

Electrophoretic Mobility of Microparticles in Concentrated Suspensions Probed by Dynamic Ultrasound Scattering Techniques

動的超音波散乱法による濃厚系微粒子懸濁液の電気泳動

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1. Introduction

Colloidal suspensions are utilized in various fields, such as cosmetics, ink, food and medical applications. Among various considerations about the functionality and properties, the colloidal stability is considered to be an important factor to control the quality of products.

So far, we have developed ultrasound scattering techniques called dynamic ultrasound scattering (DSS)¹ and ultrasonic spectroscopy (US)² in order to evaluate the size and elasticity of particle in liquid in range several tens of nanometers to several tens of micrometers. In addition to these standard technologies, a novel technique called electrophoretic ultrasound scattering (ESS) technique is introduced here to evaluate the surface properties of particle, such as the electrophoretic mobility and zeta potential.

Such information is considered to be an important property to produce particle suspensions since the repulsive forces associated with the electrostatic interaction lead to homogeneous dispersion of particle in liquid. Therefore, electrophoretic mobility or zeta potential can be a measure of the colloidal stability. The electrophoretic light scattering (ELS) technique has been utilized to evaluate the zeta potential. However ELS requires sufficient dilution to allow transmission of light source particularly for the concentrated and/or non-transparent suspension. In the present study, the ESS technique, which is an acoustic analog of ELS is employed to evaluate the zeta potential without dilution of concentrated suspensions. In this study, the dynamics of micron and submicron sized particles are examined to understand the dependence of the electrophoretic mobility on the ionic strength, pH, and particle concentration of silica and mullite particles.

2. Materials and Samples

Silica (nominal average diameter $d = 3 \mu\text{m}$ and 500 nm) and mullite ($d = 1.63 \mu\text{m}$) particles were purchased from Sekisui Chemical, Nippon Shokubai

and Kyoritu Material, Japan. A longitudinal wave pulse was generated by a negative spike pulser equipped with a 20MHz water-immersion transducer to irradiate the sample with ultrasound. The scattered waves were recorded by a high-speed digitizer. The electric field was applied to the sample cell in order to induce electrophoresis of the particles.

3. Experiments and Results

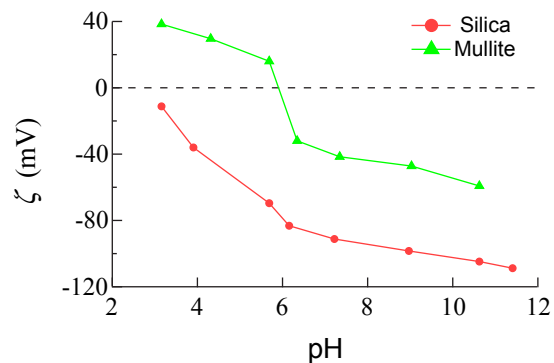


Figure 1. The pH dependence of the zeta potential of the silica and mullite micron-sized particles.

First, the ESS experiments were carried out for the micron-sized particles because of the large signal-to-noise ratio of the scattered signal. Figure 1 shows the pH dependence of the zeta potential obtained for the silica and mullite microparticles with the concentration $c = 0.5\text{wt}\%$. Since the silica particle contains negative charges above $\text{pH} = 3$, all the zeta-potential exhibited negative values. The concentration of potassium chloride was fixed to be 1 mM. The magnitude of the electrophoretic mobility and the corresponding zeta potential increased with pH. The isoelectric point (IEP) of the silica particle was evaluated to be $\text{pH} = 2.9$. These data showed good agreement with the values reported by Xu et al.³ On the other hand, the mullite particles showed either positive or negative potential depending on pH. Since mullite particle was composed of aluminum oxide and silica particle, the IEP of mullite particle was about $\text{pH} = 6$. For $\text{pH} > 6$, the magnitude of zeta potential increased with

pH, similar to the case of the silica particles. On the other hand, for $\text{pH} < 6$, the electrophoretic mobility showed the positive mobility (zeta potential) opposite to the case of the larger pH. The IEP was found to be $\text{pH} = 5.8$ for the mullite particles.

Figure 2 shows the relationship between the applied voltage and the electrophoretic mobility. The sign of the electrophoretic mobility is defined based on the direction of the positive electrode. Therefore, the mobility of the mullite particle is positive for $\text{pH} < 6$, and negative for $\text{pH} > 6$. The relationship between the electrophoretic mobility and the applied electric field was clearly demonstrated in the figure.

