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Experimental Investigation of Coupling Effects between Particle Size and Temperature on the Thermal Conductivity of Alumina Nanofluids

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Key Words: Nanofluids(나노유체), Thermal Conductivity(열전도도), Particle Size(입자크기), Temperature(온도)

Abstract

This study investigates the effects of nanoparticle size and temperature on the thermal conductivity enhancement of water-based alumina (Al_2O_3) nanofluids, using the centrifuging method and relative centrifugal forces of differing magnitude to produce nanofluids of three different particles without involving any dispersants or surfactants. We determined the coupling dependency in thermal conductivity enhancement relative to nanoparticle size and temperature of the alumina nanofluids and also experimentally showed that the effect of temperature on thermal conductivity is strongly dependent on nanoparticle size. Also, our experimental data presented that the effective medium theory models such as the Maxwell model and Hasselman and Johnson model are not sufficient to explain the thermal conductivity of nanofluids since they cannot account for the temperature- and size-dependent nature of water-based alumina nanofluids.

Nomenclature

a_K : Kapitza radius [m]
a_p : Radius of particle [m]
d : Mean particle diameter [m]
d_p : Particle diameter [m]
K_B : Boltzmann constant [J/K]
k_{BF} : Thermal conductivity of base fluid [W/m·K]
k_{NF} : Thermal conductivity of nanofluid [W/m·K]
k_p : Thermal conductivity of particle [W/m·K]
l_{BF} : Mean free path [m]
R_b : Kapitza resistance [Km^2/W]
T : Temperature [K]
V_{BR} : Brownian velocity [m/s]
ϕ : Volume fraction [%]
μ_{BF} : Dynamic viscosity of fluid [N·s/m^2]

1. Introduction

Considerable research on the thermal conduction in nanofluids, solid nanoparticles dispersed in liquids, has been conducted to investigate the main heat conduction mechanism up to now(1-13) since some experimental study on their thermal conductivity show nanofluids have high thermal conductivity exceeds the predictions of effective medium theory.(1-5, 7, 12) Mechanisms of the enhanced thermal conductivity in nanofluids are inconclusive and debated due to the inconsistencies in the reported thermal conductivity data from different research groups. For example, in the case of alumina-water nanofluids, several researchers have discovered that nanofluids have strongly temperature-dependent thermal conductivity(3-4), while other show there are no temperature dependency(6, 9), although experiments were performed with similar volume concentration (~4 vol.%). Nanoparticle size effects on the thermal conductivity of alumina-water

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nanofluids also have been investigated; however, there are also contradictory data(4, 7-10, 12). Moreover, the postulation that the effective medium theory can predict and explain the heat conduction in nanofluids has suggested(14-15). One of the suggestions comes from the molecular dynamics simulations[14] and another comes based on experimental data with order-of-magnitude analysisx(15). However, the experiments performed only as a function of volume fraction at room temperature and did not consider the variations of temperature and particle size(15). Therefore, more systematic experiments are needed to investigate the particle size and temperature effects on heat conduction in nanofluids. This can lead the development of the fundamental mechanisms for heat conduction in nanofluids.

In this paper, our systematic process involved using the centrifuging method and relative centrifugal forces to manufacture water-based alumina nanofluids with particles of three different sizes and without any dispersants or surfactants. Using these nanofluids, we further the knowledge of the thermal conductivity behaviors of nanofluids by reporting the effect of nanoparticle size and temperature on the thermal conductivity of water-based alumina nanofluids.

There are two main points in our experiments. First, to investigate the effects of particle size on thermal conductivity, we produced three nanofluids, each containing nanoparticles of a different size, using the centrifuging method and no dispersants or surfactants. The centrifuging method uses relative centrifugal force to eliminate large, aggregated nanoparticles and produce nanofluids with more uniform particle size. Second, in order to capture an accurate picture of the thermal conductivity of alumina nanofluids, we took measurements over a broad range of temperatures (10°C to 80°C), which is unusual in thermal conductivity measurements of water-based alumina nanofluids. The experimental results showed that 1) the effects of both size and temperature on the thermal conductivity can occur simultaneously and 2) the effect of temperature on thermal conductivity is strongly correlated to the effect of particle size. On comparing our data with the effective medium theory (EMT) models, we found that the EMT model cannot explain the results we obtained. Moreover, the experimental results were compared to the calculated Brownian velocity to evaluate the feasibility of the Brownian motion can be the main contribution factor which affects the thermal conductivity enhancement.

