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BROADBAND SCATTERING MEASUREMENTS BY ACOUSTIC SPECTROMETER

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ABSTRACT

A widebandwidth near-plane-wave ultrasonic spectrometer is used to measure absorption and propagation velocity in monodispersions of polystyrene spheres in water. The results show good agreement with theoretical calculations based on Allegra and Hawley [J. Acoust.Soc.Am. 51(5), 1545-64, (1972)] and could be used as the basis for an industrial process monitoring instrument.

1. INTRODUCTION

In many industrial processes, there is a need to characterise solid/liquid and liquid/liquid suspensions in terms of the suspended particle size distribution, volume fraction and other factors. Ultrasound is considered to be the most likely method to achieve characterisation on an industrial scale because the relevant technology is well known and relatively cheap, and ultrasonic radiation can penetrate and interact with mixtures that are optically opaque. The parameters most commonly measured are near-plane-wave absorption and propagation velocity as functions of frequency. Both are also functions of the physical properties of the test medium and the theoretical relationship between these and the measureable quantities forms the basis of the characterisation process. This relationship is generally very complicated and the characterisation process requires that the measured data (absorption and propagation velocity, say) be the basis for fitting parameters of the theoretical models of propagation. The result of the minimised error fit will then be the required measurement - particle size distribution, volume fraction etc. The development of measurement processes such as these requires that

- i) Theoretical models must be verified as appropriate to the colloidal system being studied.
- ii) Multiple frequency or broadband measurements of wave absorption and speed be available.
- iii) The physically measured data be digitised so that the model fitting process can be carried out by computer

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on line.

Whilst it is now relatively easy to digitise radio frequency signals, most techniques for the measurement of absorption and wave velocity are narrowband and quite slow, requiring changes of transducer driving frequency, changes of the transducers themselves, or even a change of cavity or vessel containing the test medium.

Our recent development^{1,2} of a broadband ultrasonic transmission spectrometer has contributed greatly to the solution of this problem. In the present work we have investigated its use in the measurement of wave absorption and propagation velocity in suspensions of monodisperse spheres of polystyrene in water; we have compared our broadband results with theoretical predictions based on the discussion of Allegra and Hawley³ and have shown good agreement. In the sections of this paper which follow we summarise our spectrometric techniques and theoretical computations, and present our wide bandwidth absorption and velocity results for the dispersed polystyrene particles. Excellent agreement is obtained between theory and experiment.

2. THE SPECTROMETER

The basis of the instrument^{1,2} is a 30 ml cube-shaped hollow cell in which the test liquid is placed. Acoustically thick⁴ transducers are fixed into two opposite faces, one acting as transmitter and one as receiver. The transmitter is excited by a short electrical pulse (500 v x 10 ns) and the electrical signals at the receiver terminals are amplified by a linear integrated amplifier of 200 MHz bandwidth and digitised at 200 MHz (Le Croy TR 8828) for subsequent processing and analysis by minicomputer (DEC, LSI 11-73). Signal processing procedures applied at present are coherent averaging to improve signal to noise ratio, and approximate deconvolutions for the radiation coupling response of the field between the two transducers, and for the insertion properties of the transducers themselves. The output of the instrument is absorption ($\alpha, \alpha\lambda$, or α/f^2) and propagation velocity as a function of frequency in the range 1 MHz to 60 MHz. An optional final stage in the computation is to fit a theoretical propagation model to the measured data, using an algorithm based on Marquardt's method⁵.

3. THEORY

Allegra and Hawley³ give a clear summary of relevant theory. The acoustic field equations yield wave equations for three

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potentials: compression, shear and thermal. A compression wave incident on a suspended sphere results in compression, shear and thermal waves reflected back from the sphere and transmitted into the sphere. A solution to the equation for each of these waves gives a set of seven equations expressed in spherical polar coordinates. Only single interactions between waves and spheres area assumed. These seven equations are then solved simultaneously in a far field limit to give a series of so-called scattering coefficients which can be combined to give wave velocity and attenuation. Following the discussion of McClements and Povey⁶ the complex propagation constant for the axial direction between the transducers, B , is given by

$$\left(\frac{B}{k_c}\right)^2 = 1 + \frac{3\phi}{j k_c^3 r^3} \sum_{n=0}^{\infty} (2n+1) A_n + M \quad (1)$$

