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# Effective viscosities and thermal conductivities of aqueous nanofluids containing low volume concentrations of  $Al_2O_3$  nanoparticles

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### Abstract

Aqueous nanofluids containing low volume concentrations of  $Al_2O_3$  nanoparticles in the 0.01–0.3 vol.% range were produced and characterized. Measurements of zeta potential and TEM micrograph of the alumina nanoparticles in the  $A_1O_3$ -water nanofluids show that the alumina nanoparticles can be best dispersed and stabilized in DI water with little evidence of aggregation at 5 h of ultrasonic vibration. Viscosity measurements show that the viscosity of the  $A1_2O_3$ -water nanofluids significantly decreases with increasing temperature. Furthermore, the measured viscosities of the  $A<sub>1</sub>O<sub>3</sub>$ -water nanofluids show a nonlinear relation with the concentration even in the low volume concentration (0.01%–0.3%) range, while the Einstein viscosity model clearly predicts a linear relation, and exceed the Einstein model predictions. In contrast to viscosity, the measured thermal conductivities of the dilute  $Al_2O_3$ –water nanofluids increase nearly linearly with the concentration, agree well with the predicted values by the Jang and Choi model, and are consistent in their overall trend with previous data at higher concentrations.

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## 1. Introduction

For more than 100 years, many scientists and engineers have made great efforts to improve the inherently poor thermal conductivities of traditional heat transfer fluids, such as water, oil, and ethylene glycol. Numerous theoretical and experimental studies on thermal conductivities of suspensions that contain solid particles have been conducted since the theoretical work of Maxwell [\[1\]](#page-5-0). However, studies of thermal conductivity of suspensions have been confined to millimeter- or micrometer-sized particles. The outstanding problems with the use of these conventional large particles include their rapid settling in fluids, abrasion, clogging, and fouling. To overcome these problems Choi [\[2\]](#page-5-0) pioneered nanofluids, a new class of heat transfer fluids that are produced by dispersing metallic or nonmetallic nanoparticles with typical length scales on the order of 1–100 nm in traditional heat transfer fluids.

Considerable research has gone into nanofluids containing  $Al_2O_3$  nanoparticles. Lee et al. [\[3\]](#page-5-0) formulated waterbased nanofluids containing  $Al_2O_3$  nanoparticles without any chemical dispersants and performed experiments to show that the  $Al_2O_3$ -water nanofluids they formulated have good suspension and dispersion characteristics and high thermal conductivities at the concentrations from 1 to 5 vol.%. Since their work, experimental data [\[4–7\]](#page-5-0) for the effective thermal conductivity of nanofluids with  $Al<sub>2</sub>O<sub>3</sub>$  nanoparticles suspended in water or ethylene glycol have been reported. As shown in [Fig. 1,](#page-1-0) most researchers

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measured the thermal conductivity of  $Al_2O_3$ -water nanofluids at  $Al_2O_3$  nanoparticle concentrations greater than or equal to 1% by volume. However, little experimental data exist on the effective thermal conductivities and viscosities of nanofluids with low volume concentrations  $\leq 1$  vol.%) of  $A<sub>1</sub>Q<sub>3</sub>$  nanoparticles. This motivated us to do this study. In addition, Patel et al. [\[8\]](#page-5-0) showed that the enhancement of the effective thermal conductivity of toluene-based nanofluids containing Au is significant (by as much as 14%) at very low concentration of Au nanoparticles in the 0.005– 0.011 vol.% range. Therefore, the objective of the present study is to measure the effective viscosities and thermal conductivities of  $Al_2O_3$ -water nanofluids at low volume concentrations in order to see how  $Al_2O_3$ —water nanofluids would behave in the very low concentration range from 0.01 to 0.3 vol.%  $Al_2O_3$  nanoparticles. For this purpose, a two-step method with ultrasonic dispersion of alumina nanoparticles was used to make  $Al_2O_3$ -water nanofluids. Measurements of zeta potential and TEM micrograph of the alumina nanoparticles in the  $Al_2O_3$ -water nanofluids were used to characterize the dispersion characteristics of the  $Al_2O_3$ -water nanofluids. The effective viscosities of the  $Al_2O_3$ -water nanofluids were measured as a function



Fig. 1. Thermal conductivities of nanofluids containing  $Al_2O_3$  nanoparticles as a function of concentration, normalized to the thermal conductivity of the base fluid.

of temperature and particle volume concentration using an oscillation type viscometer. The effective thermal conductivities of the  $Al_2O_3$ -water nanofluids were measured as a function of nanoparticle volume concentration using a transient hot wire apparatus.

