Sound absorption by suspensions of nonspherical particles: Measurements compared with predictions using various particle sizing techniques

Simon D. Richards[a)]
Marine & Acoustics Centre, QinetiQ, Winfrith Technology Centre, Dorchester Dorset, DT2 8XJ, United Kingdom

Timothy G. Leighton and Niven R. Brown[b)]
Institute of Sound and Vibration Research, University of Southampton, Highfield, Southampton SO17 1BJ, United Kingdom

(Received 27 January 2003; revised 15 July 2003; accepted 28 July 2003)

Knowledge of the particle size distribution is required in order to predict ultrasonic absorption in polydisperse particulate suspensions. This paper shows that the method used to measure the particle size distribution can lead to important differences in the predicted absorption. A reverberation technique developed for measuring ultrasonic absorption by suspended particles is used to measure the absorption in suspensions of nonspherical particles. Two types of particulates are studied: (*i*) kaolin (china clay) particles which are platelike in form; and (*ii*) calcium carbonate particles which are more granular. Results are compared to theoretical predictions of visco-inertial absorption by suspensions of spherical particles. The particle size distributions, which are required for these predictions, are measured by laser diffraction, gravitational sedimentation and centrifugal sedimentation, all of which assume spherical particles. For a given sample, each sizing technique yields a different size distribution, leading to differences in the predicted absorption. The particle size distributions obtained by gravitational and centrifugal sedimentation are reinterpreted to yield a representative size distribution of oblate spheroids, and predictions for absorption by these spheroids are compared with the measurements. Good agreement between theory and measurement for the flat kaolin particles is obtained, demonstrating that these particles can be adequately represented by oblate spheroids.
[DOI: 10.1121/1.1610449]

PACS numbers: 43.30.Es, 43.35.Bf [DKW]

I. INTRODUCTION

The propagation of high frequency (tens to hundreds of kHz) sound in shallow, littoral seas is complicated by many phenomena, including variable bathymetry and sound speed profiles, seabed and sea surface interactions, tides, currents, turbulence, ambient noise and the effects of bubbles and suspended mineral particles throughout the water column.

The last of these, the effect of suspended mineral particles, has been shown[1,2] to be important for high frequency sonar performance in turbid environments. In order to account for this effect in sonar performance predictions it is necessary to calculate the absorption coefficient in particulate suspensions. A reverberation time technique has been developed for the laboratory measurement of absorption in dilute suspensions and measurements made with spherical glass particles have been shown[3] to be in good agreement with predictions which assume the particles to be homogenous spheres.

Natural particles are not generally spherical, and there have been a number of studies of attenuation by irregular particles. A coupled-phase theory[4] has been modified[5] to include the effects of particle shape using an effective radius approach. Another approach[6] treats the suspension as a fractal medium, with the Reynolds number as the fractal dimension, and this has been shown[7] to give good agreement with measured data for airborne suspensions. This approach is of limited use since it requires that the fractal dimension be determined by fitting the model to observations at a given frequency. In this paper Urick's[8] formula for visco-inertial absorption by spherical particles has been used, along with an extension[9] to that model which accounts for spheroidal particles.

In order to characterize absorption by natural marine particles measurements in representative suspensions are required. This paper addresses that issue by comparing the measured absorption in suspensions of nonspherical particles with predictions for both spheres and spheroids. Particular attention is paid to the interpretation of the measurements of particle size distribution, which are required as inputs to the theoretical predictions.

II. EXPERIMENT

A. Protocol

The experimental method used in this study employs measurements of reverberation time to infer the acoustic ab-

[a)]Electronic mail: sdrichards@qinetiq.com
[b)]Present address: Industrial Research Limited, PO Box 2225, Auckland 1015, New Zealand.

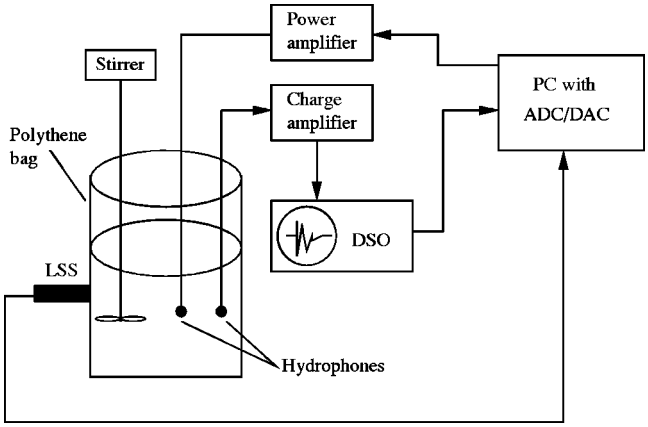

FIG. 1. A schematic of the experimental apparatus used to measure absorption in dilute suspensions over the frequency range 50 to 150 kHz. The stirrer is removed during acoustic measurements.

sorption in the system. The use of reverberation time measurements has been credited[10] to the work of Meyer and Skudrzyk. Relative measurements of the reverberation decay rate between clear water and water containing suspended particles are used to determine the absorption due to the particles. Initial experiments showed the importance of reducing boundary losses in the system.[3,11] This resulted in the development of the current system in which 0.016 m^3 of water is contained within a thin-walled plastic membrane, suspended in air by means of fine wires. This configuration approximates well to a pressure-release boundary condition surrounding the volume, thus minimizing the losses at the boundary. It may be noted that, even with the system optimized for minimum boundary losses in this way, the absorption due to a dilute suspension of particles is a small contribution to the total attenuation in the system. For example, a small increase (O(mm)) in the depth of the hydrophones was observed to produce a significant change in the reverberation time of the system. Experiments conducted with a third hydrophone[11] demonstrated that this was caused by additional attenuation due to the increased length of submerged hydrophone cable, thus illustrating the challenging nature of the measurements. The apparatus is shown schematically in Fig. 1, and Fig. 2 shows a photograph of the apparatus.

The signal generation, data acquisition and signal processing were all performed under the LabVIEW laboratory instrument management system, running on a personal computer (PC). The signal from the PC's digital-to-analog converter (DAC) board was fed to a power amplifier driving a Brüel and Kjær (B&K) 8103 hydrophone. A second B&K 8103 hydrophone was used for the receiving transducer and the signal from this was amplified and fed to a LeCroy digital storage oscilloscope (DSO). The DSO was connected to the PC via a GPIB interface, and the data were transferred to the computer for storage and post processing. A mechanical stirring device (propeller) was used to resuspend particles that had settled out of suspension. This was removed from the water when the acoustic measurements were performed, as additional absorbing/reflecting surfaces complicate the acoustic system. The temporal variation in the concentration of the suspension was monitored using a Sea Tech light scattering sensor (LSS). The LSS was mounted horizontally on the outside of the bag, with its window in close contact with the side of the bag. The mating faces of the LSS window and the side of the bag were wetted to ensure good optical coupling. This configuration ensured that any spatial integrating inherent in the LSS measurements would be in the horizontal direction, not in the vertical where the concentration would be expected to be spatially varying after a finite period of settling.