2. Experimental Methods

The alumina nanoparticles were purchased from Alfa Aesar (99.5% purity; nominal powder size is 40-50 nm) and distilled water was purchased from J.T. Baker. The centrifuging method of preparing our water-based alumina nanofluids with three different sizes consisted of the following process: First, dry alumina nanoparticles were dispersed in distilled water at room temperature. The volume fraction (vol.%) was set to 6 vol.%. The container with the 6 vol.% alumina nanofluids was immersed in the ultrasonic bath (40 kHz and 300W) for 5 hours to form a stable suspension(16). Half of the sonicated 6 vol.% alumina nanofluids were centrifuged with 7155 RCF (relative centrifugal force) for 30 minutes, and then the supernatant of centrifuged alumina nanofluids was decanted. This decanted alumina nanofluid is referred to as AN1 in this letter. The measured volume concentration of AN1 was 0.51 vol.%. The other half of the sonicated alumina nanofluids with 6 vol.% was centrifuged with 2,795 RCF for 30 minutes, and the supernatant of centrifuged alumina nanofluids was also decanted. Distilled water was added to this decanted nanofluid to adjust the volume fraction to 0.51 vol.%. This alumina nanofluid is referred to as AN2. The uncentrifuged 0.51 vol.% alumina nanofluid was also prepared in the ultrasonic bath (40 kHz, 300W) for 5 hours. This nanofluid is referred to as AN3. All three of alumina nanofluids were produced without any surfactants and dispersants. Fig. 1 shows the mean particle size of decanted alumina nanoparticles that remained in the supernatants, as a function of RCF when centrifugations were applied. Mean particle diameter was measured by the dynamic light-scattering (DLS) technique using

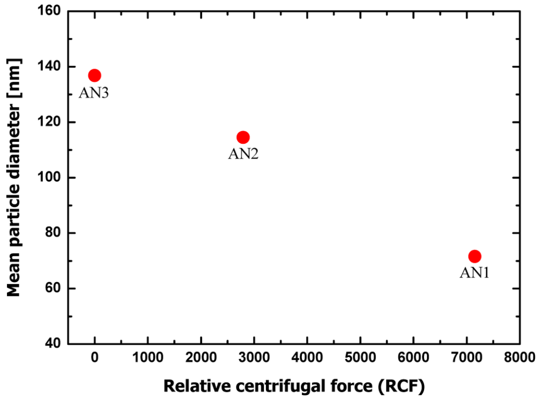

Fig. 1 Mean particle diameter of alumina nanoparticles suspended in water using the centrifuging method plotted as a function of applied relative centrifugal force (RCF). Mean particle diameter was measured by the dynamic light-scattering (DLS) method

the Zetasizer Nano S90 (Malvern Instruments). The mean particle size of decanted nanofluids decreases with increasing applied RCF, as shown in Fig. 1, because small particles settled evenly when a great RCF was applied. Also, to observe the dispersion form and particle-size distribution, transmission electron microscope (TEM) images of the nanoparticles were also taken by JEOL JEM-2100 F TEM.

The thermal conductivity of alumina nanofluids was measured using the custom-made transient hot wire system, whose design was based on the transient hot wire method developed by Nagasaka and Nagashima(17). In our transient hot wire system, Teflon was employed as an insulating material, since it has high resistance to chemical reactions, corrosion and stress-cracking at elevated temperatures. Consequently, a 50 μm diameter platinum wire with a Teflon insulation coating of 25 μm thickness (i.e., total 100 μm hot wire), manufactured by A-M Systems, Inc., has been used as the hot wire. Soldered spots are also insulated by silicon after soldering to avoid electrical disturbances. The wire tension can be maintained and adjusted by the top side of a tension spring and both side of supporters. The XYZ linear translation stage employed to adjust the wire tilting angles aimed to reduce the natural convection effects. Measured thermal conductivity values of distilled water were within 1% of literature values, and relative uncertainties of thermal conductivity were less than 1.5%. The thermal chamber was used to adjust temperature and had a temperature range of -20°C to 120°C. Using the device, we measured the thermal conductivity of water-based alumina nanofluids over a broad range of temperatures from 10°C to 80°C.

