

The Importance of Suspension Stability for the Hot-wire Measurements of Thermal Conductivity of Colloidal Suspensions.

M. Chiesa¹ A. J. Simonsen²

¹Department of Mechanical Engineering
Massachusetts Institute of Technology, Cambridge, Ma 02139-4307 USA

²Sintef Materials and Chemistry
SINTEF, Trondheim, 7491 Norway

Abstract

The development of advanced heat transfer fluids with enhanced thermal conductivity is essential to improve the effective heat transfer behavior of conventional coolants. Thermal conductivity enhancement has been reported for colloidal suspensions formulated with different nanoparticles. Reliable thermal conductivity measurement requires that the suspension is stable against sedimentation during the measurement since particle sedimentation may effect the experimental results. The present study proposes a criterion to ensure that thermal conductivity data is independent of the particle sedimentation. A measuring device based on the transient hot-wire method is designed to allow vertical and horizontal thermal conductivity data collection. Our investigation shows that the thermal conductivity measurements in the horizontal and vertical configurations are almost identical when the colloidal suspension is stable while they differ for unstable systems.

Introduction

Fluids such as air, water, ethylene glycol and mineral oils are typically used in applications like power generation, chemical production, automobiles, computing processes, air conditioning and refrigeration. However their heat transfer capability is limited by their very low thermal conductivity. These fluids have almost two orders of magnitude lower thermal conductivity compared to metals, resulting in low heat removal efficiencies. First attempts to improve the thermal conductivity involved dispersing micron-sized particles in these liquids. Ahuja and Liu et al [1] studied the heat transfer augmentation and rheology of slurries. One of the drawbacks associated with the use of slurries is the abrasive action of the particles causing erosion of the components. Secondly micron-sized particles tend to rapidly fall out of the suspension due to their large mass and thus can cause fouling of the components thereby clogging the flow path and increasing the pressure drop. Decreasing the particle size to the nanometer range offers the potential to overcome these drawbacks. Masuda et al [5] first reported the thermal conductivity and viscosity of liquids containing nanometer sized particles. Choi [2] also investigated the heat transfer properties of such colloidal suspensions and coined the term "nanofluids". In addition to overcoming the drawbacks associated with the use of micron-sized suspensions, nanofluids have often exhibited thermal conductivity enhancement substantially higher than predicted by the Maxwell-Garnett effective medium theory [4]. Over the past decade, a significant amount of data has been gathered on the thermal conductivity of nanofluids. Typical materials used for nanoparticles include metals like copper, silver and gold, metal oxides like alumina, titania and iron oxide. Carbon nanotubes have also been used to enhance the thermal conductivity of liquids. Experimental data on the thermal conductivity of nanofluids varies widely and mechanisms responsible for the thermal conductivity enhancement are un-

der debate, as summarized in recent reviews. There is clearly a need to resolve the differences and confirm the repeatability of data obtained. This can be achieved by reporting the exact components of the nanofluid tested like surfactants, particle material, base fluid, method of synthesis of nanoparticles, and any possible contamination with other agents. Also the data obtained by different groups need to be verified for repeatability. Reaching a consensus in regard with the experimental data is critically important for an accurate physical model to be developed to explain the anomalous enhancement. Recently, Liu et al. [3] reported a maximum increase in thermal conductivity of water of about 23.8% with a volume fraction of 0.1 vol% copper nanoparticles. The reported thermal conductivity increase was strongly dependent on the time after sonication after which the measurements were carried out. The thermal conductivity enhancement decayed to nearly zero after about 10 minutes of sonication. It was not clear if the measured enhancement was due to the instability of the nanofluid. In the present study, a measuring apparatus based on the transient hot-wire method has been developed. The apparatus is designed to allow measurements in different configurations rotated with respect to a vertical base configuration. Horizontal and vertical measurements of the thermal conductivity of different systems comprising various colloidal suspensions and their base fluids are carried out. The investigation shows that the thermal conductivity measurements in the horizontal and vertical configurations is almost identical when the colloidal suspension is stable. On the other hand they differ strongly when the suspension is not stable. A stability criterion for the colloidal suspension based on the measurements obtained from two different configurations is proposed. Such a stability criterion guarantees that the measured is not effected by particle sedimentation.

