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INVESTIGATIONS ON THE NANOLAYER HEAT TRANSFER IN NANOPARTICLES-IN-LIQUID SUSPENSIONS

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ABSTRACT

Experiments have shown that nanoparticles-in-liquid suspensions (nanofluids) have higher thermal conductivities compared to the base fluids. Thus, applications of nanofluids hold enormous promise for industrial thermal energy management and similar functions. Possible parameters responsible for this increase were reviewed here leading to the development of a new correlation for the effective thermal conductivity of the nanofluid. Results show that although the thermal conductivity of the nanolayer significantly contributes to the effective thermal conductivity of the nanofluid, the nature of its variation in the nanolayer is not significant to the contribution. Results using the correlation were compared to experimental results and correlations by other researchers. A parametric study was also performed to understand how a number of factors affect thermal conduction in nanofluids. Significant factors that influence the thermal conductivity of nanofluids were determined.

Keywords: nanofluids, heat transfer, thermal conductivity, nanolayer, nanoparticles.

INTRODUCTION

A nanofluid is produced by dispersing solid nanoparticles in a liquid. Experiments have shown that nanofluids have higher thermal conductivities than the base fluids. Thus, their exploitation has potential remarkable impact in applications such as energy systems, and drug delivery in body tissues. Nanofluids can be described as colloids since a colloid is a substance made up of a system of particles that is insoluble yet remains in solution and dispersed in another liquid medium. The concept of enhancing the thermal conductivity of liquids by suspending solid particles in them could be traced to the theoretical work by Maxwell [1]. Other studies on suspensions of solid particles (millimeter and micron-sized) in liquids for heat transfer and other applications have been reported. Lee *et al*., [2] produced copper oxide and aluminum oxide nanofluids and measured their thermal conductivities using the transient hotwire method. The results showed that nanofluids containing a small amount of nanoparticles have demonstrated higher thermal conductivities than the same liquids without nanoparticles. Studies by Eastman *et al*., [3] using copper particles and oxide particles in ethylene glycol indicated that nanofluids exhibit superior heat transfer properties compared to conventional heat transfer fluids. Other experimental studies, which have shown that nanofluids exhibit better heat, transfer characteristics than traditional heat transfer fluids include [4-11].

Studies have been performed to explain the reasons for the enhanced thermal performance of nanofluids. These reasons include thermal transport in the nanoparticles, nanoparticle aggregation, Brownian dynamics and thermophoresis. Early attempts to predict the experimentally measured values of the thermal conductivity of nanofluids were made with existing theories such as Maxwell [1] and Hamilton-Crosser [12]. However, these theories predict lower values compared to experimental measurements. Theories such as the nanolayer theory, Yu and Choi [10], the average polarization theory, Xue [13] and Brownian induced nano-convection [14-16] have also been reported. The study by Yu and Choi [10] determined that the solid-liquid interfacial layer in the nanofluid plays an important role in the enhanced thermal conductivity. Yu and Choi modified the model by Maxwell [1] to include the effect of the nanolayer. The study proposed that the solid-like nanolayer behaves like a thermal bridge between a solid particle and bulk liquid. Using effective medium theory, the equivalent thermal conductivity of the equivalent particle was calculated and used to modify the Maxwell equation. Timofeeva *et al*., [17] indicated that agglomeration state, geometry and surface resistance of nanoparticles are the main variables that control the thermal conductivity enhancement of nanofluids. Other studies on the underlying behavior of thermal energy transfer in nanofluids include [18-26].

Explanations and theories underlying the heat transfer mechanisms in nanofluids and previously developed correlations have not completely predicted the anomalous increase in the thermal conductivity. It is clear that more research is needed to arrive at comprehensive and suitable theories. In this present study, the heat conduction in the nanolayer region was studied. A new correlation for the effective thermal conductivity of the nanofluid was developed. Comparison of the results obtained for the nanofluids studied shows that the correlation proposed is close to experimental results. Results predicting the thermal conductivity of nanofluids using the new correlation were compared to experimental results as well as studies by other researchers. Further studies were performed to understand how some factors affect the thermal conductivity of nanofluids. A number of important factors that influence the thermal conductivity of nanofluids were determined.

