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Ultrasound Scattering Studies on Pickering Emulsion

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1. Introduction

Ultrasound spectroscopy (US) is a non-contacting and nondestructive method, enabling us to monitor acoustic properties of materials. In the previous work[1)], we studied rigid and core-shell particle suspensions. Subsequently, US was applied to monitor a reaction process of hollow silica particles[2)]. In this study, effects of nanometer-sized silica particles on Pickering emulsion were investigated.

Pickering emulsion was known as an emulsion stabilized by localization of colloid particles at the oil/water interface[3)]. The Pickering emulsion was believed to be more stable than conventional emulsions with surfactants due to the higher adsorption energy of particles. Such a structure of emulsion coated by solid particles may be regarded as a model core-shell system for ultrasound study. Therefore, in this study, the ultrasound analysis for Pickering emulsion was carried out to investigate the structure and the stability of emulsions.

2. Experiments and Results

Silica particles (SP) were synthesized by the so-called Stöber method[5)], followed by surface modification using dimethyldiethoxysilane (DMDES) according to the procedures reported in the literature[6)]. First, DMDES was hydrolysized in a hydrochloric acid solution for 15 minutes before adding into the silica suspension in order to ensure the hydrolysis of the alkoxide. The obtained particles showed monodisperse size distribution as shown in **Fig. 1a**. The mean diameter was estimated to be 254 ± 9 nm by FE-SEM. After modification, the paticle size increased about 7 nm because of the DMDES chain growth on the silica surfaces. The reaction yield of the surface modification was characterized by TGA (Thermogravimetric analysis) as shown in **Fig. 1b** where the derivative of the weight loss was ploted as a funciton of temperature. The degradation temperature of organic part was found to be 350ºC. It is noted that the dispersability of the silica particles in hexadecane was better than in water showing that particles could be localized in the hexadecane phase rather than the water phase. Hence, an W/O emulsion would be formed using this stabilizer[7)].

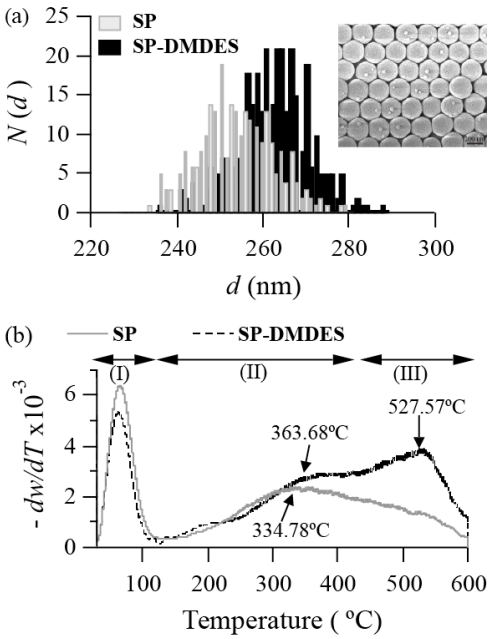

Fig. 1 (a) Size distribution of hydrophobic silica particles and (b) TGA results with and without surface modification.

The particles were dispersed in *n*-hexadecane. Then, Pickering emulsion was made using a homogenizer and SPG membrane with 2wt% of hydrophobically modified silica particles as a stabilizer. The formation of W/O type emulsion was confirmed by dropping the emulsion into a hexadecane solution as shown in **Fig. 2.**

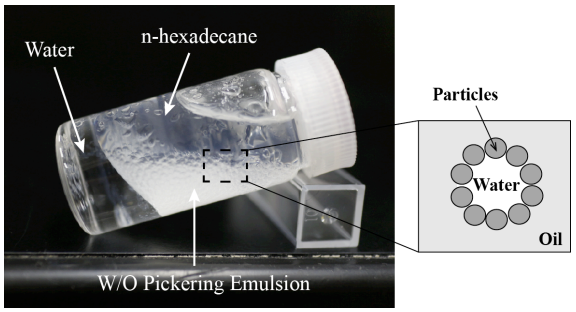

Fig. 2 Pickering emulsion stabilized by hydrophobic SP dispersed well in oil phase

Fig. 3 shows the acoustic properties of SPs. The ECAH theory[8)] was used to evaluate the attenuation coefficient and the phase velocity.

