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Simultaneous evaluation of size distribution and mechanical properties of microparticles in suspensions by ultrasound spectroscopy

Nguyen Thao Tran [1‡], Hideyuki Nakanishi[1] , Tomohisa Norisuye[1] and Qui Tran-Cong-Miyata[1]
([1]Grad. School of Sci. & Tech., Kyoto Institute of Technology)

1. Introduction

Ultrasound spectroscopy is a technique to evaluate the attenuation coefficient and sound velocity in materials. In this technique, the sample is placed between transmitting and receiving transducers to measure the amplitude and phase of the transmitting signals in the frequency domain. Although the technique has been utilized to investigate purity or composition of a binary mixture, for most cases, what obtained are the average properties of the two components. However, such an additive role does not hold for systems containing objects with a finite size such as particle suspensions because of scattering. When ultrasound is considered as a wave accompanying diffraction and scattering, static and dynamic scattering approach similar to light or X-ray scattering technique could be applied. On the other hand, ultrasound is propagation of force or displacement so that the viscoelasticity of the sample can be evaluated. As a matter of fact, ultrasound has been utilized for rheological measurements although the corresponding elastic moduli were obtained at frequencies in the range of megahertz. In this study, scattering of ultrasound waves was fully utilized to identify the particle structure such as hard spheres or core-shell particles, followed by evaluation of the elastic information on specific parts of the structure (e.g., the shell part of microcapsules) dispersed in a liquid.

2. Experiments and Results

The frequency dependence of the attenuation coefficient, α, and sound velocity, c, for polydivinylbenzne (PDVB) particles with the volume fraction ϕ = 0.3% having various particle diameters is shown in Fig. 1. The solid line indicates the theoretical calculation based on a dispersion relation and the Faran's scattering theory where the scattered amplitude is calculated by solving three equations with boundary conditions in terms of normal stress, tangential stress, and normal displacement at the surface of a particle in liquid. The theory contains information about the longitudinal, v_L, and shear velocity, v_S in addtion to the viscosity of solvent, η, allowing the evaluation of the shear modulus and bulk modulus. Therefore, the mechanical information of microparticles can be obtained without neither contacting nor dilution of the suspension. It was found that the α and c of the linear polystyrene particles could be reproduced by the theory without any adjustable parameters provided that the particle size distribution, density and viscosity of liquid were calibrated prior to the analysis. The analysis also revealed that our PDVB particles were 40% more rigid than the PS particles. Further analysis on the crosslinked density will be performed in future studies.

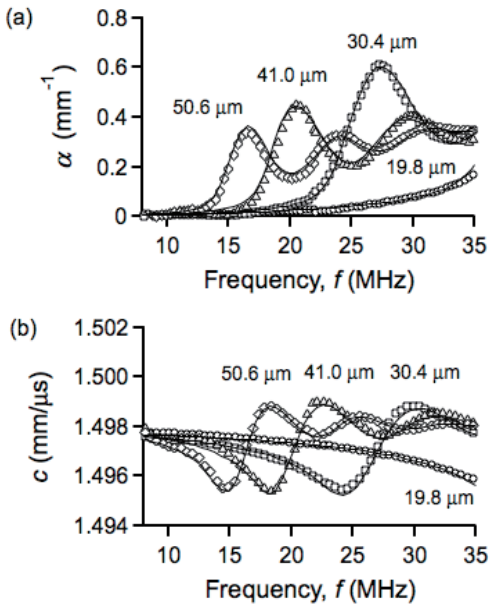

Fig. 1 The frequency dependence of α (a) and c (b) obtained for the particle suspensions of the PDVB particles with different diameters.

In the case of hollow particles, a 6 x 6 matrix is solved to obtain the scattered amplitude because of the additional boundary conditions at the inside and outside of the hollow particle. The

obtained results of the α and c of borosilicate glass are shown in Fig. 2. In contrast to the simple rigid spheres, the shell thickness is an additional unknown parameter for the analysis of microcapsules. In order to reduce the uncertainty of analysis, we carried out a SEM analysis first to obtain the particle size and the thickness distribution with the fixed elastic moduli. As the results, the experimental data were well reproduced by the Goodman's scattering theory as indicated by the solid line. The meaning of the dashed line will be discussed below.

