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Temperature Dependence of Thermal Conductivity Enhancement for Nanofluids

Usual heat transfer fluids with suspended ultra fine particles of nanometer size are named as nanofluids, which have opened a new dimension in heat transfer processes. The recent investigations confirm the potential of nanofluids in enhancing heat transfer required for present age technology. The present investigation goes detailed into investigating the increase of thermal conductivity with temperature for nano fluids with water as base fluid and particles of Al_2O_3 or CuO as suspension material. A temperature oscillation technique is utilized for the measurement of thermal diffusivity and thermal conductivity is calculated from it. The results indicate an increase of enhancement characteristics with temperature, which makes the nanofluids even more attractive for applications with high energy density than usual room temperature measurements reported earlier. [DOI: 10.1115/1.1571080]

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Introduction

Heat transfer technology stands at the cross roads of miniaturization on one hand and astronomical increase in heat flux on the other. The usual enhancement techniques for heat transfer can hardly meet the challenge of ever increasing demand of heat removal in processes involving electronic chips, laser applications or similar high energy devices. The factors which limit the usual techniques are many folded. One major limitation is the poor thermal characteristics of usual heat transfer fluids. They are about two orders of magnitude less efficient in conducting heat compared to metals. This inherent inadequacy of these fluids makes the heat removal mechanism less effective even with the best utilization of their flow properties.

The idea of increasing thermal conductivity of fluids with conducting particles suspended on them is not new. Ahuja [1] and Liu et al. [2] carried out the studies on practical implication of hydrodynamics and heat transfer of slurries. However, the usual slurries with suspended particles of the order of micro to millimeters suffer from a number of drawbacks. The abrasive action of the particles causes erosion of components, clogging becomes a major problem in small flow passages and their requirement of momentum transfer increases the pressure drop considerably. In addition to these the tendency of micro/millimeter size particles to settle under gravity brings fouling and related problems. Thus eventhough the slurries have higher conductivities, they are hardly useable as heat transfer fluids from other technological considerations.

The above bottleneck of slurries with micron or bigger size particles can be removed by using particles of nanometer dimensions. The concept was first materialised by series of research works at the Argonne National Laboratory and probably Choi [3] was the first to call the fluids with particles of nanometer dimension suspended in them as "nano-fluids," which has gained popularity.

The subsequent study by Lee et al. [4] was a detail measurement of thermal conductivity of two types of nano particles ${\rm Al_2O_3}$ and CuO. The average weighted particle diameter they used were 23.6 nm for CuO and 38.4 nm for ${\rm Al_2O_3}$. These particles were used with two different base fluids—water and ethylene glycol to

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get four combinations of nanofluids. Using transient hot wire method, the thermal conductivities of the fluids were measured which showed significant improvement. The phenomenal increase in the thermal conductivities of the liquids with the addition of small volume of nano particles has created tremendous interest in this technique of heat transfer enhancement. The reason for this lies in the fact that due to small particle size and their small volume fraction problems such as clogging and increase in pressure drop become insignificant. The large surface area of the nano particles not only reduces the non equilibrium effect between fluid and solid but also increases the stability of the particles and sedimentation problem is greatly reduced. A more recent study by Xuan and Li [5] has shown that even with particles as large as 100 nm (which is rather sub-micron particles rather than nano particles) the stability can be achieved by using very meagre amount of laurate salt.

Thus the problems of traditional slurries can be eliminated by reducing the particles to nanometer dimensions. It must be kept in mind that the enhancement that is talked about in the above studies is only that of thermal conductivity. The real enhancement of heat transfer capability of these fluids when used under convective condition is expected to be much higher as the studies of Ahuja [1], Sohn and Chen [6] for laminar flow and that of Liu et al. [2] for turbulent flow indicate. This makes nanofluids a prospective candidate for cooling application such as energy intensive laser and X-ray applications, super conducting magnets, high speed computing systems, fibre manufacturing processes and high-speed lubrication applications.

The need of explaining the results observed above can be fulfilled by theorising them or fitting them to existing theories. The classical theory of thermal conductivity of fluid with suspended solid particles is from Maxwell [7] more than a century back which assumes the shape of particles to be spherical. This theory was subsequently modified for non-spherical particles by Hamilton and Crosser [8] for solid to liquid thermal conductivity ratio of more than 100. This formula was further confirmed by Wasp [9] for spherical particles. However measurement of Lee et al. [4] confirmed that even though the model of Hamilton and Crosser [8] agrees well with Al_2O_3 -water or ethylene glycol nanofluid, it fails in the case of nanofluids containing CuO nanoparticles. Thus, it can be said that simple model using the concept of suspensions of millimeters to micrometer dimensions are doubtful for application to nanofluids. The reason for the failure of Hamilton and

