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Sedimentation Dynamics of Concentrated Silica Suspensions by Means of Dynamic Ultrasound Scattering Method

動的超音波散乱法による高濃度シリカ微粒子懸濁溶液の沈降ダイナミクス解析

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1. Introduction

Sedimentation field of microparticle suspensions is highly heterogeneous. The heterogeneities are origingated from the long-ranged hydrodynamic intereactions which induce cooperative domain structures consisting of microparticles having the same velocity within a blob. In our previous study, we reported that the velocity fluctuations of charged partciles are larger than the neutral systems and the electrostatic interactions could be suppressed by addition of a small amount of salt. Although the velocity flucutuations of micron-sized particles can be employed as a signature of the stability of particle suspensions, the concentration dependence, particulary at the high concentration regime is still uncovered. In this study, we examined the velocity fluctuations of settling charged particles over 30 vol% to investigate the role of electrostatic intereaction on the dynamics of charged particles at fairly high concentration regime.

2. Theory

As an ultrasound pulse propagates through a cell containing a suspension of microspheres, four reflected echoes A1, A2, A3, A4 from the cell walls are observed as shown in **Fig. 1**. If there are noticeable scattering contributions from the microspheres, the complicated scattering patterns could be observed between A2 and A3 as well. The pulse wave ψ for the scattering component may be written as:

$$\psi(t) = A(t)\cos\left[2\pi f_C t + \phi(t)\right] \tag{1}$$

where t is the field-time, f_c is the central frequency, A and Φ are respectively the amplitude and phase of the temporal pulse. In the case of the backscattering geometry, t contains the spatial information of the scatters along the beam direction enabling us to obtain the information on the location of the particles as indicated by the arrow A in **Fig. 1**. The time-evolution of the pulse can be visualized as an image of the sound field by successive recording of pulses at an interval ΔT(～ms). As the results, we obtain the time fluctuations of the scattered signals along the axis

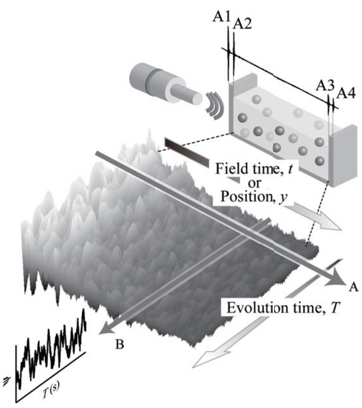

Fig. 1 Schematic illustration of DSS setup.

of the evolution-time, T, as indicated by the arrow B. The characteristic time of the fluctuations may be evaluated by the time correlation function $g_y^{(1)}(\tau)$ defined by:

$$g_y^{(1)}(\tau) = \exp\left(-\frac{1}{2}q^2\left\langle \delta V_y^2 \right\rangle \tau^2\right) \tag{2}$$

3. Experimental

3.1 Samples

Monodisperse silica microspheres with the particle diameters, 3 – 10 μm (purchased from Sekisui Chemical Co. Ltd) were used. Two types of suspensions were prepared: (1) Silica particles in distilled water without any surfactant and (2) silica particles in 1 mM sodium chloride aqueous solution. Disposable polystyrene rectangular vessels with the dimension 10 x 10 x 40 mm^3 and the wall thickness 1 mm were used as the sample cells.

3.2 Apparatus

Negative impulse emitted from a pulser/receiver (iSL Pulser) was transferred to a 5 or 16

MHz-longitudinal plane wave transducer (B5K6I or 25C6I respectively) immersed in a water bath to generate broadband ultrasound pulses. The same transducers received the reflected or scattered ultrasound waves. The obtained signals were then amplified by the receiver, followed by successive recording with a 14bit high-speed digitizer (Compuscope CS14200) at the sampling rate 200Ms/s.

4. Results and discussions

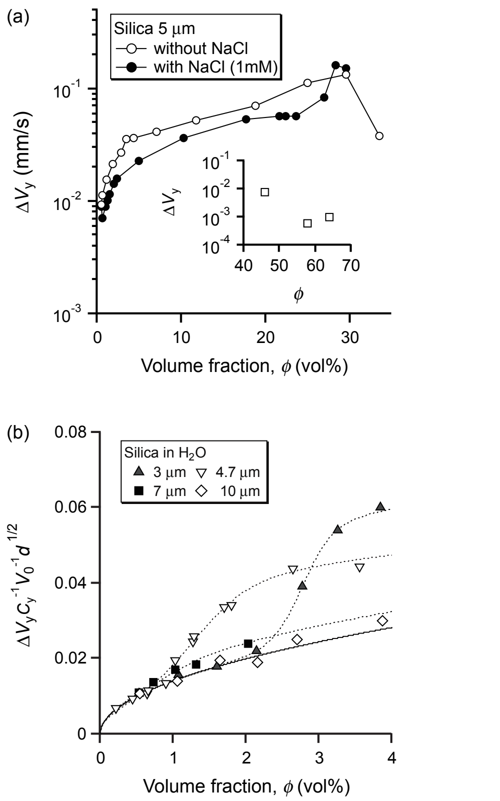

Fig. 2 (a) the volume-fraction dependence of the velocity fluctuations. (b) similar ϕ dependence reported previously.

Fig. 2(a) shows the volume-fraction dependences of the velocity fluctuations observed for the silica particles with d = 5 μm dispersed in water (open circle) and in 1 mM sodium chloride aqueous solution (solid circle) where the standard deviation of the horizontal velocity is abbreviated as $\langle \delta V_y^2 \rangle^{1/2} \equiv \Delta V_y$.

For ϕ < 25 vol%, the velocity fluctuations of silica particles in water were larger than that with salt. As we reported in the previous work, velocity fluctuations followed the Caflisch-Luke model at the low concentrations and the effect of charge became noticeable at the higher concentration as shown in Fig 2(b). As the particle size becomes larger, the inflection point shifted to the lower volume fractions. Therefore the velocity fluctuations for silica particle (d = 5 μm) without salt were larger than that with salt in our experimental condition.

While Caflisch-Luke theory predicted that the velocity fluctuations become higher as volume-fraction increased, reduction of the fluctuations for charged system was confirmed for the first time. On the other hand, in the presnece of salt, an upturn of the velocity fluctuations was observed around at ϕ = 30 vol%. Since the mobility of particle should be highly restricted, this may be attributed to the rapid exchange of the occupying space of particles to minizing the effects of intermolecular potential and electronic charges. Furthermore, although it is not shown here, we also confirm the frequency-dependence of the velocity fluctuations by frequency-domain DSS.[2)] In the light of this results, there are temporal and hierarchial structures coresponding to the length scale of ultrasound order.

5. Conclusions

Sedimentation dynamics of the silica suspensions having particle diameters 3 - 10 μm was investigated by means of the DSS method. The velocity fluctuations of the silica suspensions with and without salt both increased with the volume fraction, while subsequent decrease was only found for the suspension without salt. Although further investigation, particularly for the suspension with salt, is required to fully understand the dynamics of charged micron-sized particles, this study revealed the volume fraction dependence of the velocity fluctuations for micron-sized silica particles over 30 vol%.

While the effect of electrostatic interactions are significant for the system without salt over the wide range of concentrations, subsequent reduction of the velocity fluctuations due to structural restriction was confirmed as often observed for neutral systems. Although the velocity fluctuations could be highly suppressed by addition of salt, they exceeded the value of suspensions without salt at above 25%. This could be explained by formation of larger aggregates due to presence of salt in high concentration of unstable particles.

References

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