DEVELOPMENT OF THE CORRELATION AND PARAMETRIC STUDIES

Figure-1 illustrates a spherical nanoparticle of radius r_p overlapped with a nanolayer of thickness δ. Spherical particles and room temperature conditions were assumed for the development of the model. The thermal conductivity of the nanolayer varies between that of the particle k_p and the liquid k_f. It is assumed that the thermal conductivity of the nanolayer very close to the particle is k_p and at the outer boundary close to the liquid is k_f. It can thus be said that, at $r = r_p$, $k = k_p$ and at $r = r_p + \delta$, $k = k_f$. Thus there is a variation in the thermal conductivity within the nanolayer from k_f to k_p. There are currently not much previously reported studies on how the thermal conductivity varies in the nanolayer. Yu and Choi [10] assumed the thermal conductivity in the nanolayer to be a constant and equal to k_p. Xie *et al*., [21] and Ren *et al*., [27] considered the thermal conductivity to vary linearly between k_p and k_f. Tillman and Hill [28] derived an expression for the nanolayer thickness by manipulating three heat conduction regions. The nanolayer thickness was considered to be in the range of 19% to 22% of the nanoparticle radius. It was considered in this present study that since the thermal conductivity of a solid is much higher than that of a liquid, the effect will be more pronounced in the nanolayer towards the solid side as one proceeds from the solid side to the liquid side. It was therefore assumed that the thermal conductivity in the nanolayer region varied logarithmically to reflect this assumption. Figure-2 illustrates the variation. The thermal resistance R of the nanolayer is given by the equation

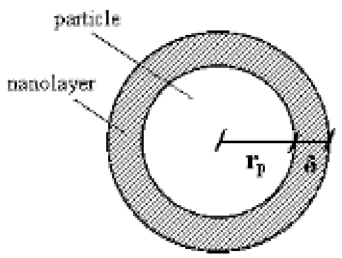

Figure-1. Diagram of a nanoparticle of radius r_p with a nanolayer of thickness δ.

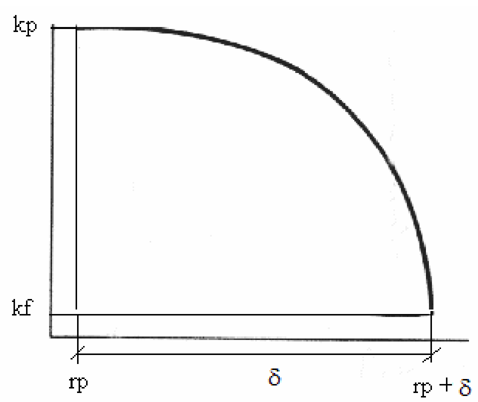

Figure-2. Variation of the thermal conductivity in the nanolayer.

$$R = \int_{r_p}^{r_p+\delta} \frac{dr}{4\pi r^2 k(r)} \tag{1}$$

R can also be expressed in terms of the average thermal conductivity (k_l) of the nanolayer as

$$R = \frac{1}{4\pi k_l}\left(\frac{1}{r_p} - \frac{1}{r_p+\delta}\right) \tag{2}$$

Combining these equations results in the expression for k_l as

$$k_l = \frac{\delta}{r_p(r_p+\delta)\int_{r_p}^{r_p+\delta}\frac{dr}{r^2 k(r)}} \tag{3}$$

Using the three equations it can be shown that the radial variation of k, i.e. $k(r)$ can be expressed as:

$$k(r) = k_f + \frac{k_p - k_f}{\delta}\left(\sqrt{\delta^2 - (r - r_p)^2}\right) \tag{4}$$

The resulting expression for k_l is

$$k_l = \frac{\delta}{r_p(r_p+\delta)\int_{r_p}^{r_p+\delta}\frac{dr}{r^2\left[k_f + \frac{k_p-k_f}{\delta}\left(\sqrt{\delta^2-(r-r_p)^2}\right)\right]}} \tag{5}$$

The effective thermal conductivity of the nanofluid k_{eff} can be related to the fluid and the solid particle through decomposition of the heat flux into the contributions from the fluid, the nanoparticle and the nanolayer.

