To the Editor:

Thermal Conductivity of Nanofluids: Effect of Brownian Motion of Nanoparticles

The model of Xuan et al. (2003) for the thermal conductivity of nanofluids in which Brownian motion effect is added to the classical Maxwell's equation is discussed. The model is revised. Also, a different model is given and found to yield the same expression for the effective thermal conductivity after amending Xuan et al.'s model. The findings do not support the claim that Brownian motion of nanoparticles has a significant impact on thermal conductivity. Also, nanoparticles clustering is found to have a very minor effect on the effective thermal conductivity of nanofluids; however, the analysis may not be appropriate to draw conclusions about the impact of clustering.

Introduction

In a previous article by Xuan et al.,[1] entitled "Aggregation structure and thermal conductivity of nanofluids," the authors provided experimental data for water-copper nanofluid thermal conductivity using 10 and 50 nm nanoparticle radii, along with a model for the calculation of thermal conductivity. The objective of this letter is to discuss the validity of the theoretical model,[1] present a different model along with a revised one, then compare both models. In addition, we show the implications on the importance of Brownian motion of nanoparticles and clustering on the thermal conductivity of nanofluids. The model[1] is reviewed first (Eqs. 3 and 9−12 in Ref. 1), then a different model is presented, along with an amended Xuan et al.'s model showing the rational for the change, followed by a comparison.

Thermal Conductivity of the Nanofluid: Model of Xuan et al.[1]

Maxwell's equation provides the effective thermal conductivity of composite material made of dispersed solid substance in a continuous solid phase. The use of Maxwell's equation was found to underestimate the thermal conductivity of water-copper nanofluid,[1] as it strictly applies to solid materials. The effective thermal conductivity was assumed to be the sum of a term representing Maxwell's contribution and the other due to the random nanoparticles motion.[1]

$$\frac{k_{\text{eff}}}{k_{\text{f}}} = \frac{k_{\text{eff,m}}}{k_{\text{f}}} + \frac{k_{\text{eff,f}}}{k_{\text{f}}} \tag{1}$$

The conductivity ratio from Maxwell's equation is given by Eq. 9 in Ref. 1

$$\frac{k_{\text{eff,m}}}{k_{\text{f}}} = \frac{k_{\text{p}} + 2k_{\text{f}} - 2\varphi\left(k_{\text{f}} - k_{\text{p}}\right)}{k_{\text{p}} + 2k_{\text{f}} + \varphi\left(k_{\text{f}} - k_{\text{p}}\right)} \tag{2}$$

where $k_{\text{eff,m}}$ denotes the effective thermal conductivity, k_{f} and k_{p} are the thermal conductivities of the fluid and nanoparticles, respectively, and φ is the dispersed solid particles volume fraction.

Using Langevin's equation, the mean square displacement was found to be given by Ref. 1 (Eq. 3 in Ref. 1)

$$\langle x^2 \rangle = \frac{k_B T}{3\pi r_{\text{c}} \eta} t \tag{3}$$

where k_{B} is Boltzmann constant, r_{c} denotes[1] the "mean radius of gyration," T is temperature, and η represents the fluid viscosity.

Based on the above equation, the "displacement per second" was considered to be given by Eq. 10 in Ref. 1

$$l = \sqrt{\frac{k_B T}{3\pi r_{\text{c}} \eta}} \tag{4}$$

Using a procedure similar to the one in Ref. 2, the authors concluded[1] that the heat flux due to the particle clusters' irregular motion is (Eq. 11 in Ref. 1)

$$q_{\text{f}} = -\frac{1}{2} n_{\text{c}} m_{\text{c}} c_{\text{p}} l \frac{\partial T}{\partial x} \tag{5}$$

where n_{c} is defined as[1] the number of nanoparticles clusters, m_{c} is the cluster's mass, and c_{p} is the nanoparticle heat capacity. Substituting for l from Eq. 3 while using Fourier's law (Eq. 12) yielded[1]

$$k_{\text{eff,f}} = \frac{1}{2} \rho_{\text{p}} \varphi\, c_{\text{p}} \sqrt{\frac{k_B T}{3\pi r_{\text{c}} \eta}} \tag{6}$$

where ρ_{p} is the nanoparticles density.

Need for a Revised Model

From Eq. 2, it is clear that $\sqrt{\langle x^2 \rangle}$ is proportional to $t^{1/2}$ and Eq. 3 is incorrect as $\sqrt{\langle x^2 \rangle}$ is not proportional to time t. Also, the dimensions of the left and right sides of Eq. 6 are clearly inconsistent.

