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Effect of interfacial nanolayer on the effective thermal conductivity of nanoparticle-fluid mixture

Huaqing Xie, Motoo Fujii, Xing Zhang *

Institute for Materials Chemistry and Engineering, Kyushu University, Kasuga 816-8580, Japan

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Abstract

Nanofluids, containing metal or nonmetal particles with nanometer sizes, exhibit much greater thermal conductivity than predictions. It has been proposed that interfacial structures formed by liquid molecule layering might play role. We investigated the impact of this interfacial nanolayer on the effective thermal conductivity of nanofluid. An expression for calculating enhanced thermal conductivity of nanofluid has been derived from the general solution of heat conduction equation in spherical coordinates and the equivalent hard sphere fluid model representing the microstructure of particle/liquid mixtures. The effects of nanolayer thickness, nanoparticle size, volume fraction, and thermal conductivity ratio of particle to fluid have been discussed. The predicted results are in good agreement with some recent available experimental data.

Keywords: Thermal conductivity; Nanofluids; Interfacial layer

1. Introduction

Nanofluids, produced by dispersing nanoparticles into conventional heat transfer fluids such as water, glycol, or oil, have been proposed to work as high efficiency heat exchange media with various expected superior properties relative to those of fluids without particles or of suspensions with large-size particles [1]. Recently, researchers have demonstrated that nanofluids, containing a small amount of metal (e.g., Cu) or nonmetal (e.g., SiC, Al_2O_3, and CuO) nanoparticles, exhibit substantially enhanced thermal conductivities compared to those of the base fluids [2–5]. An enhancement in thermal conductivity up to 38% was observed in the study for pump oil based suspensions containing alumina particles with specific surface areas (SSAs) of 25 $m^2 g^{-1}$ at a volume fraction of 0.05 [4]. Intriguingly, for a nanofluid consisting of ethylene glycol (EG) and only 0.3 vol% Cu nanoparticles, the thermal conductivity was shown to be increased to 40% [2]. Furthermore, the thermal conductivity enhancement appears highly dependent on the morphologies (the particle size and the shape) of the included nanoparticles and the thermal conductivity of the base fluid.

For particle-fluid mixtures, numerous theoretical studies have been conducted dating back to the classic work of Maxwell [6]. Most of the existing understanding, e.g. Maxwell model [6], Hamilton and Crosser model [7], and Davis model [8], of the effective thermal conductivity of composites and mixtures are derived

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* Corresponding author. Tel./fax: +81 92 583 7638.
E-mail address: xzhang@cm.kyushu-u.ac.jp (X. Zhang).

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Nomenclature

k	thermal conductivity
n	particle number per volume
$\vec{q}$	heat flux vector
r	diameter
R	thermal resistance
S	surface area
T	temperature
V	volume
Greek symbols	
δ	thickness of nanolayer
ε	reduced thermal conductivity
γ	thickness ratio
ϕ	volume fraction
Subscripts	
eff	effective
f	fluid
l	nanolayer
p	nanoparticle

from continuum-level phenomenological formulations that typically incorporate only the particle shape and volume fraction as variables and assume diffusive heat transport in both liquid and solid phase; no effects of particle size and interfaces are taken into account. Some theoretical attempts addressing the interfacial characteristics between inclusion and matrix have been made [9–12]. Hasselman and Johnson [9], based on an equivalent inclusion concept, extended Maxwell's theory to systems of spherical inclusion with contact resistance. Meanwhile, Chiew and Glandt [10] extended Jeffrey's results [13] to the case of contact resistance problems. Lu and Song [11,12] investigated coated or debonded inclusion and developed a more general model to predicting the effective thermal conductivity of composites. However, the experimental results have shown that the enhanced thermal conductivities of nanofluids are much higher, even up to an order of magnitude greater, than those values predicted by the traditional models which are satisfied for computing the effective thermal conductivity of mixture consisting of fluid and micro- or larger-sized solid particles. The weakness of traditional models in predicting the thermal conductivity of nanofluid indicates some mechanisms may be missed. Therefore, nanofluids not only offer an opportunity for upgrading traditional thermal engineering, but also present a theoretical challenge to explain their heat transport mechanisms. Keblinski et al. [14] investigated the possible factors of enhancing thermal conductivity in nanofluids, and proposed that the size effect, the clustering of nanoparticles, and the nanolayer at solid/liquid interface could be the major contributions of the enhancement. Recently Yu and Choi [15] reported a modified Maxwell model, in which a nanolayer with thickness of a few nanometer has been proposed to exist at the interface between particle and fluid and a formula for calculating the effective thermal conductivity has been derived based on effective medium theory and Maxwell model. This model's predictions have shown that measurable enhancement of the effective thermal conductivity can be expected when nanolayers are accounted for.