Following the approach of Shin-Yuan Li [29], Lu and Song [30] and Xie *et al*., [21] the equation for the effective thermal conductivity of the nanofluid can be written as:

$$k_{eff} = k_f\left[1 + \left(3F\phi(1+\beta)^3 + \frac{3F^2[\phi(1+\beta)^3]^2}{1 - F[\phi(1+\beta)^3]}\right)\right] \tag{6}$$

Where

$$F = \frac{lf\left[(1+\beta)^3 - \left(\frac{pl}{fl}\right)\right]}{(1+\beta)^3 + 2(lf)(pl)} \quad and \quad \beta = \frac{\delta}{r_p} \tag{7}$$

The expressions for lf, pl and fl are given respectively as follows:

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$$lf = \frac{k_l - k_f}{k_l + 2k_f} \tag{8}$$

$$pl = \frac{k_p - k_l}{kp + 2k_l} \tag{9}$$

$$fl = \frac{k_f - k_l}{k_f + 2k_l} \tag{10}$$

Equation (6) is thus the expression for the effective thermal conductivity of the nanofluid with the parameters *F, β,* k_l*, lf, pl*, and *fl* defined by equations (7), (5), (8), (9) and (10), respectively.

In terms of the temperature field *T*, the heat transport in a multi-component material is described by the equation

$$\frac{\partial T}{\partial t} = \alpha \nabla^2 T \tag{11}$$

where α is the thermal diffusivity defined as

$$\alpha = \frac{k}{\rho c_p} \tag{12}$$

and ρ is density, *k* is thermal conductivity and c_p is specific heat. Research has shown that amongst other factors, the effective thermal conductivity k_{eff} of a nanofluid depends on the thermal conductivity of the base fluid k_f, the thermal conductivity of the solid particles k_p and the particle volume fraction ϕ.

The models that were compared to this study are described as follows: The effective thermal conductivity of a fluid with particles dispersed in it is given by Maxwell [1] as

$$k_{eff} = k_f \left[\frac{k_p + 2k_f + 2\phi(k_p - k_f)}{k_p + 2k_f - \phi(k_p - k_f)} \right] \tag{13}$$

where *n* is the empirical shape factor. Yu and Choi [10] showed a modified Maxwell model for the effective thermal conductivity of a homogenous suspension as

$$k_{eff} = k_f \left[\frac{k_{pe} + 2k_f + 2\phi(k_{pe} - k_f)(1+\beta)^3}{k_{pe} + 2k_f - \phi(k_{pe} - k_f)(1+\beta)^3} \right] \tag{14}$$

where k_{pe} is equivalent thermal conductivity of the nanolayer based on effective medium theory and β is the ratio of the nanolayer thickness to the particle radius. An alternative expression for calculating the effective thermal conductivity of fluids with particles dispersed in it is the model by Hamilton and Crosser [12] expressed as

$$k_{eff} = k_f \left[\frac{k_p + (n-1)k_f - (n-1)\phi(k_f - k_p)}{k_p + (n-1)k_f + \phi(k_f - k_p)} \right] \tag{15}$$

The model by Jeffrey [31] is

$$\frac{k_{eff}}{k_f} = 1 + 3\beta\phi + \left(3\beta^2 + \frac{3\beta^3}{4} + \frac{9\beta^3}{16}\frac{\alpha+2}{2\alpha+3} + \frac{3\beta^4}{26} + .. \right)\phi^2 \tag{16}$$

In this equation, $\beta = (\alpha - 1)/(\alpha + 2)$ where α is the ratio of the thermal conductivity of the particle to the thermal conductivity of the base fluid.

The model by Maxwell-Garnett reported by Bu-Xuan *et al*., [32] is

$$\frac{k_{eff}}{k_f} = \frac{(1-\phi)(k_p + 2k_f) + 3\phi k_p}{(1-\phi)(k_p + 2k_f) + 3\phi k_f} \tag{17}$$

The model by Wasp [33] is

$$\frac{k_{eff}}{k_f} = \frac{k_p + 2k_f - 2\phi(k_f - k_p)}{k_p + 2k_f + \phi(k_f - k_p)} \tag{18}$$

Computations show that the results obtained from studies by Jeffery [31], Maxwell-Garnett [32] Wasp [33] and Maxwell [1] are about the same. Thus in Figure-3 which shows comparisons with this study (labeled "Study" in the Figure); the values shown for Maxwell also basically represents those for Jeffery, Maxwell-Garnett and Wasp. The experimental results used for the comparisons are those given by Yu and Choi [10] which used experimental results obtained by Lee *et al*., [2] and Eastman *et al*., [3].