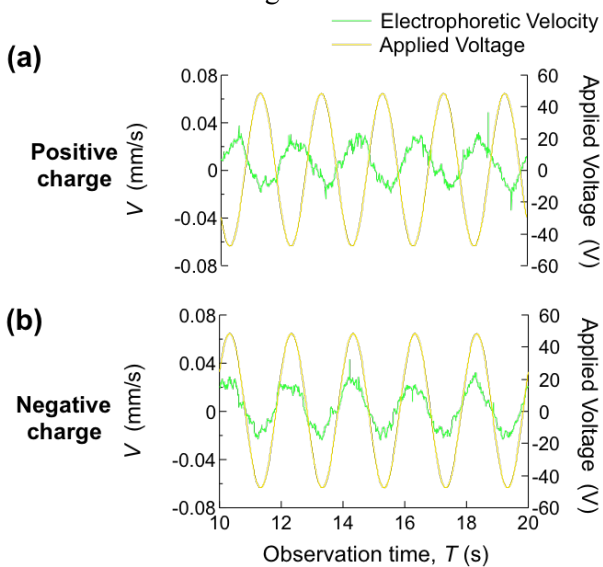


Figure 2. The relationship between the applied voltage and the electrophoretic mobility (a) $\text{pH} = 4.30$ (b) $\text{pH} = 6.33$.

Second, we tried to evaluate the pH dependence of the zeta potential for smaller silica particles, $d = 500$ nm with the particle concentration $c = 5\text{wt}\%$. Figure 3 shows the pH dependence of the zeta potential. The IEP was estimated to be $\text{pH} = 1.92$. The difference of IEP between Silica $3 \mu\text{m}$ and 500 nm may be ascribed to the salt concentration.

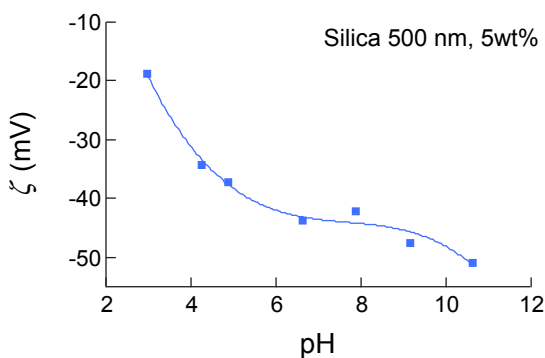


Figure 3. The pH dependence of the zeta potential of sub-micron silica particles.

The result also showed good agreement with the literature reported by Franks et al.⁴ in terms of the pH dependence. Figure 4 shows the image of scattering intensity in the suspension. The submicron particles showed extremely low scattering intensities compared with the particles with $d = 3 \mu\text{m}$. As shown in Figure 4 (b), when submicron particles have sufficient amount of charge to disperse without formation of aggregates, the scattering intensity must be weak and should not exhibit significant scattering. However, the strong scattering was observed at $\text{pH} = 2.98$. This finding is presumably attributed to the existence of aggregates in the suspension.

The existence of aggregates was consistent with the result in figure 3. Namely, the silica suspension contains the weak repulsive force at $\text{pH} = 2.98$. In summary, the ESS method could allow us to investigate not only the zeta potential but also the stability of suspension.

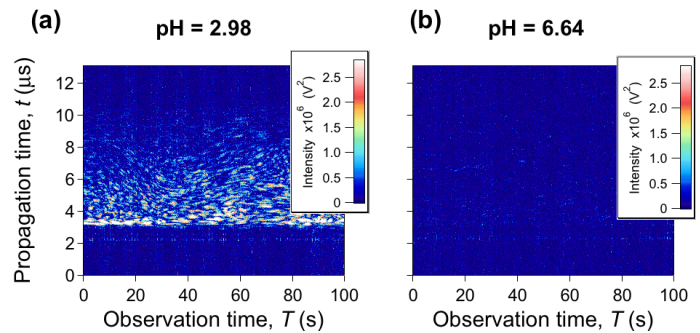


Figure 4. The image of the scattering intensity (a) $\text{pH} = 2.98$ and (b) 6.64 .

4. Conclusion

We showed that the potential of ESS to evaluate the stability of micron- and submicron-sized particle in highly turbid suspension through the measurements of zeta-potential. As the results, the pH dependence of the zeta potential showed a good agreement between the experiment and the literature for the silica suspensions. Now, the applicable particle size was extended to the submicron regime. Since the ESS technique employs the homodyne correlation function approach and the phase imaging method, discrimination of the aggregates from the primary particle is satisfactory carried out.

References

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