Where k_c is the compression wave number in the suspending medium, ϕ the volume fraction occupied by the suspended particles and r their radius. j is $(-1)^{1/2}$ and A_n are the so-called scattering coefficients. The middle term gives the contribution to B from single scattering processes and M represents the (here neglected) contributions from multiple scattering processes. The propagation velocity and attenuation can be obtained by equating the real and imaginary parts on either side of the equality in Eq1, and noting that

$$B = \frac{\omega}{c(\omega)} + j\alpha(\omega) \quad (2)$$

Where w , c and α have their usual meanings. Only the low order coefficients (A_0 , A_1 , A_2) make significant contributions to attenuation in the systems we are studying. They can be interpreted thus :

If $A_0 = A_{0,1} + A_{0,2}$, then $A_{0,1}$ represents the change in attenuation and velocity that results from the substitution of the suspending material by the material of the particles. $A_{0,2}$ represents thermal conduction and scattering loss due to the difference in the parameter $(B/\rho C_p)$ for the two materials. B is the coefficient of thermal expansion, ρ the density and C_p the specific heat at constant pressure. A_1 represents the contribution from so-called visco-inertial loss which occurs when a particle is physically moved to and fro in a liquid due to its different density, and radiates a secondary sound field. A_2 , whilst not described explicitly in earlier publications, behaves as though it represents re-radiation due to resonance of the particles themselves. The computations given in section 3 of this paper

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illustrate these A_n contributions to attenuation in graphical form.

For the purpose of comparison, we have also carried out computations based on the non-scattering mixture-substitution theory of Urlick⁷, in which effective compressibility and density depend simply on the volume fraction, and the properties of the particles, and of the suspending medium. Following this model, with subscripts 1 and 2 for the medium and particles respectively, we have, for effective density

$$\rho = \phi \rho_2 + (1 - \phi) \rho_1 \quad (3a)$$

and for effective compressibility

$$k = \phi k_2 + (1 - \phi) k_1 \quad (3b)$$

Wave velocity is independent of frequency, given by

$$c = (1/k\rho)^{1/2} \quad (4)$$

4. COMPUTATIONS AND MEASURED RESULTS

Fig. 1 shows the calculated absorption (α/f^2) and compression wave velocity versus frequency for monodisperse polystyrene particles in water. For the absorption data the contributions of A_0 , A_1 and A_2 , and their sum, are shown by solid lines and our experimental results are shown by dotted lines. For the velocity data the solid lines show theoretical calculations with A_0 , A_1 and A_2 contributing, whereas the dotted lines show measured results. From these results we note that A_0 represents the major contribution to the absorption process, and that there is good agreement between measurement and theory. The form of the predicted velocity curve followed the measured function of frequency in all cases but the actual values differed by less than 1.5 ms^{-1} .

Fig. 2 shows our results for the 2 MHz components taken from spectrometer data, plotted versus volume fraction (crosses), irrespective of particle size. The actual sizes in the experiment range from 100 nm to 308 nm. The solid curves show predictions for two extreme particle sizes, whilst the dotted line shows expectation on the basis of Urlick's⁷ theory.

Fig. 3 shows the 2 MHz data of Barrett-Gultepe⁸ plotted against expectation for the Urlick model (dotted line) and the Allegra-

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Hawley's scattering model (solid line) described earlier.

5. CONCLUSION

The wide bandwidth spectrometer can be used to study the frequency dependence of attenuation and wave velocity in solid-in-liquid suspensions. Good agreement was observed between measured results and predictions based on a single scattering model incorporating only the first three scattering coefficients, A_0 , A_1 and A_2 , although the calculated result was dominated by the A_0 term (thermal scattering and material substitution.) Finally, the Urlick' model gave a very poor prediction of velocity as a function of volume fraction, and this casts serious doubts on the discussion of Barrett-Gultepe et al⁸.

6. REFERENCES

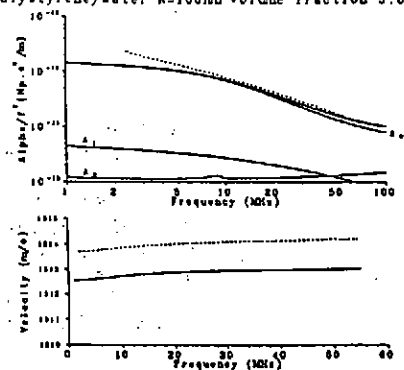
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2. R.E. Challis , A.K. Holmes, J.A. Harrison and R.P. Cocker. Ultrasonics, 25, 5-15 (1990)
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7. FIGURES