3. Results and Discussion

Figure 2 shows thermal conductivity ratio, defined as k_{NF}/k_{BF}, where k_{NF} and k_{BF} are thermal conductivity of the alumina nanofluids and the base fluid, respectively, as a function of temperature and experimental data were compared with the prediction of the Maxwell model.(18) The thermal conductivity ratio of the three different alumina nanofluids presents similar trends; increases with increasing temperature, however, the rate of increment in thermal conductivity was different. For the AN1, the thermal conductivity increases sharply with increasing temperature compared to other two alumina nanofluids with a maximum increment of about 4.7% from the prediction of the Maxwell model at 80°C. On the other hand, the thermal conductivity enhancement of the AN2 is smaller in the overall temperature range compared to the AN1 with a maximum increment of about 0.85%

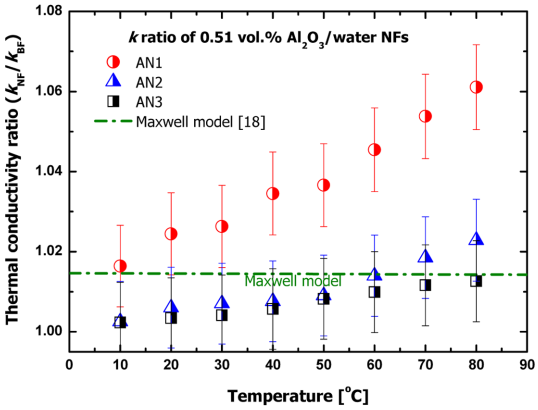

Fig. 2. Thermal conductivity ratio (k of nanofluids to that of water) of three 0.51 vol.% Al_2O_3/water nanofluids as a function of temperature

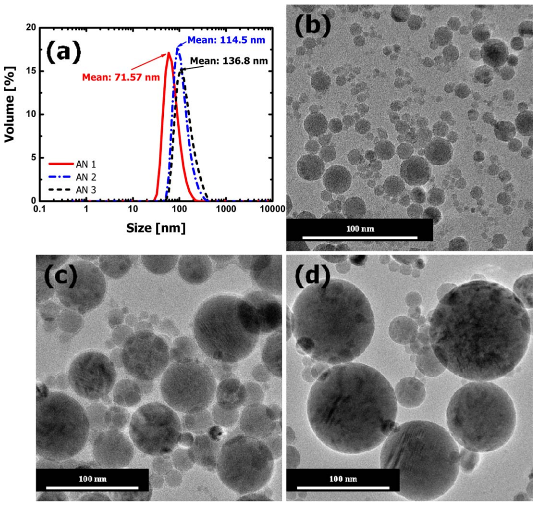

Fig. 3 Distribution of particles and particle sizes in water-based alumina nanofluids: (a) Particle-size distribution of three alumina nanofluids by the DLS; (b) TEM image of AN1, (c) TEM image of AN2, (d) TEM image of AN3. The scale bar of (b), (c), and (d) is 100 nm

from the prediction of the Maxwell model at 80°C. Moreover, in case of the AN3, the thermal conductivity enhancement is even lower than the prediction of the Maxwell in the overall temperature range.

The difference in the thermal conductivity increments of water-based alumina nanofluids can be explained by distribution and size of particles. Fig. 3(a) shows the particle size distribution and mean particle size in water-based alumina nanofluids measured by DLS. The particles in nanofluids AN1, AN2, and AN3 have mean diameters $d_{AN1} = 71.6$ nm, $d_{AN2} = 114.5$ nm, and $d_{AN3} = 136.8$ nm. The TEM images shown in Figs. 3(b)-3(d) demonstrate that the nanoparticles range in size from 3-27 nm (AN1), 10-76 nm (AN2), and 10-111 nm (AN3), although the size distribution is not monodispersed. However, there is clearly a difference in particle size among the three nanofluids. Based on the results, Fig. 4 demonstrates the effect of mean particle size on thermal conductivity, by recording the thermal conductivity ratio (*k* of nanofluids to that of water) of the nanofluids as a function of mean diameter of nanoparticles.

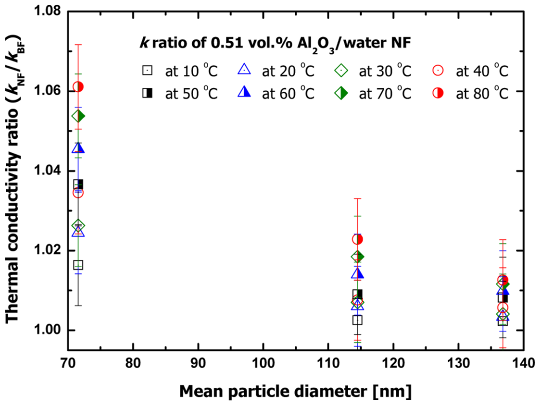

Fig. 4 Thermal conductivity ratio (*k* of nanofluids to that of water) of water-based alumina nanofluids as a function of mean particle diameter

As shown in Figs. 2-4, we found that particle size and temperature affect the thermal conductivity of water-based alumina nanofluids, although the particles are not monodispersed by size. We also found that the temperature effect depends on the size effect. The smaller the nanoparticle size, the stronger the temperature effect on thermal conductivity. This allowed us to determine the coupling dependency on nanoparticle size and temperature in heat conduction enhancement of alumina nanofluids.