## 2. Production and dispersion characteristics of  $Al_2O_3$ -water nanofluids

A two-step method is used to produce  $Al_2O_3$ -water nanofluids with low concentrations of  $Al_2O_3$  nanoparticles from 0.01 to 0.3 vol.% without any surfactant. The twostep method used in this study can be divided into two stages. The first stage is to mix  $Al_2O_3$  nanoparticles with a nominal particle size of  $30 \pm 5$  nm in DI water. The  $Al_2O_3$  nanoparticles used in the present work were manufactured and supplied by Nanotechnologies Inc. Since ultrasonic vibration breaks down agglomerates in the mixture, the next stage is to homogenize the mixture using ultrasonic vibration at sound frequencies of 30–40 kHz.

Uniform dispersion and stable suspension of nanoparticles in liquids is the key to most applications of nanofluids since the final properties of nanofluids are determined by the quality of the dispersion and suspension. Because the surface of  $Al_2O_3$  nanoparticles is positively charged, measurement of the surface charge or zeta potential of the nanoparticles is critical for making stable nanofluids. For example, it is well-known that a suspension with zeta potential (absolute value) above 30 mV are physically stable and above 60 mV show excellent stability while a suspension below 20 mV has limited stability and below 5 mV undergoes pronounced aggregation [\[9\].](#page-5-0) In this study we are interested in the effect of ultrasonic vibration on zeta potential. Zeta potential was measured using a Zetasizer Nano ZS manufactured by Malvern Instruments Ltd. Because this technique requires extremely dilute suspensions (as low as 0.1 vol.%), four dilute  $Al_2O_3$ -nanofluids at a fixed concentration of 0.1 vol.% were made and sonicated for 0 h, 5 h, 20 h and 30 h, respectively. Zeta potential was measured for these  $Al_2O_3$ -nanofluids at 25 °C and a pH of 6.04.

[Fig. 2](#page-2-0) shows the measured zeta potential of a 0.1 vol.%  $Al_2O_3$ -nanofluids as a function of sonication time. First of all, we observe that the  $Al_2O_3$ -water nanofluids prepared

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Fig. 2. Effect of sonication time (h) on zeta potential of the  $Al_2O_3$ nanoparticles dispersed in water.

using ultrasonic vibration have higher zeta potentials than the same  $Al_2O_3$ -nanofluids without ultrasonic vibration. More important observation is that zeta potential of 0.1 vol.% alumina nanofluids reaches a maximum value of 34 mV after 5 h of sonication and then decreases with increasing sonication. Therefore, the  $0.1$  vol.%  $Al_2O_3$ nanofluids are most stable when they are kept under ultrasonic vibration for 5 h. This shows that sonication is needed for long  $(\sim 5)$  h to improve the particle dispersion.

The  $Al_2O_3$  nanoparticles were further characterized with a TEM (transmission electron microscope). A TEM micrograph in Fig. 3 shows that most  $Al_2O_3$  nanoparticles are smaller than  $30 \pm 5$  nm and that there are few agglomerated  $Al_2O_3$  nanoparticles. Zeta potential measurements and a TEM micrograph show that 5 h of ultrasonic vibra-



Fig. 3. TEM micrograph of  $Al_2O_3$  nanoparticles dispersed in water.

tion provides a good dispersion/suspension of  $Al_2O_3$  nanoparticles with little aggregates in DI water.

#### 3. Effective viscosity

The viscosity and thermal conductivity of nanofluids are important transport properties for applications of nanofluids as a new class of heat transfer fluids in thermal devices or systems such as heat exchangers or cooling systems. However, experimental data for the viscosity of nanofluids are rare compared with their thermal conductivity. Therefore, in addition to the effective thermal conductivities of  $Al_2O_3$ -water nanofluids, the effective viscosities of dilute nanofluids at low concentrations of alumina nanoparticles  $(0.01 - 0.3 \text{ vol.})$ % were measured.