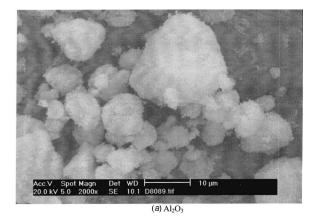
Crosser [8] model to CuO nanofluid may lie in the fact that the particles of Al₂O₃ used in the experiment were of 38.4 nm size while that of CuO was 23.6 nm. It is well known that the oxides of metals are thermally insulators when compared to pure metals. Hence even though difficult to produce, the recent efforts are towards using pure metal nano powders such as Cu-ethylene glycol or Cu-transformer oil nanofluids. Xuan and Li [5] could enhance the thermal conductivity of water by using Cu particles of comparatively large size (100 nm) to the same extent as has been done by oxide particles of Cu of much smaller dimension (36 nm). The more recent breakthrough was again from Argonne National Laboratory by Eastmann et al. [10] which brought out the astonishing finding that by using pure Cu nanoparticles of less than 10 nm size a phenomenal 40% increase in thermal conductivity can be attained with only 0.3% volume fraction of the solid. This indicates very clearly that Maxwell [7] or Hamilton Crosser [8] model breaks down completely with decreasing particles size in the nanometer range. Thus as has been felt by Eastman et al. [10] that the "anomalously increased" effective thermal conductivity of nanofluids with smaller particles cannot be explained by existing theories because they do not take into account the phenomenal increase of surface to volume ratio of particles with decreasing size.

All the investigation mentioned above measured thermal conductivity at room temperature and hence failed to have an insight to probable enhancement mechanism. Apart from this in all the cases above measurements were made by a particular variation of transient hot-wire method. Even though it has been claimed that this method is suitable for electrically conducting fluids, no discussion has been presented on the possible concentration of ions of conducting fluids around the hot wire due to the electrical field there. Thus, the present study aims at throwing some light on the enhancement mechanism of heat transfer by experimentally evaluating the temperature effect on thermal conductivity. The experimental method used here is based on the oscillation method proposed by Roetzel [11] and further developed by Czarnetski [12]. The method is purely thermal and the electrical components of the apparatus are away from the test sample which does not influence the ion movements. Finally the enhancements of conductivity of Al₂O₃ and CuO based water nano fluids at higher temperature will give more clear idea about the behavior of nano fluids at the practical range of application since all the single phase cooling application uses temperatures between room and saturation temperature.

Preparation and Characterization of Nanofluids

The preparation of nanofluid must ensure proper dispersion of nanoparticles in the liquid and proper mechanism such as control of pH value or addition of surface activators to attain the stability of the suspension against sedimentation. In the present experiment ultrasonic vibration is used for dispersing the particles. The nano particles were produced (by Nanophase Technologies Corporation) using physical vapor synthesis method. Under atmospheric condition these particles form loose agglomerates, which are of the order of micrometers as can be seen in the TEM photographs shown in Fig. 1. However they can be dispersed in the fluid quite successfully which results in breaking of the agglomerates to some extent giving particles of nanometer range as shown in Fig. 2. The particles of the nano powder show a lognormal size distribution. The size distribution of a typical sample is given in Fig. 3. The volume weighted average values of particle diameter for Al₂O₃ was 38.4 nm while that for CuO was 28.6 nm. As can be seen from Fig. 2 the agglomerates of nanoparticles after dispersion are much smaller compared to that for the power, which confirms a good dispersion in the liquid. However to compare the results with Lee et al. [4] no effort has been made to further break up the smaller agglomerates.

The dispersion of the particles was done by first mixing the required volume of the powder in the chemical measuring flask



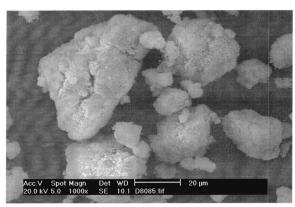
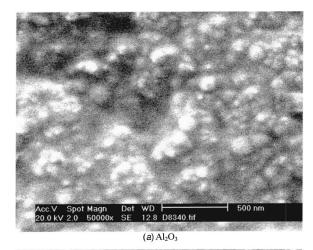