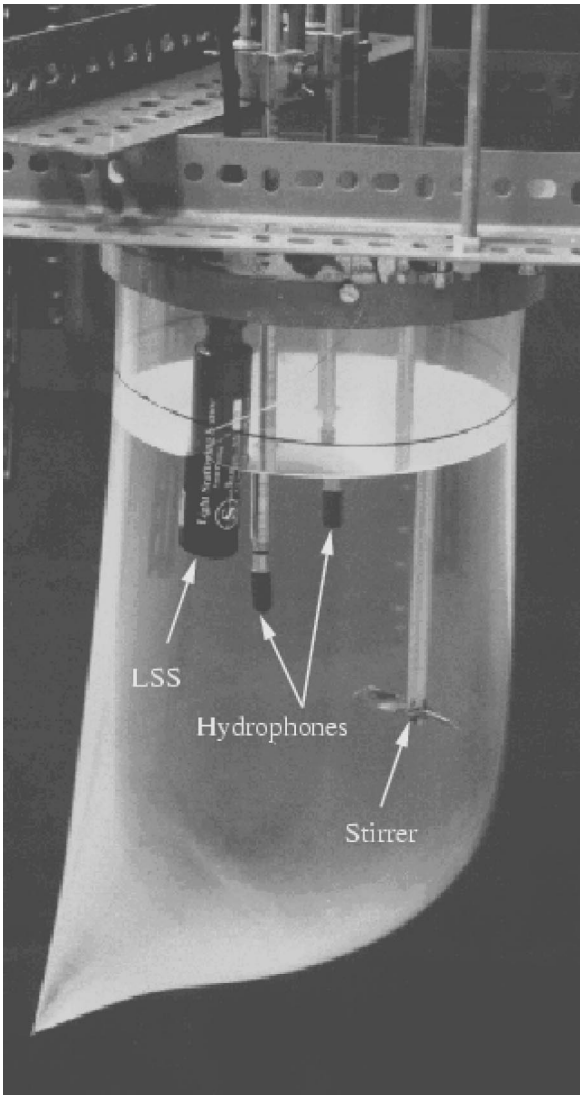

FIG. 2. Photograph of the experimental apparatus. The bag containing the test fluid is surrounded by air, resulting in a boundary condition which is close to pressure-release. This photograph is illustrative only, as the LSS and stirrer were removed during acoustic measurements. The majority of experiments were carried out with the LSS mounted horizontally on the outside of the bag (see text). Other instrumentation included temperature and dissolved oxygen probes (not shown). The hydrophones are mounted in rigid tubes to prevent movement.

The photograph (Fig. 2) shows the polythene bag suspended from its mechanical support by means of fine wires attached to a metal hoop. The items dipping into the water are, from left to right, the LSS used to monitor the concentration of the suspended particles (shown here mounted vertically, rather than in the more usual horizontal configuration as described above), the transmitting and receiving hydro-

phones and the propeller used to stir the water in order to suspend the particles. The diameter of the bag at the water surface is about 235 mm.

Prior to acoustic measurements the water was passed through a reverse osmosis system and then filtered to remove particulate matter. This ensured that no particles of diameter greater than 0.22 μm remained in the water. The water was then degassed under partial vacuum in order to avoid the presence of bubbles which may be acoustically significant. The temperature of the water was measured using a Jenway 3071 pH and temperature meter and this measurement was used to calculate the speed of sound in the ambient water. The level of dissolved oxygen was monitored throughout the experiments (between acoustic runs) using a Jenway 9010 dissolved oxygen probe attached to the 3071 meter. This was to ensure that the addition of the particles or the stirring process did not cause significant gas entrainment. The dissolved oxygen content was found to vary from 51% to 65% (expressed as a percentage of the saturation level) over the course of a measurement period, and no bubbles were observed.

In each series of experiments, reverberation time measurements were first made in particulate-free water in order to provide a reference measurement. Particles were then added in stages, in known quantities by mass, to enable a series of measurements to be made at varying concentrations. The concentration was taken to be the spatial average over the volume, i.e., the mass of particles added divided by the volume of water.

Before each series of acoustic measurements, the suspension was stirred by the mechanical stirrer until the particles were evenly distributed in the suspension. The time taken for this to occur was shown, by measurements made with the LSS, to be of the order of a few seconds. Care was taken to ensure that the particles that collect in the corners formed by the bottom seam of the bag were resuspended by the stirring. As the stirring takes place just before each set of acoustic measurements, the clear water reference measurements were also made on stirred water. This ensured that the reference signal used was obtained under conditions which most closely represented those experienced during the measurements made with the particles in suspension. It may be noted that stirring induces turbulence which can, in principle, lead to absorption. The mechanism for this absorption is the interaction between two phenomena: the perturbation of the turblence field by the acoustic wave, leading to anisotropic Reynolds stresses; and the redistribution of turbulent kinetic energy as it cascades from the large scale to the dissipation scale. As a result of these two effects the Reynolds stress is not in phase with the acoustic field and there is a net transfer of energy from the acoustic field to turbulent kinetic energy. This absorption mechanism has been shown[12] to be negligible for the current measurements.

The acoustic measurement itself involved first generating the sound field, then switching off the sound source and recording the decay of the reverberant sound field.

Two techniques for generating the sound field were investigated; an impulse and a long burst (20 ms) of uniform white noise. Both of these methods produce a broadband sound field. The advantage of using the long burst technique is that it gives sufficient time for a steady-state sound field to build up before the source is switched off, and the decaying sound field is also less prone to large perturbations resulting from direct reflections and particular modes of the volume. For these reasons the long burst generation method was used for the results presented in this paper.

Typically a series of measurements consisted of emitting ten noise bursts from the transmitting transducer and recording their responses via the receiver hydrophone, with the entire sequence lasting approximately 35 s. During this period there was no observable decrease in the concentration according to the estimates obtained using the LSS.

The decaying, reverberant sound field measured by the receiving hydrophone demonstrates fluctuations resulting from the interference between the many modes within the reverberating volume. These fluctuations limit the accuracy with which the decay rate of the reverberant field may be determined from the measured decay curve. In order to improve the accuracy of the estimate of the reverberation time of a reverberating volume, it is usual practice to repeat the measurement many times and average the decay rates obtained from the individual measurements. This method is, however, inefficient owing to the large number of measurements which must be made in order to obtain an accurate estimate of the decay rate or reverberation time. This averaging also obscures any ping-to-ping variations in the decay rate and any temporal variation within the integration time, which can be long due to the requirement for a large number of measurements.