In this paper, we considered an interfacial nanolayer with linear thermal conductivity distribution and attempted to deduce an expression for calculating the enhanced thermal conductivity of nanofluid, which includes the impact of nanolayer. The effects of nanolayer thickness, nanoparticle size, volume fraction, and thermal conductivity ratio of particle to fluid have been discussed. The comparisons between some currently available data and predicted values have been made.

2. Nanolayer structure

In particle-fluid mixtures, the liquid molecules close to a particle surface are known to form layered structures and behave much like a solid [16]. The thickness of this aligned solid-like layer of liquid molecules at the interface is at a magnitude of nanometer, but this nanolayer might play an important role in heat transport from solid to adjacent liquid [5,14,15]. For particles with micrometer size, the surface areas are small, e.g., for alumina powders with an average diameter of 10 μm, their SSA is only $0.15\ \mathrm{m}^2\ \mathrm{g}^{-1}$. The effect of interfacial nanolayer is negligible. However, nanoparticles have very large specific surface area, e.g., for alumina powders with an average diameter of 10 nm, their SSA is as great as $151\ \mathrm{m}^2\ \mathrm{g}^{-1}$, much larger than that of micro-sized particles. Therefore, the aligned solid/liquid interfacial shell in nanoparticle suspension would make heat transfer across the interface effective.

In order to include the effect of nanolayer, let us consider a nanoparticle-liquid mixture with monosized spherical inclusions of radius r_p and particle volume concentration ϕ. Fig. 1 denotes the schematic structures of a nanoparticle with an interfacial nanolayer when the nanoparticle is dispersed in a fluid. The alignment of the liquid molecules inside the solid-like interfacial

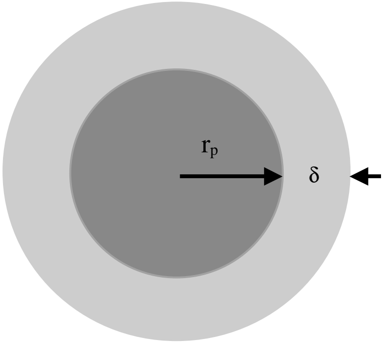

Fig. 1. Schematic structures of nanoparticle and interfacial nanolayer. Particle: dark; layer: gray.

nanolayer of thickness δ is more ordered than that of bulk liquid. The thickness, the microstructures, and the physicochemical properties of this nanolayer are highly depended on the suspended nanoparticle, the base fluid, and the interaction between them. Therefore, the thermophysical behaviors of this interfacial nanolayer may be investigated by carefully analyzing the above mentioned factors. Although there is no available expression for calculating the thermal conductivity of the nanolayer on the surface of the nanoparticle, the solid-like nanolayer would be expected to have an intermediate thermal conductivity between that of the bulk liquid (k_f) and that of the nanoparticle (k_p) because the layered molecules are in an intermediate physical state between a bulk liquid and a solid [15]. If the distribution of the thermal conductivity inside the nanolayer is $k(r)$ ($r_p \leqslant r \leqslant r_p + \delta$), the thermal resistance presented by this layer, R_l, is

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

R_l can also be expressed by the average thermal conductivity of the nanolayer, k_l, as

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

From Eqs. (1) and (2), we have

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

To decide $k(r)$ is clear a nontrivial problem due to the complexity of physicochemical interactions between nanoparticle and fluid. For simplifying, we assume linear variation of $k(r)$, that is

$$k(r) = \frac{k_f - k_p}{\delta} r + \frac{k_p(r_p+\delta) - k_f r_p}{\delta}. \tag{4}$$

