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Effective thermal conductivity of nanofluids containing spherical nanoparticles

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Abstract

A theoretical model which includes considerations of the effects of an interfacial nanolayer formed by liquid molecule layering on the particle/liquid interface and of micro-convection caused by thermal motion of nanoparticles has been proposed to calculate the effective thermal conductivity of nanofluids. This model accounts for the enhancement in effective thermal conductivity of a nanofluid with respect to the suspended nanoparticle size, volume fraction, temperature and thermal conductivities of the nanoparticle and base fluid. The predicted results are in good agreement with some recently available experimental data.

Adding particles into heating and cooling fluids has long been regarded as a promising approach to enhance heat transfer. However, the applications of the particle-fluid mixtures are held back due to the poor stability of those suspensions containing milli-/micro-sized particles. In the past decade, a novel kind of particle suspensions, nanofluids [1], have been developed and have attracted increasing attention because of their intriguing properties such as high thermal conductivity, stability and prevention of clogging in micro-channels. Researchers have demonstrated that nanofluids, containing a small amount of metal (e.g. Cu) or nonmetal (e.g. SiC, Al₂O₃ and CuO) nanoparticles, exhibit substantially enhanced thermal conductivities compared with those of the base fluids [2–5]. The enhanced thermal conductivities have been found to be much higher, even up to an order of magnitude greater, than those values predicted by the traditional models, e.g. Maxwell model [6], Hamilton and Crosser model [7] and Davis model [8], which are satisfactory for computing the effective thermal conductivity of mixture consisting of fluid and micro- or largersized solid particles. The weakness of traditional models in predicting the thermal conductivities of nanofluids indicates that 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. We attempt here to investigate theoretically the mechanism of the effective thermal conductivity of nanofluids.

In particle-fluid mixtures, the liquid molecules close to a particle surface are known to form layered structures and behave much like a solid [9]. The thickness of this aligned solid-like layer of liquid molecules at the interface is at a magnitude of nanometre, but this nanolayer might play an important role in heat transport from solid to adjacent liquid [5, 10, 11]. In building up our model, we consider the nanoparticle and the attached nanolayer as a complex nanoparticle. These complex nanoparticles undergo restless motion arising from the random thermal agitation of the molecules in the surrounding liquid: these smaller entities act both as the driving force for the nanoparticle fluctuations (through the impact of the liquid molecules on the nanoparticles) and as a means of damping these motions (through the viscosity experienced by the nanoparticles) [12]. The nanoparticle motions cause micro-convection of the suspending fluid, and thus enhance the overall heat transport in nanofluids. There exist four heat transport modes in a nanofluid, that is by the base fluid, by the nanoparticle, by the nanolayer and by the micro-convection. Figure 1 schematically shows the four involved heat transport modes.



Figure 1. Four heat flux modes in a nanofluid. $\vec{q}_t, \vec{q}_p, \vec{q}_l, \vec{q}_c, \vec{q}_f$ represent the total heat flux, heat flux through the nanoparticle, heat flux through the nanolayer, heat flux caused by micro-convection and heat flux through the base fluid.

The overall heat flux can be expressed as

$$\langle \vec{q}_{\rm t} \rangle = \langle \vec{q}_{\rm f} \rangle + \langle \vec{q}_{\rm e,p} \rangle + \langle \vec{q}_{\rm e,l} \rangle + \langle \vec{q}_{\rm e,c} \rangle, \tag{1}$$

where $\langle \rangle$ denotes the volume-averaged value, \vec{q} is the heat flux and the subscripts t, f, e, p, e, l and e, c represent the total value, base fluid, extra value of nanoparticle, extra value of nanolayer and extra value of micro-convection. $\langle \vec{q}_{\rm f} \rangle$, $\langle \vec{q}_{\rm e,p} \rangle$ and $\langle \vec{q}_{\rm e,l} \rangle$ can be written as [13]

$$\langle \vec{q}_{\rm f} \rangle = -k_{\rm f} \langle \nabla T \rangle,$$
 (2a)

$$\begin{aligned} \langle \vec{q}_{\mathrm{e,p}} \rangle &= \frac{1}{V} \frac{k_{\mathrm{p}} - k_{\mathrm{f}}}{k_{\mathrm{p}}} \int_{V_{\mathrm{p}}} \vec{q} \, \mathrm{d}V \\ &= \frac{1}{V} \frac{k_{\mathrm{p}} - k_{\mathrm{f}}}{k_{\mathrm{p}}} \int_{S_{\mathrm{p}}} \vec{r} \vec{q} \cdot \vec{n} \, \mathrm{d}S, \end{aligned} \tag{2b}$$

and

$$\langle \vec{q}_{e,l} \rangle = \frac{1}{V} \frac{k_l - k_f}{k_l} \int_{V_l} \vec{q} \, \mathrm{d}V$$

