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Verification of longitudinal and shear multiple scattering models in concentrated suspensions of silica nanoparticles

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1. Introduction

The ultrasonic spectroscopy (US) method is an effective method for analyzing the physical properties of liquid droplets and solid particles dispersed in liquid. Ultrasonic waves transmitted through a particle suspension and a reference were compared to evaluate the attenuation coefficient and the phase velocity. By analyzing the frequency spectra of magnitude and phase of the signals using ultrasonic scattering theories, the physical properties of the particle suspension can be evaluated as a function of frequency. The ultrasonic scattering theory used in the analysis has developed over a long history. Typical examples are the ECAH theory proposed by Epstein, Carhart, Allegra, and Hawley [1,2]. It is a theory of single scattering that considers the scattering due to heat and viscosity and is valid for sufficiently dilute dispersions. In other words, it does not hold for finite concentrations because it neglects wave interference between particles. This is because when the suspension becomes concentrated, multiple scattering occurs, where waves scattered by one particle are re-scattered by other particles. Therefore, various multiple scattering theories were proposed by Foldy, Waterman-Truell, Lloyd-Berry[3].

However, when ultrasonic waves are incident on a particle, some of the longitudinal waves are converted to shear or thermal waves by mode conversion. These attenuate in a few hundred nm, but as the suspension becomes concentrated and the distance between particles becomes smaller, the effects of the shear and thermal waves become non-negligible. The theory of multiple scattering, which includes viscous and thermal effects, was developed by Luppé, Conoir, and Norris (LCN)[4]. In fact, it has been confirmed that the longitudinal attenuation coefficient decreases after a certain concentration, because of a shear mode reconversion[5]. However, verification of the multi-mode multiple scattering theory for nano particle below 100 nm has not been conducted.

In this study, measurements were performed using silica particles of various sizes, including nanoparticles below 100 nm to validate the multi-mode multiple scattering models.

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2. Experiment

2.1 Sample

Silica particles (Snowtex series) manufactured by Nissan Chemical Co., Ltd., Japan were used as a model particle. The nominal particle sizes are 12, 100, and 450 nm. The suspensions were diluted with distilled water and purified using a 200 nm membrane filter. The densities of these particles were determined by density matching method using sodium metatungstate solution and turned out to be 2.040 g/cm^3, 2.159 g/cm^3, 2.168 g/cm^3 respectively.

2.2 Ultrasonic Spectroscopy

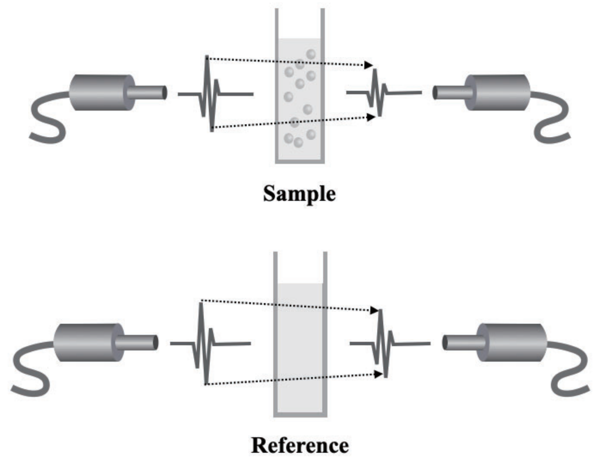

Fig. 1 Schematic representation of the experimental setup for ultrasonic transmission spectroscopy.

A broadband pulser receiver was connected to a longitudinal plane wave transducer to generate ultrasonic pulses. A schematic of the equipment is shown in **Fig. 1**. The transducer had a nominal center frequency of 20 MHz and was placed face to face in a thermostatic water bath controlled at 25 ± 0.005°C, with the sample cell placed between them.

A disposable plastic cell approximately 1 mm thick, with dimensions of 10×10×40 mm^3 was employed. Alignment of two transducers was carefully performed using a homemade stainless steel stage equipped with a microstage. To minimize the effect of edge waves, the distance between transducers was adjusted to the optimal distance, and spike waves of approximately –300 V were applied to the transducer. The signal was received by a high-speed digitizer board, and the waveform was Fourier transformed using a computer. The waves transmitted through the sample were compared with those transmitted through the reference, and the frequency dependences of the attenuation coefficient and the phase sound velocity were calculated from the amplitude ratio and phase difference.