Lee *et al*., prepared oxide nanofluids by producing and dispersing nanometer-sized solid particles into the liquid in a mixing chamber. The nanoparticles were characterized before dispersion in the liquids and after dispersion in the liquids by using transmission electron microscopy techniques. Eastman *et al*., used a one-step method to produce and disperse the nanoparticles into the liquid. The thermal conductivity of the nanofluid was measured by means of the transient hot-wire method.

A parametric study was performed to investigate the dependence of thermal conductivity of nanofluid on other parameters. Functions such as nanolayer thickness, volume fraction, radius of nanoparticles and different nanoparticle-liquid combinations were studied. Keeping other factors constant, parameters were varied for different nanoparticle/liquid combinations. The nanofluids studied include (1) Copper Oxide in Ethylene Glycol, (2) Titanium Oxide in Ethylene Glycol, (3) Aluminum Oxide in Water, and (4) Iron in Ethylene Glycol.

Figure-4 shows results for the variation of the effective thermal conductivity of the nanofluid with particle radius for Titanium Oxide in Ethylene Glycol. Li and Peterson [34] studied the effect of particle size on the effective thermal conductivity of Al_2O_3-water nanofluids. Results from that study also showed a non-linear relationship between the particle size and thermal conductivity.

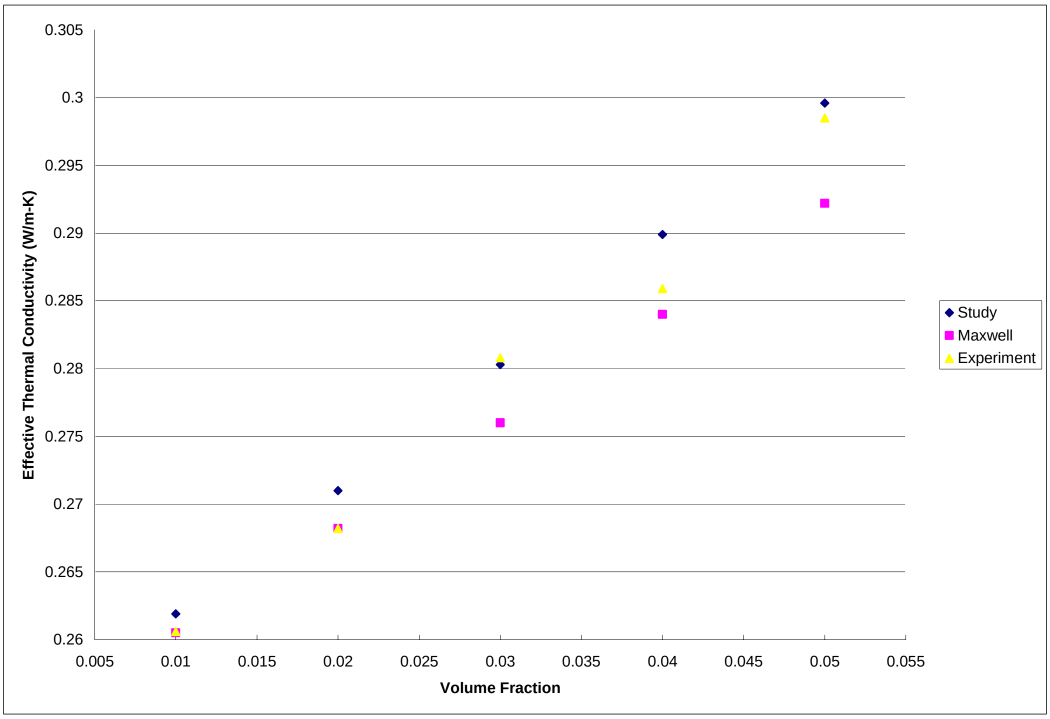

Figure-3. Comparison of the effective thermal conductivities of Al_2O_3–Ethylene glycol mixture.