The decay rates were therefore determined by applying the method of integrated impulse response[13] (IIR) to the sound field from the time that the driving signal was switched off. This method was used, even for signals derived from the noise burst, as it gives a smooth decay curve.

The raw data were filtered using second-order Butterworth bandpass filters in 10-kHz bands over the frequency range 50 to 150 kHz, and the IIR analysis was performed in each band. A second-order Butterworth filter has a roll-off of 12 dB per octave outside of the pass band, and it is therefore recognized that there will be some out of band data in each of the 10-kHz bands. The data were also reduced into time bins representing the rms value of the signal for a user-defined number of samples, typically 100. The sampling rate of the DSO was 500 kHz.

Errors in the determination of the decay rate in such an experiment can arise as a result of spatial variations in the sound field. There are two approaches to mitigating against this problem: (i) the measurements should be made at many points and the spatial average determined; or (ii) the measurements must be made under diffuse field conditions, in which the average energy density is the same throughout the entire volume and all directions of propagation are equally probable. The diffuse field approach was adopted for this study, and this is discussed later.

It will be noted that while these experiments are broadband, the theory referred to Sec. III is obtained for a single insonifying frequency. However, under the assumption that the individual frequency components are linearly indepen-

dent, the single-frequency models may be applied to the interpretation of the broadband measurements in each narrow band. The results of frequency-binned measurements made with spherical particles were found to be in very good agreement with single-frequency predictions,[3] thus validating the assumption that the frequency components may be considered as linearly independent.

B. Interpretation

In the experiments described in this paper the total acoustic loss in a volume of fluid containing suspended particles was inferred from measurements of the reverberation time of the system. The attenuation due to the suspended particles may thus be estimated by comparing the reverberation times of the system with and without the particles present. Making measurements relative to the clear water case in this way partially corrects for the effects of the boundary losses and other contributions to the total attenuation in the system, such as absorption by the hydrophone mounts and the hydrophones themselves. However, it should be noted that when the suspended particles' contribution to the total attenuation is small (i.e., at low concentration) the effect of the relatively large losses due to the boundary and the hydrophones will be to give large errors in the relative measurement of the influence of the suspended particles on the reverberation time.

The reverberation time of a reverberating volume is defined as the time taken for the sound pressure level to fall by 60 dB after the sound source is removed and may be given by[14]

$$t_{60}=\frac{55.26\ V_{\mathrm{enc}}}{c(S_{\mathrm{enc}}\bar{a}+8\zeta V_{\mathrm{enc}})}, \tag{1}$$

where V_{enc} is the volume of the enclosure, S_{enc} is the surface area, c is the compression wave speed, $\bar{a}$ is the average Sabine absorptivity and ζ is the absorption coefficient of the fluid in Nepers per meter. The average Sabine absorptivity is defined

$$\bar{a}=\frac{A_{\mathrm{enc}}}{S_{\mathrm{enc}}}, \tag{2}$$

where A_{enc} is the total sound absorption of the enclosure, expressed in units of m^2, such that $\bar{a}$ is dimensionless.

If t_{60} is the reverberation time of a volume of particulate-free water, then t'_{60} is the corresponding reverberation time in water containing suspended particles, differing only from the definition given in Eq. (1) in that ζ is replaced by ζ'. It is then clear from Eq. (1), and the relation $\alpha=20\log(e)\zeta$, that the difference in the attenuation coefficients of the two fluids, expressed in dB m^{-1}, is given by

$$\Delta\alpha=20\log(\mathrm{e})\frac{55.26}{8c}\left(\frac{1}{t'_{60}}-\frac{1}{t_{60}}\right)=\frac{60}{c}\left(\frac{1}{t'_{60}}-\frac{1}{t_{60}}\right), \tag{3}$$

provided that the addition of the particles does not significantly affect (i) the sound speed in the medium; (ii) the volume of fluid; or (iii) the absorption at the boundaries. It should be noted that while $\Delta\alpha$ is defined in terms of the reverberation times t_{60} and t'_{60}, in practice it is determined from the rate of decay of the sound field rather than from a direct measurement of t_{60}.

The change in sound speed resulting from the introduction of the particles at concentrations used in the experiments is sufficiently small[12] (less than a few tens of $\mathrm{cm\,s}^{-1}$ even at the highest concentrations) not to invalidate Eq. (3). Similarly, the volume change due to the addition of the particles at the concentrations used in these experiments is sufficiently small (volume fractions are typically much smaller than 10^{-3}), and it may be assumed that the particles in suspension have little effect on the absorption properties of the walls. Particles that settle to the bottom of the volume may, however, affect the boundary losses, but attempts were made to make measurements with the great majority of the particles in suspension.

As discussed above, the reverberation time should be determined from the decay of a diffuse sound field. The onset of a diffuse sound field in an enclosure can be described by the Schroeder cut-off frequency, which gives an indication of the lowest frequency at which the modal density, i.e., the number of modes per unit bandwidth, is sufficient to constitute a diffuse field. The Schroeder cut-off frequency, f_s, may be written[15]

$$f_{\mathrm{s}}=\left(\frac{c^3}{4\ln 10}\right)^{1/2}\left(\frac{t_{60}}{V_{\mathrm{enc}}}\right)^{1/2}. \tag{4}$$

Ideally then, all experiments should be carried out at frequencies well above the Schroeder cut-off in order to ensure that the measurements are not influenced by modal structure in the sound field. For the experimental system described in this paper the Schroeder cut-off frequency was typically around 50 to 75 kHz. To test the assumption that the field is diffuse, measurements of reverberation time were made with the hydrophones in different positions.[11] It was found that the change in reverberation time occurring when the hydrophones were moved vertically was dominated by the change in absorption owing to the difference in length of submerged hydrophone cables. It was therefore not possible to investigate experimentally the variation in reverberation time in the vertical direction. However, measurements made in a number of hydrophone positions in the horizontal plane, i.e., with the hydrophones at a constant depth such that the amount of submerged cable is unchanging, showed a 4% standard deviation in the reverberation time. This is the dominant source of experimental error in the system and is taken into account in the error analysis.

C. Particle concentration and size distributions

The attenuation depends on both the absolute concentration of particles in suspension, and the size distribution of the particles. As described previously, the concentration is determined by carefully weighing the dry particles before they are added to the volume and ensuring that the acoustic measurements are made with all of the particles in suspension.

Because the particles settle out of suspension over time, the LSS was used to monitor the concentration to ensure that

it did not change significantly over the time taken for a series of acoustic measurements to be carried out.

