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# Dynamics of Carbon-black Suspension Probed by Dynamic Ultrasound Scattering Techniques
動的超音波散乱法によるカーボンブラック懸濁液のダイナミクス

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## 1. Introduction

Dynamic ultraSound Scattering (DSS) techniques are non-contacting and non-destructive analysis method for evaluation of dynamics of microparticles in suspension. With the aid of hydrodynamic theories, the relaxation time of the correlation function constructed by the time evolution of the scattered signal is converted into the hydrodynamic radius. With this technique, it is possible to get the particle motion, such as diffusive motion for nanometer-sized particles and settling motion for micrometer-sized particles.

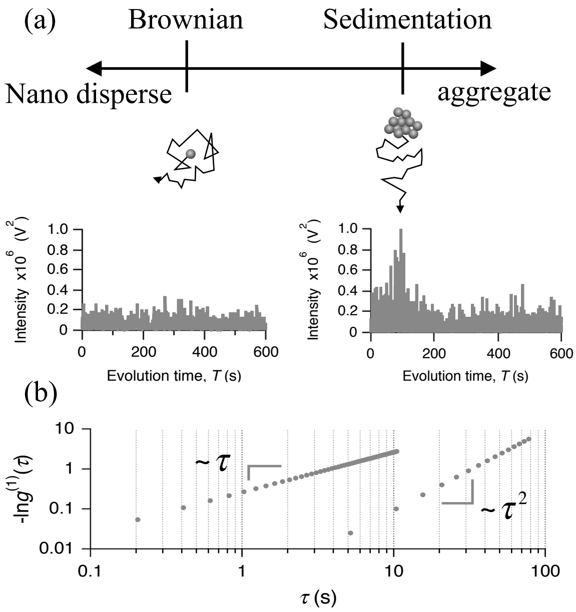


**Fig. 1** (a) a schematic diagram of the possible particle motion in nanometer to micrometer range. (b) double logarithmic plot of the time-correlation functions for diffusive (left) and settling particles (right).

As nano-particles form a larger aggregate in a micrometer range, sedimentation of the aggregates dominates the dynamics. If the nano-particles are stable, clear diffusive motion of the independent particle could be observed. **Fig. 1(a)** shows a schematic diagram of the possible particle motion in nanometer to micrometer range. While the scattering intensities of nano-particles are weak, that of micrometer size particles has a strong contribution. In addition, the DSS technique allows us to discriminate the type of particle motion by the analysis of time-correlation function as shown in **Fig. 1(b)**. In this study, this technique was utilized to investigate the stability of carbon black suspensions which have potential applications in fuel cells and particle-reinforced rubbers.

## 2. Experiments and Results

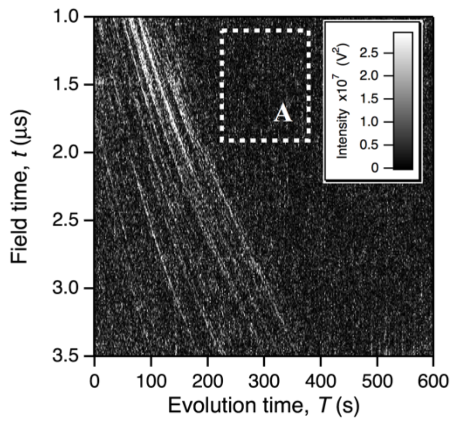


**Fig. 2** Image of the scattering intensity obtained for a suspension of carbon black in a mixture of water and 1-propanol without Nafion.

50% compressed acetylene carbon black (CB) was purchased from Sterm Chemical. The CB particles with the volume fraction $\varphi$=0.14% was dispersed in a solvent with or without Nafion as a stabilizer. Two types of Nafion solutions, DE521 and DE1021, were purchased from Wako Chemical.

For DE521, Nafion was dissolved in a mixed solvent, with the ratio Nafion: water: 1-propanol = 5: 45: 50. DE1021 is an aqueous solution containing 10% of Nafion without alcohol. Hereafter the suspension of CB dispersed in DE521 is regarded as an alcohol rich system, while that in DE1021 is called as a water rich system.