Substitute Eq. (4) into (3), one can derive the following expression:

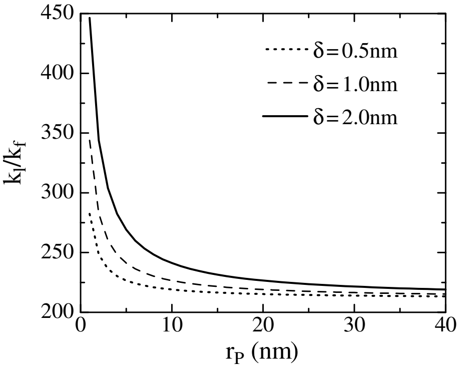

Fig. 2. Thermal conductivity ratio of nanolayer as a function of r_p.

$$k_l = \frac{k_f M^2}{(M-\gamma)\ln(1+M) + \gamma M}, \tag{5}$$

with $M = \varepsilon_p(1+\gamma) - 1$, where $\varepsilon_p = k_p/k_f$ is the reduced thermal conductivity of nanoparticle and $\gamma = \delta/r_p$ is ratio of the nanolayer thickness to the original particle radius. In Eq. (5), the average thermal conductivity of nanolayer depends on the thermal conductivity of fluid, the reduced thermal conductivity of nanoparticle and ratio of the nanolayer thickness to the original particle radius. Fig. 2 shows the thermal conductivity ratios of the nanolayers, k_l/k_f, in ethylene glycol (EG) based nanofluids containing copper nanoparticles. k_l/k_f is shown to be strongly dependent on the particle size and the thickness of nanolayer. With an increase in the thickness of nanolayer or a reduction in particle size, k_l/k_f increases. A much steeper change can be seen at small particle size range, which indicates that the impact of nanolayer would be more effective when the particle is small whilst the nanolayer is thick. The detailed relations of the effective thermal conductivity of nanofluid to the particle size and the thickness of nanolayer will be discussed in Section 4.

The total volume fraction of the original nanoparticle and nanolayer, ϕ_T, is

$$\phi_T = \frac{4}{3}\pi(r_p+\delta)^3 n = \phi(1+\gamma)^3, \tag{6}$$

where $\phi = \frac{4}{3}\pi r_p^3 n$ is the original volume fraction of nanoparticle and n is the particle number per volume.

3. Effective thermal conductivity expression

Nanofluid consisting of base fluid and nanoparticles with nanolayers described in Section 2 is statistically homogeneous and isotropic. Its effective thermal conductivity, k_{eff}, is defined following Fourier's law of heat conduction:

$$\langle\vec{q}\rangle = -k_{\text{eff}}\langle\nabla T\rangle. \tag{7}$$

Here, $\langle\vec{q}\rangle$ and $\langle\nabla T\rangle$ are the volume average heat flux and temperature gradient vector, respectively. To relate k_{eff} to other system parameters, the average heat flux can be decomposed into contributions coming from the fluid, nanoparticles, and nanolayers as follows [11]:

$$\langle\vec{q}\rangle = -k_{\text{f}}\langle\nabla T\rangle + \langle\vec{q}_{\text{p}}\rangle + \langle\vec{q}_{\text{l}}\rangle, \tag{8a}$$

with

$$\begin{aligned}\langle\vec{q}_{\text{p}}\rangle &= \frac{1}{V}\frac{k_{\text{p}}-k_{\text{f}}}{k_{\text{p}}}\int_{V_{\text{p}}}\vec{q}\,\mathrm{d}V\\ &= \frac{1}{V}\frac{k_{\text{p}}-k_{\text{f}}}{k_{\text{p}}}\int_{S_{\text{p}}}\vec{r}\vec{q}\cdot\vec{n}\,\mathrm{d}S\end{aligned} \tag{8b}$$

