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Dynamics of Suspended-Microspheres Probed by Scattering Phase of Ultrasound
超音波の散乱位相を利用した懸濁微粒子のダイナミクス解析

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1. Introduction

There has been great attention to polymer microspheres because of the versatile applications, such as inks, cosmetics, fillers, spacers, catalytic support, and standard calibration. In-situ monitoring the particle suspension is sometimes required to be observed as is. This is particularly important if the systematic quality control is required for the subsequent manufacture process.

In the previous study[1], we demonstrated that high frequency dynamic ultrasound scattering (DSS), an acoustic analogue of dynamic light scattering (DLS), allowed us to investigate the sedimentation dynamics of microspheres with the diameter ranging from several to several tens of micrometers. In addition to the potential of dynamics analysis in turbid solutions (where neither conventional optical velocimetry nor DLS does not work efficiently), there are further advantages on the fluid dynamics characterization. For example, when ultrasound is emitted from the top of the sample (z-direction), the sedimentation velocity can be probed by a backscattering setup, allowing the determination of the particle size via the Stokes relation. If settling is hindered by particle collision or other external force, the velocity fluctuations are simultaneously observed. On the other hand, the velocity fluctuations can be solely investigated from a horizontal setup (y-direction) because of the average component being zero. This enables us to investigate the long-range dynamic fluctuations originated from the size distribution and/or hydrodynamic interactions. While the average velocity of the sedimentation particles is well understood, its fluctuations are still of interests in the field of fluid dynamics. Spreading of the sedimentation front, screening of hydrodynamic interactions and hindered sedimentation are the examples. All these must be crucial for studying the velocity fluctuations observed by dynamic ultrasound scattering. Once the complicated hydrodynamics is elucidated, the technique may be utilized for characterization of soft matter such as surface modified micro beads, hollow spheres, metal-polymer hybrids, micro bubbles and so on.

Besides the fascinating amplitude analysis, scattering phase contains rich information inherent in ultrasound scattering[2]. Therefore, the motivation of this study is to develop a phase mode - dynamic ultrasound scattering technique, which enables us to investigate the sedimentation dynamics by the spatio-temporal image constructed by successive phase difference of the particles.

2. Experiments

Standard Latex microspheres with different diameter (3 ~ 45μm) were supplied by courtesy of Sekisui Chemical Co. Ltd. The particles were dispersed in an aqueous solution containing 0.2 % sodium dodecyl sulfate (SDS) to obtain a suspension with desired volume fraction in range $0.5 < \phi < 3$ %, followed by a brief immersion in a low power ultrasonic bath prior to DSS experiments in order to avoid aggregation. Disposal polystyrene rectangular vessels with the dimension 10 x 10 x 40 mm and the wall thickness 1 mm were used as the sample cells. The maximum scattering path was 10 mm for all the experiments. The ultrasonic transducer and the cell container were carefully aligned by using a custom-made stainless stage prior to the DSS experiments. SEM micrographs (Hitachi S-3000N) were also taken to verify the particle size and its distribution. A 25 mL Gay-Lussac pycnometer was used to measure the densities to three places of decimals. All the experiments were performed at 20.0 ± 0.05 °C.

