LIMITS FOR THERMAL CONDUCTIVITY OF NANOFLUIDS

by

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Nanofluids have offered challenges to thermal engineers and attracted many researchers over the past decade to determine the reasons for anomalous enhancement of thermal conductivity in them. Experiments on measurement of nanofluid thermal conductivity have ended in a large degree of randomness and scatter in their values. Hence in this paper, lower and upper limits for thermal conductivity of nanofluids are developed. The upper limit is estimated by coupling heat transfer mechanisms like particle shape, Brownian motion and nanolayer while the lower limit is based on Maxwell's equation. Experimental data from a range of independent published sources is used for validation of the developed limits.

Key words: *nanofluids, Brownian motion, nanolayer, thermal conductivity, volume concentration.*

Introduction

The thermal loads are increasing in a wide variety of applications like microelectronics, transportation, lighting, utilization of solar energy for power generation *etc*. Micro electromechanical systems (MEMS) technology and nanotechnology are also rapidly emerging as a new revolution in miniaturization. Hence the management of high thermal loads in these systems offers challenges and the thermal conductivity of heat transfer fluid have become vital. Traditional heat transfer fluids such as water, engine oil, and ethylene glycol (EG) are inherently poor heat transfer fluids with low thermal conductivities of 0.613, 0.145, and 0.253 W/mK, respectively, and thus major improvements in cooling capabilities have been constrained. To overcome this limited heat transfer capabilities of these traditional heat transfer fluids, micro/millimeter sized particles with high thermal conductivity suspended in them were considered by Ahuja [1]. Heat transfer fluids containing suspended particles of micro/millimeter sizes suffered from numerous drawbacks like erosion of the components by abrasive action, clogging in small passages, settling of particles and increased pressure drop. Hence they are not accepted as suitable candidate for heat transfer enhancement and the search for new heat transfer fluids continued. Nanotechnology has come to rescue by providing opportunities to process and produce materials of sizes in nanometer range which

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can be suspended in traditional heat transfer fluids to produce a new class of engineered fluids with high thermal conductivity. In addition, due to small size of the nanoparticles, the problems of clogging, settling and increased pressure drop can also be eliminated. This new class of heat transfer fluids with nanoparticles in suspension is called nanofluid.

Masuda *et al*. [2] were the first to conduct experiment to show that there was alteration in the values of thermal conductivity and viscosity of liquids containing dispersed ultra fine particles of 13 nm size. However, the concept of nanofluids was first materialized by Choi [3] after performing a series of research works at Argonne National Laboratory of USA. Subsequent researches [4-6] have showed that the nanofluids containing Al_2O_3, CuO, Cu, and TiO_2 nanoparticles in water, ethylene glycol, engine oil exhibited higher thermal conductivity even for low concentration of suspended nanoparticles. Hence recently, many theoretical studies have been carried out to predict the anomalously increased thermal conductivity of nanofluids. A detailed summary of all classical and recently developed models for the prediction of the effective thermal conductivity of nanofluids has been provided by Murshed *et al*. [7]. A long list of physical phenomena has been proposed for explaining the experimentally observed enhancement of effective thermal conductivity of nanofluids which include size and shape effects, fluid temperature, agglomeration, clustering of particles, interfacial resistance, Brownian motion of nanoparticles resulting in micro convection, phonon dispersion, and liquid layering at the particle surface. Some authors [8-11] state Brownian motion of particles as a prime factor of the thermal enhancement while others [12, 13] have considered the effect of the interfacial layer between the fluid and the particle. Only a few results have reported so far regarding the dependence of the effective thermal conductivity of nanofluids on temperature. Das *et al*. [14] experimentally investigated the effect of temperature on the thermal conductivity of nanofluids and reported a two to four fold increase in thermal conductivity enhancement for nanofluids over a temperature range of 21-51 °C. Recently, Mintsa *et al*. [15] have presented some new experimental data on the temperature dependence of the thermal conductivity of alumina and copper oxide based nanofluids. Results clearly suggest that there is a relative increase in thermal conductivity of nanofluids at higher temperatures as well as with smaller diameter particles.

Recent review on experimental studies on thermal conductivity of nanofluids clearly showed that there exists inconsistency in the reported results of various research groups [7]. The experimental observations have led to the conclusion that even well-established classical models of Maxwell and Hamilton Crosser (HC) are not capable of explaining the thermal conductivity enhancements of nanofluids. The results of the existing theoretical models for calculating nanofluid thermal conductivity seem to agree with experimental data of a certain group of authors is found to disagree with data and correlations of other authors. It is also understood that due to large level of scatter and inconsistency in the published data, the development of a comprehensive model which can explain all the trends is a difficult task at the present time. Hence in this paper, we have developed more restrictive lower/upper limits for the thermal conductivity of nanofluids and compared them with the published experimental data.

