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An explanation of the Al_2O_3 nanofluid thermal conductivity based on the phonon theory of liquid

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ABSTRACT

In the present work a systematic investigation on several mechanisms affecting the thermal conductivity of Alumina based nanofluid, such as layering, Brownian motion, clustering, ballistic phonon motion, thermal boundary resistance and mass difference scattering, is presented. The effect of mass difference scattering is for the first time suggested and studied in the present work. Both theoretical and experimental approaches have been carried out in order to analyze the competition of these phenomena and to identify the most relevant. This was obtained by comparing micrometric and nanometric particles suspended in liquid water (293 K), frozen water (253 K) and diathermic oil (293 K). Each of the above-mentioned conditions was selected to make dominant only one of the mechanisms that affect nanofluid thermal conductivity. The main results of this investigation concern the mass difference scattering, which has been found to be the most intensive mechanism reducing the nanofluid thermal conductivity with respect to the microfluid one.

1. Introduction

Adding solid particles to improve thermal conductivity of liquids, widely used in heat transfer applications (e.g. water, ethylene glycol, oil, etc.), is a problem of obvious practical importance. This issue was first studied by Maxwell [1], since 1881, who provided a mathematical model able to describe the thermal conductivity of randomly distributed and non-interacting homogeneous spheres in a homogeneous medium. Due to sedimentation and erosion phenomena, related to bulk dimension of particles, this idea has not found application for a long time. These problems are, nowadays, overcome by means of nanometric particles (*NPs*) [2], which, if added to a base fluid (*BF*), give rise to a mixture, named nanofluid (*NF*) by Choi in 1995 [3].

According to Mahian *et al.* [4,5], the development and use of nanofluids have found a wide range of applications in consumer products, nanomedicine, energy conversion, and microsystem cooling. Furthermore, of special interest is the use of *NF* flow for enhanced convection heat transfer to achieve rapid cooling of high heat-flux devices [6,7].

Nevertheless, nanometric scale involves an interesting effect if compared to micrometric one: experimental thermal conductivity of *NFs* is lower than microfluids (*MFs*) and often is not predictable by means of the theoretical Maxwell model. Actually, Beck *et al.* [8] measured the thermal conductivity enhancement in seven nanofluids, containing 8÷282 nm diameter Alumina *NPs* in water and ethylene glycol, demonstrating a direct correlation with particle size: the thermal conductivity enhancement in these nanofluids decreases as the particle size decreases. Özerinç *et al.* [9] showed an experimental data set prepared by combining the results of five different research groups and compared with the predictions of two theoretical models. As seen from this study, there is significant discrepancy in experimental data, but the general trend is increasing thermal conductivity with increasing particle size. Lomascolo *et al.* [10] refer that the effect of nanoparticle size on thermal properties of *NFs* is still today a question, which is far from being answered, since the results reported in the literature are contradictory, but for the water-based Al_2O_3 *NF*, thermal conductivity seems to increase as nanoparticle size increases.

A rational approach to solve this issue should be based on knowledge of complex phenomena, which influence thermal conductivity of *NFs*.

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Nomenclature

A	surface [m^2]
c	average velocity of sound in a medium [m/s]
C_v	volumetric heat capacity [J/m^3 K]
d	average diameter of the particles [m]
h_{Bd}	thermal boundary conductivity [W/m^2 K]
$\hbar$	reduced Planck constant [m^2 kg/s]
K_B	Boltzmann constant [J/K]
G_∞	instantaneous shear modulus [N/m^2]
N_{NP}	numbers of atoms in the *NP*
N_{NPj}	density of phonons in the medium *NP*
$\dot{Q}$	thermal power [W]
r	average radius of the particles [m]
R_k	Kapitza resistance [m^2 K/W]
T	temperature [T]
V	volume [m^3]
X_v	volumetric fraction of the suspension
Γ_{NPj}	parameter, whose expression is evaluated by Eq. (6)
η	liquid viscosity [N s/m^2]
λ	thermal conductivity [W/m K]
λ_{ph}	phonons wavelength [m]
τ_{BF}	relaxation time of phonons in base fluid [s]
τ_C	combined relaxation time [s]
τ_D	Debye relaxation time [s]
τ_F	Frenkel relaxation time [s]
τ_P	contribution of relaxation time due to the mass-difference impurity related to the presence of particles (scatters) [s]
ω	frequency [1/s]
ω_D	Debye frequency [1/s]
ω_{NP}^{Debye}	Debye cutoff frequency in the *NP* [1/s]
ω_F	Frenkel frequency [1/s]

