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Aggregation based model for heat conduction mechanism in nanofluids

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ABSTRACT

A new mathematic model for heat conduction of nanofluid is developed based on the experimental findings. Effective medium theory, nanolayer of liquid molecule around solid particle, aggregation, nano-convection due to the Brownian motion of nanoparticle, and interfacial thermal resistance are included to elucidate the heat conduction mechanism in nanofluids. The analytical result fits well with the experimental data and the maximum deviation is obtained to be 1.52% for SiO₂ nanofluid. The effects of aggregate shape (i.e., ellipsoid, sphere and fiber) and its size are investigated to evaluate the thermal conductivity of the nanofluids. The prediction shows that nano-convection induced by the movement of aggregates, is leading the main contribution for thermal conductivity enhancement at a low concentration of ~0.1 vol%. Thermal conductivity of aggregate becomes crucial to affect the static contribution for the enhancement. In addition, it is found that the interfacial thermal resistance and nanolayer have little effect on the thermal conductivity enhancement of nanofluids at a very low concentration of nanoparticles.

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1. Introduction

Nanofluids are produced by dispersing nanoparticles in the conventional fluid and generally which will be processed by ultra-sonication vibration or by adding chemical surfactant to improve the dispersion stability $[1]$. In the past decades, thermal conductivity of nanofluids as the essential heat transfer property has been attracted the most widely attention due to its great theoretical and practical interests to scientists and engineers. Significantly enhanced thermal conductivity of nanofluids has been reported in a number of experimental results just as described in the review literatures $[2-5]$. Lee et al. $[4]$ have summarized the previous thermal conductivity data, and the advanced features of thermal conductivity in nanofluids were proposed, such as anomalously high enhancement at low nanoparticle concentration, nonlinear relationship between the enhancement and the concentration, temperature-/size- and pH-dependent thermal conductivity enhancement. The specific parameters that would affect the enhancement were reviewed as well [\[5,6\]](#page--1-0), which are (1) nanoparticle material, shape, size and concentration; (2) basefluid, temperature and pH. However, most of the available thermal conductivity data from different contributors could not agree well with each other because of the various experimental conditions and the measurement deviations. Thus, theoretical investigation on the enhanced thermal conductivity of nanofluids becomes strongly important to understand and to improve the heat transfer enhancement in practice.

To elucidate the mechanism of heat conduction in nanofluids, investigations by numerical model and visualization of nanoscale level have been extensively reported. Keblinski et al. [\[7\]](#page--1-0) in 2002 explored four possible explanations for the anomously enhanced thermal conductivity, which are the Brownian motion of nanoparticles, molecular-level layering of the liquid at the liquid/particle interface, the nature of heat transport in the nanoparticles, and the effect of nanoparticles clustering. They finally concluded that the ballistic combined with direct or fluid-mediated clustering effects were the main contributions to explain the significant enhancement of thermal conductivity. Yu and Choi [\[8\]](#page--1-0) introduced the concept that a nanolayer acted as a thermal bridge between nanoparticles and bulk liquid, and developed a renovated Maxwell model for the effective thermal conductivity of nanofluids containing spherical nanoparticles with an ordered nanolayer. In 2004, they extended the concept of the thermal bridge nanolayer to non-spherical nanoparticles and renovated the Hamilton–Crosser model [\[9\].](#page--1-0) Generally, the Brownian motion in nanofluids would be categorized into collision between nanoparticles and convection induced by Brownian nanoparticles. However, the former Brownian motion should be negligible because Brownian nanoparticle diffusion is much slower than thermal diffusion [\[7\].](#page--1-0) Furthermore, aggregation which is induced by the random movement of nanoparticles is one of the mostly discussed mechanisms for enhanced heat conduction in nanofluids [\[4,5\]](#page--1-0).

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In our previous study, we firstly developed the methanol based $SiO₂$ nanofluids and measured the thermal conductivity for $CO₂$ absorption enhancement in synthetic natural gas (SNG) systems [\[10–12\]](#page--1-0). The aggregation of nanoparticles was observed in nanofluids by measuring the particle size, and the result showed that significant thermal conductivity enhancement was obtained to be 10% for SiO₂ nanofluids at a nanoparticle concentration of 0.1 vol%. In the present work, a new model is built which is based on the effective medium theory and includes the effect of liquid molecule nanolayer, aggregation induced by the Brownian nanoparticles and the formative nano-convection. In the present aggregation based model, which takes into account of the effects of interfacial thermal resistance (Kapitza resistance), aggregate shape and size, and the non-linear phenomenon of thermal conductivity enhancement is included as well. Reason for the present work accomplished at low nanoparticle concentrations, less than 0.1 vol%, is due to consider the importance of dispersion stability of nanofluids in the industrial application. The problem of particle sedimentation is too hard to control at a high particle concentration, and which will result in some non-negligible drawbacks like erosion of the components, clogging in small passages and increased pressure drop [\[13,14\].](#page--1-0) In addition, the heat conduction mechanisms in nanofluids could be categorized into static and dynamic mechanisms. Static mechanisms include nanolayer, natural thermal transport in aggregate, interface thermal resistance, and fractal geometry, while dynamic mechanisms include the induced nano-convection $[4]$. In this study, the distributions of static and dynamic contribution proportions for methanol based $SiO₂$ nanofluids would be evaluated, furthermore, the thermal conductivity of aggregates would be investigated as well which would affect the static contribution.

2. Heat conduction mechanism and mathematical model

2.1. Aggregation of nanoparticles

Nanoparticles suspended in liquid would aggregate with each other to form chain structure which has been confirmed experimentally $[15-18]$. Prasher et al. $[19]$ studied the effect of aggregation on thermal conductivity enhancement by the analysis of aggregation kinetics of nanoscale colloidal solutions combined with the physics of thermal transport. They have reported that the aggregation time constant decreases rapidly with decreasing nanoparticle size and the thermal conductivity enhancement increases with increasing level of aggregation, leveling off after the optimum level of aggregation is reached. Evans et al. [\[20\]](#page--1-0) revealed that clustering could result in fast transport of heat along relatively large distances since heat could be conducted much faster by solid particles. They also proposed that the effective thermal conductivity increased with an increasing of cluster size. Gao et al. [\[21\]](#page--1-0) experimentally investigated the enhancement mechanism by thermal conductivity measurements and structural analysis for the same materials in both liquid and solid states and have strongly suggested that the clustering holds the key contribution for the enhancement. Guided by this in-sight, Wang et al. [\[22\]](#page--1-0) used graphite flakes as additives and developed stable graphite suspensions in water and oil to discuss the heat conduction mechanisms in nanofluids. Thermal percolation phenomenon was observed and explained based on combined optical and AC impedance spectroscopy studies.

Following well-established understanding of the fractal morphology of nanoparticle aggregate (clusters) in colloids [\[23\],](#page--1-0) a fractal aggregate is embedded within a sphere and is composed of a few approximately linear chains, which span the whole aggregate and side chains. The linear chains which span the whole aggregate are called the backbone. The other particles, which do not span the whole aggregate, are called dead ends [\[20\].](#page--1-0) The backbone plays a significant role in the rheology of colloids because it is the only structure that can transfer elastic forces between aggregate. Due to its connectivity, the backbone is also expected to play a crucial role in thermal conductivity. Fig. 1 describes the schematics of the independent single aggregate which includes the backbone and dead-end particles. To further understand the effect of aggregation on the fluid properties the thermal conductivity are modeled using fractal theory. Therefore, the thermal conductivities of aggregates are estimated by separating them into two components, the percolation contributing backbone, and non-percolation contributing dead-end particles. