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Technical Note

Thermal conductivity enhancement of nanofluids in conjunction with electrical double layer (EDL)

Jung-Yeul Jung, Jung Yul Yoo *

School of Mechanical and Aerospace Engineering, Seoul National University, Seoul 151-742, Republic of Korea

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ABSTRACT

A novel expression for the thermal conductivity of nanofluids is proposed, which incorporates the kinetic theory to describe the contribution of the Brownian motion of the nanoparticles with a more realistic definition of the mean free path, and additionally to consider the contribution of the interparticle interaction due to the existence of the electrical double layer (EDL). It is shown that this model is applied to Au/water nanofluids satisfactorily with respect to temperature, volume fraction and particle size. In the case of dense Al_2O_3/water nanofluids, the effect of the interparticle interaction due to EDL on enhancing the thermal conductivity is more prominent than in the case of dilute Al_2O_3/water nanofluids. The model proposed in this paper shows that interparticle interaction due to EDL is the most responsible for the enhancement of thermal conductivity of nanofluids.

1. Introduction

Thermal conductivity of nanofluids (liquids with suspended nanoparticles) is significantly higher than that of the base liquid even for negligible nanoparticle concentrations. Therefore, nanofluids are attracting extensive attention as the next-generation heat transfer media. As is well known, Maxwell first proposed that liquids with suspended solid particles exhibit enhanced thermal conductivity [1]. However, it was Choi who practically led the way to utilize the enhanced thermal conductivity with stable suspensions of nanofluids [2].

Since then, various models have been proposed to explain the mechanism of thermal conductivity enhancement of nanofluids with increasing temperature and decreasing nanoparticle size [3–6]. By using molecular dynamics simulations, Vladkov and Barrat [7] showed that in the absence of collective effects, the heat conductivity of the nanofluid is well described by the classical Maxwell–Garnet equation model. Through an order-of-magnitude analysis, Prasher et al. showed [8,9] that the localized convection caused by the Brownian motion of the nanoparticles is the key mechanism for enhancing the effective thermal conductivity of nanofluids. Furthermore, the relationship between the Brownian motion and enhanced thermal conductivity was observed with the optical visualization of the particle motions and experimental measurement of thermal conductivity [10]. Recently, Lee et al. showed that surface charge states are mainly responsible for enhancing the thermal conductivity of nanofluids [11].

From the microscopic point of view, thermal conductivity of nanofluids is a very complex thermodynamic property which involves non-equilibrium behavior, so that the classical Maxwell model cannot explain its dependence on temperature. In fact, the kinetic theory reveals that the effective thermal conductivity of the particle is directly proportional to its mean velocity. Thus, the objective of the present study is to propose a theoretical model to which the kinetic theory is applied with a more realistic definition of the mean free path of the nanoparticle demonstrating the Brownian motion than in a previous study, and (more importantly) to which the kinetic theory is applied, for the first time, to take into account of the interparticle interaction due to the existence of EDL.

2. Theoretical approach

The model proposed in this study consists of the stationary mode (based on classical theory), the single particle motion mode (based on thermal conductivity of nanofluids deduced from the kinetic theory regarding the Brownian motion), and the interparticle interaction mode (based on thermal conductivity of nanofluids deduced from the kinetic theory regarding the particle motions induced by EDL).

2.1. Stationary mode

We start our argument by assuming that stationary thermal conductivity of nanofluids obeys the Maxwell model [1]. Therefore,

* Corresponding author. Tel.: +82 2 880 7112.
E-mail address: jyyoo@snu.ac.kr (J.Y. Yoo).

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Nomenclature

A	Coulomb constant (9.0×10^9 N m^2 C^{-2})
b	outer radius of virtual shell (m)
c	derivative constant (J m^{-2} K^{-1})
$\hat{c}_v$	specific heat (J K^{-1})
d	diameter of nanoparticle (m)
D	particle diffusion constant (m^2 s^{-1})
F	force (N)
k	thermal conductivity (kJ s^{-1} m^{-1} K^{-1})
k_B	Boltzmann constant (1.3807×10^{-23} J K^{-1})
l	mean free path of molecules or nanoparticles (m)
m	mass (kg)
N	number of atoms
n	particle concentration (m^{-3})
q	electric charge (C)
r	particle radius (m)
t	time (s)
T	absolute temperature (K)
T_D	Debye temperature (K)
$\bar{u}$	mean velocity (m s^{-1})