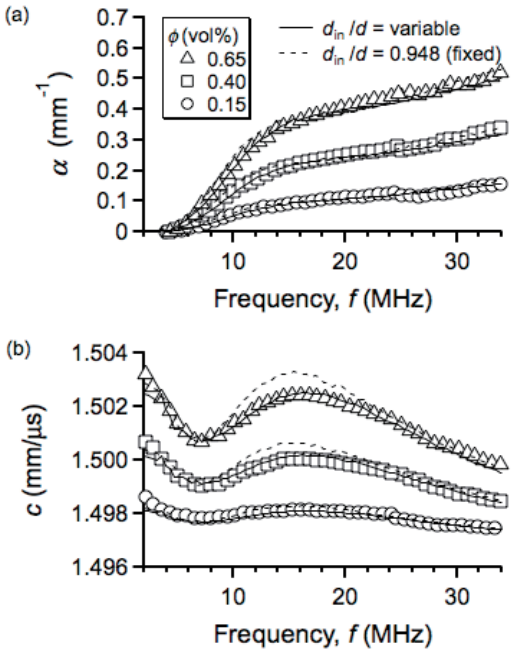

Fig. 2 The frequency dependence of α (a) and c (b) obtained for the borosilicate particle suspensions with different concentrations.

Fig. 3 (a) shows the SEM histogram of the particle size. The ratio of the inner and outer diameters (d_{in}/d) is also shown in Fig.3 (b) as a function of the diameter. Here, the open circles indicate the result of SEM analysis, while the solid circles show a moving average every 10 μm for clarity. As seen from the figure, it was found that the d_{in}/d was not a constant. In order to show the effect of thickness distribution on the acoustic data, the calculated results based on d_{in}/d = 0.948 is also demonstrated by the dashed line in Fig. 2, suggesting the importance in the shell - thickness distribution calibrated by SEM to the frequency dependence of the data. However, the values of α and c are sufficiently sensitive to the employed fitting parameters so that the evaluation of the mechanical properties by ultrasound spectroscopy could be uniquely carried out with a good accuracy even if the curve has slight deviation from the experimental data.

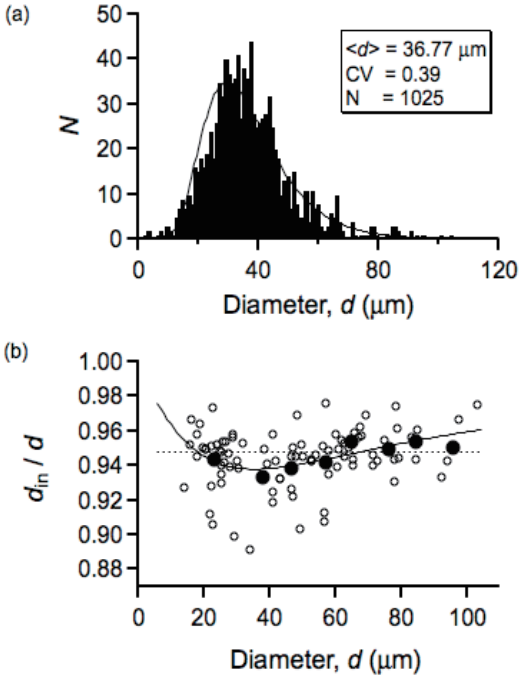

Fig. 3. (a) The particle size distribution and (b) The diameter dependence of d_{in}/d calibrated by SEM for the borosilicate particle.

3. Conclusions

The frequency dependences of ultrasound attenuation and sound velocity obtained for the microsphere suspensions could be successfully carried out by combining the scattering theories with the dispersion relation. The elastic moduli were quantitatively evaluated by the density and speed of sound obtained by the fitting analysis. Since these values agreed well with the literature values, it was concluded that the ultrasound spectroscopy method could be a good candidate for evaluation of mechanical properties and structures of micron-sized particles without dilution of suspension. In addition, it opens a new route to monitor the change in elastic moduli without contacting and disturbing the systems and may be utilized in practical applications, such as drug delivery systems.

References

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