The large enhancement was observed at the high temperature region than the low temperature region in aqueous alumina nanofluids as shown in Fig. 2 and this result led suggests that there are other effects which play an important role on thermal conductivity enhancement beyond the conventional effective medium theory (EMT). Our experimental data show the particle size and temperature dependence in thermal conductivity enhancement and cannot be explained by the classical EMT model such as the Maxwell model.(18) Hasselman and Johnson(19) derived an expression for the effective thermal conductivity of composites taking into account the thermal barrier resistance at the interface between the materials and the relations for insertion shapes for spherical, cylindrical and flat plate for low concentration of dispersions. The resulting expression for spherical particles can be arranged as

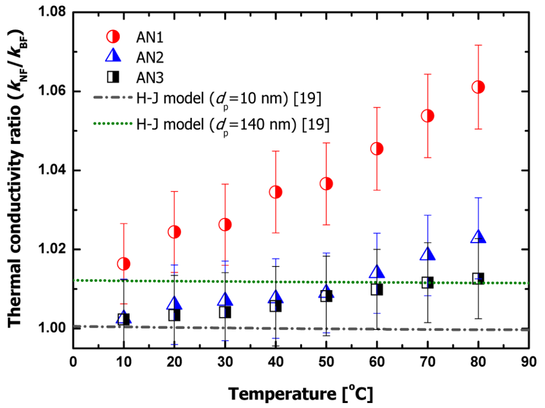

Fig. 5 Experimental data comparison with Hasselman and Johnson (H–J) model as function of temperature

$$\frac{k_{NF}}{k_{BF}} = \left[\frac{k_p(1+2\alpha)+2k_{BF}+2\phi(k_p(1-\alpha)-k_{BF})}{k_p(1+2\alpha)+2k_{BF}-\phi(k_p(1-\alpha)-k_{BF})}\right] \tag{1}$$

where k_{NF}, k_{BF}, k_{p}, and ϕ are thermal conductivity of the nanofluids, thermal conductivity of the base fluid, thermal conductivity of the particle and volume fraction of the nanoparticle within the base fluid, respectively. In equation (1), α denotes a dimensionless parameter defined as $\alpha = a_K / a_{\mathrm{p}}$, where a_K is the so-called Kapitza radius and a_{p} is the radius of nanoparticle. Kapitza radius a_K defined as $a_K = R_b k_{\mathrm{BF}}$, where R_b is the Kapitza resistance (thermal boundary resistance). The contribution of the thermal boundary resistance on effective thermal conductivity was evaluated using the Hasselman and Johnson (H-J) model(19) by assuming all the particles are spherical and particle sizes are within 10-140 nm based on the DLS and TEM results, and R_b is assumed to be R_b 0.77×10^{-8} Km^{2}W^{-1} for water(20). Fig. 5 shows the predictions of the H-J model along with the experimental data as a function of the temperature. According to the H-J model prediction by assuming particle sizes are within 10-140 nm, the prediction values can be regarded as the upper bound of the thermal conductivity enhancement when the mean particle diameter (d_{p}) is 140 nm, while the lower bound is calculated with $d_{\mathrm{p}} = 10$ nm. This trend is the contradictory to our experimental result since thermal conductivity enhancement increases with decreasing the particle size in our data. Moreover, in case of the AN1, the thermal conductivity enhancement is above the upper bound in the overall temperature range. Hence, H-J model cannot explain the size effect on the thermal conductivity enhancement in our experimental data and we confirmed that thermal boundary resistance does not have significant influence on the thermal conductivity of the effective medium as pointed out in refs 11 and 15.