The LSS only provides a relative measurement of the total concentration of suspended particles in the measurement volume, and does not provide any information on the sizes of the particles present. Since the particle size is an important parameter influencing the acoustic absorption by suspended particles, it is important to know the size distribution of the particles in suspension. While this is possible with spherical particles, it is not a simple matter where nonspherical particles are used. This is discussed in the following sections.

The particle size distribution was determined using three standard techniques: (*i*) laser diffraction analysis; (*ii*) gravitational sedimentation; and (*iii*) centrifugal sedimentation. These measurements were carried out independently and it was not possible to measure the particle size distribution dynamically during the acoustic measurements.

1. Light scattering sensor

It is important here to note the limitations of the LSS device for quantitative measurements of suspended particle concentration. The LSS device emits light in the forward direction and detects the light which is backscattered from the medium. While the intensity of the backscattered light depends upon the concentration of particles in suspension, it also depends upon the properties of the particles, such as their size relative to the wavelength of the laser light, shape and complex refractive index. The LSS may be calibrated using a known concentration of the suspension of interest. However, in cases where the suspension contains particles with a range of sizes, larger particles will settle out of suspension more quickly. The size distribution will therefore be time-varying, thus invalidating the calibration. It should be noted that the LSS was not used to obtain quantitative measurements of particle concentration for normalization of the attenuation measurements. This was done by weight, as previously described.

2. Laser diffraction analysis

Laser diffraction analysis uses the diffraction pattern of laser light scattered by a sample of particulate in suspension to infer the particle size distribution. The instrument used in this study measured particle diameters in the range 0.4 to 1000 μm.

It is instructive to describe briefly the laser diffraction method of particle sizing, which exploits the fact that small particles in a laser beam scatter light in a characteristic pattern, i.e., the diffraction pattern. The details of the diffraction pattern depend upon the distribution of particle sizes contributing to the light scattering. Information about the particle size distribution can thus be inferred from the details of the light flux pattern.

The simplest diffraction pattern due to a collection of spheres is that from a monomodal dispersion. It is the familiar Airy pattern consisting of a central bright spot surrounded by concentric light and dark rings, the intensity of which diminish further from the center of the diffraction pattern, that is to say at higher scattering angles. The scattering angles at which the diffraction maxima and minima occur depend on the size of the particles relative to the laser wavelength, with smaller particles leading to higher scattering angles. The particle size in such a monomodal dispersion of spheres can therefore be simply inferred from the scattering angles.

These diffraction patterns obey the principle of linear superposition, meaning that the total scattering pattern for a mixture of two or more monomodal dispersions is obtained by adding the intensities of the scattering patterns from each constituent monomodal dispersion. This allows the possibility of inferring the particle size distribution from sufficiently accurate measurements of the scattering pattern due to a multimodal dispersion of spherical particles.

The interpretation of laser diffraction measurements of particle size distribution becomes complicated when nonspherical particles are involved. The standard analysis yields the size distribution of spherical particles which would give the observed diffraction pattern. In principle it would be possible to infer the size distribution of particles of a different shape by inverting a model for the diffraction pattern obtained from forward scattering by such particles. However, for highly irregular particles and suspensions containing many different particles, this becomes impractical. A more fundamental question is what exactly we mean by the size when discussing nonspherical particles. In general nonspherical particles cannot be described by a single number, such as the diameter of a sphere. The laser diffraction technique may be considered to yield an effective spherical diameter for optical scattering. Other particle sizing techniques are available which also yield effective spherical diameters for nonspherical particles. In order to compare with the measurements made by laser diffraction, measurements have also been made using two alternative techniques: (*i*) gravitational sedimentation and (*ii*) centrifugal sedimentation.

3. Gravitational sedimentation

The gravitational sedimentation technique is based on the measurement of the steady-state settling velocity of the particles as they fall through a fluid under the influence of gravity. This method yields the Stokes diameter of the particle, defined as the diameter of a sphere which has the same density and the same free falling velocity in a given fluid as the particle, within the viscous flow regime. From Stokes' equation the Stokes diameter may be written

$$d_{\mathrm{st}}=\left(\frac{18\eta h_{\mathrm{s}}}{(\rho'-\rho)gt}\right)^{1/2}, \tag{5}$$

where η is the molecular viscosity of the suspending fluid, h_{s} is the distance the particle falls in time t, ρ' and ρ are the densities of the particle and fluid, and g is the acceleration due to gravity. Typically, in gravitational sedimentation measurements, the time taken for the particle to reach its terminal velocity is negligible.[16] The free fall velocity is therefore taken to be $v_{\mathrm{s}}=h_{\mathrm{s}}/t$.

Stokes' equation is only valid in the region of viscous flow, which sets an upper limit on the particle size which may be determined by this technique. This limit is determined by the Reynolds number

$$\mathrm{Re}=\frac{\rho v_s d_{st}}{\eta}. \tag{6}$$

The Reynolds number should not exceed 0.25 if the error in the Stokes diameter is not to exceed 3%.[16] Setting $\mathrm{Re}=0.25$ and equating Eq. (5) and Eq. (6) yields the following expression for the limiting Stokes diameter:

$$d_{st}|_{\max}=\left(\frac{4.5\eta^2}{(\rho'-\rho)\rho g}\right)^{1/3}. \tag{7}$$

For example, for silica ($\rho'=2650\ \mathrm{kg\,m^{-3}}$) settling in water ($\rho=1000\ \mathrm{kg\,m^{-3}}$; $\eta=0.001$ Pa s), the limiting Stokes diameter is 65.3 μm.

The lower size limit is determined partly by the long settling times experienced by small particles, and partly by other motions which may be significant compared to the small settling velocities, such as Brownian motion, diffusion and convection currents which may be set up over long integration periods. For these reasons the use of gravitational sedimentation is not usually recommended for particles smaller than about 1 μm.

4. Centrifugal sedimentation

Some of the difficulties associated with the use of gravitational sedimentation for fine particles may be reduced by speeding up the settling time. This can be achieved through the use of centrifugal sedimentation techniques.

As in the case of gravitational sedimentation the Stokes diameter is determined from Stokes' law, but now the acceleration due to gravity is replaced by the centrifugal acceleration and the free fall velocity is replaced by the radial settling velocity to give

$$d_{st}=\left(\frac{18\eta}{(\rho'-\rho)\omega_c^2 r_c}\frac{\mathrm{d}r_c}{\mathrm{d}t}\right)^{1/2}, \tag{8}$$

where r_c is the radial distance of the particle from the axis of the centrifuge, $\mathrm{d}r_c/\mathrm{d}t$ is the radial settling velocity and ω_c is the rotation speed of the centrifuge in radian $\mathrm{s^{-1}}$.

