_n n(n+1) h_n(z_s) = \\ \tilde{A}_n \tilde{z}_c j_n'(\tilde{z}_c) + \tilde{B}_n \tilde{z}_T j_n'(\tilde{z}_T) - \tilde{C}_n n(n+1) j_n(\tilde{z}_s) \end{aligned} \quad (32)$$

$$\begin{aligned} j_n(z_c) + A_n h_n(z_c) + B_n h_n(z_T) - C_n (h_n(z_s) + z_s h_n'(z_s)) = \\ \tilde{A}_n j_n(\tilde{z}_c) + \tilde{B}_n j_n(\tilde{z}_T) - \tilde{C}_n (j_n(\tilde{z}_s) + \tilde{z}_s j_n'(\tilde{z}_s)) \end{aligned} \quad (33)$$

$$b_c [j_n(z_c) + A_n h_n(z_c)] + B_n b_T h_n(z_T) = \tilde{A}_n \tilde{b}_c j_n(\tilde{z}_c) + \tilde{B}_n \tilde{b}_T j_n(\tilde{z}_T) \quad (34)$$

$$\tau (z_c b_c [j_n'(z_c) + A_n h_n'(z_c)] + B_n b_T z_T h_n'(z_T)) = \tilde{\tau} (\tilde{A}_n \tilde{b}_c \tilde{z}_c j_n'(\tilde{z}_c) + \tilde{B}_n \tilde{b}_T \tilde{z}_T j_n'(\tilde{z}_T)) \quad (35)$$

$$\begin{aligned} (-i\omega\mu) \left(\left[(z_s^2 - 2z_c^2) j_n(z_c) - 2z_c^2 j_n''(z_c) \right] + A_n \left[(z_s^2 - 2z_c^2) h_n(z_c) - 2z_c^2 h_n''(z_c) \right] \right. \\ \left. + B_n \left[(z_s^2 - 2z_T^2) h_n(z_T) - 2z_T^2 h_n''(z_T) \right] + C_n 2n(n+1) [z_s h_n'(z_s) - h_n(z_s)] \right) \\ = \tilde{A}_n \left[(\omega^2 \tilde{\rho} a^2 - 2\tilde{\mu} \tilde{z}_c^2) j_n(\tilde{z}_c) - 2\tilde{\mu} \tilde{z}_c^2 j_n''(\tilde{z}_c) \right] + \tilde{B}_n \left[(\omega^2 \tilde{\rho} a^2 - 2\tilde{\mu} \tilde{z}_T^2) j_n(\tilde{z}_T) - 2\tilde{\mu} \tilde{z}_T^2 j_n''(\tilde{z}_T) \right] \\ + \tilde{C}_n 2\tilde{\mu} n(n+1) [\tilde{z}_s j_n'(\tilde{z}_s) - j_n(\tilde{z}_s)] \end{aligned} \quad (36)$$

$$\begin{aligned} (-i\omega\mu) (z_c j_n'(z_c) - j_n(z_c) + A_n [z_c h_n'(z_c) - h_n(z_c)] + B_n [z_T h_n'(z_T) - h_n(z_T)] \\ - (C_n/2) [z_s^2 h_n''(z_s) + (n^2 + n - 2) h_n(z_s)]) \\ = \tilde{\mu} \left(\tilde{A}_n [\tilde{z}_c j_n'(\tilde{z}_c) - j_n(\tilde{z}_c)] + \tilde{B}_n [\tilde{z}_T j_n'(\tilde{z}_T) - j_n(\tilde{z}_T)] - (\tilde{C}_n/2) [\tilde{z}_s^2 j_n''(\tilde{z}_s) + (n^2 + n - 2) j_n(\tilde{z}_s)] \right). \end{aligned} \quad (37)$$

Here, b_c and b_T are given by

$$b_c = \frac{(1-\gamma)\omega^2}{\beta c^2}, \quad b_T = -\frac{\gamma}{c_1^2 \beta} \left[\omega^2 - \left(\frac{c_1^2}{\gamma} - \frac{4i\omega\mu}{3\rho} \right) k_T^2 \right], \quad (38)$$

with β the thermal expansion coefficient and c_1 the liquid-equivalent of the speed of sound for spherical compressional waves in an elastic isotropic solid $\tilde{c}_1 = \sqrt{(\tilde{\lambda} + 2\tilde{\mu}/3)/\tilde{\rho}}$. The Lamé constant $\tilde{\lambda}$ is not really needed when the speed of sound (c) of longitudinal compressional waves is specified, as we can also write $c_1^2 = c^2(1 - 4\tilde{\mu}/(3\rho c^2))$. The above equations have also been given by Epstein and Carhart⁵ and Allegra and Hawley⁶. However, in both there are typographical errors: in Ref. 5, the last $j'_n(\tilde{z}_s)$ in (33) is erroneously replaced by $h'_n(\tilde{z}_s)$; in Ref. 6 the signs of both $(n^2 + n - 2)$ -terms is wrong, while the last $h_n(z_s)$ on the left-hand side of (37) has the argument \tilde{z} instead and the first z_s after C_n is replaced by \tilde{z}_s . The influence of the errors in Ref. 6 is significant.