= $\frac{1}{V} \frac{k_l - k_f}{k_l} \int_{S_p + S_l} \vec{r} \vec{q} \cdot \vec{n} \, \mathrm{d}S.$ (2c)

Here ∇T is temperature gradient vector, *V* the total volume of the observed composite system, \vec{r} the position vector and \vec{n} the unit outward normal vector of the bounding surface. S_p and S_l denote the inner and outer bounding surface of the nanolayer. k_p and k_f represent the thermal conductivity of the nanoparticle and the base fluid, respectively.

To build up the effective thermal conductivity model, we consider a nanoparticle-liquid mixture with monosized spherical inclusions of radius r_p and particle volume concentration ϕ . As aforementioned, the alignment of the liquid molecules inside the solid-like interfacial nanolayer of thickness d is more ordered than that of the bulk liquid. The thickness, the microstructures and the physicochemical properties of this nanolayer are highly dependent on the suspended nanoparticle, the base fluid, and the interaction between them. Therefore the thermophysical behaviour of this interfacial nanolayer may be investigated by carefully analysing these factors. Although there is no available expression for calculating the thermal conductivity of the nanolayer on the surface of the nanoparticle, the solidlike nanolayer would be expected to have an intermediate thermal conductivity between that of the bulk liquid and that of the solid because the layered molecules are in an intermediate

physical state between the bulk liquid and the solid [11]. If the distribution of the thermal conductivity inside the nanolayer is k(r) ($r_p \le r \le r_p + d$), the thermal resistance presented by this layer, R_1 , is

$$R_{\rm l} = \int_{r_{\rm p}}^{r_{\rm p}+d} \frac{{\rm d}r}{4\pi r^2 k(r)}.$$
 (3)

 R_1 can also be expressed by the average thermal conductivity of the nanolayer, k_1 , as

$$R_{\rm l} = \frac{1}{4\pi k_{\rm l}} \left(\frac{1}{r_{\rm p}} - \frac{1}{r_{\rm p} + d} \right). \tag{4}$$

From equations (3) and (4), we have

$$k_{\rm l} = \frac{d}{r_{\rm p}(r_{\rm p}+d) \int_{r_{\rm p}}^{r_{\rm p}+d} ({\rm d}r/r^2 k(r))}.$$
 (5)

To decide k(r) is clearly 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

$$k(r) = \frac{k_{\rm f} - k_{\rm p}}{d}r + \frac{k_{\rm p}(r_{\rm p} + d) - k_{\rm f}r_{\rm p}}{d}.$$
 (6)

Substituting equation (6) into (5), one can derive the following expression

$$k_{\rm l} = \frac{k_{\rm f} M^2}{(M - \gamma) \ln(1 + M) + \gamma M},$$
(7)

with $M = \varepsilon_p(1 + \gamma) - 1$, where $\varepsilon_p = k_p/k_f$ is the ratio of the thermal conductivity of the solid to that of the base fluid and $\gamma = d/r_p$ is the ratio of the nanolayer thickness to the original particle radius. The total volume fraction of the original nanoparticle and nanolayer, ϕ_T , is

$$\phi_{\rm T} = \frac{4}{3} n \pi (r_{\rm p} + d)^3 = \phi (1 + \gamma)^3, \tag{8}$$

where $\phi = (4n\pi r_p^3)/3$ is the original volume fraction of nanoparticle and *n* is the particle number per volume.