Fig. 1 TEM photographs of agglomerated powder of nano particles

with distilled water and then using Ultrasonic vibration to disperse it. It may be mentioned here that the true density of particles are more than 50 times the apparent density. Hence the volume of the solid was determined by calculating the equivalent weight the solid using the true density (neglecting the weight of air trapped inside) and using that weight the suspension was made. After making the proper mixture the flask was kept under ultrasonic vibration for 12 hours. After this no sedimentation was observed for the fluids for about next 12 hours and thereafter minor sedimentation were observed for 3% and 4% (volume) suspensions and none for 1% and 2% (volume) suspensions. Even though in practical applications it is expected to stabilize the particles with suitable third agent such as oleic acid or laurate salts, in the present case this has not been done. This is because of the fact that the time required for the experiments were much less than that required for the first sedimentation to occur and the addition of the third agent may influence the thermal conductivity of base fluid itself and thus the real enhancement by using nanoparticles may be over shadowed. To keep consistency with this, for each experiment freshly vibrated fluid was used so that the experimental time of 1.5 to 2 hours does not bring out sedimentation. As a cross check, the density of nanofluids was measured and was also calculated from the weight of powder alone and the volume of the suspension assuming the rest of the volume to be of water. The two calculations agreed excellently excluding any possibility of agglomerates with non wetted space in between. Thus nano fluids were prepared which are usable for conductivity measurement without any stabilising agent.

Measurement of Thermal Conductivity

Principle of Measurement. The measurement of thermal diffusivity and the thermal conductivity is based on the energy equation for conduction given by



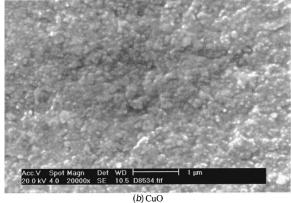


Fig. 2 TEM photographs of dispersed nano particles

$$\frac{1}{\alpha} \frac{\partial T}{\partial t} = \nabla^2 T \tag{1}$$

In the present case this equation is applied with the assumption that the test fluid is isotropic and the thermophysical properties are uniform and constant with time throughout the entire specimen volume. The cylindrical fluid volume considered for analysis with its boundaries is shown in Fig. 4. At the surfaces A and B, periodic temperature oscillations are generated with an angular velocity given by

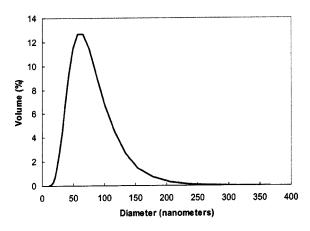


Fig. 3 Volume weighted particle size distribution for ${\rm Al_2O_3}$ particles

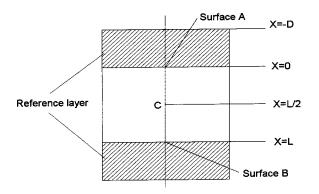


Fig. 4 The fluid volume for analysis

$$\omega = \frac{2\pi}{t_p} \tag{2}$$

Thus with the nondimensional space and time co-ordinates

$$\xi = x \sqrt{\frac{\omega}{\alpha}} \tag{3}$$

$$\tau = \omega \cdot t$$
 (4)

the governing Eq. (1) in its one dimensional form can be reduced to

$$\frac{\partial^2 T}{\partial \xi^2} = \frac{\partial T}{\partial \tau} \tag{5}$$

For the general case of input oscillations with same main frequency but different amplitude and phase at surfaces A and B, the boundary conditions are given by

$$T(\xi_0 = 0, \tau) = T_m + u_0 \cos(\tau + G_0) \tag{6}$$

$$T(\xi_L = L\sqrt{\omega/a}, \tau) = T_m + u_L \cos(\tau + G_L)$$
 (7)

Under steady periodic conditions the solution of the differential Eq. (5) with boundary condition given by Eqs. (6) and (7) can be obtained by using method of Laplace transform. The solution can be written in complex form as

$$T(\xi,\tau) = T_m + \frac{u_L e^{iG_L} \sinh(\xi \sqrt{i}) - u_0 e^{iG_0} \sinh(\sqrt{i}(\xi - \xi_L))}{\sinh(\xi_L \sqrt{i})} e^{i\tau}$$
(8)

The complex amplitude ratio between the mid point of the specimen and the surface can be given by

$$B^* = \frac{2u_L e^{iG_L}}{u_L e^{iG_L} + u_0 e^{iG_0}} \cosh\left[\frac{L}{2} \left(\frac{i\omega}{\alpha}\right)^{1/2}\right]$$
(9)

The real measurable phase shift and amplitude ratio can be expressed as

$$\Delta G = \arctan\left(\frac{\operatorname{Im}(B^*)}{\operatorname{Re}(B^*)}\right) \tag{10}$$

And

$$\frac{u_L}{u_{L/2}} = \sqrt{\text{Re}(B_R^*)^2 + \text{Im}(B_R^*)^2}$$
 (11)

By measuring phase and amplitude of temperature oscillation at the two surfaces as well as at the center (point C), the thermal diffusivity can be determined either from Eq. (10) or from Eq. (11). To measure the thermal conductivity directly from experiment, we must consider the temperature oscillation in the reference layer at the two boundaries of the test fluid. The frequency of temperature oscillation in this layer is also same as the frequency

generated in the Peltier element (described later) and that in the test fluid. The one dimensional heat conduction in the reference layer is given by

$$\frac{\partial^2 T_R}{\partial \zeta^2} = \frac{\partial T_R}{\partial \tau} \tag{12}$$

where
$$\zeta = x \sqrt{\frac{\omega}{\alpha_R}}$$
. (13)