Formulation and Thermal Conductivity Measurement of Colloidal Suspensions

Formulation of Colloidal Suspensions

Colloidal suspensions were formulated by mixing Al₂O₃ alumina nanoparticles with a mean particle size of 20nm with different base fluids: a high purity mineral oil made of a single alkane hydrocarbon, 99.9% C₁₀H₂₂ also referred as decane, and a highly branched isoparaffinic polyalphaolefin (PAO). The particles were stabilized by adding a constant amount of surbitan monolurate 0.25 volume % to both base fluids before adding the particles. The alumina nanopowder was evenly dispersed in the base fluid through ultrasonic disruptor. Sonication was performed in a ice bath to maintain the temperature of the suspension constant. The thermal conductivity variation of the base fluid due to addition of the surfactants is taken into account.

Thermal Conductivity Measurement set-up

Thermal conductivity measurements of the alumina suspensions were carried out by means of the transient hot wire method

developed by Nagasaka and Nagashima [6]. Fig. 1 illustrates the set up used in the present work. It consists of a cylindrical pipe of internal diameter 19mm and length 190mm. A thin platinum wire (25 μm) coated with an insulation (1.5 μm thick isonel layer) is suspended between two copper electrodes in the center of the pipe. The length of the wire is 150mm. The leakage of electrical current from the electrodes to the surrounding fluid is minimized in order to increase the reliability of our measurements. This is especially important when the based nanofluids is a conductive medium. The wire is immersed in the nanofluid and a constant current is passed through it. The temperature rise of the wire is measured as a function of time. The thermal conductivity of the nanofluid can be calculated from the obtained data by using the equation,

$$k_{nf} = \frac{Q}{4\pi L \frac{dT}{d \ln t}} \quad (1)$$

Here k_{nf} is the thermal conductivity of the nanofluid, Q is the total power dissipated in the wire, L is the length of the wire, T is the wire temperature and t is the time. The constant current used in the measurement served two purposes, one to act as a heat source and second to enable the temperature measurement of the wire. In order to measure the temperature rise, the hot wire was made part of a Wheatstone bridge. Before starting the experiment the bridge was balanced. During the experiment, the change in wire temperature caused a change in wire resistance leading to an imbalance in the bridge. The change in wire resistance was measured by measuring the voltage imbalance in the bridge. The temperature coefficient of resistance of the wire was measured carefully by measuring the resistance as a function of the temperature of the wire. By knowing the temperature coefficient of resistance of the wire, the temperature rise of the wire was calculated from the change in wire resistance data. By measuring the slope of the temperature rise versus log of time curve and using Eq. 1, the thermal conductivity of the nanofluid was measured. Our measurement setup allows mea-

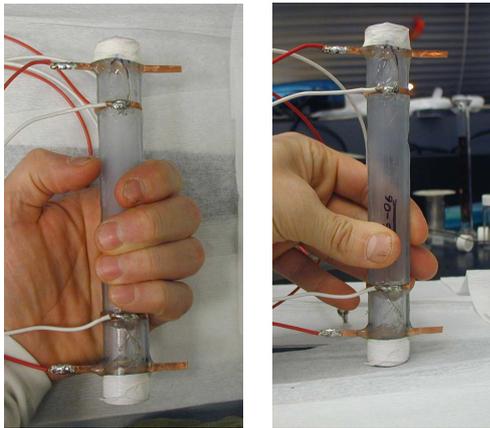


Figure 1: Thermal conductivity measuring apparatus designed to allow measurements in different configurations rotated with respect to a vertical base configuration.

surements to be carried out in different configurations rotated with respect to a vertical base configuration. The experimental set up was calibrated by comparing the measured value of thermal conductivity for ethylene glycol against the literature value. The literature value was reproduced with an uncertainty of 0.5%. Thermal conductivity measurements in the horizontal and vertical configuration were performed for different base fluids and for a couple of stable nanofluids systems. Fig. 2 shows that the measurements in the two configurations provide very

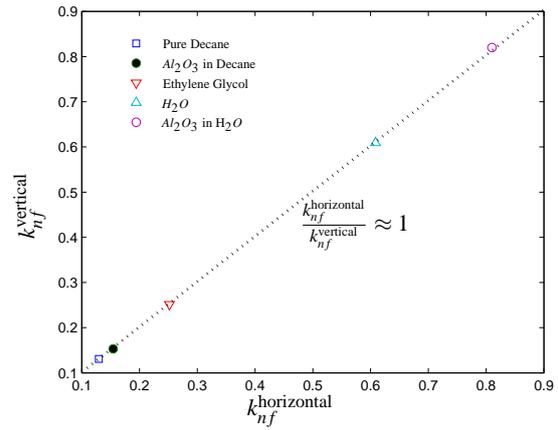


Figure 2: Thermal conductivity measurements carried out in the horizontal and vertical configuration.