Development of limits for nanofluid thermal conductivity

Lower limit

Maxwell model [16] was developed to determine the effective electrical or thermal conductivity of liquid-solid suspensions. This model is applicable to statistically homogeneous and low volume concentration liquid-solid suspensions, with randomly dispersed and uniformly sized noncontacting spherical particles. It is given as:

$$\frac{k_{\mathrm{eff}}}{k_{\mathrm{l}}} = \frac{k_{\mathrm{p}} + 2k_{\mathrm{l}} + 2\phi(k_{\mathrm{p}} - k_{\mathrm{l}})}{k_{\mathrm{p}} + 2k_{\mathrm{l}} - \phi(k_{\mathrm{p}} - k_{\mathrm{l}})} \tag{1}$$

Experiments report thermal conductivity enhancement of nanofluids beyond the Maxwell limit of 3ϕ. In the limit of low particle volume concentration (ϕ) and the particle conductivity (k_{p}), being much higher than the base liquid conductivity (k_{l}), eq. (1) can be reduced to Maxwell 3ϕ limit as:

$$k_{\mathrm{low}} = \frac{k_{\mathrm{eff}}}{k_{\mathrm{l}}} = 1 + 3\phi \tag{2}$$

Equation (1) represents the lower limit for the thermal conductivity of nanofluids and it can seen that in the limit where $\phi = 0$ (no particles), eq. (2) yields $k_{\mathrm{low}} = 1$ as expected.

Upper limit

An upper bound for thermal conductivity of nanofluid is established by coupling the heat transport mechanisms like particle shape, nanolayer thickness in the particle fluid interface and Brownian motion which are expected to enhance the thermal conductivity of nanofluid.

Brownian motion by which particles move through liquid and possibly collide, thereby enabling direct solid-solid transport of heat from one to another particle can be expected to increase the thermal conductivity of the nanofluids. It is believed that the Brownian motion contribution to thermal conductivity increases with rising temperature and decreasing particle size. Shukla *et al*. [17] proposed the following equation based on the Brownian motion of the nanoparticles in a homogeneous liquid and Maxwell model:

$$k_{eff} = k_{\mathrm{l}} \frac{k_{\mathrm{p}} + 2k_{\mathrm{l}} + 2\phi(k_{\mathrm{p}} - k_{\mathrm{l}})}{k_{\mathrm{p}} + 2k_{\mathrm{l}} - \phi(k_{\mathrm{p}} - k_{\mathrm{l}})} + C\frac{\phi(T - T_0)}{\mu a^4} \tag{3}$$

where C is a constant whose value is fitted with experimental data and adjusted to $7\cdot 10^{-36}$, and T_0 is the reference temperature equal to 294 K. Reasonable agreement is found by them between the predicted values and the experimental data. It can be noted from eq. (3) that the first term represents the contribution due to macroscopic Maxwell model whereas the second term represents the contribution due to Brownian motion of nanoparticles.

The ordered layering of liquid molecules at the solid particle surface is commonly referred as nanolayer. This layer acts as a thermal bridge between the solid nanoparticles and the base liquid. Hence it may become an important mechanism in enhancing the thermal conductivity. Yu *et al*. [13, 18] modified Maxwell and HC model to account for the effect of nanolayer. They replaced the thermal conductivity and volume concentration of nanoparticles, respectively, in the Maxwell model with the thermal conductivity and volume fraction of equivalent particles (*i. e.* particle with nanolayer). On this basis, Avsec *et al.* [19] have derived the following equation based on HC model for thermal conductivity of nanofluids.

$$\frac{k_{\mathrm{eff}}}{k_{\mathrm{l}}} = \frac{k_{\mathrm{p}} + (n-1)k_{\mathrm{l}} + (n-1)(1+\beta)^3\phi(k_{\mathrm{p}} - k_{\mathrm{l}})}{k_{\mathrm{p}} + (n-1)k_{\mathrm{l}} - (1+\beta)^3\phi(k_{\mathrm{p}} - k_{\mathrm{l}})} \tag{4}$$

The shape of the nanoparticles is taken in account in the form of an empirical shape factor ($n = 3/\psi$) while calculating the thermal conductivity of nanofluids. The ratio of the surface area of a sphere with a volume equal to that of the particle to the surface area of the particles is defined as sphericity (ψ). For spherical particles, sphericity takes the value of 1 whereas for non spherical particles it may vary from 0.5 to 1. Thus n takes the value of 3 for spherical particles and with no nanolayer (the ratio of nanolayer thickness to particle radius, $\beta = 0$), eq. (4) reduces to Maxwell's equation given in eq. (1).

