3P2-10

Accurate Ultrasound Scattering Analysis of the Size Distribution and Mechanical Properties of Microparticles in Liquid

液体中の微粒子のサイズ分布と弾性率の精密超音波散乱解析

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1. Introduction

Ultrasonic transmission Spectroscopy (US) is a powerful tool to evaluate the mechanical properties and/or size distribution of particle dispersed in liquid. When the mechanical properties of patticle are well known, the size distribution may be easily determined by acoustic spectra. On the other hand, when the elastic properties are unknown, there is a serious uncertainty to determine the size distribution of particle. For this reason, the fundamental physical properties such as the attenuation coefficient, phase velocity, and density of particle should be accurately determined prior to the particle scattering analysis. Especially, in the case of a concentrated suspension or a submicron-sized particles-suspension, it could be an important factor to reproduce the acoustic spectra. It is noted that the acoustic properties must be evaluated as a function of frequency. In this syudy, an analysis method for determination of the attenuation coefficient, phase velocity, and density of solid plates was explored. In addition, particles having a physical properties equivalent to the solid plate were prepared by remolding the plate to study the concentarted particle suspensions with the propertes evaluated by the plate analysis.

2. Experiments

Acrylite™, an acrylic resin mainly composed of poly(methyl methacrylate) (PMMA), was purchased from Mitsubishi Chemical Co. Japan. PMMA particles were prepared by an emulsification/deswelling method[1], using a PMMA/acetone/toluene solution in water.

A spike pulse emitted from a pulser was transferred to a longitudinal plane wave transducer immersed in a water bath to generate ultrasound pulses. The transmitted and reflected signals were respectively received by another transducer and the same transducer, followed by successive recording with a high-speed digitizer. The signals utilized in the plate analysis were schematically illustrated in **Fig. 1**. In order to obtain the frequency dependences of the intensity attenuation coefficient

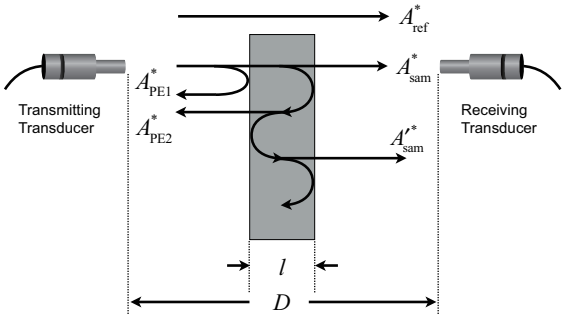

Fig. 1 Schematic illustration of transmission and reflection signals utilized in the plate analysis.

α, the phase velocity c, density ρ, and thickness l were analyzed using the relations[1],

$$c_{sam} = c_{water}\left[1 - \frac{2\left(\Delta\theta - \arg\left[1-\left(r_{10}^*\right)^2\right]\right)}{\Delta\theta' - \arg\left[\left(r_{10}^*\right)^2\right]}\right]$$

$$l = \frac{c_{water}}{4\pi f}\left[\Delta\theta' - \arg\left[\left(r_{10}^*\right)^2\right] - 2\left(\Delta\theta - \arg\left[1-\left(r_{10}^*\right)^2\right]\right)\right]$$

$$\alpha_{sam} = \alpha_{water} - \frac{2}{l}\left[\ln\left|\frac{A^*_{sam}}{A^*_{ref}}\right| - \ln\left|1-\left(r_{10}^*\right)^2\right|\right]$$

$$\rho_{sam} = \frac{Z_{water}}{c^*_{sam}}\frac{1-\left(r_{10}^*\right)}{1+\left(r_{10}^*\right)}, \quad \left(\left(r_{10}^*\right)^2 = \frac{\left(A'^*_{sam}/A^*_{sam}\right)}{\left(A'^*_{sam}/A^*_{sam}\right)-\left(A^*_{2PE}/A^*_{1PE}\right)}\right)$$

where A is the amplitude, θ is the phase of the transmitted pulse and r_{10} is the reflection coefficient between water 0 and sample 1, and the subscript "sam" and "ref" respectively refer to the transmitted pulse for the sample and reference.

3. Results

Fig.2 shows the frequency dependence of the attenuation coefficient α, the phase velocity c_L, density ρ, and thickness l, obtained for the PMMA plate. Four pairs of transducers (5, 10, 20 and 30 MHz) were used to acquire the spectra in the wide range of frequencies. The density was evaluated to be 1.187 ± 0.008 (g/cm^3), which was close to the independent experiment obtained by floating test in a reference liquid calibrated by a pycnometer (1.182 (g/cm^3)). The ultrasonically evaluated value of thickness was in good agreement with the value