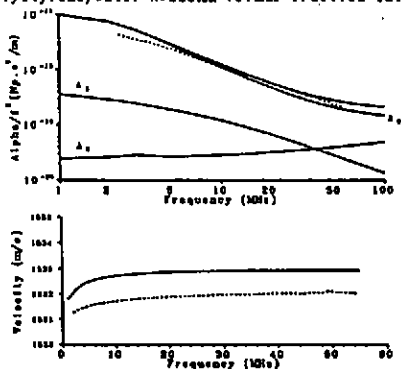
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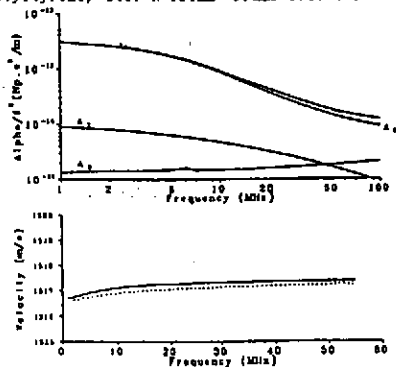
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Polystyrene/water R=225nm volume fraction 12.1%



Polystyrene/water R=104nm volume fraction 7.03%



Polystyrene/water R=308nm volume fraction 8.25%

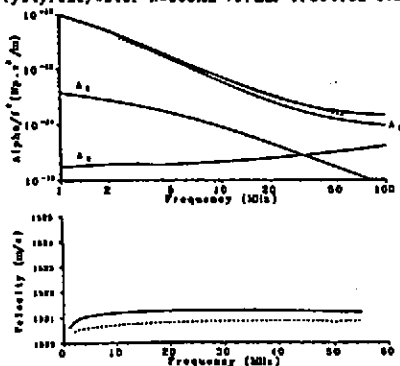


Fig.1. Calculated absorption (α/f^2) and compression wave velocity for monodisperse polystyrene particles in water. The contributions due to A_0 , A_1 and A_2 are shown, as well as the total. The dotted lines show experimental results obtained with the spectrometer.

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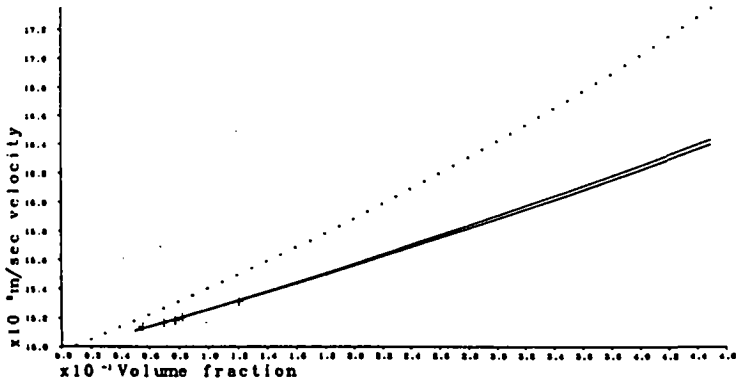


Fig.2. Velocity results at 2MHz taken from the curves of Fig.1 plotted versus volume fraction (crosses), irrespective of particle size. The solid lines show predictions at the two extreme particle sizes (100nm, 308nm) whilst the dotted line gives expectation from the Urlick⁷ model.

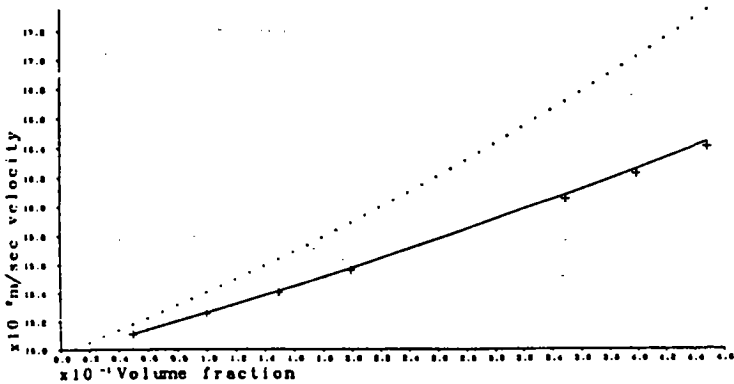


Fig.3. 2MHz velocity data of Barrett-Gultepe⁸ plotted against expectation for the Urlick model (dotted line) and the Allegra & Hawley³ model (solid line).

