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Ultrasound Scattering Studies on Silica Micro-particle Suspensions

シリカ微粒子懸濁液の超音波散乱解析

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1. Introduction

Ultrasound spectroscopy has been utilized to investigate the structures and properties of micro-particles dispersed in liquid. Since sound waves propagate through the material with its mechanical contrast, the reduction of amplitude and the difference of phase respectively give the viscous and elastic information of the sample. Therefore, the attenuation and velocity of ultrasound pulse allow us to evaluate the elastic moduli of particles dispersed in suspension. So far, the validity of the technique was shown by analyzing the acoustic properties obtained for micrometer sized polydivinylbenzene (PDVB) particles[1)]. On the other hand, for charged particles, ultrasound propagation is considered to be more or less affected by the presence of charge at the surface of particles. However, its effect on the frequency dependencies of the attenuation coefficient and the phase velocity still remains unclear. Therefore, in this study, a small amount of salt is added to the suspensions to investigate the effect of electrostatic interactions on the attenuation and velocity.

2. Experiments and Results

Standard silica and PDVB microparticles purchased from Sekisui Chemical Co. were used in this study. The aqueous suspension of the particle was injected into a polystyrene disposal cell with the dimension 10 x 10 x 40 mm^3. For the PDVB suspensions, 0.3 wt% of sodium dodecylsulfate was added as a surfactant. As for the silica particles, the suspension was made with different sodium chloride concentrations to control the electrostatic interactions (thickness of the electrostatic double layer) between the silica particles. Particle size distribution was calibrated by a scanning electron microscope prior to the ultrasound studies.

Fig. 1 shows a schematic representation of the ultrasound spectroscopy. An ultrasound pulse was generated using a broadband pulser/receiver connected to a longitudinal plane wave transducer. Various water-immersion sensors having different nominal frequencies were employed to examine

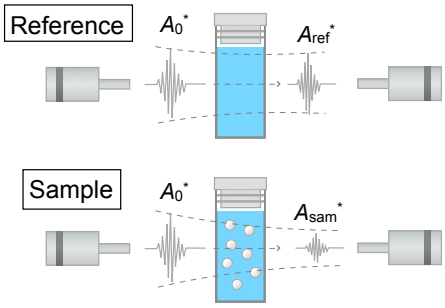

Fig. 1 Schematic representation of the experimental setup for ultrasound spectroscopy.

the frequency dependence of the sound velocity and attenuation coefficient of the materials. The transmitted pulse was received by another transducer, followed by amplification by the receiver. The signal was recorded by a high-speed digitizer with the sampling rate 1G samples/sec. The pulse repetition time was set at 0.5 ms and the pulse was averaged over 5000 times to achieve good statistics. The sample was set in a homemade thermostat bath regulated at 25 ± 0.01°C.

Fig. 2 (a) shows the frequency dependence of the ultrasound attenuation and phase velocity obtained for the PDVB particles with the average diameter d = 10 μm. The particle concentration was varied in range 0.3 - 10 wt%. In order to clarify the particle concentration dependence of the acoustic properties, the data extracted at a fixed frequency f = 27 MHz was shown in **Fig. 2 (b)**. The solid lines indicate the theoretical prediction by the ECAH theory[2, 3)] with the Waterman-Truell dispersion relation[4)], and agree well with our experimental data.

Fig. 3 (a) shows the frequency dependence of the ultrasound attenuation and phase velocity obtained for the silica particles with d = 10 μm. The particle concentration was fixed to be 5 wt% and the sodium chloride concentration, C_s, was varied in range 0 - 10 mM. As seen from the figure, the attenuation coefficient at a fixed concentration