The boundary conditions for the reference layer are

$$T_R(\zeta = 0, \tau) = T(\xi = 0, \tau)$$
 (14)

Interface temperature balance,

$$\lambda_R \sqrt{\frac{\omega}{\alpha_R}} \frac{\partial T_R}{\partial \zeta} \bigg|_{\zeta=0} = \lambda \sqrt{\frac{\omega}{\alpha}} \frac{\partial T}{\partial \xi} \bigg|_{\xi=0}$$
 (15)

Interface flux balance.

The solution of Eq. (12) along with boundary condition (14) and (15) is given by

$$T_R^*(\zeta, \tau, \xi_L) = T_m + u_0 e^{i(\tau + G_0)} \cosh(\zeta \sqrt{i}) + C[u_R e^{i(\tau + G_R)}]$$

$$-u_0 e^{i(\tau + G_0)} \cosh(\xi_L \sqrt{i}) \right] \frac{\sinh(\zeta \sqrt{i})}{\sinh(\xi_L \sqrt{i})}$$
 (16)

Where

$$C = \frac{\lambda}{\lambda_R} \sqrt{\frac{\alpha_R}{\alpha}} \tag{17}$$

The complex amplitude ratio in this case between x = -D (*D* being the thickness of reference layer) and x = 0 is given by

$$B_R^* = \cosh(\zeta_D \sqrt{i}) - C \sinh(\zeta_D \sqrt{i})$$

$$\times \left[\frac{(u_L/u_0)e^{i(G_L - G_0)} - \cosh(\xi_L \sqrt{i})}{\sinh(\xi_L \sqrt{i})} \right]$$
(18)

The real phase shift and amplitude attenuation is giving by

$$\Delta G_R = \arctan\left(\frac{\operatorname{Im}(B_R^*)}{\operatorname{Re}(B_R^*)}\right) \tag{19}$$

and

$$\frac{u_D}{u_0} = \sqrt{\text{Re}(B_R^*)^2 + \text{Im}(B_R^*)^2}$$
 (20)

the thermal diffusivity of the test fluid has already been measured as described earlier and that of the reference layer being known, the thermal conductivity of the specimen can be evaluated from above formula.

Data Reduction

The temperature oscillation generated by the Peltier element requires to be strictly periodic. The shape of the oscillation is immaterial since any periodic oscillation can be expanded by Fourier series in the form

$$T(\tau) = \frac{a_0}{2} + \sum_{k=1}^{\infty} A_k \sin(k\tau + \theta_k)$$
 (21)

where

$$a_k = \frac{1}{\pi} \int_0^{2\pi} T(\tau) \cos(k\tau) d\tau, \quad k = 1, 2, 3, \dots$$
 (22)

$$b_k = \frac{1}{\pi} \int_0^{2\pi} T(\tau) \sin(k\tau) d\tau, \quad k = 0, 1, 2, 3, \dots$$
 (23)

and

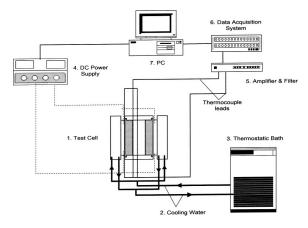


Fig. 5 Schematic of the experimental setup

$$A_k = \sqrt{(a_k^2 + a_k^2)}, \quad \tan G_k = \frac{a_k}{b_k}$$
 (24)

In the solution presented for fluid as well as the reference layer earlier the fundamental oscillation is considered and the coefficients a_k and b_k are evaluated by numerical integration at the appropriate location to yield the corresponding amplitude attenuation and phase shift.

The Experimental Set-up and Procedure

The experimental setup has been shown schematically in Fig. 5. As has been stated earlier a temperature oscillation technique which is a modification of that used by Czarnetzky and Roetzel [12] has been used. This technique required a specially fabricated test cell (1) which is cooled by cooling water (2) on both of the ends coming from a thermostatic bath (3). Electrical connection provides power to the Peltier element which is a DC power obtained through a converter (4). The temperatures are measured in the test section (discussed later) through a number of thermocouples and these responses are amplified with amplifier (5) followed by a filter which is finally fed to the data acquisition system (6) comprising of a card for logging the measured data. The data logger is in turn connected to a computer with proper software (7) for online display which is required to assess the steady oscillation and for recording data. Since in the present experiments the prime objective is to observe the effect of temperature on the enhancement of thermal conductivity, the control of temperature of the fluid is important which is effected by proper adjustment of the cooling water from the thermostatic bath. However for higher temperatures it is sometimes necessary to increase the input voltage to attain the required temperature level which is then fine tuned to the required temperature by control of cooling water temperature.