and

$$\begin{aligned}\langle\vec{q}_{\text{l}}\rangle &= \frac{1}{V}\frac{k_{\text{l}}-k_{\text{f}}}{k_{\text{l}}}\int_{V_{\text{l}}}\vec{q}\,\mathrm{d}V\\ &= \frac{1}{V}\frac{k_{\text{l}}-k_{\text{f}}}{k_{\text{l}}}\int_{S_{\text{p}}+S_{\text{l}}}\vec{r}\vec{q}\cdot\vec{n}\,\mathrm{d}S,\end{aligned} \tag{8c}$$

where V is the total volume of the observed composite system, $\vec{r}$ is the position vector, and $\vec{n}$ is the unit outward normal vector of the bounding surface (particle/nanolayer or nanolayer/liquid interface). S_{p} and S_{l} denote the inner and outer bounding surface of the nanolayer. $\langle\vec{q}_{\text{p}}\rangle$ and $\langle\vec{q}_{\text{l}}\rangle$ are clearly the extra heat fluxes resulting from the presence of the nanoparticle and interfacial nanolayer, respectively. Once $\langle\vec{q}_{\text{p}}\rangle$ and $\langle\vec{q}_{\text{l}}\rangle$ are determined, the effective thermal conductivity can be obtained by matching Eqs. (7) and (8).

To determine $\langle\vec{q}_{\text{p}}\rangle$ and $\langle\vec{q}_{\text{l}}\rangle$, and thus k_{eff}, the temperature distributions inside nanoparticle-fluid mixture should be evaluated. Assuming that the mixture system is subjected to steady state heat conduction, the temperature fields within nanoparticle, nanolayer, and fluid are governed by steady state heat conduction equations. To one specified particle, regardless the spatial arrangement of the remaining particles, the general solutions of these equations can be represented by the following expressions [11]:

$$\begin{aligned}&T_{\text{f}}(r,\theta,\varphi)\\ &\quad=\sum_{j=0}^{\infty}\sum_{i=0}^{j}\left[a_{ij}r^{j}P_{j}^{i}(\cos\theta)\cos i\varphi + a'_{ij}r^{j}P_{j}^{i}(\cos\theta)\sin i\varphi\right]\\ &\qquad+\sum_{j=0}^{\infty}\sum_{i=0}^{j}\Big[b_{ij}r^{-(j+1)}P_{j}^{i}(\cos\theta)\cos i\varphi\\ &\qquad+b'_{ij}r^{-(j+1)}P_{j}^{i}(\cos\theta)\sin i\varphi\Big],\end{aligned} \tag{9a}$$

$$\begin{aligned}T_{\text{p}}(r,\theta,\varphi) &= \sum_{j=0}^{\infty}\sum_{i=0}^{j}\Big[c_{ij}r^{j}P_{j}^{i}(\cos\theta)\cos i\varphi\\ &\qquad+c'_{ij}r^{j}P_{j}^{i}(\cos\theta)\sin i\varphi\Big],\end{aligned} \tag{9b}$$

$$\begin{aligned}T_{\text{l}}(r,\theta,\varphi) &= \sum_{j=0}^{\infty}\sum_{i=0}^{j}\Big[e_{ij}r^{j}P_{j}^{i}(\cos\theta)\cos i\varphi\\ &\qquad+e'_{ij}r^{j}P_{j}^{i}(\cos\theta)\sin i\varphi\Big]\\ &\qquad+\sum_{j=0}^{\infty}\sum_{i=0}^{j}\Big[f_{ij}r^{-(j+1)}P_{j}^{i}(\cos\theta)\cos i\varphi\\ &\qquad+f'_{ij}r^{-(j+1)}P_{j}^{i}(\cos\theta)\sin i\varphi\Big],\end{aligned} \tag{9c}$$

where T_{f}, T_{p}, and T_{l} are the temperature fields in the fluid, nanoparticle, and nanolayer, respectively, (r,θ,φ) are the spherical coordinates, a_{ij}, a'_{ij}, b_{ij}, b'_{ij}, c_{ij}, c'_{ij}, e_{ij}, e'_{ij}, f_{ij}, and f'_{ij} are unknown coefficients to be determined based on relevant boundary conditions, and $P_{j}^{i}(\cos\theta)$ are the associated Legendre functions of order j and degree i with argument $\cos\theta$. The continuities of temperature and normal heat flux across the bounding surfaces need to be satisfied in Eq. (9).