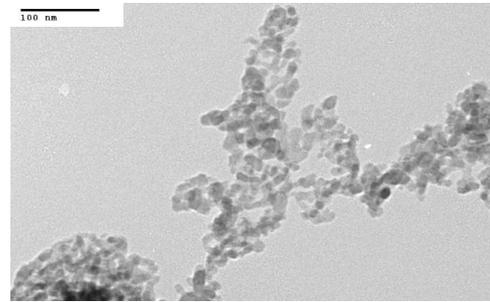


Figure 3: TEM image of a stable alumina in decane nanofluid.

similar results as one would expect. On the x-axis of Fig. 2 one reports the thermal conductivity measured in the horizontal configuration while on y-axis one has the thermal conductivity measured in the vertical configuration. The thermal conductivity for each data point was measured 15 times in both the horizontal and the vertical configuration over a period of about 60mins. The average of the 15 experimental data is reported in this article. The thermal conductivity measurement did not vary appreciably over the 60min interval in both the vertical and horizontal configuration. The typical standard deviation involved was 0.5%.

Thermal Conductivity Measurements

The thermal conductivity of the Al_2O_3 in decane and PAO nanofluids was measured in less than a day after dispersing the nanoparticles in the base fluid as explained earlier. Fig. 3 shows a TEM image of a stable alumina in decane nanofluid. The nanoparticles seem to be well distributed. Fig. 4 shows the thermal conductivity enhancement of the Al_2O_3 in decane and PAO nanofluid for volume fractions ranging from 0.1% to 1.5%. The values for the thermal conductivity plotted in Fig. 4 are obtained in the vertical setup configuration. The thermal conductivity ratio predicted by the Maxwell Garnett theory [4] is given by

$$\frac{k_{nf}}{k_f} = \frac{k_p + 2k_f - 2\phi(k_f - k_p)}{k_p + 2k_f + \phi(k_f - k_p)} \quad (2)$$

Here k_f , k_p and k_{nf} are the thermal conductivity of the base fluid, particle and nanofluid respectively and ϕ is the volume fraction of the nanoparticles. The above expression for effective thermal conductivity does not take into account the thermal interface resistance between the particle and the fluid and is ap-

plicable only for spherical particles. The thermal conductivity ratio prediction based on Eq.2 is plotted in Fig. 4. One can observe that the measured thermal conductivity ratio was higher than that predicted by the Maxwell Garnett theory in both PAO and decane based nanofluids for all the volume fractions studied.

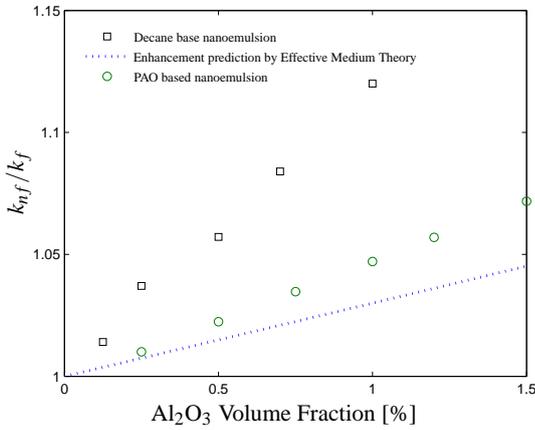


Figure 4: Thermal conductivity enhancement vs Alumina volume fraction in a PAO and Decane based nanofluid.

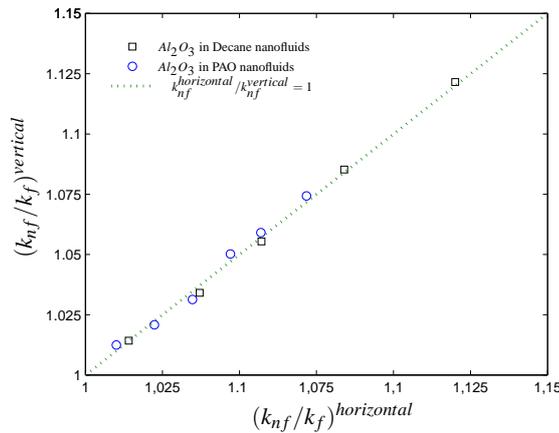


Figure 5: Thermal conductivity enhancement

Thermal conductivity measurements are carried out both in the horizontal and vertical configuration. No significant change in the thermal conductivity enhancement measured at different volume fraction is obtained as indicated in Fig. 5. The x-axis in Fig. 5 represents the thermal conductivity enhancement obtained in the horizontal configuration $(k_{nf}/k_f)^{horizontal}$, while the y-axis represents the thermal conductivity enhancement obtained in the vertical configuration $(k_{nf}/k_f)^{vertical}$.