Centrifugal sedimentation is usually used for particles up to a few microns in diameter, although this can easily be extended using more viscous suspending fluids and longer settling times. The lower limit on particle size is determined by the consideration that the radial displacement of the particles by Brownian motion during sedimentation should be much smaller than the displacement due to centrifugal motion. For a typical example[17] the minimum Stokes diameter is less than 0.01 μm.

For the measurements of the size distributions of clay-like particles presented in this paper, all samples were dispersed in the suspension media and insonified with ultrasound (normally for one minute) to assist in dispersal and breaking up of agglomerates. Additionally, the sample portions used for the centrifugal sedimentation were dispersed using more prolonged ultrasonics in an attempt to further disseminate the aggregates and obtain a particle size distribution more representative of the discrete particles. This additional deflocculation was only used for the centrifugal sedimentation samples, in order to exploit the ability of that technique to characterize smaller particles.

III. THEORY

Urick[8] obtained the following expression for the visco-inertial attenuation coefficient by employing the expression for the viscous drag on an oscillating sphere developed by Stokes:[18]

$$\alpha_v=20\log(\mathrm{e})\frac{\phi k(\sigma-1)^2}{2}\left[\frac{s}{s^2+(\sigma+\tau)^2}\right]\quad \mathrm{dBm^{-1}}, \tag{9}$$

where

$$\tau=\frac{1}{2}+\frac{9}{4}\left(\frac{\delta_v}{a}\right) \tag{10}$$

and

$$s=\frac{9}{4}\left(\frac{\delta_v}{a}\right)\left(1+\frac{\delta_v}{a}\right). \tag{11}$$

Here ϕ is the volume fraction of suspended particles, k is the acoustic wavenumber, σ is the ratio of the densities of the solid and fluid phases, a is the particle radius and $\delta_v=\sqrt{2\eta/(\omega\rho)}$ is the skin depth for shear waves, where ω is the acoustic frequency. Polydisperse suspensions may be accounted for by summing the contributions from each size bin.

Implicit in Eq. (9) is the assumption that the absorption coefficient in a suspension of similar particles is linearly proportional to the volume fraction, ϕ, i.e., the process is linearly additive. This assumption is valid for dilute suspensions, in which interparticle interactions may be neglected. Urick[8] showed experimentally that this linear relationship between the viscous absorption coefficient and concentration holds for volume fractions of up to 8% to 9% for kaolin particles at MHz frequencies. Note that a volume fraction of 8% corresponds to a mass concentration of approximately 200 $\mathrm{kg\,m^{-3}}$ for kaolin particles. This is far higher than concentrations found in the natural environment, except perhaps in the boundary region near the seabed.

Equation (9) may be extended to account for spheroidal particles by employing suitable expressions[9] for τ and s, employing a shape factor[19] and inertia coefficient[20] for the drag on spheroidal bodies. The application of these equations to the calculation of attenuation by the suspensions under investigation in this paper was first described by the current authors in another paper[21] and the equations will not therefore be reproduced here. The only additional parameters introduced in this analysis are the semimajor and semiminor axes of the spheroids.

The absorption due to spheroidal bodies in suspension depends on the orientation of the body with respect to the insonifying wave. The model used in this work accounts for

particles having the two orthogonal orientations: with the axis of symmetry orientated parallel to the direction of motion, and with the axis perpendicular to the direction of motion. For comparison with the measurements made using the reverberation technique it is appropriate to assume that the particles are orientated randomly with respect to the acoustic field. That is not to say that they are randomly orientated with respect to the gravitational field. Indeed, there may well be a preferred orientation for particles settling slowly under gravity. However, under diffuse field conditions all propagation directions are equally probable and so it may be assumed that there is no preferred particle orientation with respect to the insonifying sound waves.

When one is considering attenuation by suspended particles in a directional sonar beam, the dependence on orientation should be taken into account if the particles are expected to have a preferred orientation. This will be dealt with in a forthcoming paper.

IV. RESULTS

The results of measurements made with spherical glass particles were found to be in very good agreement with the predictions of Eqs. (9)–(11).[3] In this paper results are presented for two types of nonspherical particle. The first is a form of kaolin (china clay) with the trade name Speswhite, and the second is a form of calcium carbonate with the trade name Polcarb. Both are well characterized industrial samples obtained from English China Clays International (ECCI, now Imerys). These particulates were chosen because they are nonspherical particles representative of some of the types of particles found in the marine environment, but with well known properties, avoiding the problems of working with poorly characterized natural samples. Results from natural marine sediment particles will be presented in a future paper.

Samples of both types of particles were examined using a scanning electron microscope.[12,21] The Speswhite particles were found to take the form of very thin, flat plates, whilst the Polcarb particles were more angular in form. The densities of the Speswhite and Polcarb particles are, respectively, 2600 and 2700 $\mathrm{kg\,m^{-3}}$.

A. Particle size distributions

Figures 3 and 4 show particle size distributions for the Speswhite and Polcarb particles, respectively, measured using laser diffraction analysis, gravitational sedimentation and centrifugal sedimentation.

Clearly there are very significant differences between the size distributions obtained using these different methods. This serves as an illustration of the fundamental difficulties of characterizing irregular particles. As described in Sec. II C, the laser diffraction technique may be considered to yield an effective optical scatterer dimension, since it gives the size distribution of spherical particles which would give the observed optical diffraction pattern. Both the gravitational and centrifugal sedimentation techniques use Stokes' law to determine particle size, and hence yield an effective Stokes diameter.

In the case of Speswhite in particular the centrifugal sedimentation measurement shows a strong bias towards smaller particles. Since samples used for the centrifugal sedimentation measurements were treated with prolonged ultrasound to break up aggregates this might possibly be an indication that there was some degree of aggregation of the Speswhite particles. This is consistent with scanning electron microscope images of the Speswhite particles,[12] which do show some clumping of particles.

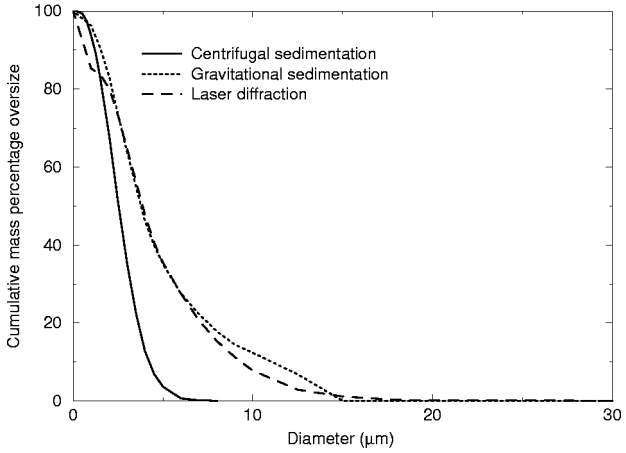

FIG. 3. Size distribution of Speswhite particles measured by centrifugal sedimentation, gravitational sedimentation and laser diffraction.