The effective viscosities of the  $Al_2O_3$ -water nanofluids were measured using a viscometer of oscillation type, VM-10A manufactured by CBC Co., Ltd. Before measuring the viscosity of  $A<sub>1</sub>O<sub>3</sub>$ -water nanofluids, we measured the viscosities of DI water. A comparison of our data with the reference data presented by Incropera [\[10\]](#page-5-0) shows that the uncertainty of the viscosity measurements is within 1.8%. The effective viscosities of  $Al_2O_3$ -water nanofluids with low concentrations from 0.01 to 0.3 vol.% were measured at the temperature range from 21 to 39 °C. Fig. 4 shows the effective viscosities of  $Al_2O_3$ -water nanofluids with various concentrations of  $Al_2O_3$  nanoparticles as a function of temperature. As shown in Fig. 4 the effective viscosities of the dilute  $Al_2O_3$ -water nanofluids significantly decreases with increasing temperature and slightly increases with increasing volume fraction. [Fig. 5](#page-3-0) shows the increment in the effective viscosity of dilute  $Al_2O_3$ – water nanofluids as a function of nanoparticle volume fraction. For comparison with measured viscosity increment we also show theoretical predictions from the Einstein model of the effective viscosity of dilute suspensions [\[11\]](#page-5-0)

$$
\frac{\mu_{\text{eff}}}{\mu_{\text{BF}}} = 1 + 2.5f\tag{1}
$$



Fig. 4. Effective viscosities of  $A_1O_3$ —water nanofluids with low concentrations from 0.01 to 0.3 vol.% as a function of temperature.

<span id="page-3-0"></span>

Fig. 5. Increment in the effective viscosity of dilute  $Al_2O_3$ -water nanofluids as a function of nanoparticle volume fraction.

Table 1

Enhancement in the effective viscosities of  $A1_2O_3$ -nanofluids at various nanoparticles concentrations and at 21 °C

Concentration $(\% )$	$0.01$ $0.02$ $0.03$ $0.1$ $0.2$		03
Enhancement $(\% )$	0.08 0.11 0.89 1.74 2.51 2.90		
$\left(\frac{\mu_{\rm nanofluids}-\mu_{\rm water}}{\mu_{\rm water}}\times\,100\right)$			

where  $\mu_{\text{eff}}$ ,  $\mu_{\text{BF}}$ , and f are the effective viscosity, the viscosity of the base fluid, and the volume fraction, respectively. Fig. 5 and also Table 1 show that the increment in the measured viscosity of the  $Al_2O_3$ -water nanofluids is greater than predicted by the Einstein model and increases up to 2.9% at the concentration of 0.3 vol.%. Moreover, Fig. 5 shows that the measured viscosity of the  $Al_2O_3$ -water nanofluids is nonlinear with the  $Al_2O_3$  nanoparticle volume concentration, while the Einstein model predicts a linear relationship. The Einstein viscosity equation is known to be valid for particle concentrations less than 2 vol.%. What's unexpected is that, for the  $Al_2O_3$ -water nanofluids, the nonlinear viscosity behavior occurs at very low particle concentrations far below 2 vol.%. Very limited experimental data such as He et al. [\[12\]](#page-5-0) also show nonlinear increase in the viscosity with the nanoparticle concentration. Nonlinear behavior implies that there are particle–particle interactions which invalidate the Einstein equation developed for dilute suspensions. It can be argued that the nanofluids sonicated for 5 h are very different from those sonicated for only a few hours or less in that the interparticle distance is shorter for nanofluids with uniformly dispersed nanoparticles than for those with agglomerated nanoparticles. Therefore, it is reasonable to expect more particle–particle interactions in nanofluids with uniform dispersion of nanoparticles than with aggregates. Also, it can be argued that this nonlinear behavior could be related to the increased surface of well-dispersed nanoparticles in the nanofluids sonicated for 5 h and used in the measurement of the viscosities because the flow behavior of a solid–liquid suspension depends on the hydrodynamic force acting on the surface of solid particles. However, presently it is challenging to understand the physical mechanisms for the discrepancy in the magnitude and concentration-dependency of the viscosity of the  $Al_2O_3$ -water nanofluids at very low concentrations below 0.3 vol.%. Therefore, this nonlinear viscosity behavior of the nanofluids needs further experimental and theoretical studies.