The effective medium theory (EMT) models such as the Maxwell model(18) and the Hasselman and Johnson (H-J) model(19) cannot apply to predict the thermal conductivity enhancement as aforementioned. Thus, we estimated the effect of Brownian motion on effective thermal conductivity using the Brownian velocity of nanoparticles based on the Einstein diffusion theory(21). The root-mean-square (rms) velocity of the nanoparticles also have used as the Brownian velocity or convection velocity(15, 23-24). However, the order of rms velocity is unreasonable to regard as the Brownian velocity of nanoparticles(15). The Brownian velocity based on the Einstein diffusion theory(21) used in our paper and defined as

$$V_{BR} \equiv \frac{K_B T}{3\pi\mu_{BF} d_p l_{BF}} \tag{2}$$

where K_B is the Boltzmann constant, defined as 1.3807×10^{23} J/K, μ_{BF} is the viscosity of the base fluid, d_{p} is the particle diameter, and l_{BF} is the mean free path of the base fluid and assumed a constant value of 0.17 nm for water(4), respectively. The Brownian velocity of nanoparticles are commonly regarded as the important factor when established prediction model based on the Brownian motion(4, 22-24) since the defined Brownian velocity equation is the functions of particle size and temperature. Our experimental data completely show the temperature- and size-dependency in the thermal conductivity enhancement, thus, it is physically reasonable to use the concept of the Brownian velocity to evaluate the effect of the particle size and temperature on the heat conduction of alumina nanofluids.

Figure 6 shows the comparison of the calculated

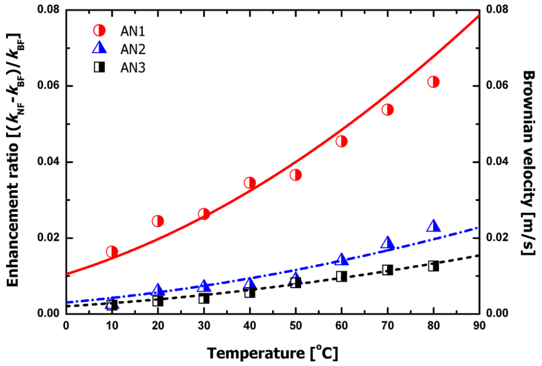

Fig. 6 Comparison of the Brownian velocity of nanoparticles in water with the enhancement ratio of thermal conductivity of three aqueous alumina nanofluids as a function of temperature

Brownian velocity of nanoparticles in water using the equation (2) with the enhancement ratio of thermal conductivity of alumina nanofluids, defined as $(k_{NF}-k_{BF})/k_{BF}$, as a function of temperature. In calculating the Brownian velocity of nanoparticles, the proportionality constants were used as *Constant* × V_{BR}. This is intended to examine the tendency comparison of the increment rate of the Brownian velocity with the enhancement ratio of thermal conductivity of nanofluids since the thermal conductivity enhancement in nanofluids may not be only the function of the Brownian velocity. The proportionality constants used for these calculations are 0.56 for AN1, 0.26 for AN2, and 0.21 for AN3. The increment rate of the calculated Brownian velocity is in accordance with the enhancement ratio of the thermal conductivity as shown in Fig. 6. Moreover, the fitted proportionality constants for each case decrease with increasing mean particle diameter and this tendency may suggest that there are other size effects beyond the Brownian velocity on the heat conduction mechanism of nanofluids. Thus, it can be concluded that the Brownian velocity can be the factor of the temperature- and size-dependent thermal conductivity data for alumina nanofluids and it is suggested that the Brownian-motion-induced convection from multiple nanoparticles or Brownian motion can be the main reason for the observed thermal conductivity enhancement shown in Fig. 6.

4. Conclusions

In conclusion, we have presented the effects of particle size and temperature on the thermal conductivity enhancement of water-based alumina nanofluids through systematic experiments. Using one kind of nanoparticle from a single provider, we manufacture three different water-based alumina nanofluids, each containing three different sizes of nanoparticles, using the centrifuging method. To observe particle distribution and size in nanofluids, we performed DLS and TEM measurements. We measured thermal conductivity from 10°C to 80°C to examine the effects of nanoparticle size and temperature. Based on the experimental results, we first found that the temperature effect strongly depends on the size effect. So we showed the coupling dependency on nanoparticle size and temperature in heat conduction enhancement of alumina nanofluids. Also the experimental results clearly show that thermal conductivity enhancement is strongly correlated to size and temperature. Our studies indicate that the EMT models are not sufficient to explain the thermal conductivity of nanofluids because the models cannot account for the temperature- and size-dependent nature of water-based alumina nanofluids. Moreover, Brownian velocity can be the factor of the temperature- and size-dependent thermal conductivity data for alumina nanofluids.

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