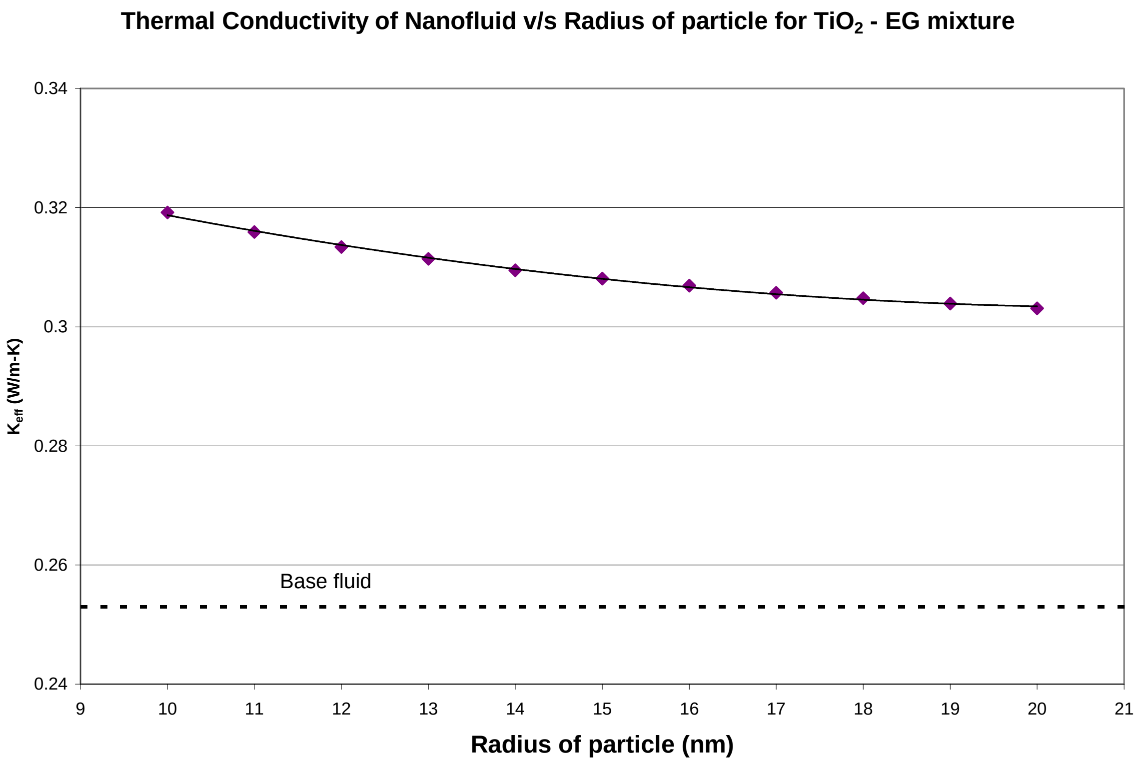

Figure-4. Effective thermal conductivity of the nanofluid versus radius of particle.

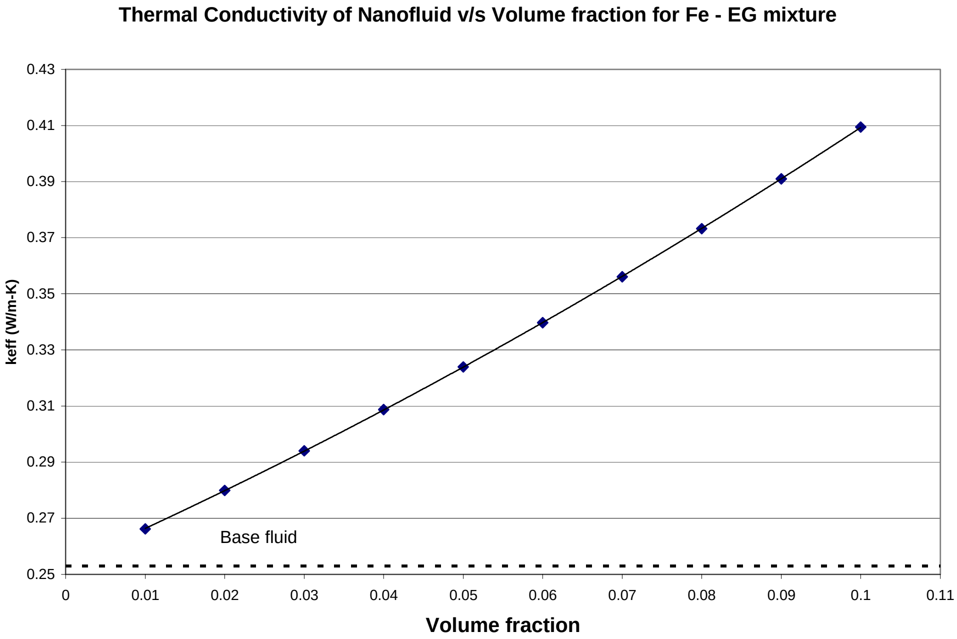

Figure-5. Effective thermal conductivity of the nanofluid versus volume fraction.