Fig. 3 shows a schematic cross-section of homogeneous nanofluid that consists of nanoparticles, base fluid, and nanolayers. For convenience, we apply linear temperature field and unit ambient temperature gradient vector, $\mathbf{\Omega}$, along a specified direction to the system to find k_{eff} (Fig. 3). Lu and Song [12], adopting equilibrium hard sphere fluid model to represent the microstructure of mixtures and considering two interacting particles, investigated the effective thermal conductivity of coated and debonded inclusion problems. Following these approaches and after algebraic derivation [12], an expression for representing k_{eff} of nanoparticle-fluid mixtures is given as follows:

$$\frac{k_{\text{eff}}-k_{\text{f}}}{k_{\text{f}}} = 3\Theta\phi_{T} + \frac{3\Theta^{2}\phi_{T}^{2}}{1-\Theta\phi_{T}} + F(r_{\text{p}},\delta,\phi,k_{\text{f}},k_{\text{p}},k_{\text{l}}), \tag{10a}$$

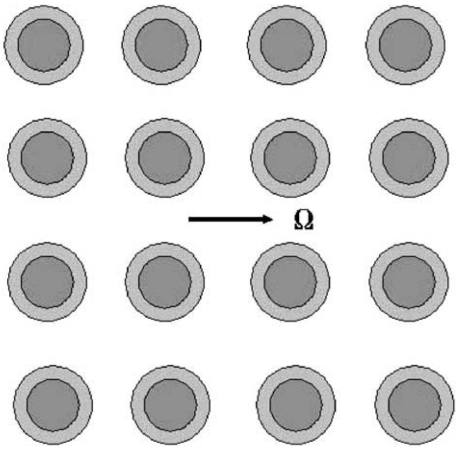

Fig. 3. Schematic cross-section of nanoparticles in a homogeneous nanofluid.

with

$$\Theta = \frac{\beta_{lf}\left[(1+\gamma)^3 - \frac{\beta_{pl}}{\beta_{fl}}\right]}{(1+\gamma)^3 + 2\beta_{lf}\beta_{pl}}, \tag{10b}$$

where $\beta_{lf} = \frac{k_1-k_f}{k_1+2k_f}$, $\beta_{pl} = \frac{k_p-k_1}{k_p+2k_1}$, $\beta_{fl} = \frac{k_f-k_1}{k_f+2k_1}$, and $F(r_p, \delta, \phi, k_f, k_p, k_1)$ is the higher order pair interactions related to the detailed microstructure of a specified nanofluid. It is clear that the sum of the first two terms of the right side in Eq. (10a) is with a truncation error of $O(\phi^2)$. For simplifying, if we consider nanofluids at low particle loadings, $F(r_p, \delta, \phi, k_f, k_p, k_1)$ is negligible. Eq. (10a) is reduced to

$$\frac{k_{eff} - k_f}{k_f} = 3\Theta\phi_T + \frac{3\Theta^2\phi_T^2}{1 - \Theta\phi_T}. \tag{11}$$

Eq. (11) relates the effective thermal conductivity of nanoparticle-fluid mixture to the system parameters such as the thermal conductivities of fluid, nanoparticle, and nanolayer, the volume fraction, the particle size of nanoparticle, and the thickness of nanolayer. This equation is the proposed model deduced for evaluating the effect of nanolayer on the effective thermal conductivity of nanoparticle-fluid mixtures.

4. Analysis and discussions

As expected, to a nanoparticle-fluid mixture consisting of solid particle with high thermal conductivity and fluid with low thermal conductivity, its effective thermal conductivity is substantially augmented. Fig. 4 shows the dependence of the calculated enhanced thermal conductivity ratio $(k_{eff} - k_f)/k_f$ on particle fractions. Here, the base fluid is chosen as EG and the dispersed phase is Cu nanoparticle with an average diameter of 10 nm. It is clearly seen that $(k_{eff} - k_f)/k_f$ increases with the particle fraction and the thickness of nanolayer. The enhancement is nonlinear at a higher concentration and thicker nanolayer.