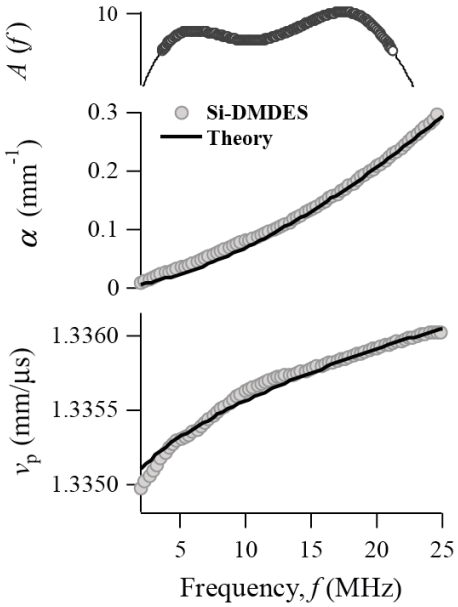

Fig. 3. Attenuation coefficient and phase velocity of hydrophobic silica particles fitted by the ECAH theory.

Because the particles size were in a nanometer range, the thermal effect played an important role in the acoustic scattering. By taking into account the effect, the experimental data was reproduced by the theoretical calculation.

Once the fitting parametters of SP were determined, they were used for the calculation of the attenuation coefficient and the phase velocity of the Pickering emulsion as shown in **Fig. 5**. Here, two scattering-function models, (1) Anderson[9] for liquid droplets and (2) Goodman[10] for core-shell particles were employed to reveal the structure of emulsion. Because the scattering analysis is so sensitive to the presence of the large particles, which disappear easily due to sedimentaion, a lognormal function was built to take the main component below 30 μm (**Fig. 4**). As shown in **Fig. 5**, the attenuation coefficeint and the phase velocity seemed to be nicely reproduced by the core-shell mode of Goodman. From the anlysis, it was found that the water droplet was sucessfully surrounded by the silica particles to form the Pickering emulsion.

3. Conclusions

Pickering emulsions were successfully made and stabilized by hydrophobic silica nanoparticles. The presence of the core-shell structures was confirmed by the anlysis of ultrasound scattering. This could be utilized to investigate the stability of emulsion as well as its structure having shell part to apply to drug delivery system, chemical reaction, and evaporation of solvent.

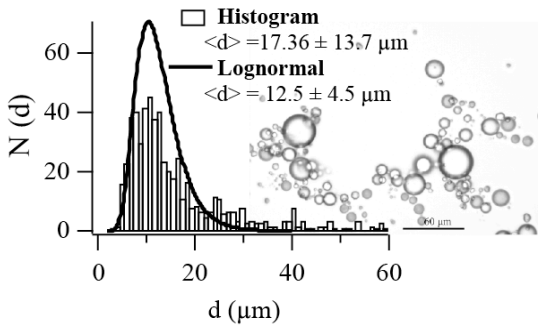

Fig. 4. Size distribution of Pickering emulsion obtained by a microscope; Histogram (bars) and fitted-lognormal distribution (solid line)

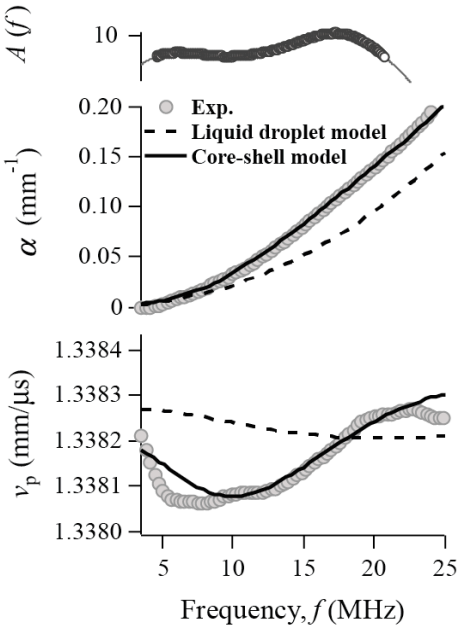

Fig. 5. Comparison of phase velocity and attenuation of Pickering emulsion with two theorical models: (1) Anderson and (2) Goodman

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