



# Thermal conductivity of suspension fluids of fine carbon particles: Influence of sedimentation and aggregation diameter



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## ABSTRACT

Nanoparticle suspension fluids, called nanofluids, have attracted much attention. It is not straightforward to predict effective thermal conductivity of the nanoparticle suspension fluids. One of that reasons is that aggregation may transform particle shape and diameter, and cause particle sedimentation in the fluid. In the present study, fine carbon particles of graphite and carbon black were mixed with surfactant-added water. The fine carbon particles were nanoscale in the primary particle diameter. Time variation of thermal conductivity of the fine-particle suspension fluids was evaluated with transient hot-wire method. In addition,  $\zeta$  potential of the fine particles, the aggregation diameter and degree of the sedimentation in the fluids were evaluated. In the present study, it turned out that thermal conductivity of the graphite particle suspension decreased as the particle sedimentation proceeded, and it was characterized by Hamilton-Crosser model. Moreover, we found that thermal conductivity of the carbon black suspension decreased with time due to sedimentation, and was lower than that of the graphite suspension. The present experimental results indicated that the difference between the graphite and carbon black suspensions arose from the functional groups formed on the particles surface that determined particle surface states and influenced the particle aggregation.

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## 1. Introduction

Nanoparticle suspension fluids, called nanofluids, have attracted much attention since Choi et al. [1] reported that the nanofluids could change their own physical properties from their base fluid. As for carbon-based materials, carbon nanotubes [2,3], fullerene [3], graphite [4,5], and carbon black [5] have been employed for suspending materials of nanofluids. Including those materials, heat conduction mechanism in the nanofluids [6,7] also have been investigated and discussed until now. It is not straightforward to predict effective thermal conductivity of the nanoparticle suspension fluids. One of that reasons is that particles may aggregate to transform shape and diameter of aggregation particle diameter and cause particle sedimentation in the fluid. Aggregation influence on the effective thermal conductivity has been not fully elucidated yet. Some previous papers reported that the aggregation enhanced the effective thermal conductivity of the suspension (e.g. [8]). One of the examples is that the magnetite nanoparticles formed a chainlike structure in a fluid under an influence of an external applied magnetic field [9]. Both natural and artificial aggregation are supposed to be the mechanism of the

thermal conductivity enhancement of the suspension. On the other hand, others reported that the aggregation enhanced sedimentation due to the increase in the aggregate size resulting in a decrease in the effective thermal conductivity of the suspension [10].

The aggregation-led enhancement of thermal conductivity depends on the aggregation diameter and structure. Adding surfactant into water of base fluid and adjusting pH prevent the particles in the suspension from aggregation [11–13]. In the present study, we investigated influences of aggregation and sedimentation on thermal conductivity of fine particles suspension by employing two different carbon-based particles of graphite and carbon black. They were employed since those particles were nanoscale in the primary particle diameter. Carbon black has some functional groups of carboxyl and hydroxyl groups on their surface. On the other hand, graphite has no such functional groups. That difference may cause a difference in particle wettability and aggregation. In the present study, a time variation of thermal conductivity of the fine-particle suspension was evaluated with transient hot-wire method. In addition,  $\zeta$  potential of the particles, the aggregation diameter and degree of the sedimentation in the suspensions were evaluated. Based on them, the present study aimed to investigate dominant factors to characterize the suspension thermal conductivity.

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## 2. Sample and characterization

### 2.1. Sample preparation

We employed a carbon black from Mitsubishi Chemical (Product number: MA100). Its nominal diameter is 24 nm. Besides, we employed graphite nanoparticles from Sigma Aldrich (Product number: 63310). Its nominal diameter is 100 nm. The graphite nanoparticles were produced by a laser method. In a previous study, Raman spectroscopy confirmed that the nanoparticles had a graphitic nature in its structure [14]. The graphite nanoparticles morphology observed by scanning electron microscopy was shown in Ref. [14].

To enhance the carbon particles suspension, we dissolved 2.5 wt% surfactant of polyvinylpyrrolidone (PVP) into a distilled water and added a few drops of ammonia solution or acetic acid so that each of those base fluids was at different pH if approximately 3.2, 6.6, and 9.2. Surfactants are able to make suspending particle surface hydrophilic and prevent the particle aggregation. In addition, some surfactants are able to change the electric charge on the particle surface, which determines repulsive electrostatic force and the particle aggregation. PVP is a non-ionic surfactant, and thus we expected that PVP has a weak capability to change the surface electric charge on the particles. Furthermore, surfactants accumulate on the particle surface and form a molecular layer. The surfactant layer causes a repulsive force between the surfactant-layered particles due to the steric structure of the layer. Especially when particles were nanoscale, the surfactant more significantly influences the particle dispersion stability [15]. PVP is a polymer solubilizer (see Fig. 1). In the case of carbon black suspension without PVP at particle weight volume of 3.0 wt%, most of the particles deposited on a container bottom after 10 min. However, in the case of carbon black suspension with PVP, the suspension maintained opaque even after 60 min. Based on this, we confirmed that PVP enhanced the particle dispersion in the suspension. We mixed a certain amount of sample particles with the base fluid at a particle weight fraction of 3.0 wt% and stirring the fluids to enhance the particle suspension for 30 min.