Greek letters

ε_0	permittivity of vacuum (8.8542×10^{-12} F m^{-1})
ε_{bf}	dielectric constant of medium
ζ	zeta potential (mV)
κ	Debye–Hückel parameter (m^{-1})
ϕ	volume fraction of nanoparticle
μ	viscosity of medium (N s m^{-2})
τ	relaxation time (s)

Subscripts

Br	Brownian motion
EDL	electrical double layer
eff	effective
bf	base fluid
Mw	Maxwell
np	nanoparticle

enhanced thermal conductivity by the Maxwell model is expressed as

$$k_{Mw} = k_{bf}\left(\frac{k_{np} + 2k_{bf} + 2(k_{np} - k_{bf})\phi}{k_{np} + 2k_{bf} - (k_{np} - k_{bf})\phi}\right). \quad (1)$$

This model is valid only when the thermal energy transport in nanofluids takes place as a diffusive process and cannot apparently consider other effects such as particle size, concentration, and temperature of nanofluids considered.

2.2. Single particle motion mode

The Brownian motion which represents the behavior of nanoparticles in colloids is characterized by the Stokes–Einstein relation in the form of $D = k_B T/3\pi\mu d$, so that the mean velocity of nanoparticles due to the Brownian motion can be calculated as

$$\bar{u}_{Br} = \frac{k_B T}{3\pi\mu d_{np} l_{bf}}. \quad (2)$$

We assume that a nanoparticle freely moves over the length of the mean free path of the base fluid molecules [4].

The effective thermal conductivity of the particle is directly proportional to its mean velocity according to the kinetic theory [12]. Thus the thermal conductivity due to the particle motion can be expressed as

$$k_{Br} = c\bar{u}_{Br}. \quad (3)$$

An estimate using the kinetic theory shows that $c = nl_{np}\hat{c}_v/3$, where $\hat{c}_v$ is the specific heat per particle which is estimated by the Debye model for the particle. In the present study, $\hat{c}_v = 3Nk_B$ for Au particle or $\hat{c}_v = 3Nk_B\frac{4\pi^4}{5}\left(\frac{T}{T_D}\right)^3$ for Al_2O_3 particle [12]. Therefore, the thermal conductivity due to the Brownian motion can be written as

$$k_{Br} = \frac{nl_{np}\hat{c}_v}{3}\frac{k_B T}{3\pi\mu d_{np} l_{bf}}. \quad (4)$$

However, without giving any detailed account, Kumar et al. simply assumed that l_{np} of nanoparticles was on the order of 1 cm to obtain high values of thermal conductivity [13]. The gold particles used were 17-nm in mean diameter and 0.00026 vol%, so that the distance between the particles was approximately 1.24 μm (which was approximately 73 times longer than the particle diameter). This assumption is not only unphysical but also inconsistent with their own experimental conditions, as will be shown later. On the other hand, in the present study, we used $l_{np} = 1/\left(\sqrt{2}n\pi d_{np}^2\right)$ as the mean free path of the nanoparticle based on the assumption that *all particles move about with a Maxwellian velocity distribution* [12], which is physically more viable.

2.3. Interparticle interaction mode

In the light of the fact that most colloidal particles are surrounded by electrical double layers (EDLs), it can be readily assumed that nanoparticles suspended in the base fluid in the form of colloidal particles are also surrounded by EDLs. General characteristics of the colloid are introduced in Ref. [14]. Dual forces, i.e. repulsion due to Coulomb force and attraction due to van der Waals force, respectively, are exerted on nanoparticles in the colloid. Coulomb force prevents coalescence of the particles, but van der Waals force induces coalescence if the distance between the particles exceeds the energy barrier. Due to the existence of the EDL, sufficient Coulomb force acts on nanoparticles in stable nanofluids [14]. Therefore, we can derive an expression for thermal conductivity enhancement of nanofluids by using the concepts of the interparticle interaction and the kinetic theory [12]. In this study, the interparticle interactions are considered by using the Kuwabara's cell model [15], as shown in Fig. 1, where each sphere is enclosed by a virtual shell of outer

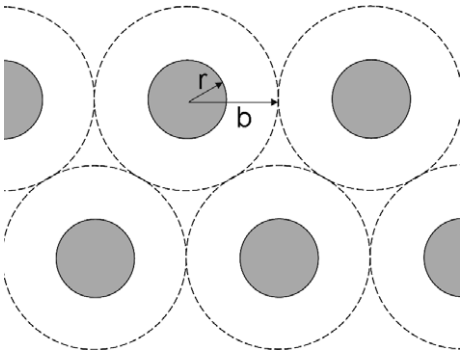

Fig. 1. Spherical particles of radius r in concentrated suspensions in the cell model [15].