The test section is a flat cylindrical cell as shown in Fig. 6. The cell is mounted with its axis in horizontal position. The frame of the cell is made of POM (Polyoxymethylene), which can be machined accurately and simultaneously acts as the first layer of insulation. The frame consists of the main part with a 40 mm hole in it which makes the cavity to hold the test fluid, as well as the two end plates which sandwiches the water cooler and the Peltier element at both ends. The hole in the main frame is closed from both sides with disc type reference material of 40 mm diameter and 15 mm thickness. Thus the space for the test fluid is formed which has a dimension of 40 mm dia and 8 mm thickness. The fluid is filled through a small hole in the body of the cell. The measurement of temperatures are made at three locations—at the interface of the Peltier element and the reference layer, at the interface of the reference layer and test fluid and the central axial plane of the test fluid. For this purpose Ni-CrNi thermocouples of 0.1 mm diameter were used at the interfaces and 0.5 mm diameter

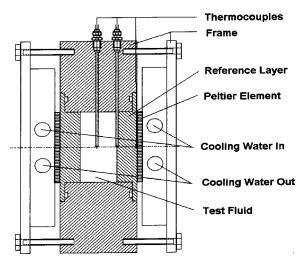


Fig. 6 The test cell construction

at the central plane from its stability consideration. The thermocouples at the interfaces are put in small groove and welded at the tip while that at the center hangs from the wall. Before putting the end reference plates, the central position of the thermocouple was ensured through a precision measurement. The entire cell was insulated further. The temperature of reference material was given periodic oscillation by two Peltier elements (40 mm \times 40 mm square) from two ends. The temperature oscillation in this element are controlled to obtained two objectives:

- 1) The oscillation amplitude is adjusted to be kept small enough (of the order of 1.5 K) within the test fluid to retain constant fluid properties and to avoid natural convection on one hand and on the other it is not allowed to decrease too much so that the accuracy of the measurement is affected. The Grashof number was calculated to be 850, which is below the asymptotic limit for onset of natural convection. The measurement with pure water of known conductivity reconfirmed that no natural convection was present.
- 2) The smaller amplitude and accurate adjustment of the mean temperature of oscillation ensures that for the conducting fluid, test is made at the sought temperature.

For example a typical temperature oscillation recorded at the locations after steady oscillation are reached is shown in Fig. 7. It can be seen that the amplitude of the temperature oscillation produced by the Peltier element gets attenuated and its phase gets shifted as it crossed the reference material. It is further attenuated and shifted as it reaches the center of the test fluid. The theoretical principle presented earlier reveals that it is possible to evaluate the

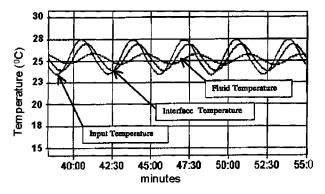


Fig. 7 Amplitude attenuation of source temperature oscillation in reference layer and in test fluid

thermal diffusivity of the fluid very accurately by considering amplitude attenuating of thermal oscillation from the boundary (fluid reference material interface) to the center of the fluid. However for direct measurement of thermal conductivity one has to consider the attenuation at the reference material as well. Since the reference material has been worked upon and the microcracks and inhomogenity of material brings out uncertainty in its thermal conductivity value, direct evaluation of thermal conductivity of fluid is less accurate. Hence, in the present measurement the value of thermal diffusivity for the nanofluid is evaluated from experiment. Subsequently the density has been measured and specific heat is calculated from handbook, as

$$C_{p,nf} = \frac{m_s C_{p,s} + m_w C_{p,w}}{m_s + m_w}$$
 (25)

Finally the thermal conductivity is calculated from

$$\lambda_{nf} = \alpha_{nf} \rho_{nf} C_{p,nf} \tag{26}$$

Error Estimates

For measurement of thermal diffusivity, the main sources of experimental uncertainty are the accuracy of thermocouples and the location of temperature measurement. In the present measurement thermocouples with an accuracy of 0.1 K were used and location was determined with an accuracy 1.25%. Over the measured temperature range the accuracy of temperature measurement came out to be 3.4%. This limits the combined uncertainty of measurement within an acceptable limit of 5%. It was observed that for using method to determine thermal conductivity directly one has to take care of uncertainty of the thermal conductivity of the reference layer, which is more than 5%. This makes the measurement more inaccurate and hence has not been used here. As a safeguard against experimental error the experimental setup described above was first calibrated by measuring thermal diffusivity of demineralised and distilled water over a wide range of temperature. It was found that over the temperature range of 20°C to 50°C the average deviation of thermal diffusivity from the standard values of handbook (VDI Wärmeatlas [13]) was 2.7%. Over the range from 20 to 30°C the maximum error was found to be limited to 2.11%. The maximum error was limited to 7% up to temperature 50°C. This gave an indication about the accuracy of the measurement and showed that over the entire range of measurement an acceptable range of accuracy existed. The values of error in thermal conductivity calculated from error analysis in the previous section also matches well with the observed values. The enhancement thermal conductivity values of nano fluids are moderate increase of 2% to 36% over the base value of water and hence the present ranges of accuracy is acceptable for the present