Abbreviations

BF	base fluid
NP	nanoparticle
MP	microparticle
NF	nanofluid
MF	microfluid
MFP	mean free path [m]
MDS	Mass Difference Scattering

The results reported in the present work, obtained on Alumina *NFs*, showed that the high number of *NPs* makes mass difference scattering in *BF* an authoritative hypothesis for the explanation of the observed anomalous thermal behavior in *NFs*.

2. Potential mechanisms that influence thermal conductivity of *MFs* and *NFs*

Thermal conductivity of micrometric and nanometric Al_2O_3 suspensions can be influenced by different phenomena, such as layering, Brownian motion, clustering, ballistic phonon motion, thermal boundary resistance and mass difference scattering, the latter for the first time suggested in this work. The relative weight of these phenomena can change deeply going from micrometric to nanometric scale, as it will be demonstrated next.

2.1. Layering

Many authors proposed the formation of a stratified fluid layer with peculiar thermal properties surrounding the nanoparticles (*layering*) as a phenomenon responsible for the anomalous thermal behavior of *NFs*.

An analytical way to study the features of layered structures was proposed by Lee [11] who applied colloidal science to obtain an equation for thickness and thermal conductivity of layer, directly connected with the surface-charge-induced electrical double layer. According to the above said model, thickness should depend on *BF* temperature and pH. The *NF* thermal conductivity is instead limited between *BF* and *NP* ones and expressed as a power of the total charged surface density and ion density in the electrical double layer formed at the *NP-BF* interface.

Yu *et al.* [12] developed a modified Maxwell model able to consider layering. Nevertheless, in this model thickness and thermal conductivity of layer were only assumed and not experimentally evaluated.

Another model comes from a study of Leong *et al.* [13]. In this case the main features of stratified structures around *NP* (layers) are taken into account by means of a model based on *NP* dimension, thickness of layer (assumed equal to 1 nm) and thermal conductivity of layer (assumed equal to 2−3 λ_{BF} according to Yu *et al.* [12]). Even though the prediction agrees well with Alumina *NF* experimental data, the authors conclude explicating the need of a theoretical model to determine layer thickness and its thermal properties.

Huaqing *et al.* [14] considered the layer as an intermediate physical state between bulk liquid and *NP*, with continuous thermal conductivity distribution from liquid to solid. Then, the theoretical value of *NF* thermal conductivity was evaluated, starting from a general solution of heat conduction equation and assuming thickness of layer equal to 2 nm. In this way, numerical and experimental data (available in literature) were compared, getting a quite good prediction.

A numerical way to study the features of layered structures is represented by molecular dynamics simulation. Li *et al.* [15], with an equilibrium molecular dynamics simulation, estimated the thickness of the layer around *NP* equal to 0.5 nm, hence lower than the thickness supposed in the previous discussions. This value of thickness, applied to the above-mentioned mathematical models, decreases significantly the weight of layering phenomenon.

This conclusion is in accord with Keblinski *et al.* [16], which assuming thermal conductivity of layer cannot be greater than the *NP* one, concluded that layering cannot influence thermal behavior of *NFs*. A similar result has been obtained by Xue *et al.* [17], which by means of non-equilibrium molecular dynamics simulations found no effect on *NF* thermal conductivity, suggesting that the experimentally observed large enhancement of thermal conductivity in *NFs* cannot be explained by altered thermal transport properties of the layered liquid.

A recent study of the layering phenomenon comes from Milanese *et al.* [18], who compared molecular dynamics simulation results to experimental thermal conductivity values for metal (Cu) and metal oxide (CuO) *NPs* in water. The results of this work highlight the presence of layering in metal based *NFs* only, in contrast to the metal oxide case, where no stratified structures were detected. From the above-mentioned literature it is possible to conclude that the weight of layering phenomenon on Al_2O_3 *NF* thermal conductivity can be considered negligible. Besides, all studies are in agreement to establish this phenomenon produces an