Influence of stability issue on hot-wire measurements: a numerical study

The formulated nanofluids have various stability. The PAO based nanoparticles are extremely stable, no sedimentation has been observed for more than 3 months, while the decane base nanofluids have a shorter stability and already after a couple of days showed signs of sedimentation. Both systems can anyway be considered stable during the time the measurements were performed so that the data obtained are reliable. In this respect a criterion that can be employed to assess whether the thermal

conductivity enhancement measurements are independent of the stability of the suspension is that the ratio between the values obtained from the vertical and horizontal setup should be close to one. On the other side when the ratio is not close to one the obtained data is most probably affected by a stability problem and it is therefore not reliable. Figure 6 shows a schematic representation of the hot-wire experimental setup in both configurations. Let L and D be the length and the diameter of the hot-wire cell. When the suspension is not stable one can achieve sedimentation of the particles, illustrated in Fig. 6 as the light blue area, while the dark blue area represents the suspension phase with higher thermal conductivity and h_x and h_y represent the extension of the suspension phase in the horizontal and vertical set up. A numerical study of the thermal conductivity measurement that one may obtain in case of an unstable system is investigated by varying h_x and h_y . The time necessary for the suspension to sediment is the same in the two configurations so that $D - h_x = L - h_y$. One may intuitively understand that in the horizontal case the hot-wire may eventually not be covered by the suspension and this affects the thermal conductivity results. The numerical model is discretized with 222800 elements. The heat equation is solved to second order accuracy and in double precision. The wire domain is set to an initial temperature of 293K. A heat flux $= 7500W/m^2$ from the wire is considered. The effect of time steps on the solution is investigated and the highest allowable time step that does not influence neither convergence or the numerical result is used.

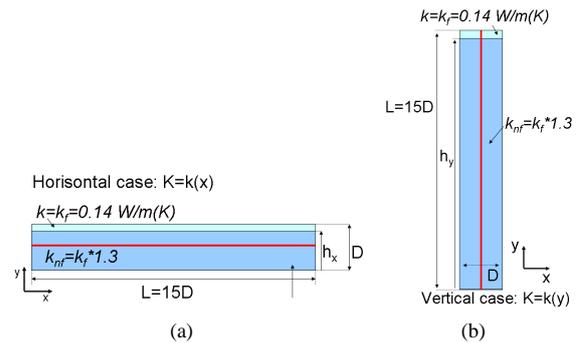


Figure 6: schematic representation of the hot wire experimental set up in both configurations

The nanofluid is described as a fluid with bulk thermal conductivity k_{nf} 30% higher than its base fluid k_f hence $k_{nf} = 1.3k_f$. The temperature change as a function of time in the vertical and horizontal configuration is compared at different sedimentation fraction h_x/D and h_y/L see Fig. 7 and 8. The extreme cases are $h_x/D = 0$ which represents a case for which the particles have completely sedimented out of the suspension. In this case the measurements belong to the base fluid. On the other end when $h_x/D = 1$ no particle sedimentation has taken place and the measurements belong to the nanofluid. In the vertical configuration the thermal conductivity is calculated from the curves obtained in Fig. 7 by means of Eq. 1 in the same way as we do in our experimental setup. The thermal conductivity varies linearly with the fraction h_y/L . The same is repeated for the horizontal configuration. The conductivity is calculated from the curves obtained in Fig. 8 by means of Eq. 1 showing that it varies in a step-like manner. The calculated conductivity is very dependent on how close to the wire the conductivities change between the suspension and the base fluid is. This could probably be explained by the area effect on the heat flux. The normalized conductivity for different fractions of sedimentation is plotted in Fig. 9. From Fig. 9 one can observe how in the case

of unstable system one expects a difference between the horizontal and vertical measurements. On the x-axis we plot the fraction of sedimentation express as $\frac{D-h_x}{D} = \frac{L-h_y}{D}$. The time necessary for sedimentation to occur is the same and for this reason the $D-h_x=L-h_y$. The results shown in Fig. 9 indicates how the results from the vertical setup are less dependent on sedimentation issues. Due to this observation one can stress that the requirement for which one should gather thermal conductivity enhancements data from both configurations and their ratio should be close to one, would ensure that experimental data is not dependent on the stability of the suspension.