Results and Discussion

To begin with measurements were done at room temperature for ${\rm Al_2O_3}$ and CuO nanofluids of water at various particles volume concentrations. The results presented in Fig. 8 show an excellent agreement with the measurement of Lee et al. [4] considering the fact that minor differences in particles size existed between them. The room temperatures were also not identical in the two cases, still the present measurements confirm the same level of enhancement of thermal conductivity as observed by the Argonne group using an entirely different measurement technique.

Subsequently measurements were made for the nano fluids of Al_2O_3 -water and CuO-water with different particles concentration and most importantly at different temperatures, which is the main objective of the present work. Figure 9 shows the enhancement of thermal conductivity of Al_2O_3 based nanofluids with temperature. It is interesting to see both for 1% and 4% (volume) particle concentrations there is a considerable increase in the enhancement from $21^{\circ}C$ to $51^{\circ}C$. With 1% particles at room temperature ($21^{\circ}C$)

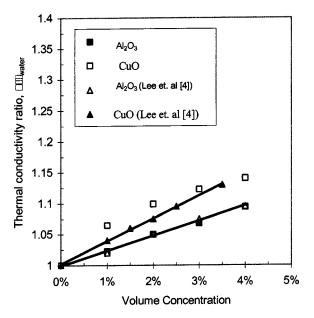


Fig. 8 Enhancement of thermal conductivity at room temperature

the enhancement in only about 2%, but at 51°C this value increases to about 10.8%. Thus the present measurement shows that in practical heat transfer application the enhancement achieved by adding small volume of nano particles is considerably higher compared to that believed at present [4]. The measurement with 4% concentration shown in the same figure (Fig. 9) shows one more interesting feature. Here the enhancement goes from 9.4% to 24.3% with temperature rising from 21°C to 51°C . The average

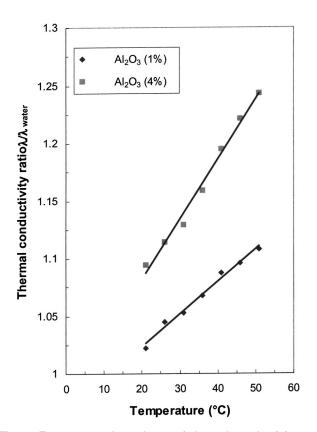


Fig. 9 Temperature dependence of thermal conductivity enhancement for water— Al_2O_3 nanofluids

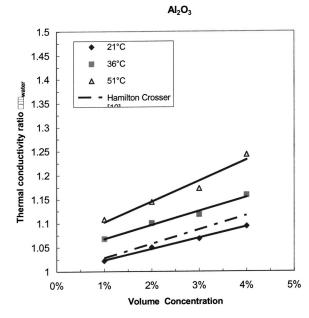


Fig. 10 Enhancement of thermal conductivity of water—Al₂O₃ nanofluids against particles concentration and temperature and comparison with Hamilton-Crosser [8] model

rate of increase of enhancement in this case in much higher compared to that of the 1% nanofluids, which can be observed from the increased slope of the fitted line of the two nano fluids shown. Thus it can be said that the enhancement of thermal conductivity shows a dramatic increase with temperature and the rate of this increase depends on the concentration of nano particles. As has been expressed by Lee et al. [4], the Hamilton–Crosser [8] model given by effective conductivity in the form

$$\frac{\lambda_{w}}{\lambda_{w}} = \frac{\lambda_{s} + (n-1)\lambda_{w} - (n-1)\phi(\lambda_{w} - \lambda_{s})}{\lambda_{a} + (n-1)\lambda_{w} + \phi(\lambda_{w} - \lambda_{s})}$$
(27)

is agreed upon by Al₂O₃ based nano fluids at room temperature. This is somewhat contrary to the results of CuO based nanofluid which did not agree with the model. Figure 10 shows that this agreement for Al₂O₃-water is somewhat accidental because the agreement is only good at room temperature. At elevated temperature even Al₂O₃-water nanofluid disagree with Hamilton-Crosser model [8] because this model hardly changes the effective conductivity with temperature (the change in λ/λ_w curve with temperature for this model [8] cannot be distinguished). This brings out some insight to the mechanism for thermal conductivity enhancement in nanofluids. In ordinary slurries the thermal conductivity is increased due to higher thermal conductivity of the solid particles and hence the combine effective conductivity is some kind of average of the liquid and solid conductivity. The Hamilton-Crosser [8] is a kind of such average, weighted according to the particles shape.