Negative impulse emitted from a pulser/receiver (Olympus, model 5800PR) was transferred to a 20 MHz-longitudinal plane wave transducer (Olympus, model V317, Bandwidth 40 %, 0.25 inch in diameter) immersed in a water bath to generate broadband ultrasound pulse. The reflected or scattered ultrasound wave was received by the same transducer for the backscattering experiments. The sample-to-detector distance was 11 mm. The obtained signals were then amplified by the receiver, followed by successive recording with a 14 bit high-speed digitizer (GaGe, Compuscope CS14200) at the sampling rate 200 MS/s. The pulse repetition rate and the number of sampling points for successive pulse were tunable.
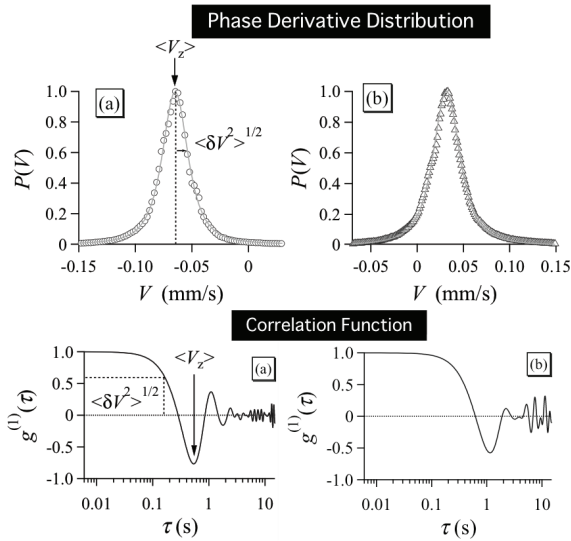
Fig.1   Velocity   distributions   and   normalized correlation functions of the particles showing (a) floatation and (b) sedimentation.

3. Results and Discussion

Fig.1 shows the velocity distributions (upper) and the normalized correlation functions (lower) of the microparticles. The details of the analysis methods are descibed elsewhere[1–2].   Since the densities of the microspheres with (a) the diameter $d = 32\mu m$ ($\rho = 1.18g/cm^3$) and (b) $d = 45\mu m$ ($\rho = 1.05g/cm^3$) were respectively higher and lower than the density of D_2O ($\rho = 1.10g/cm^3$), these could be the model systems for studying sedimentation and floatation. While the time correlation functions gave only the magnitude of the average velocity, the phase distribution functions successfully provided the velocity with the positive or negative sign, allowing us to investigate the direction of the particle motion.

Fig.2 demonstrates the quantitative agreement of the velocity fluctuations between the two methods for the different particle diamaters. Note that the solid and dotted lines show the theoretical prediction with size-dependent and -independent
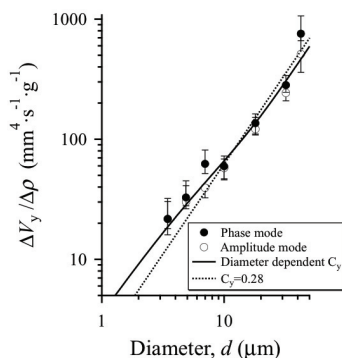

Fig.2   Velocity   fluctuations   of   the   particles evaluated by the scattered phase compared with those of the correlation function.
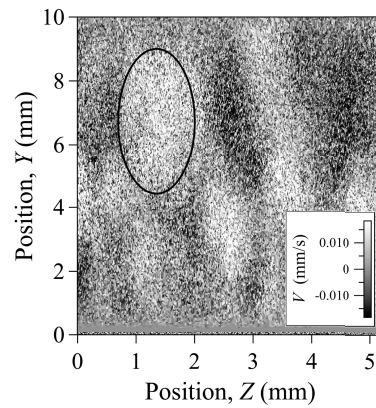

Fig.3   2D-velocity image.

coefficient C_y, where C_y is a coefficient appeared in a theory[3-4] and contains the information on the structure along the y-direction.

Since the signed velocity was sucessfully evaluated from the phase difference approach, the method was utilized to investigate the spatio-temporal field of the particle velocity. **Fig.3** shows the 2D-velocity image of the microshperes with $d = 10\mu m$ ($\phi = 2\%$).   The image suggested a collective particle motion driven by vortex formation. Althoug the vortex structure was reported earlier by particle image velocimentry experiments or computer simulations, the present method could be utilized for highly opaque samples without dilution of the samples. The spatial correlatin length obtained from the image analysis also revealed the decrease of the swirl size with increasing the concentration, indicating the possibility to carry out more versatile analysis to elucidate the complex fluid dynamics by ultrasound.

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