A nanofluid consisting of base fluid and complex nanoparticles is statistically homogeneous and isotropic. Its effective thermal conductivity, k_{eff} , is defined following Fourier's law of heat conduction:

$$\langle \vec{q}_{\rm t} \rangle = -k_{\rm eff} \langle \nabla T \rangle. \tag{9}$$

The effective thermal conductivity can be obtained by matching equations (9) and (1). To determine $\langle \vec{q}_{e,p} \rangle$ and $\langle \vec{q}_{e,l} \rangle$, the temperature distributions inside nanoparticle–fluid mixture should be evaluated. Lu and Song [13] investigated the heat conduction in a suspension with random arrays of coated or debonded spherical inclusions by adopting equivalent hard sphere fluid model to represent the microstructure of mixtures and considering two interacting particles. Following the same approaches of Lu and Song, the total volume-averaged heat fluxes through the base fluid, the nanoparticles and the nanolayers can be expressed as

$$\langle \vec{q}_{\rm f} \rangle + \langle \vec{q}_{\rm e,p} \rangle + \langle \vec{q}_{\rm e,l} \rangle = -k_{\rm f} \langle \nabla T \rangle \\ \times \left[1 + 3\Theta \phi_{\rm T} + \frac{3\Theta^2 \phi_{\rm T}^2}{1 - \Theta \phi_{\rm T}} + G(r_{\rm p}, d, \phi, k_{\rm f}, k_{\rm p}, k_{\rm l}) \right]$$
(10)

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with

$$\Theta = \frac{\beta_{\rm lf} [(1+\gamma)^3 - (\beta_{\rm pl}/\beta_{\rm fl})]}{(1+\gamma)^3 + 2\beta_{\rm lf}\beta_{\rm pl}},\tag{11}$$

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

$$\langle \vec{q}_{\rm f} \rangle + \langle \vec{q}_{\rm e,p} \rangle + \langle \vec{q}_{\rm e,l} \rangle = -k_{\rm f} \langle \nabla T \rangle \left[1 + 3\Theta \phi_{\rm T} + \frac{3\Theta^2 \phi_{\rm T}^2}{1 - \Theta \phi_{\rm T}} \right].$$
(12)

Equation (12) describes the heat fluxes through the base fluid and the complex nanoparticles in a nanofluid. Now let us go one more step further to include the effect of micro-convection caused by the thermal motion of complex nanoparticles. From the molecular theory of heat, we can calculate the mean value of the instantaneous velocity which a particle may have at the absolute temperature T. Thus the kinetic energy of the motion of the centre of gravity of a particle is independent of the nature of its environment, e.g. the liquid in which the particle is suspended. This kinetic energy is equal to that of a monatomic gas molecule. Therefore, the mean velocity \bar{u} of the complex nanoparticle is determined by the equation [14]

$$\bar{u} = \sqrt{\frac{3kT}{m_{\rm c}}},\tag{13}$$

where m_c is the mass of the complex nanoparticle, and $k = 1.38 \times 10^{-23} \text{ JK}^{-1}$ is Boltzman constant. For the complex nanoparticle, m_c can be calculated as

$$m_{\rm c} = \frac{4}{3} \rho_{\rm p} \pi r_{\rm p}^3 \left\{ \frac{\rho_{\rm l}}{\rho_{\rm p}} \left[\left(1 + \frac{d}{r_{\rm p}} \right)^3 - 1 \right] + 1 \right\}, \qquad (14)$$

where ρ_p and ρ_l are the densities of the nanoparticle and the nanolayer, respectively. The relative motion between the suspended complex nanoparticle and the suspending base fluid brings about micro-convection around it, and thus augments the overall heat transport. Gupte *et al* [15] investigated the role of this micro-convection in the enhancement of the heat transfer at a macroscopic scale and established an expression to describe the extra heat flux. The formula is given as

$$\langle \vec{q}_{\mathrm{e,c}} \rangle = -k_{\mathrm{f}} \langle \nabla T \rangle F(Pe),$$
 (15)

where *F* is a function of Peclet number *Pe* and is given in [15] as $F(Pe) = 0.0556Pe + 0.1649Pe^2 - 0.0391Pe^3 + 0.0034Pe^4.$

Here Pe is defined as

$$Pe = \frac{\bar{u}L}{\alpha_{\rm f}} \phi_{\rm T}^{0.75},\tag{17}$$

where α_f is the thermal diffusivity of the base fluid and *L* is the specific length. *L* can be calculated from the following relation

$$L = (r_{\rm p} + d) \sqrt[3]{\frac{4\pi}{3\phi_{\rm T}}}.$$
 (18)



Figure 2. Dependence of the thermal conductivity enhancement on the reciprocal of nanoparticle radius.