#### 4. Effective thermal conductivity

A transient hot wire system was designed and manufactured in-house to measure the effective thermal conductivity of  $Al_2O_3$ -water nanofluids with low concentrations from 0.01 to 0.3 vol.%. The schematic of the experimental apparatus and electric circuit is shown in Fig. 6. In the Wheatstone bridge,  $R_w$ ,  $R_1$ ,  $R_2$ , and  $R_3$  are the resistance



Fig. 6. Schematic of a transient hot wire apparatus for measuring the thermal conductivities of  $A_1O_3$ –water nanofluids.

of a platinum hot wire, 5 k $\Omega$  resistor, 5 k $\Omega$  potentiometer, and 5  $\Omega$  resistor, respectively. The resistance at 0 °C, length and diameter of the Platinum hot wire manufactured by OMEGA are 10.144  $\Omega$ , 210 mm, and 50.8 µm, respectively. The temperature coefficient of resistivity for the platinum wire is  $0.0039092$ /°C [\[13\]](#page-5-0). Instead of a two-wire compensation system [\[14\]](#page-5-0) which is used to eliminate axial conduction at both ends of the wire a hot wire with high aspect ratio (wire length-to-diameter ratio of  $\approx$ 4100) was used in this study as a sensor to minimize conduction loss at the end of the wire. In addition, a constant temperature chamber was used to reduce the thermal disturbance due to variations in room temperature. When experimental data are obtained from the transient hot wire shown in [Fig. 6](#page-3-0), the effective thermal conductivity of  $Al_2O_3$ -nanofluids is calculated from

$$
k = \frac{q}{4\pi (T_2 - T_1)} \ln\left(\frac{t_2}{t_1}\right) \tag{2}
$$

where  $k$  and  $q$  are the thermal conductivity and applied power per unit length of the wire, respectively and  $T_2 - T_1$  is the temperature rise of the wire between times  $t_1$  and  $t_2$ . To validate the transient hot wire system the thermal conductivity of DI water was measured. We found that the uncertainty of the thermal conductivity measurements is within 0.5%.

The effective thermal conductivities of  $Al_2O_3$ -water nanofluids with low concentrations from 0.01 to 0.3 vol.% were measured at 21 °C. Fig. 7 shows the enhancement in the effective thermal conductivities of  $Al_2O_3$ -water nanofluids as a function of nanoparticle volume fraction. Fig. 7 shows that the enhancement in the measured thermal conductivities of the  $Al_2O_3$ -water nanofluids increases with increasing concentration and that the maximum enhancement is 1.44% at the concentration of 0.3 vol.%. These results show that the enhancement in the effective thermal conductivity of very dilute  $Al_2O_3$ -water nanofluids is not as remarkable as that of toluene-based



Fig. 7. Enhancement in the effective thermal conductivities of  $\text{Al}_2\text{O}_3$ water nanofluids as a function of nanoparticle volume fraction.



Fig. 8. Comparison between measured and predicted thermal conductivities of  $Al_2O_3$ -water nanofluids, normalized to the thermal conductivity of the base fluid.

nanofluids containing extremely low concentrations of Au nanoparticles in the range of 0.005–0.011 vol.% [\[8\].](#page-5-0) Fig. 7 also shows that, in contrast to viscosity, the measured thermal conductivities of the dilute  $Al_2O_3$ -water nanofluids increase nearly linearly with the  $Al_2O_3$  nanoparticle volume concentration. However, it is interesting to see in Fig. 8 that the present data is very consistent with the data from the previous studies conducted at higher concentrations of  $Al_2O_3$  nanoparticles [\[3–6\]](#page-5-0). Furthermore, Fig. 8 shows that the Jang and Choi model [\[15,16\]](#page-5-0) can be used to estimate the effective thermal conductivity of  $A<sub>1</sub>Q<sub>3</sub>$ -water nanofluids over a very wide concentration range from 0.01 to 5 vol.%, which is more than two orders of magnitude.

## 5. Summary and conclusion

In this paper, we experimentally investigated the effective viscosities and thermal conductivities of water-based nanofluids containing very low concentrations of  $Al_2O_3$ nanoparticles  $(Al_2O_3$ -water nanofluids). For this purpose, we produced  $Al_2O_3$ -water nanofluids with various concentrations from 0.01 to 0.3 vol.% using a two step method with ultrasonication and without any surfactant. To examine the suspension and dispersion characteristics of the  $Al_2O_3$ -water nanofluids, we studied the zeta potential and TEM micrograph of the  $Al_2O_3$  nanoparticles. Measurements of zeta potential and TEM micrograph of the alumina nanoparticles in the  $Al_2O_3$ -water nanofluids show that 5 or more hours of ultrasonic vibration can uniformly disperse the alumina nanoparticles in DI water with little evidence of aggregation. Viscosity measurements show that the viscosity of the  $Al_2O_3$ -water nanofluids significantly decreases with increasing temperature. We have also observed that the alumina nanofluids have a nonlinear relation between their viscosity and the nanoparticle concentration even at very low  $(0.01-0.3 \text{ vol.})$ % concentrations,

<span id="page-5-0"></span>while the Einstein viscosity equation clearly predicts a linear relation.