B. Absorption measurements

Figure 5 shows the measured attenuation coefficient for the Speswhite (kaolin) particles as a function of frequency, normalized with respect to concentration. The symbols indicate measurements made at different concentrations. The measurements are binned at 10 kHz intervals over the range 50–150 kHz, but are shown offset slightly so that the individual error bars can be resolved.

Also shown on this graph is the attenuation predicted by Eqs. (9)–(11) for spherical particles using the three size distributions shown in Fig. 3. These three predictions are in surprisingly close agreement with each other, given the apparent differences displayed by the size distributions measured by the different techniques. However, it must be remembered that the absorption is dominated by those particles

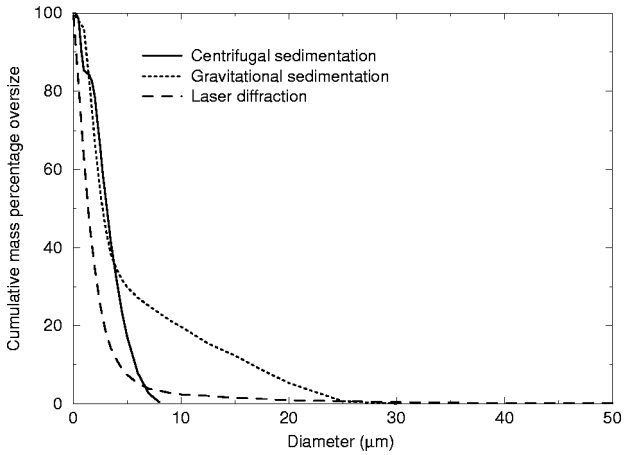

FIG. 4. Size distribution of Polcarb particles measured by centrifugal sedimentation, gravitational sedimentation and laser diffraction.

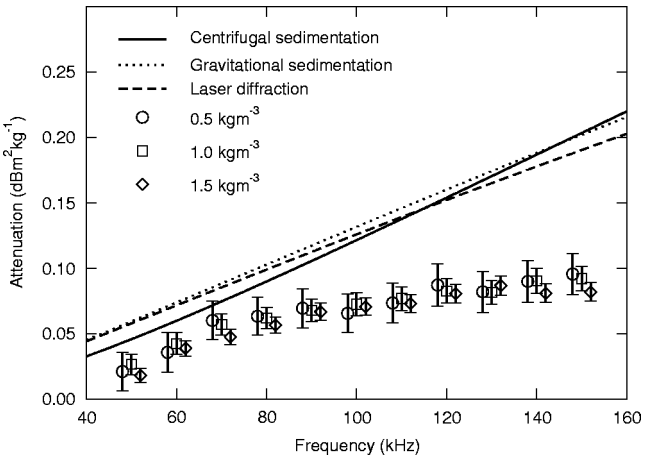

FIG. 5. Normalized attenuation coefficient for Speswhite particles: experimental data and theoretical predictions assuming spherical particles. Data points have been offset in frequency to show individual error bars (see text).

whose size is of the order of the skin depth for shear waves. At 100 kHz, for example, the skin depth in water is approximately 2 μm, and 80% of the absorption is due to particles smaller than 6.5 μm. As expected for these highly nonspherical particles, the prediction of the theory for spherical particles does not agree well with the measured attenuation.

Figure 6 shows the results of the measurements of attenuation with Polcarb (calcium carbonate) particles. Again, the symbols indicate measurements made at different concentrations, and the data have been normalized with respect to concentration. In this case the theoretical predictions using the size distributions yielded by the different sizing techniques show greater differences than in the case of the Speswhite. This is because the size distributions have greater differences in the particle size range contributing most to the absorption, although this is not immediately apparent from visual inspection of the curves showing cumulative mass percentage oversize.

The measurements for Polcarb show much better agreement with the theoretical predictions than was the case for Speswhite. Although the theoretical curves do not have the same form for the frequency dependence as suggested by the

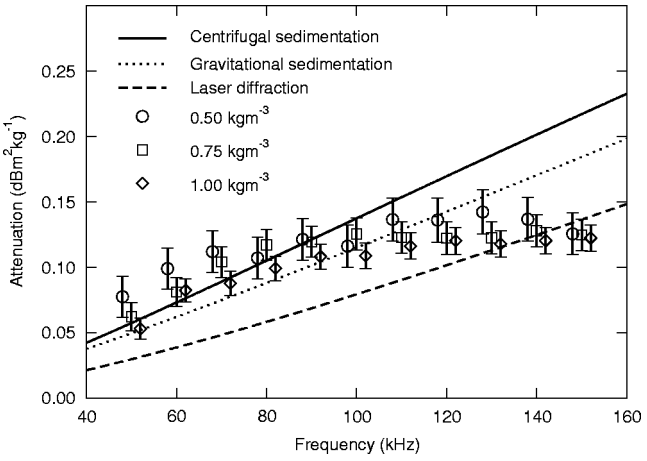

FIG. 6. Normalized attenuation coefficient for Polcarb particles: experimental data and theoretical predictions assuming spherical particles. Data points have been offset in frequency to show individual error bars (see text).

data, the magnitude of the predicted absorption is close to the measured absorption over much of the frequency range. The fact that the Polcarb measurements agree better with the predictions of the spherical particle theory than the Speswhite measurements is not surprising, since inspection of electron microscope images shows the Speswhite particles to be highly platelike whereas the Polcarb particles, although certainly irregular and nonspherical, are closer to spheres in aspect ratio.

These figures show the comparison between the predictions of Eq. (9) with Eqs. (10) and (11), which assumes spherical particles, and measurements made with nonspherical particles. These results demonstrate that the spherical model is of some limited use in providing estimates of the attenuation from the Polcarb particles. In the case of the highly nonspherical Speswhite particles, the spherical theory significantly overpredicts the attenuation.

A method for calculating the attenuation due to suspensions of oblate spheroids was referred to in Sec. III. Since the degenerate form of an oblate spheroid is a thin circular disk it is appropriate to approximate the platelike Speswhite particles as oblate spheroids.

In order to apply this method it is necessary to know the size distribution of the spheroids representing the particles. It is not appropriate to use the size distributions shown in Figs. 3 and 4 as these are distributions of equivalent spheres. Therefore these distributions have been used to derive new distributions for spheroids.