Substituting equations (12) and (15) into equation (1), we arrive at

$$\langle \vec{q}_{\rm f} \rangle = -k_{\rm f} \langle \nabla T \rangle \left[1 + F(Pe) + 3\Theta \phi_{\rm T} + \frac{3\Theta^2 \phi_{\rm T}^2}{1 - \Theta \phi_{\rm T}} \right].$$
(19)

To compare equation (19) with equation (9), the expression for k_{eff} can be obtained as

$$k_{\rm eff} = k_{\rm f} \left[1 + F(Pe) + 3\Theta\phi_{\rm T} + \frac{3\Theta^2\phi_{\rm T}^2}{1 - \Theta\phi_{\rm T}} \right].$$
(20)

Equation (20) relates the thermal conductivity enhancement of a nanofluid to the system parameters such as the thermal conductivities of the base fluid, nanoparticle and nanolayer, the volume fraction, the particle size of nanoparticle, the thickness of nanolayer and the tempera-This equation is the proposed model deduced for ture. evaluating the effects of the nanoparticle presence and the particle motion on the effective thermal conductivity of a Since the thermal motion of the suspended nanofluid. nanoparticles shows a strong dependence on temperature, it is expected that the thermal conductivity of a nanofluid will vary remarkably with the temperature. Das et al [16] reported similar temperature-dependent thermal conductivity enhancement tendency for water-based nanofluids containing 38.4 nm Al₂O₃ nanoparticles. Nanofluids utilize fully the size effect of nanoparticles. To evaluate this effect, we calculated the effective thermal conductivity of Cu-EG nanofluids with copper nanoparticle concentration of 5.0%. In the calculations, the thermal conductivities were taken as $398\,W\,m^{-1}\,K^{-1}$ for Cu and $0.258 \text{ W m}^{-1} \text{ K}^{-1}$ for EG. The results reveal that the enhancement in the effective thermal conductivity increases with the reciprocal of the nanoparticle radius (figure 2). Due to the contributions of the nanolayer and the particle motion, the effective thermal conductivity augments with very large slope at small particle sizes. It is further shown in figure 2. that an increase in nanolayer thickness leads to a larger enhancement in the effective thermal conductivity. 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 nanoparticle-fluid mixtures through nanoparticle surface process.

The comparisons of the computed values by the present model to some available experimental data are shown



Figure 3. Comparisons between the calculated and the experimental data.

in figure 3. The observed nanofluids are copper nanoparticles in EG (Eastman et al 2001), copper oxide nanoparticles in EG (Lee et al 1999) and alumina nanoparticles in water (Masuda et al 1993). 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 because the thickness of the nanolayer is at a magnitude of nanometre. The thermal conductivities were taken as $32.9 \text{ W m}^{-1} \text{ K}^{-1}$ for CuO, $36.0 \text{ W} \text{ m}^{-1} \text{ K}^{-1}$ for Al_2O_3 and $0.610 \text{ W} \text{ m}^{-1} \text{ K}^{-1}$ for water. It is illustrated in figure 3 that the present model including the effects of nanolayer and nanoparticle motion predicts these experimental data quite well. Figure 3 further presents the values calculated by the Hamilton and Crosser (HC) model [7]. For a suspension containing spherical particles, the HC model has the following expression

$$\frac{k_{\rm eff}}{k_{\rm f}} = \frac{\varepsilon_{\rm p} + 2 - 2(1 - \varepsilon_{\rm p})\phi}{\varepsilon_{\rm p} + 2 + (1 - \varepsilon_{\rm p})\phi}.$$
(21)

This formula gives an effective thermal conductivity which is independent of the particle size. It is shown that the experimental data and the predicted values of our model are much larger than those values calculated by the HC model, indicating that the traditional model which gives reasonable predictions for the effective thermal conductivity of mixtures consisting of fluids and micro- or larger-sized particles cannot be used to predict the effective thermal conductivity of a nanofluid containing nanoscale particles. The larger the size of the suspended particles, the weaker appear the effects of the nanolayer and the thermal motion. When the average particle size reaches 1000 nm, the prediction of the current model is the same as that of a traditional model (HC model) without considering the interfacial layer and the particle motion (figure 3).

In this paper, we developed a theoretical model which includes the effects of the interfacial nanolayer and the particle motion for explaining the enhancement in the effective thermal conductivity of nanofluids. The parameters of a nanofluid system, such as the temperature, nanoparticle size, volume fraction, nanolayer thickness and thermal conductivities of the nanoparticle and the base fluid, have been shown to play roles in the enhanced thermal conductivity ratios. The calculated values fit quite well with some currently available experimental data.

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