In nanofluid the main mechanism of thermal conductivity enhancement can be thought as the stochastic motion of the nano particles. Presumable this Brownian like motion will be dependent on fluid temperature and so this amount of enhancement with temperature is quite explicable for Al₂O₃ since the particles size (both for Lee et al. [4] and present study) was bigger. At low temperature this motion was less significant giving the characteristics of normal slurries which rapidly changed at elevated temperature bringing more nanoeffect in the conducting behavior of the fluid. This explanation also indicates the reason for the "anomalously increased" conductivity of nano fluid containing Cu particles of less than 10 nm as observed by Eastman et al. [10]. The main mechanism of enhancement of thermal conductivity there is the increased stochastic motion of the nanoparticles

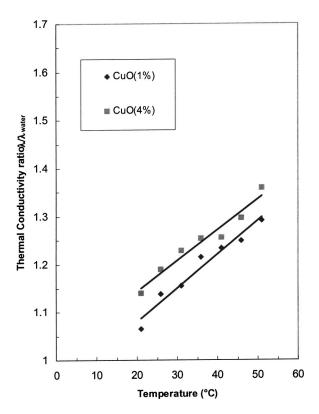


Fig. 11 Temperature dependence of thermal conductivity enhancement for water—CuO nanofluids

rather than the thermal conductivity of Cu because the volume fraction used (0.5%) was too small to bring about such an effect by any kind weighted average. Thus the present results indicate that it is possible to have a threshold temperature corresponding to each particle size at which the effective thermal conductivity of nanofluids starts deviating from that of usual slurries and the enhancement through stochastic motion of the particles start dominating. The measurements with CuO-water nanofluid show nano effect even at room temperature and in this case the enhancement of the conductivity is much higher as shown in Fig. 11. The Hamilton-Crosser [8] model gives a value less than that measured at room temperature. The measurement of enhancement of conductivity with particle concentration at different temperature presented in Fig. 12 confirms this and indicates the necessity for better theoretical model for these for the entire range of temperature. In this case the effect of temperature on thermal conductivity enhancement is even more dramatic as it climbs from 6.5% to 29% (for 1% particle concentration) and from 14% to 36% (for 4% particle concentration). However, in the case of CuO water nanofluid the rate of change of enhancement with temperature did not change as much with concentration as that observed for Al₂O₃-water nanofluid in the present measurements. At this point, we would like to express that the Hamilton Crosser model for CuO as calculated by Lee et al. [4] seems to be incorrect since the curve given by them corresponds to a particle conductivity of less than 2 W/m K which is much smaller than value in literature $(\sim 17.65 \text{ W/m K})$ [14]. For convenience we have indicated both the curves in Fig. 12.

In general, the effect of particle concentration was found to be less for CuO water nanofluid compared to $\mathrm{Al_2O_3}\text{-}\mathrm{water}$ nanofluids which can be attributed to the larger particle size of $\mathrm{Al_2O_3}$. On the other hand the effect of temperature was found to be more predominant in CuO water fluid which is due to the stochastic motion of the particles which arguably will be more mobile with smaller particle size.

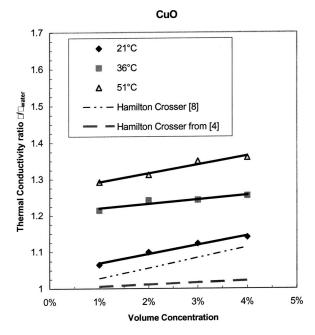


Fig. 12 Effect of particle concentration on the thermal conductivity enhancement of water-CuO nanofluids at different temperature and comparison with Hamilton-Crosser [8] models of [4] and calculated by us

Conclusion

The temperature effect of thermal conductivity enhancement in nanofluids has been presented through an experimental investigation. A purely thermal method consisting of temperature oscillations at the fluid sample has been used for this purpose. The method has been found to be appropriate for nano fluids both from accuracy and prevention of natural convection point of view. The measurement confirmed the level of thermal conductivity enhancement at room temperature as observed by others. It was further observed that a dramatic increase in the enhancement of conductivity takes place with temperature. It is observed that a 2 to 4 fold increase in thermal conductivity enhancement of nanofluids can take place over a temperature range of 21°C to 51°C. This finding makes nanofluids even more attractive as cooling fluid for devices with high energy density where the cooling fluid is likely to work at a temperature higher than the room temperature. It has been observed that nanofluids containing smaller CuO particles show more enhancement of conductivity with temperature. However the enhancement is considerably increased for nanofluids with Al₂O₃ as well. The effect of particle concentration was observed to be more for the Al₂O₃-water system. The measurements indicate that particle size is an important parameter for the observed behavior and the usual weighted average type of model for effective thermal conductivity is a poor approximation of the actual enhancement particularly at the higher temperature range. The results indicate that a stochastic motion of nano particles can be a probable explanation if future theoretical studies can confirm it.