The starting point for this process is the distribution of Stokes diameters obtained from gravitational sedimentation measurements. These were originally derived by applying Eq. (5) to measurements of the fraction of particles which settle out of suspension as a function of time. Although the original time-domain data were not available, it was possible to recalculate them by inverting Eq. (5) and applying it to the distributions of Stokes diameters. This gives the fraction of particles which settle out of suspension as a function of time. Now, if the settling velocity of spheroids is known as a function of their major and minor radii, these data can be used to derive a particle size distribution for the spheroids.

The steady-state settling velocity, v_s, for spheroids with shape factor[19] K_{sf} (which depends on the aspect ratio) may be obtained by equating the drag force on a spheroid:

$$F_0 = 6\pi\eta K_{sf} a' v_s \tag{12}$$

with the gravitational force and rearranging to give

$$v_s = \frac{4\pi a' b' (\rho' - \rho) g}{18\pi\eta K_{sf}}, \tag{13}$$

where a' and b' are, respectively, the semi-major and semi-minor axes for oblate spheroids and the semi-minor and semi-major axes for prolate spheroids.

In this way the size distribution for Speswhite particles derived by gravitational sedimentation, shown in Fig. 3, was used to calculate the size distribution of spheroids of given aspect ratio, $h = b'/a'$, which would give the same measured settling time history. A similar analysis is used for the centrifugal sedimentation measurements.

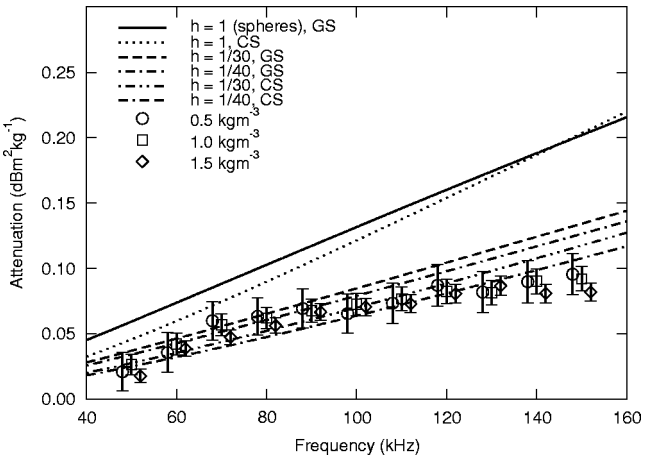

FIG. 7. Normalized attenuation coefficient for Speswhite particles: experimental data and theoretical predictions using the model for spheroidal particles. The predictions are based on size distributions derived from gravitational sedimentation (GS) and centrifugal sedimentation (CS) measurements.

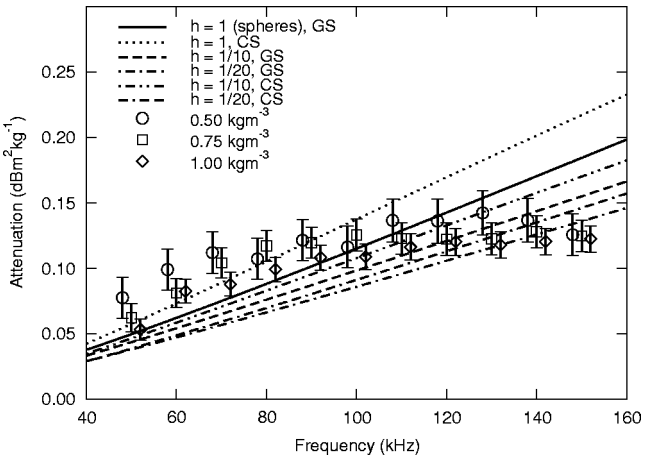

FIG. 8. Normalized attenuation coefficient for Polcarb particles: experimental data and theoretical predictions using the model for spheroidal particles. The predictions are based on size distributions derived from gravitational sedimentation (GS) and centrifugal sedimentation (CS) measurements.

These particle size distributions were then used to calculate the attenuation spectrum due to the suspension of spheroids, using the approach referred to in Sec. III. The attenuation measurements, as described in Sec. II, employ an approximately diffuse field in which all directions of propagation are equally probable. Ideally, then, these calculations would be integrated over all particle orientations. However, the method only yields solutions for the two orthogonal cases. In the case of oblate spheroids the three independent spatial directions may be resolved into the broadside direction and two edgewise directions, and the results presented below were therefore obtained assuming that two-thirds of the particles were orientated edgewise to the sound field and one-third orientated broadside.

Figure 7 shows the comparison between the attenuation predicted using the oblate spheroid model and the measured attenuation for Speswhite particles. The range of typical aspect ratios of the Speswhite particles as quoted by the suppliers is $h=\frac{1}{30}$ to $h=\frac{1}{40}$. The figure therefore, shows predictions for these values of aspect ratio, together with the curve for $h=1$, i.e., for spherical particles. The predictions for spheres, using the spheroidal model with $h=1$, are in agreement with the predictions of the spherical model shown in Fig. 5, and thus overestimate the attenuation. The predictions for the aspect ratios which are representative of the Speswhite particles, however, show much better agreement with the measured attenuation. It is notable that this agreement is achieved without using any *a priori* knowledge of the attenuation measurements. While the predictions of the spheroid model show significantly improved agreement with the data, it is noted that the exact form of the frequency dependence suggested by the data is not predicted by the model. In particular there is a prominent increase in attenuation over the range 50 to 70 kHz which is not predicted. This is in the frequency range of the Schroeder cut-off and this is suggested as a possible explanation for this feature of the data. It is also noted that the predictions based on the centrifugal sedimentation measurements of particle size distribution appear to be slightly closer to the measurement than those based on gravitational sedimentation, but the data do not really support any definitive conclusions on this matter.

The spheroidal approach has also been applied to the Polcarb particles in exactly the same manner. However, inspection of the scanning electron microscope images[12,21] showed that they are very angular and irregular. The spheroidal approach was therefore not expected to yield significantly improved agreement over the spherical model. Furthermore, no information was available on the aspect ratio, other than perhaps what could be inferred from the microscope images. Therefore calculations were performed for a number of different aspect ratios and those which gave predictions which were reasonably close to the measured data are shown in Fig. 8. All that can really be concluded from this result is that the predictions of the spheroidal model for $0.05 \leqslant h \leqslant 1$ are neither better nor worse than the predictions of the spherical model. This is consistent with expectations, given the nature of the particles.

Particle size data based on laser diffraction analysis were also presented in Sec. IV A. It is not, however, appropriate to use these data in the spheroidal analysis, as was done using the gravitational and centrifugal sedimentation data. That is because the analysis relies on equating the steady-state settling velocities of spheres and spheroids. To do that with the laser diffraction data would require that the effective optical scatterer diameter be equated to the Stokes diameter, a step which would be both arbitrary and nonphysical.