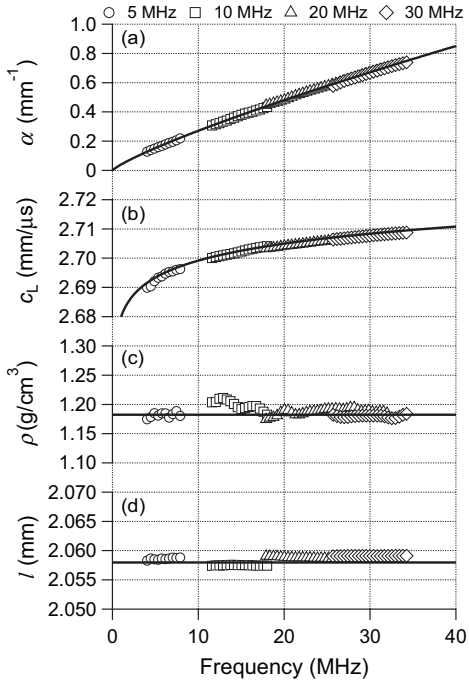

Fig. 2 The frequency dependence of (a) α, (b) c_L, (c) ρ and (d) l of the PMMA plate obtained by using 5, 10, 20 and 30 MHz transducers.

measured by a thickness gauge as indicated by the solid line in **Fig. 2(d).** The f (Hz)-dependent values of the attenuation and phase velocity, $\alpha = 0.47 \times f^{0.82}$ (mm^{-1}) and $c_L = 2.566 \times f^{0.0031}$ (mm/μs) at 4 -34 MHz, were given by the nonlinear least-squared fitting. These coefficients are subsequently used in the scattering analysis of suspensions as the known mechanical properties of particles. In addition, the shear velocity $c_S = 1.264 \times f^{0.0046}$ (mm/μs) was evaluated by the mode conversion technique.

Fig. 3 shows the frequency spectra of (a) attenuation coefficient α, and (b) phase velocity c, obtained for the PMMA particles dispersed in water with a small amount of surfactant, sodium dodecyl sulfate (SDS). The open markers represent the data acquired at the different particle concentration and the solid lines indicate the theoretical curves reproduced by the ECAH theory[1], [2] with the dispersion relation of Lloyd-Berry. As shown in the figure, the experimental results were satisfactory reproduced by the ECAH theory with given size distribution (the average diameter d and the standard deviation are respectively 15.0 μm and 2.64 μm) evaluated by an optical microscope.

Finally, the analysis method was applied to the particle sizing of the concentrated nanoparticle suspensions with d = 150 nm where given frequency spectra of the attenuation coefficient, velocities and density were employed to reproduce the suspension data. Thus evaluated d was shown by the open circles in **Fig 4**. In contrast, when the fixed parameters were used, d was seriously overestimated as shown by the open squares.

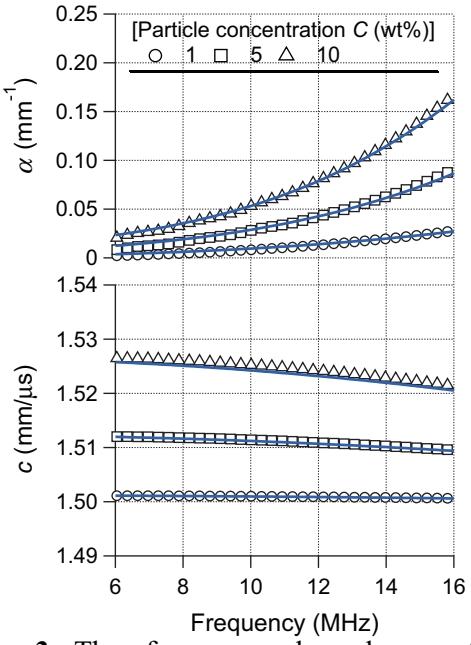

Fig. 3 The frequency dependence of (a) attenuation coefficient and (b) phase velocity obtained for the PMMA particle suspensions. Experimental data (open markers) and theoretical reproduction (solid lines).

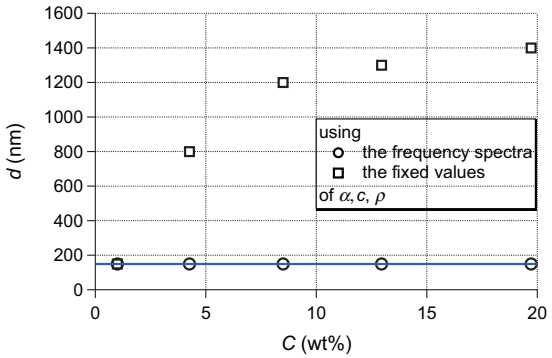

Fig. 4 The average diameter evaluated by the conventional method (open square) and the new analysis method (open circle) at each concentration.

4. Conclusions

The acoustic spectra of the particle suspensions could be successfully reproduced by the ultrasound scattering theories without any adjustable parameters up to 20wt%. This opens a new route to understand the acoustical scattering of concentrated suspension and to perform nanoparticle sizing in the concentrated regime.

References

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