**Fig. 2** shows an image of scattered intensity as functions of the field time and observation time. The sample position, $x$, can be obtained by $x=ct/2$ where $c$ is the speed of sound. The figure shows the presence of extraordinary large aggregates showing sedimentation motion of the CB particles. The sedimentation velocity could be estimated by the gradient of line in **Fig. 2**. Subsequently, the sedimentation velocity was calculated and converted into the diameter of particle using the Stokes equation. The diameter was evaluated to be 4 μm. In addition to the noticeable aggregates in **Fig 2**, the particle motion could be statically analyzed by the time-correlation function approach for the region A as indicated in **Fig. 2**. The correlation function may be given by,

$$g^{(1)}(\tau) = \cos(q\langle V_z\rangle\tau)\exp\left(-\frac{1}{2}q^2\langle\delta V_z^2\rangle\tau^2\right) \quad (1)$$

for the settling particles, and

$$g^{(1)}(\tau) = \exp(-Dq^2\tau) \quad (2)$$

for the diffusive nano-particles where $q$ is the magnitude of the scattering vector, $\tau$ is the lag time, $\langle V_z\rangle$ and $\langle\delta V_z^2\rangle$ are respectively the average sedimentation velocity and the variance. In this region, the presence of smaller particles is expected. When the ultrasound experiments were carried out at the early stage of sedimentation, the undershoot in the correlation function was observed. This indicates the presence of large aggregates. Therefore, the average velocity and the variance were evaluated by fitting the data to eq (1). Using the Stokes equation, the particle size was found to be 3 μm. On the other hand, after 180 min, most of the micrometer sized particles settled down. Since the time correlation function showed a diffusive nature as shown in **Fig. 3(b)**, non-least squared curve fitting was carried out to evaluate the diffusion coefficient of the CBs in the suspension. After the trivial calculation using the Stokes-Einstein formula, the particle size $d$ = 450 nm was obtained for the CBs suspend in the DE521 solution. Such a clear diffusive motion could not be observed without the DE521 solution, suggesting the presence of Nation plays an important role to stabilize CB particles in alcohol.

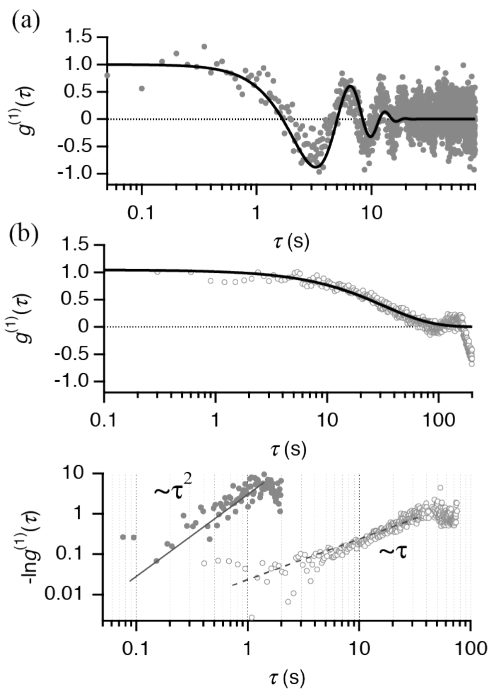


**Fig. 3** Correlation function observed from the direction along sedimentation (a) in the presence and (b) in the absence of aggregates.

## 3. Conclusions

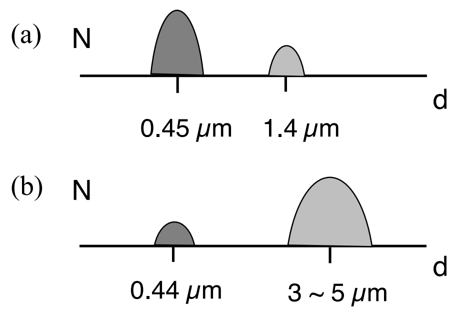


**Fig. 4** Schematic representation of carbon black suspension for DE521 (a) with and (b) without Nafion.

**Fig. 4** summarizes the information obtained in this study. Nafion was found to have a significant role to stabilize the carbon black (CB) and suppressed the formation of large aggregates. DSS could be used not only to evaluate the particle radius, but also as a potential tool to distinguish the type of particle motion, such as well dispersed nano-particles and micrometer-sized aggregates from the time-correlation function analysis.


**References**
1. K.Igarashi, et. al., J. Appl. Phys, **115** (2014) 203506-1-8
2. M.Kohyama, et. al., Macromolecules **42(3)** (2009) 752-759
3. M.Kohyama, et. al., Polym. J **40** (2008) 398-399