The upper limit for thermal conductivity can now be established by replacing the first term of eq. (3) which does not include the particle shape and nanolayer with eq. (4). Thus the upper limit on effective thermal conductivity can be established as:

$$k_{\mathrm{upp}} = \frac{k_{\mathrm{eff}}}{k_1} = \frac{k_{\mathrm{p}} + (n-1)k_1 + (n-1)(1+\beta)^3\phi(k_{\mathrm{p}} - k_1)}{k_{\mathrm{p}} + (n-1)k_1 - (1+\beta)^3\phi(k_{\mathrm{p}} - k_1)} + C\frac{\phi(T - T_0)}{\mu a^4 k_1} \tag{5}$$

Note that the first term of the above equation accounts the effect of particle shape and the nanolayer while the second term accounts the effect of Brownian motion. It was found that for $\psi = 0.7$, the theoretical estimation of thermal conductivity of nanofluid was close to the experimental data [20]. Assuming a nanolayer thickness of 1 nm and particle radius of 10 nm, the ratio of nanolayer thickness to particle radius (β) can be calculated as 0.1 [21]. Hence ψ and β are assigned with the values of 0.7 and 0.1, respectively, in the present work.

Discussion

Figures 1(a)-(f) show the comparison between the developed lower/upper limits for thermal conductivity, that is, eq. (2) and eq. (5) with experimental data. The data are collected from several sources and categorized based on the nanoparticle material and the base fluids, which include Al_2O_3, CuO, and TiO_2 particles dispersed in water and ethylene glycol over a range of the volume concentration (0-5%), particle size (13-38.4 nm), and temperature (21-51 °C). It is interesting to observe that the majority of the experimental data lie between the lower and upper limits of thermal conductivity. This indicates that the limits developed in this paper are capable of providing a tight and narrow range for enhancement of thermal conductivity of nanofluids. This is because the developed limits include the heat transport mechanisms like particle shape, Brownian motion and nanolayer in addition to volume concentration. This also validates our assumption of considering these heat transport mechanisms in setting the upper limit for thermal conductivity of nanofluids. It is evident from fig. 1 that at 5% volume concentration, a maximum of 30-35% and 50% enhancement in thermal conductivity is possible for water and ethylene glycol based nanofluids, respectively. Figure 2 shows that the present limits are also capable of setting limits for temperature dependent nanofluid thermal conductivity. As temperature is increased, there is a drift in the values of thermal conductivity of nanofluid from lower limit to upper limit. This can be attributed to the reason that as the temperature is increased, the Brownian motion of the nanoparticle is enhanced resulting in rapid mixing within nanofluid. Also due to decrease in viscosity with an increase in temperature, the effect of second term of eq. (5) which represents the effect of Brownian motion on thermal conductivity of nanofluid becomes important leading to enhanced heat transport in nanofluids.