The main limitation of the previous and the present experiments is the non-availability of nano particles of different sizes for same material which can conclusively indicate the effect of particle size. However the present study brings out the important aspect of temperature dependence of conductivity enhancement which can be used as an important source for a comprehensive theoretical treatment in the future.

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Nomenclature

B =the amplitude ratio

 $B^* = \text{complex amplitude ratio}$

C = constant

 C_p = specific heat, kJ/kg.K

 \vec{D} = thickness of reference layer, m

G = the phase shift

L = thickness of fluid sample, m

m = mass, kg

n = shape factor

T = temperature, K

t = time, s

 $t_p = \text{period, s}$

u = amplitude, K

x = space coordinate, m

Greek Symbols

 α = thermal diffusivity, m²/s

 Δ = difference

 ξ , ζ = dimensionless space coordinates

 λ = thermal conductivity, W/(m K)

 ω = the constant angular frequency, 1/s

 ρ = the density, kg/m³

 $\tau =$ dimensionless time (angle)

Subscripts

m = mean

nf = nanofluids

R = reference layer s = solid (particle)

w =water (base fluid)

References

- Ahuja, A. S., 1975, "Augmentation of Heat Transport in Laminar Flow of Polystyrene Suspension: Experiments and Results," J. Appl. Phys., 46(8), pp. 3408–3416.
- [2] Liu, K. V., Choi, U. S., and Kasza, K. E., 1988, "Measurement of Pressure Drop and Heat Transfer in Turbulent Pipe Flows of Particulate Slurries," Argonne National Laboratory Report, ANL-88-15.
- [3] Choi, U. S., 1995, "Enhancing Thermal Conductivity of Fluids With Nanoparticles," *Developments and Applications of Non-Newtonian Flows*, D. A. Siginer and H. P. Wang, eds., FED-vol. 231/MD-Vol. 66, ASME, New York, pp. 99–105.
- [4] Lee, S., Choi, U. S., Li, S., and Eastman, J. A., 1999, "Measuring Thermal Conductivity of Fluids Containing Oxide Nanoparticles," ASME J. Heat Transfer, 121, pp. 280–289.
- [5] Xuan, Y., and Li, Q., 2000, "Heat Transfer Enhancement of Nanofluids," Int. J. Heat Fluid Flow, 21, pp. 58–64.
- [6] Sohn, C. W., and Chen, M. M., 1981, "Microconvective Thermal Conductivity in Disperse Two Phase Mixture as Observed in a Low Velocity Couette Flow Experiment," ASME J. Heat Transfer, 103, pp. 47–51.
- [7] Maxwell, J. C., 1881, A Treatise on Electricity and Magnetism, 2nd Ed., 1, Clarendon Press, Oxford, U.K., p. 435.
- [8] Hamilton, R. L., and Crosser, O. K., 1962, "Thermal Conductivity of Heterogeneous Two Component Systems," I & EC Fundamentals, 1(3), pp. 187–191.
- [9] Wasp, F. J., 1977, "Solid-Liquid Slurry Pipeline Transportation," Trans. Tech., Berlin.
- [10] Eastman, J. A., Choi, U. S., Li, S., Yu, W., and Thompson, L. J., 2001, "Anomalously Increased Effective Thermal Conductivities of Ethylene Glycol-Based Nanofluids Containing Copper Nanoparticles," Appl. Phys. Lett., 78(6), pp. 718–720.
- [11] Roetzel, W., Prinzen, S., and Xuan, Y., 1990, "Measurement of Thermal Diffusivity Using Temperature Oscillations," *Thermal Conductivity 21*, C. Y, Cremers and H. A. Fine, eds., Plenum Press, New York and London, pp. 201–207.
- [12] Czarnetzki, W., and Roetzel, W., 1995, "Temperature Oscillation Techniques for Simultaneous Measurement of Thermal Diffusivity and Conductivity," Int. J. Thermophys., 16(2), pp. 413–422.
- [13] VDI-Wärmeatlas, 1997, 8th Ed. VDI Verlag GmbH, Düsseldorf.
- [14] Bolz, R., and Tuve, G., 1970, Handbook of Tables for Applied Engineering Science, The Chemical Rubber Co.