To evaluate the impact of particle size on the effective thermal conductivity of nanofluid, we considered Cu-EG nanofluids with copper nanoparticle concentration of 5.0%. When the particle size decreases, $(k_{eff} - k_f)/k_f$ increases inversely (Fig. 5). Due to the contribution of nanolayer, $(k_{eff} - k_f)/k_f$ augments with very large slope at small particle size. This has a significant implication that exploiting the nanolayer structure might be a new way to produce nanofluids that are highly thermally conductive. One can manipulate the thermal conductivity of mixture through nanoparticle surface process.

When the inclusion changes from a thermal insulation material to a highly thermally conductive material, the thermal conductivity enhancement of the mixture increases inversely from minus to plus (Fig. 6). The results shown in Fig. 6 are calculated by choosing $r_p = 10$ nm and $\delta = 2$ nm. There has no effect of inclusion and interfacial layer if $k_p = k_f$. Although the mixture of which the inclusion with thermal conductivity lower than that of matrix is not the case for nanofluid, the results indicate that the present model is applicable to predict the effective thermal conductivities of various mixture systems, for example, emulsions containing nanosized bubble. The effect of k_p/k_f is also related to the volume fraction. To nanofluids with 1.0 vol% particle fraction, the enhanced thermal conductivity ratio is not sensitive to k_p/k_f if $k_p/k_f > 100$. But for nanofluids with 5.0 vol% particle loading, the thermal conductivity ratios still increase with an increase in k_p/k_f, even at a much larger k_p/k_f.

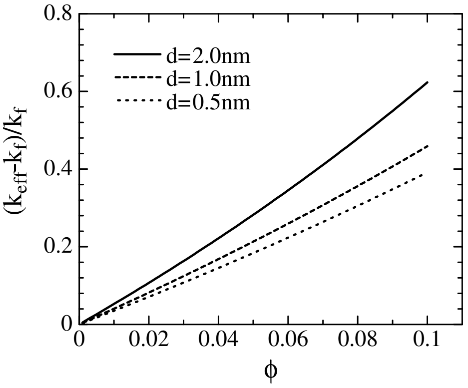

Fig. 4. Dependence of the enhanced thermal conductivity ratios on particle loadings.

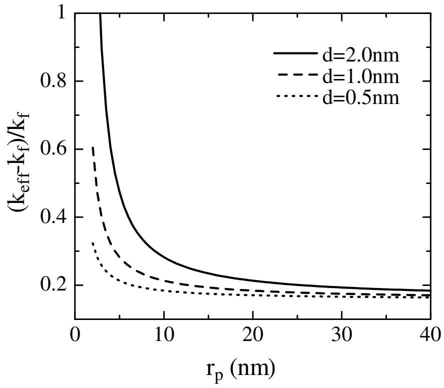

Fig. 5. Dependence of the enhanced thermal conductivity ratios on particle sizes.

To investigate the interfacial effect on the thermal conductivity of nanofluids, we considered a nanolayer at the interface between nanoparticle and fluid. The nanolayer has continuous thermal conductivity distribution, that is, it has a thermal conductivity same to that of nanoparticle at its inner surface, while at the outer

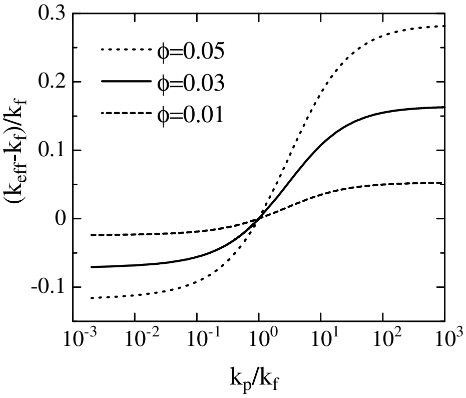

Fig. 6. Enhanced thermal conductivity ratio as a function of the reduced thermal conductivity of nanoparticle.