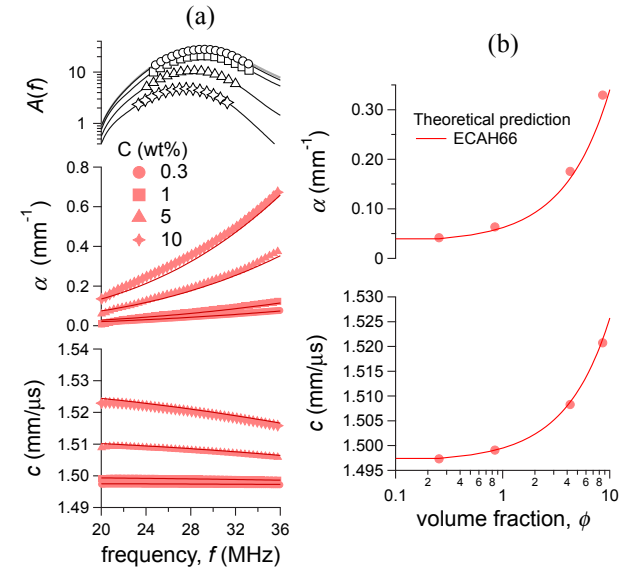

Fig. 2 Experimental results and theoretical predictions probed by ultrasound measurements. (a) Frequency dependences of the attenuation coefficient and sound velocity obtained for PDVB particles with d =10 μm. (b) Volume fraction dependence of α and c at 27 MHz.

increased with the salt concentration, followed by level-off at C_s = 4 mM. In **Fig. 3(b)**, the attenuation coefficient as a function of the salt concentration was depicted at a fixed frequency f = 26 MHz. We also carried out dynamic ultrasound scattering (DSS) experiment in order to probe the sedimentation dynamics of the silica suspensions.

In DSS, the variance of the settling velocity

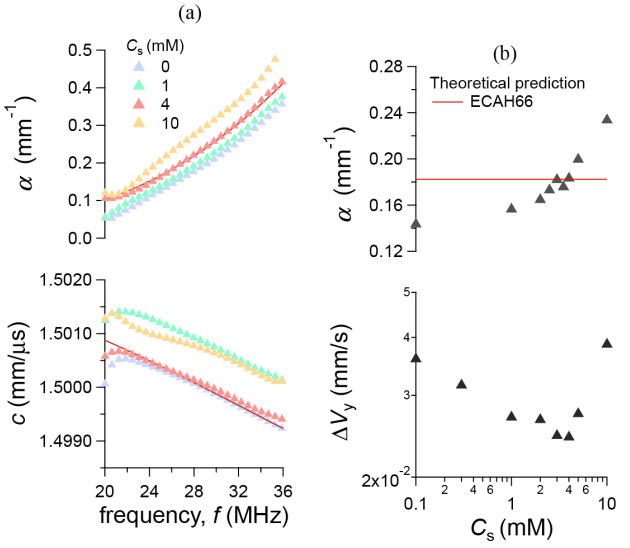

Fig. 3 Experimental results and theoretical predictions probed by ultrasound measurements. (a) Frequency dependences of the attenuation coefficient and sound velocity obtained for Silica particles with d =10 μm, C = 5 wt%. (b) Salt concentration dependence of α and ΔV_y at 26 MHz.

was investigated along the horizontal direction. Since the beam direction is perpendicular to that of the vertical sedimentation, the average velocity component is zero, resulting in the direct evaluation of the velocity flucutuations ΔV_y. The velocity fluctuations are known to be dependent on the long-range hydrodynamic interactions at the low volume fraction.[5] However, as the particle size becomes larger, the electrostatic interactions play more important role in the sedimentation fluctuations.[6] Such an interaction is also known to be screened out by addition of a small amount of salt, and it could be a good indication of the suppression of the charge effects on the acoustic properties.

As seen in the **Fig. 3(b)**, ΔV_y decreased with the salt concentration, followed by upturn due to formation of aggregates. At C_s = 3 - 4 mM, the ΔV_y showed minima. Since a plateau in attenuation coefficient was found at 4 mM in **Fig. 3(b)**, the frequnecy dependences of the attenuation and the phase velocity were fitted by the ECAH theory. Although the obtained parameters were more or less similar to that reported in the litetures[7], best fitting was achieved by choosing three times larger thermal expansion coefficient.

3. Conclusions

It was found that the salt concentration also has a large effect on the acoustic properties for the charged particles and the DSS technique could be employed as a supplementary technique to evaluate the particle size and the elastic properties of charged silica micro-particles.

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