Figure-5 shows the trend of the results for the variation of the effective thermal conductivity of the nanofluid with the volume fraction. The Figure shown is for iron nanoparticles in Ethylene Glycol.

Figure-6 shows the trend for the effect of nanolayer thickness on the effective thermal conductivity of the nanofluid. The Figure shown is for Copper Oxide in Ethylene Glycol. The study by Tillman and Hill [28] indicated that the nanolayer was in the range of 19% to 22% of the nanoparticle radius.

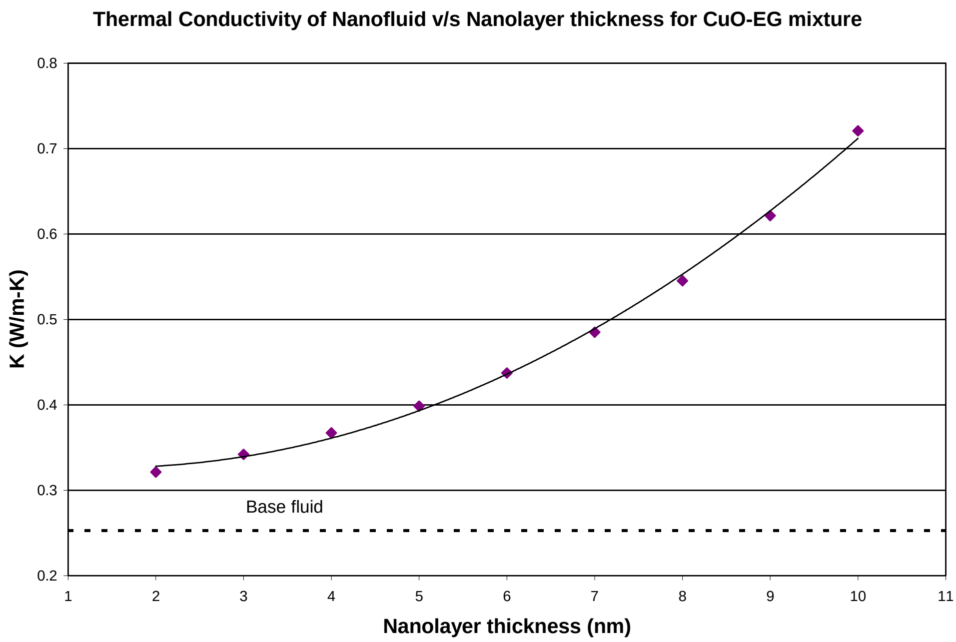

Figure-6. Effect of nanolayer thickness on the effective thermal conductivity of the nanofluid.

DISCUSSION OF RESULTS

It can be seen from Figure-3 that there is close agreement between the model developed from this study and experimental measurements. It was also found that the results are also close to the model by Yu and Choi [10] and Xie *et al.*, [21]. There is currently not much previous report on how the thermal conductivity varies in the nanolayer. Yu and Choi [10] assumed the thermal conductivity in the nanolayer to be a constant k_p. Xie *et al.*, [21] and Ren *et al.*, [27] considered the thermal conductivity to vary linearly between k_p and k_f in the nanolayer. In this present study, it was considered that since the thermal conductivity of a solid is much higher than that of a liquid, the nature of the effect will be more pronounced towards the solid side in the nanolayer as one proceeds from the liquid side to the solid side. Thus it was assumed to vary logarithmically as shown in Figure-2. It can be seen from Figure-3 that consideration of the nanolayer improves the effective thermal conductivity of the nanofluid. Based on the nanolayer logarithmic assumption in the nanolayer used in this present study, the constant solid thermal conductivity assumption in the nanolayer by Yu and Choi [10] and the linear variation assumption in the nanolayer by Xie *et al*., [21] and Ren *et al*. [27], it can be concluded that although the thermal conductivity of the nanolayer contributes to the effective thermal conductivity of the nanofluid, how it varies in the nanolayer region is not significant to the contribution.