### 2.2. $\zeta$ Potential and aggregation diameter

We evaluated  $\zeta$  potential and aggregation diameter of the carbon black and graphite suspensions by means of  $\zeta$  potential analyzer (Zetasizer Nano ZS from Malvern). In measurements of  $\zeta$  potential, water suspension at the particle volume fraction of 0.5 vol%, and at 25 °C. Due to a limitation of the analyzer specification, the particle fraction was lower than that in measurements of thermal conductivity, and PVP was not added. Although, we expected that PVP depletion did not significantly influence the surface electric charge since PVP is a non-ionic surfactant. In addition, we

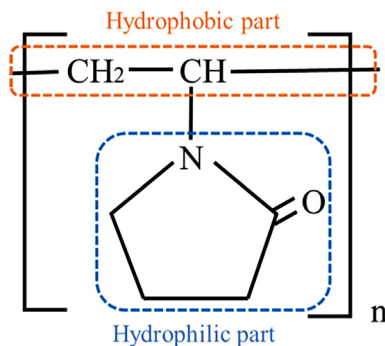


Fig. 1. Constitutional formula of PVP.

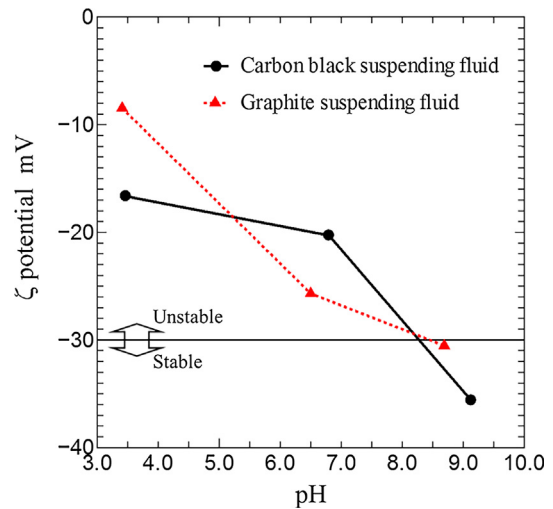


Fig. 2. Relation between  $\zeta$  potential and pH value.

expected that the surface electric charge did not significantly differ even at relatively dilute particle fraction. Fig. 2 shows a relation between  $\zeta$  potential and pH value in the case of carbon black and graphite suspensions. From this, it was found that both of the particles were negatively charged in the present range of pH. Furthermore, the negative charge increased as pH increased, which indicated that the repulsive force between the particles got stronger. When pH was approximately 9.3,  $\zeta$  potential was lower than 30 mV in both cases. The previous study [16] reported that particle aggregation was suppressed due to the repulsive electric force at greater/less than  $\pm 30$  mV. Based on this, at pH of approximately 9.3, the particle dispersion was stabilized in term of the electrostatic force.

In the measurement of particle diameter, back-scattering detection ( $\theta = 173^\circ$ ) was employed. This method accepts samples even at high particle fraction. Therefore, the sample suspensions described in Section 2.1 were tested. We measured the particle distributions with time variation at multiple times and averaged profiles were shown in Fig. 3. The measurements were performed four times every 24 h from the initial state of the suspensions, which was repeated six times for averaging. A viscosity of the base fluid was measured with a rheometer (MCR92, from Anton Paar) at 25 °C, which was employed for evaluating the particle diameters. Fig. 3 shows that the suspending particle diameter became smaller with time variation in both cases of carbon black and graphite. In the case of graphite, the highest peak was detected at around 2300–3600 nm. In the case of carbon black, the highest peak was detected at around 3000–3600 nm. They indicate that, in the suspensions, from the beginning the particles aggregated. Particle sedimentation is characterized by Stokes' law:

$$v_s = \frac{D_p^2(\rho_p - \rho_f)g}{18\mu}, \quad (1)$$

where  $v_s$  denotes terminal velocity of a particle in a fluid,  $D_p$  does particle diameter.  $\rho_p$  and  $\rho_f$  denote density of the particle and fluid, respectively.  $g$  denotes gravitational acceleration, and  $\mu$  does fluid viscosity. Based on Stokes' law, the sedimentation speed is accelerated by a factor of  $D_p^2$ . Larger particles deposited faster, and thus smaller particles stayed suspended in each of the fluid. From Fig. 3, particles of less than 10 nm diameter were detected after 24 h. That change was caused by that most of the relatively large particles deposited, and thus weaker scattering light became able to be detected.

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