V. DISCUSSION

The experimental method described in this paper has previously been shown[3] to give results for absorption by suspensions of spherical particles which are in good agreement with theory. In this paper the method has been applied to the measurement of absorption in suspensions of highly nonspherical particles of Speswhite (kaolin) and Polcarb (calcium carbonate). With these nonspherical particles the method of obtaining the particle size distribution became an important issue, and three different particle sizing techniques

were employed. Significant differences were observed in the particle size distributions yielded by the three techniques, leading to differences in the predicted attenuation.

The predictions of Urick's equation for visco-inertial absorption by spherical particles, integrated over the size distributions obtained using the three particle sizing techniques, significantly overestimate the attenuation by the Speswhite particles. This poor agreement is to be expected since these platelike particles are far from spherical. Agreement between the spherical particle model and the experimental data for Polcarb particles is much better, although the model fails to predict the observed frequency dependence in the attenuation. These particles, while still highly irregular, have aspect ratios closer to unity than the kaolin particles do. The closer agreement is therefore consistent with expectation.

A model for acoustic attenuation by dilute suspensions of spheroidal particles has been used to predict the attenuation in suspensions of the two different particles. This model is most appropriate for the platelike Speswhite particles as the degenerate form of an oblate spheroid is a circular disk. The predictions of the model for oblate spheroids, using particle size data based on gravitational sedimentation measurements, were found to be in good agreement with the measurements of attenuation by Speswhite particles. This agreement was obtained without the need for any *a priori* information about the attenuation measurements. Agreement between this model and the measurements made with Polcarb particles was less good, as expected due to the shape of these particles.

It may be noted that instruments based on the standard, spherical analysis of laser diffraction measurements are commonly used for *in situ* particle sizing.[22] The results given in this paper suggest that such measurements may be of limited use when applying the derived size distributions to the prediction of acoustic attenuation spectra.

It is concluded that the model for visco-inertial absorption by oblate spheroids used in this paper is appropriate for estimating the absorption in dilute suspensions of platelike clay particles.

ACKNOWLEDGMENTS

The authors are grateful to S. G. Foran, QinetiQ Bridgwater, for performing the particle size measurements, and to Gary Heald, QinetiQ Winfrith, for valuable discussions. TGL would like to thank the Royal Society and the Leverhulme Trust for a Senior Research Fellowship. This work was funded by Technology Group 01 of the MoD Corporate Research Program.

[1] S. D. Richards, A. D. Heathershaw, and P. D. Thorne, ''The effect of suspended particulate matter on sound attenuation in seawater,'' J. Acoust. Soc. Am. **100**, 1447–1450 (1996).
[2] S. D. Richards and T. G. Leighton, ''Acoustic sensor performance in coastal waters: Solid suspensions and bubbles,'' in *Acoustical Oceanography*, Vol. 23 of *Proceedings of the Institute of Acoustics*, edited by T. G. Leighton, G. J. Heald, H. D. Griffiths, and G. Griffiths (Bath U. P., Bath, 2001), pp. 399–406.
[3] N. R. Brown, T. G. Leighton, S. D. Richards, and A. D. Heathershaw, ''Measurement of viscous sound absorption at 50–150 kHz in a model turbid environment,'' J. Acoust. Soc. Am. **104**, 2114–2120 (1998).
[4] S. Temkin and R. A. Dobbins, ''Attenuation and dispersion of sound by particulate-relaxation processes,'' J. Acoust. Soc. Am. **40**, 317–324 (1966).
[5] J. M. Evans, ''Models for sound propagation in suspensions and emulsions,'' Ph.D. thesis, Open University, 1996.
[6] Z. W. Qian, ''Fractal dimensions of sediments in nature,'' Phys. Rev. E **53**, 2304–2306 (1996).
[7] Q. Wang, K. Attenborough, and S. Woodhead, ''Particle irregularity and aggregation effects in airborne suspensions at audio and low ultrasonic frequencies,'' J. Sound Vib. **236**, 781–800 (2000).
[8] R. J. Urick, ''The absorption of sound in suspensions of irregular particles,'' J. Acoust. Soc. Am. **20**, 283–289 (1948).
[9] A. S. Ahuja and W. R. Hendee, ''Effects of particle shape and orientation on propagation of sound in suspensions,'' J. Acoust. Soc. Am. **63**, 1074–1080 (1978).
[10] G. Kurtze and K. Tamm, ''Measurements of sound absorption in water and aqueous solutions of electrolytes,'' Acustica **3**, 34–48 (1953).
[11] T. G. Leighton, N. R. Brown, and S. D. Richards, ''Effect of acoustic absorption by hydrophone and cable on a reverberation technique for measuring sound absorption coefficient of particulate suspensions,'' ISVR Technical Report No. 299, University of Southampton, 2002.
[12] S. D. Richards, ''Ultrasonic visco-inertial dissipation in dilute particulate suspensions,'' Ph.D. thesis, University of Southampton, 2002.
[13] M. R. Schroeder, ''New method of measuring reverberation time,'' J. Acoust. Soc. Am. **37**, 409–412 (1965).
[14] L. E. Kinsler, A. R. Frey, A. B. Coppens, and J. V. Sanders, *Fundamentals of Acoustics*, 3rd ed. (Wiley, New York, 1982).
[15] A. D. Pierce, *Acoustics: An Introduction to its Physical Principles and Applications* (McGraw-Hill, New York, 1981), reprinted by the Acoustical Society of America, 1989 and 1994.
[16] BS3406 Part 2, *British Standard Methods for Determination of Particle Size Distribution. Part 2: Recommendations for Gravitational Liquid Sedimentation Methods for Powders and Suspensions*, 1984.
[17] BS3406 Part 6, *British Standard Methods for Determination of Particle Size Distribution. Part 6: Recommendations for Centrifugal Liquid Sedimentation Methods for Powders and Suspensions*, 1985.
[18] G. G. Stokes, ''On the effect of internal friction of fluids on the motion of pendulums,'' Trans. Cambridge Philos. Soc. **IX**, 8–106 (1851).
[19] J. Happel and H. Brenner, *Low Reynolds Number Hydrodynamics* (Prentice–Hall, Englewood Cliffs, NJ, 1965).
[20] H. Lamb, *Hydrodynamics*, 6th ed. (Dover, New York, 1945).
[21] S. D. Richards, T. G. Leighton, and N. R. Brown, ''Visco-inertial absorption in dilute suspensions of irregular particles,'' Proc. R. Soc. London, Ser. A **459**, 1–15 (2003).
[22] Y. C. Agrawal and H. C. Pottsmith, ''Instruments for particle size and settling velocity observations in sediment transport,'' Mar. Geol. **168**, 89–114 (2000).