Table

| | polystyrene | glass | water |
|--|----------------------|---------------------|----------------------|
| density (g/cm ³) | 1.055 | 2.3 | 1.0 |
| thermal conductivity (J/K · cm · sec) | $1.15 \cdot 10^{-3}$ | $9.6 \cdot 10^{-3}$ | $5.87 \cdot 10^{-3}$ |
| specific heat (J/g · K) | 1.19 | 0.836 | 4.19 |
| thermal expansion coefficient (1/K) | $2.04 \cdot 10^{-4}$ | $3.2 \cdot 10^{-6}$ | $2.04 \cdot 10^{-4}$ |
| attenuation coefficient per freq ² (sec ² /cm) | 10^{-15} | 10^{-15} | $2.5 \cdot 10^{-16}$ |
| sound speed (cm/sec) | $2.3 \cdot 10^5$ | $5.2 \cdot 10^5$ | $1.48 \cdot 10^5$ |
| shear viscosity (g/cm · sec ²) | - | - | $1.01 \cdot 10^{-2}$ |
| shear rigidity (g/cm · sec ²) | $1.27 \cdot 10^{10}$ | $2.8 \cdot 10^{11}$ | - |

Table 1. The values of the physical properties that are used in this paper. The properties of water and polystyrene were taken from Ref. 6; the properties of glass from various sources, most notably Ref. 15.

Figure captions

Figure 1.

Example of the dependence of attenuation on frequency f for a solid-liquid mixture. Monodispersed polystyrene particles (—) and glass particles (-.-) of $79\mu m$ radius in water.

Figure 2.

Example of the dependence of wave speed on frequency f for a solid-liquid mixture. Monodispersed polystyrene particles (—) and glass particles (-.-) of $79\mu m$ radius in water.

Figure 3.

Contributions from the first five modes n in Eqns (17)-(19) to the total attenuation (the imaginary part of k_{eff} , which is given by Eqn. (10)). Polystyrene particles in water. —, $n = 0$; - - -, $n = 1$; - · - ·, $n = 2$; · · · · ·, $n = 3$; —, $n = 4$.

Figure 4.

Attenuation divided by wavenumber as a function of the wavenumber times the particle radius in the scattering regime for monodispersed polystyrene particles (—, $a = 79\mu m$; - - -, $a = 50\mu m$).

Figure 5.

Comparison with experimental data for the attenuation as a function of frequency. Polystyrene particles of radius $a = 79\mu m$ and 0.05 volume fraction. ○, experiments; —, theory for monodispersed particles; -.-, theory for monodispersed particles with effective medium correction for finite volume fraction effects; - - -, theoretical result with a particle size distribution with a mean particle radius of $73.5\mu m$ and standard deviation of $1.7\mu m$ (this is within the range specified by the manufacturer).

Figure 6.

Attenuation by monodispersed glass particles (of $79\mu m$ radius) in water and the contributions from the first three modes n in Eqns (17)-(19) to the total attenuation (the imaginary part of k_{eff} , which is given by Eqn. (10)). —, $n = 0$; - - -, $n = 1$.

Figure 7.

Influence of fluctuations superimposed on the volume fraction distributions (a) on attenuation data (b). The distribution with fluctuations in (a) has been divided by 100. The two attenuation graphs are indistinguishable.

Figure 8.

Typical dependence of the error in the attenuation for the solved volume fraction distribution as a function of the regularization parameter ϵ . The (small) parameter ϵ should be chosen such that this error is minimized. The minimum was always found to be well-defined.

Figure 9.

Solving the inverse problem for polystyrene particles. The solid line is the volume fraction distribution used to generate attenuation data (shown in Figure 12a, with f_{\max} as indicated by a square); the dash-dotted line is the solution of the inverse problem when taking the particle radius range to be $1 - 100\mu m$ and using 50 ‘bins’ of particle sizes. The dashed line is the result after narrowing the radius range to $10 - 35\mu m$, using 30 ‘bins’ of particle sizes.

Figure 10.

Successful solution of the inverse problem for a bimodal distribution of glass particles, using 30 particle size bins. Solid line is the exact result, markers represent the inverse problem solution. The attenuation data used to solve the problem included two peaks.

Figure 11.

Solution of the inverse problem when random noise of 2% standard deviation is introduced in the attenuation (input) data. Solid line is the exact result; the broken line is the result when no noise is introduced (already shown in Figure 8); the dash-dotted curve the result after introduction of the noise. Glass particles in water.

Figure 12.

Influence of the size of the frequency range over which attenuation is specified on the solution of the inverse problem. Polystyrene particles. (a) Input-attenuation data and four different upper bounds on the frequency. (b) Results from the inverse problem from these different ranges, using the same marker type. The solid line is the exact result; \square , result when cutting off the frequency range just at the end of the first peak in the attenuation; $+$, result when cutting of the frequency range after the second change in slope of the attenuation; \circ , result when cutting off before the second change in slope.

Figure 13.

As Figure 12, but with a different size distribution.

Figure 14.

As in Figure 10, but after cutting off the frequency range over which the attenuation was given between the first and second (attenuation) peak.

Figure 15.

Too big a frequency range over which the attenuation is available for polystyrene particles also deteriorates the result: the solid line is the exact result; the dashed line the inverse problem result when using attenuation data of Figure 11a below the point marked by ‘□’; the dashed-dotted line represents the result when this end point is shifted to the the point marked by ‘◇’ and the dotted line is the result when this end point is shifted to 1000 MHz.

Figure 16.

The kernel $K(a_i, a_j)$ for polystyrene particles when using for f_{\max} the value indicated in Figure 12a by a ○ (a), + (b) and □ (c).

Figure 17.

Results for air bubbles in water. (a) Inverse problem result with a total volume fraction equal to 0.004, together with the attenuation as a function of frequency (b) and the kernel $K(a_i, a_j)$ (c).

Figure 18.

Kernel $K(a_i, a_j)$ for glass particles when using the frequency range 0.1-39.8 MHz (a) and 0.1-126 MHz (b).