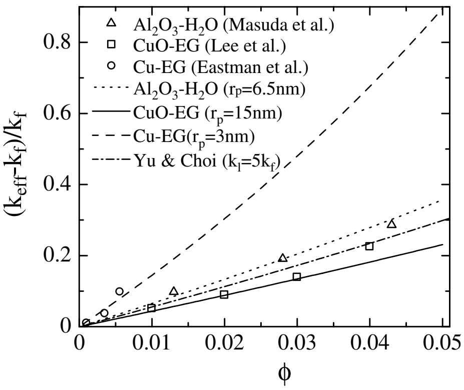

Fig. 8. Experimental data compared with predictions.

surface, its thermal conductivity equates to that of fluid. This assumption is based on the conceptions that the nanolayer is in an intermediate physical state between a bulk liquid and a solid [15,16] and that the interaction between nanoparticle and fluid molecules is continuous. In Yu and Choi' model [15], the thermal conductivity of a nanolayer is set to be a constant, without considering the interaction of nanoparticle and fluid. If we take a constant thermal conductivity for nanolayer in the present model, the predicted thermal conductivity enhancement ratios are consistent with those calculated by Yu and Choi's model using the same parameters (Fig. 7). The results shown in Fig. 7 are calculated by choosing r_p = 10 nm and δ = 2 nm. It is clearly seen in Fig. 7 that both the predictions of the present model and Yu and Choi' model are much greater than that of a traditional model (Hamilton and Crosser model [7]) not including the interfacial role. The present model's predictions have shown that measurable enhancement of the effective thermal conductivity can be expected when nanolayers are accounted for.

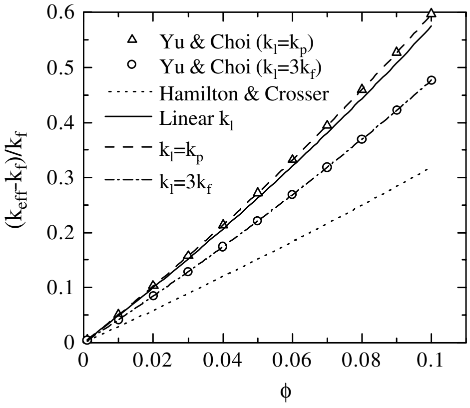

Fig. 7. Comparisons with other models.

The comparisons of the computed values to some available experimental data are shown in Fig. 8. The experimented nanofluids are copper nanoparticles in EG [2], copper oxide nanoparticles in EG [5], and alumina nanoparticles in water [17]. In this calculation, the particle sizes were taken as 3 nm for copper nanoparticle, 15 nm for copper oxide particle, and 6.5 nm for alumina nanoparticle from the measured data [2,5,17]. A 2 nm thick nanolayer was considered. An example of prediction by Yu and Choi's model is also presented (dashed-dot line in Fig. 8), in which the parameters corresponding to the above-mentioned alumina nanofluids were taken except that the thermal conductivity of nanolayer was take as five times as that of the base fluid. It is indicated in Fig. 8 that the present model including the effect of nanolayer predicts these experimental data quite well. However, it should be noted that the predicted values are inconsistent with some other currently available experimental data. For example, Eastman et al. [2] reported 40% enhancement in thermal conductivity when adding some acid to EG based nanofluid containing only 0.3 vol% copper nanoparticle. An even higher enhancement, about 160%, was measured in a nanofluid containing oil and 1.0 vol% multi-walled carbon nanotubes (MWNTs) [18]. Although our results reveal that the nanolayer is a key factor, it appears that other factors such as inclusion shape and surface chemistry should be considered.

5. Conclusions

A model including the consideration of the role of nanolayer at particle-fluid interface has been proposed for predicting the effective thermal conductivities of nanoparticle-fluid mixtures. Nanolayer thickness, nanoparticle size, volume fraction, and thermal conductivity ratio of particle to fluid have been shown to have effects on the enhanced thermal conductivity ratios. The effective thermal conductivity increases with a decrease of particle size and an increase in nanolayer thickness. Especially in small particle size range, the effects of particle size and nanolayer thickness become much more obvious, which implies that manipulating nanolayer structure might be an effective method to produce highly thermally conductive nanofluids. The calculated values fit quite well with some currently available experimental data.

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