Present Model

In the present approach, the method in Bird et al.[2] is also used. Instead of using Langevin's equation as in Ref. 1, we use the Einstein–Smoluchowski and Stokes–Einstein equations.[3]

Referring to the notations in Figure 1, we have

$$\rho_{\text{p}} \frac{A\, l\, \varphi}{2} c_{\text{p}} T\big|_{x-\frac{l}{2}} - \rho_{\text{p}} \frac{A\, l\, \varphi}{2} c_{\text{p}} T\big|_{x+\frac{l}{2}} = k_{\text{eff,f}} \left[\frac{1}{l}\left(T\big|_{x-\frac{l}{2}} - T\big|_{x+\frac{l}{2}}\right)\right] A\, t \tag{7}$$

where A is the area for heat transfer (transverse to the x-direction for heat transfer). Simplifying the above equation gives

$$k_{\text{eff,f}} = \rho_{\text{p}} \frac{l^2}{2t} c_{\text{p}} \varphi \tag{8}$$

Using the Einstein–Smoluchowski equation, we can substitute for the diffusion coefficient D as[3]

$$D = \frac{l^2}{2t} \tag{9}$$

which leads to

$$k_{\text{eff,f}} = \rho_{\text{p}} D\, c_{\text{p}} \varphi \tag{10}$$

The Stokes–Einstein equation provides the expression for diffusivity for relatively large particles at low concentrations[3] (so that interactions between the particles can be neglected)

$$D = \frac{k_B T}{6\pi \eta r_{\text{c}}} \tag{11}$$

Substituting for D into Eq. 10 yields

$$k_{\text{eff,f}} = \rho_{\text{p}} \frac{k_B T}{6\pi \eta r_{\text{c}}} c_{\text{p}} \varphi \tag{12}$$

which is significantly different from Eq. 5 (Eq. 12 in Ref. 1).

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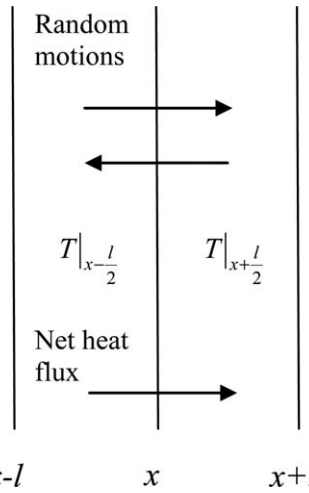

Figure 1. Schematic figure for the heat transfer associated with nanoparticles random motions.

Amended Model of Xuan et al.[1] and Comparison

Using Eq. 3 yields

$$l^2 = \frac{k_B T}{3\pi r_c \eta} t \tag{13}$$

The heat transfer per unit area during time t can be obtained from Eq. 5 as

$$q_f t = -\frac{1}{2}\frac{l\varphi\,\rho_p}{m_c} m_c c_p\, l \frac{\partial T}{\partial x} \tag{14}$$

Substituting for l^2 using Eq. 13 yields

$$q_f = -\rho_p \frac{k_B T}{6\,\pi\,\eta\,r_c}\, c_p\, \varphi\, \frac{\partial T}{\partial x} \tag{15}$$

which gives the same expression for effective thermal conductivity as the one obtained with the previous model, Eq. 12.

Results and Conclusions

We consider the copper-water nanofluid system for which experimental data[1] are available at $T = 296$ K. The physical properties values used for copper and water at $T = 293$ K are: $\rho_p = 8.92 \times 10^3$ kg/m^3, $c_p = 386.4$ J/kg K, and $\eta = 1.002 \times 10^{-3}$ kg/(m s). The thermal conductivities at $T = 300$ K are $k_p = 401$ W/(m K) and $k_f = 0.607$ W/(m K). For volume fraction φ ranging from 0.01 to 0.04 (experiments' range[1]), $k_{eff,m}/k_f$ range from 1.03 to 1.12 (also shown graphically in Ref. 1), while the contribution of Brownian motion of nanoparticles, $k_{eff,f}/k_f$, is found to be very small: 1.23×10^{-6}–4.92×10^{-6} for 10-nm copper particle size and 2.46×10^{-7}–9.84×10^{-7} for 50-nm particle radius.

If as in Ref. 1, we consider thermal conductivity to be additive (Eq. 2), it is clear that the results obtained do not support the idea that Brownian motion of nanoparticles is of significant contribution as a direct mechanism for thermal conductivity enhancement in nanofluids, which is in line with the conclusion of Keblinski et al.[4] Increasing r_c (nanoparticles clustering effect) does not affect significantly the effective thermal conductivity according to the model. However, this may not be conclusive as clustering might affect the effective thermal conductivity through mechanisms other than Brownian motions of nanoparticles.

Notation

A = area transverse to the direction of heat transfer, m^2
c_p = specific heat capacity, J/(kg K)
D = diffusion coefficient, m^2/s
k = thermal conductivity, W/(m K)
k_B = Boltzmann constant = 1.381×10^{-23} J/K
m = mass, kg
n = number
r = radius, m
t = time, s
T = temperature, K
x = Cartesian coordinate in the direction of heat transfer

Greek letters

l = mean displacement of a nanoparticle, m
φ = volume fraction of solid particles
η = viscosity, kg/(m s)
ρ = mass density, kg/m^3

Subscripts

c = cluster
eff = effective
f = fluid
m = Maxwell equation
p = solid particle

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