Figure-4 shows that within the limits of this study, the effective thermal conductivity of the nanofluid decreases with the particle radius at constant volume fraction. Kumar *et al.,* [35] also studied the effect of particle size in nanofluids using gold and Al_2O_3 nanoparticles in water and CuO nanoparticles in ethylene glycol and concluded that the effective thermal conductivity of a nanofluid is inversely proportional to the radius of the particle. Xie *et al*., [21] demonstrated that the effects of the nanolayer would be more when the particle size is small. These reported studies support the results obtained from this present study on the effect of the particle radius on the effective thermal conductivity of nanofluids.

Figure-5 shows that the thermal conductivity of the nanofluid increases as the volume fraction increases. Thus, increasing the volume fraction of the nanofluid increases the effective thermal conductivity of the nanofluid. There appears to be a linear relationship between the effective thermal conductivity and the volume fraction of the nanofluid. Xuan and Li [6] reported that as the volume fraction increases the thermal conductivity of the Cu-water nanoparticle suspension increases. Kumar *et al*., [35] also studied the effect of volume fraction on different nanofluids and reported that the enhancement in thermal conductivity is linearly proportional to the nanoparticle concentration. Eastman *et al*., [3] reported an increase in the thermal conductivity of nanofluid with increase in volume fraction for the Al_2O_3-water, CuO-water, CuO-ethylene glycol and Cu in ethylene glycol nanofluids. These results confirm the validity of the results from this study.

Figure-6 shows the effect of nanolayer thickness on the effective thermal conductivity of the nanofluids. This present study indicates that at constant volume fraction, the effective thermal conductivity of the nanofluid increases as the thickness of the nanolayer increases. Yu and Choi [10] studied nanofluids such as Copper oxide/Ethylene Glycol and copper/Ethylene Glycol and reported that nanolayer formation does play an important role in enhancing the thermal conductivity of a nanofluid. The study by Xue [13] which is related to aluminum oxide water nanofluid reported that the larger the thickness of the interfacial shell (nanolayer thickness) the larger the thermal conductivity of the nanofluid. Yu and Choi [36] reported that when the thermal conductivity of the interfacial layer is high it can increase the overall thermal conductivity of the nanofluid. Keblinski *et al*., [15] and Xue *et al*., [37] reported that nanolayer formation is one of the reasons responsible for the overall increase in the thermal conductivity of the nanofluid. The non-equilibrium molecular dynamic simulations of a simple mono-atomic liquid with imposed temperature gradient showed no effect on the thermal transport either normal to the surface or parallel to the surface.

CONCLUSIONS

A number of parameters that could be responsible for the increase in the effective thermal conductivity of nanofluids compared to the base fluid were highlighted in this study. Factors responsible for the enhanced thermal conductivity of nanofluids include nanoparticle aggregation, Brownian motion induced nanoparticle convection and thermophoresis and nanoparticle and nanolayer thermal transport in the nanofluids. Conduction in the nanolayer was combined with some parameters to formulate a new correlation for the effective thermal conductivity of the nanofluid. The correlation which amongst other factors depends on the thermal conductivities of the liquid and nanoparticle, particle size, nanolayer thickness and volume fraction closely predicts experimental results for the effective thermal conductivity of nanofluids. Results predicting the thermal conductivity of nanofluids using the correlation were compared with experimental results and studies by other researchers. Results show that taking the nanolayer effect into consideration improves the effective thermal conductivity of the nanofluid. It was also concluded that although the thermal conductivity of the nanolayer contributes to the increased thermal conductivity of the nanofluid, how the thermal conductivity varies in the nanolayer region is not significant to the contribution.

The study performed to understand how a number of factors affect thermal conduction in nanofluids included particle radius, volume fraction and nanolayer thickness. Different base fluid and nanoparticle combinations were used. Results show that the thermal conductivity of the nanofluid increases with nanolayer thickness and in general, increases linearly with the volume fraction but increases as the particle radius decreases.

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Nomenclature

F: Heat flux factor
k: Thermal conductivity (W/m ºC)
$k_{eff:}$ Overall effective thermal conductivity of a nanofluid (W/m ºC)
k(r): Variation of thermal conductivity in the nanolayer
q: Heat flux (W/m^2)
R: Thermal resistance (ºC/W)
r: Radius (m)
T: Temperature (ºC)
α: Thermal diffusivity (m^2/s)
B: ratio of the nanolayer thickness to the original particle radius
Δ: thickness of nanolayer (nm)
ρ: density (kg/m^3)
Φ: volume fraction

Subscripts

f: fluid
l: nanolayer
p: particle