In contrast to viscosity, the measured thermal conductivities of the dilute  $Al_2O_3$ -water nanofluids increase nearly linearly with the concentration. Furthermore, the measured thermal conductivities of the  $Al_2O_3$ -water nanofluids are consistent in their overall trend with previous data at higher concentrations and agree well with the predicted values by the Jang and Choi model [15,16] over a very wide concentration range from 0.01 to 5 vol.%.

This work shows that further experimental and theoretical studies are needed to understand and control the interesting transport properties of the  $Al_2O_3$ -water nanofluids at very low concentrations such as the discrepancy in the magnitude and concentration-dependency of their viscosity.

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## References

- [1] J.C. Maxwell, Electricity and Magnetism, first ed., Clarendon Press, Oxford, 1873.
- [2] S.U.S. Choi, Enhancing thermal conductivity of fluids with nanoparticles, in: D.A. Singer, H.P. Wang (Eds.), Developments and Applications of Non-Newtonian Flows, FED 231/MD 66, ASME, New York, 1995, pp. 99–105.
- [3] S. Lee, S.U.S. Choi, J.A. Eastman, Measuring thermal conductivity of fluids containing oxide nanoparticles, ASME J. Heat Transfer 121 (1999) 280–289.
- [4] X. Wang, X. Xu, S.U.S. Choi, Thermal conductivity of nanoparticle– fluid mixture, J. Thermophys. Heat Transfer 13 (1999) 474–480.
- [5] H.Q. Xie, J.C. Wang, T.G. Xi, Y. Liu, F. Ai, Q.R. Wu, Thermal conductivity enhancement of suspensions containing nanosized alumina particles, J. Appl. Phys. 91 (2002) 4568–4572.
- [6] S.K. Das, N. Putra, P. Thiesen, W. Roetzel, Temperature dependence of thermal conductivity enhancement for nanofluids, ASME Trans. J. Heat Transfer 125 (2003) 567–574.
- [7] S.M.S. Murshed, K.C. Leong, C. Yang, Thermal Conductivity of Nanoparticle Suspensions (Nanofluids), in: 2006 IEEE Emerging Technologies-Nanoelectronic Conference.
- [8] H.E. Patel, S.K. Das, T. Sundararajan, A.S. Nair, B. George, T. Pradeep, Thermal conductivities of naked and monolayer protected metal nanoparticle based nanofluids: manifestation of anomalous enhancement and chemical effect, Appl. Phys. Lett. 83 (14) (2003) 2931–2933.
- [9] R.H. Müller, Zetapotential und Partikelladung in der Laborpraxis, first ed., Wissenschaftliche Verlagsgesellschaft, Stuttgart, 1996.
- [10] F.P. Incropera, D.P. Dewitt, Fundamentals of Heat and Mass Transfer, fifth ed., Wiley, New York, 2002.
- [11] A. Einstein, Investigation on the Theory of Brownian Movement, Dover, New York, 1956.
- [12] Y. He, Y. Jin, H. Chen, Y. Ding, D. Cang, H. Lu, Heat transfer and flow behaviour of aqueous suspensions of  $TiO<sub>2</sub>$  nanoparticles (nanofluids) flowing upward through a vertical pipe, Int. J. Heat Mass Transfer 50 (2007) 2272–2281.
- [13] J.P. Bentley, Temperature sensor characteristics and measurement system design, J. Phys. E: Sci. Instrum. 17 (1984) 430–439.
- [14] H.M. Roder, Transient hot-wire thermal conductivity apparatus for fluids, J. Res. Nat. Bur. Stand. 86 (1981) 457–493.
- [15] S.P. Jang, S.U.S. Choi, Role of Brownian motion in the enhanced thermal conductivity of nanofluids, Appl. Phys. Lett. 84 (2004) 4316– 4318.
- [16] S.P. Jang, S.U.S. Choi, Effects of various parameters on nanofluid thermal conductivity, ASME Trans. J. Heat Transfer 129 (2007) 617– 623.