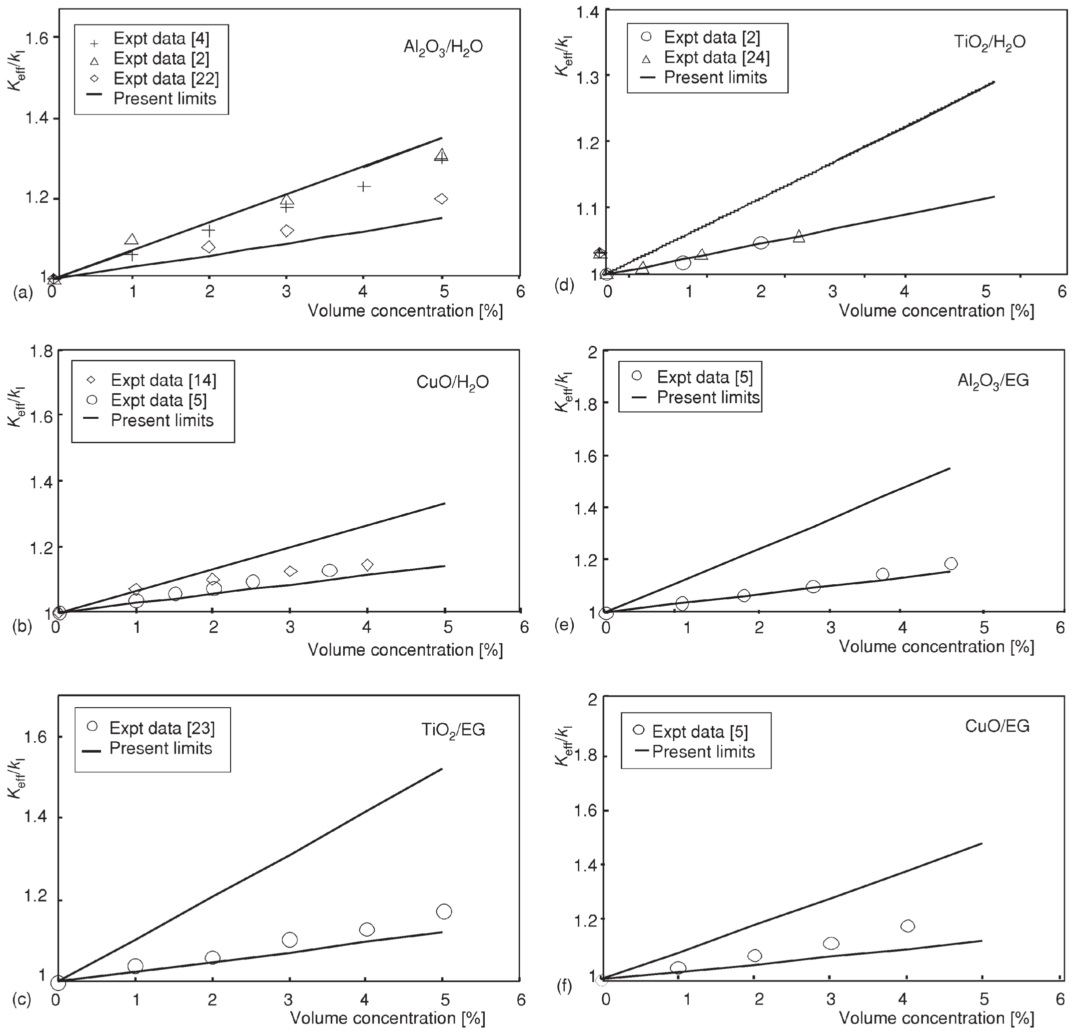

Figure 1. Comparison of present limits for thermal conductivity of various nanofluids

Conclusions

In this paper, we have developed lower/upper limits for thermal conductivity of nanofluids and compared these bounds with the published experimental data. The comparison indicates that the experimental data considered lie between the newly developed limits. Comparison also revealed that the present limits are more rigorous in placing a narrow lower and upper limit. As most of the experimental data lies within the newly developed limits, it can be concluded that particle shape, Brownian motion and nanolayer are significant in enhancing the thermal conductivity of nanofluids. With a better understanding of the role of these parameters, it will be possible to develop a more realistic theoretical model to predict the thermal conductivity of nanofluids.

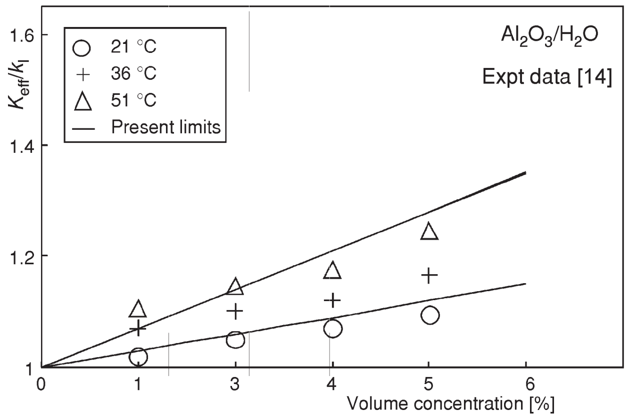

Figure 2. Comparison of the present limits with temperature dependent thermal conductivity data for Al_2O_3/water nanofluid

Nomenclature

a – particle radius, [m]
k – thermal conductivity, [$Wm^{-1}K^{-1}$]
n – empirical shape factor, [–]
T – temperature, [K]
T_0 – reference temperature, [K]

Greek letters

β – ratio of nanolayer thickness to particle radius, [–]
μ – dynamic viscosity, [Nsm^{-2}]
ϕ – volume concentration/fraction, [–]
ψ – sphericity, [–]

Subscripts

eff – effective
l – base liquid
low – lower limit
p – particle
upp – upper limit

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