3. Results

Fig. 2 shows the concentration dependence of the attenuation coefficient α and the phase sound velocity c at 20 MHz obtained by the US method.

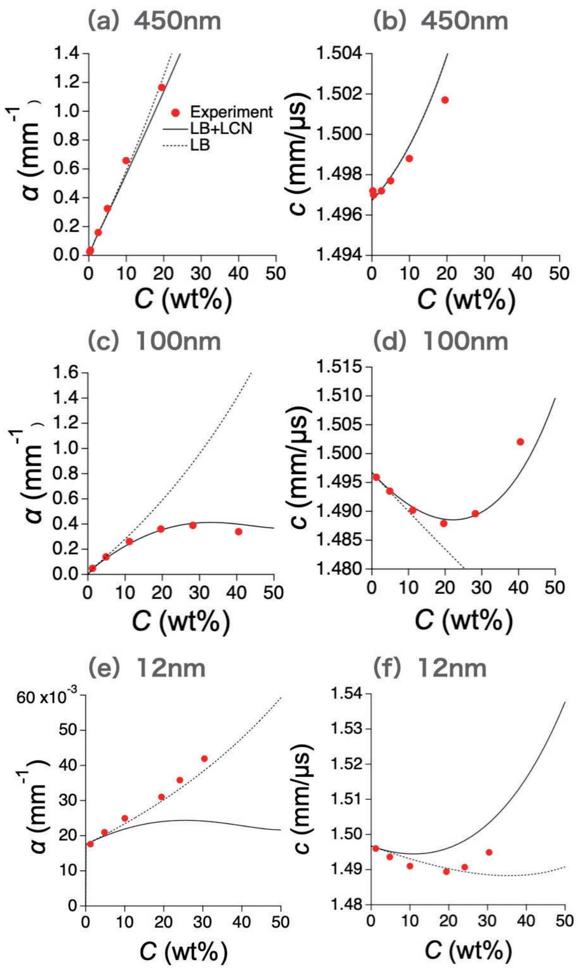

Fig. 2 The particle concentration dependences of the attenuation coefficient and the phase sound velocity obtained for the silica particles with different particle diameters.

The dotted curve represents the theoretical calculation considering only longitudinal multiple scattering proposed by Lloyd-Berry (LB), and the solid curve is the theoretical calculation with the multi-mode multiple scattering theory proposed by Luppé and Conoir, Norris (LCN)[4]. From Fig. 2(a) and (b), both theories reproduced the experimental data for the 450 nm silica particle, suggesting that the effect of shear mode reconversion is unimportant at this particle size. However, as shown in Fig. 2 (c) and (d), it was found that when the particle size approaches 100 nm, the theory cannot be reproduced without shear reconversion effect, i.e., multi-mode multiple scattering theory is required to reproduce the experimental data as pointed out by Forrester et al.[5]. As the particle size becomes comparable with the shear penetration depth and the concentration of suspension becomes higher, the shear waves do not completely attenuate and rather enter other particles, and after a certain concentration, the attenuation decreases, and the phase sound velocity begins to increase.

As shown in Fig. 2(e) and (f), however, none of the theory including the multi-mode multiple scattering theory reproduced the attenuation data for the silica particle with the particle diameter 12 nm. This may be due to overestimation of the effect of shear-mode reconversion. A recent theoretical paper[6] discussed the effect of shear-mode reconversion at the long wavelength regime where role of a coherent shear wave and an incoherent transverse wave were reexamined to take account of multi-mode multiple scattering properly. Further correction of this effect may improve the theoretical reproduction of the attenuation and phase velocity spectra for the nanoparticles.

4. Conclusions

For the silica particles in suspension measured by the US method, it was found that the longitudinal and multi-mode multiple scattering theory could reproduce the acoustical spectra of microparticles up to submicron-scale, but the analysis of attenuation coefficient and phase velocity obtained for nanoparticles required further development for the complete understanding of the multi-mode multiple scattering from nanoparticles.

References

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