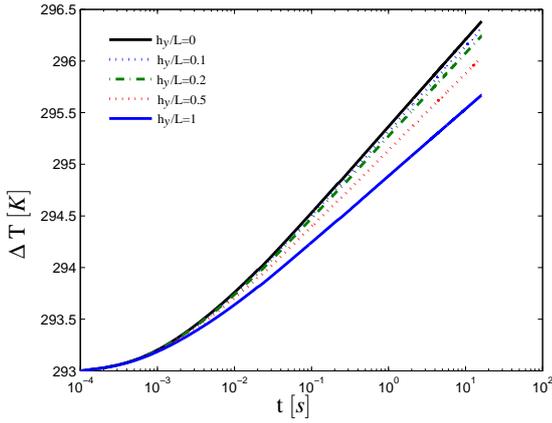


Figure 7: Average wire temperature as a function of time in the vertical configuration

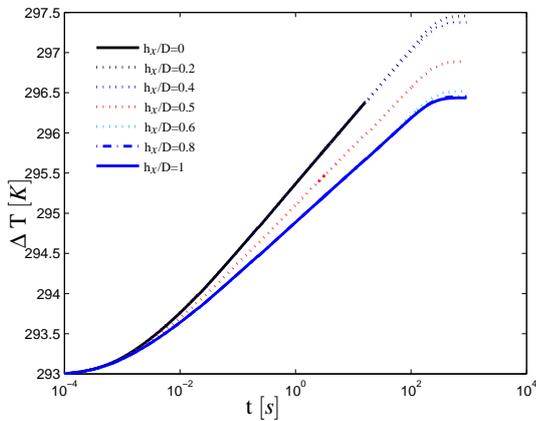


Figure 8: Average wire temperature as a function of time in the horizontal configuration

Conclusions

Reliable thermal conductivity measurements of colloidal suspensions require that the suspension is stable against sedimentation during the measurement since particle sedimentation may effect the experimental results. The present study proposes a criterion to ensure that thermal conductivity data is independent of the particle sedimentation. A measuring device based on the transient hot-wire method is designed to allow vertical and horizontal thermal conductivity data collection. Our investigation shows that the thermal conductivity measurements in the horizontal and vertical configurations are almost identical when the colloidal suspension is stable while they differ for unstable sys-

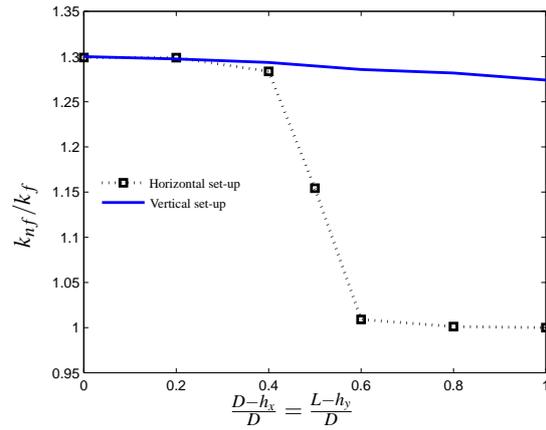


Figure 9: Normalized conductivity for different fractions of sedimentation. The thermal conductivity of the suspension is 30% greater than the thermal conductivity of the base fluid. On the x-axis we have plotted the fraction of sedimentation.

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Acknowledgements

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References

- [1] A.S. Ahuja, *J. Appl. Phys.* 46, 3408 (1975).
- [2] S.U.S. Choi, *Proceedings of the American Society of Mechanical Engineers* 66, 99 (1995)
- [3] M. Liu, M.C. Lin, C.Y. Tsai, and C. Wang, *Int. J. Heat Mass Transfer* 49, 3028 (2006).
- [4] J.C. Maxwell, *A Treatise on Electricity and Magnetism* (Clarendon, Oxford, 1891)
- [5] H. Masuda, A. Ebata, K. Teramae, and N. Hishinuma, *Netsu Bussei* 4, 227 (1993).
- [6] Y. Nagasaka, and A. Nagashima, *J. Phys. E* 14, 1435 (1981).