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Anomalous Electrostatic Interactions on the Velocity Fluctuations of Settling Microspheres

沈降マイクロ粒子の速度揺らぎに及ぼす特異的な静電的相互作用に関する研究

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1. Introduction

We have developed a novel particle-sizing technique called dynamic ultrasound scattering (DSS) method. This technique enables us to mesure the sedimentation velocity of micron-sized particles dispersed in a suspension via the time evolution of the scattering amplitude and phase of ultrasound waves.

Because of the hydrodynamic interactions, the sedimentation field is no longer uniform, resulting in the spatio-temporal distribution of the sedimentation velocities. So far, the mean sedimentation velocity was used to evaluate the particle diamater via the balance equation of buoyancy-friction relation. We also focused on the variance of the velocities, as a measure of velocity fluctuations, in order to briefly extract the size and interactions of the particles. The origin of the fluctuations were believed to be attributed to the number fluctuation of the particles having the same settling velocites(so-called the cooperative phenomena). Besides the extensive studies on the long-ranged hydrodynamic intereactions, the effects of charge on the particles were ignored in the previous studies since the particle size was much larger than the thickness of the electric double layer.

In this study, the effect of electrostatic interaction on the dynamics of settling particles in suspensions will be explored by comparing the velocity fluctuations of the negatively charged sillca particles.

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:

$$\psi(t) = A(t)\cos[2\pi f_c t + \Phi(t)] \tag{1}$$

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

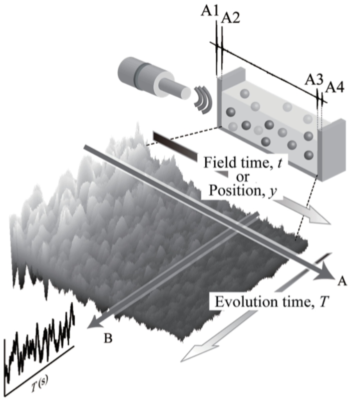

Fig. 1 Schematic illustration of DSS setup.

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

3. Experimental

3.1 Samples

Four kinds of monodisperse silica microspheres with the particle diameters, 3.05, 4.84, 7.02 or 10.1 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 20MHz-longitudinal plane wave transducer (B20K2I-M) immersed in a water bath to generate broadband ultrasound pulses. The same transducer 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

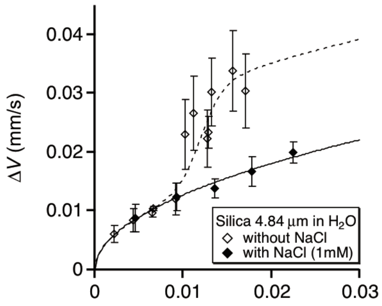

Fig. 2 Effects of salt on the volume-fraction dependence of the velocity fluctuations.

Fig. 2 shows the dependence of the velocity fluctuations obtained for the silica particles dispersed in water without any surfactant (open diamond) and in 1 mM sodium chloride aqueous solution (solid diamond) where the standard deviation of the horizontal velocity is abbreviated as $\langle \delta V_y^2 \rangle^{1/2} \equiv \Delta V$. In the case of the silica particles in pure water, the results were fairly different from those expected from the Caflisch-Luke(CL) theory [2] (solid line). While the velocity fluctuations followed the CL theory at relatively low volume fraction, i.e. $\phi < 1\%$, an anomalous upturn in the velocity fluctuations was observed at the higher volume fraction. On the other hand, addition of salts resulted in the reduction of the velocity fluctuations which were well reproduced by the CL theory. This behavior may be ascribed to the electrostatic interactions which can be eliminated by addition of small amount of salt to screen the electrostatic interactions. In order to investigate the effect of charge, the diameter dependence of the velocity fluctuations was further investigated.

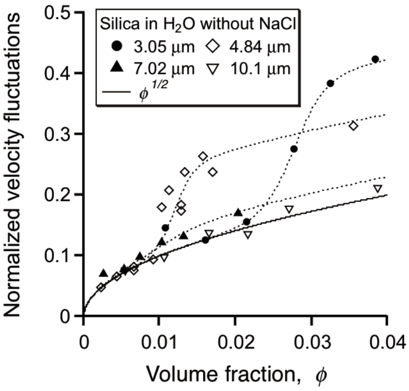

Fig. 3 The volume-fraction dependence on the normalized velocity fluctuations for different particle sizes.

Fig. 3 shows the volume-fraction dependence on the velocity fluctuations for the different particle sizes. The smallest particle showed the largest increase in the normalized velocity fluctuations at the higher volume fraction. The difference became smaller as the diameter increased, approaching the predicted curve of the CL theory(solid line). Since the charged particles have the sedimentation potentials which are caused by the separation of the counter ions from the settling particles against gravitational flow, the velocity fluctuations also affected by the interplay of the hydrodynamic and electrostatic interactions. As the origin of the velocity fluctuations are considered to be the number fluctuations of the particles in a blob, these results might be ascribed to the number fluctuation of the dipoles caused by the sedimentation potential.

5. Conclusion

Sedimentation dynamics of the suspensions of silica particles having different particle diameters ranging approximately from 3 to 10 µm was investigated by means of DSS. The dynamics significantly depended on the amount of charge. When the electrostatic interactions were screened out, the velocity fluctuations were well reproduced by the Caflisch-Luke theory. Furthermore, the diameter dependence of the interactions on the dynamics was observed. The present study demonstrated the importance of the electrostatic interactions which have been neglected in the previous studies.

References

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2. R. E. Caflisch and J. H. C. Luke: *Phys. Fluids* **28** (1985) 759-760.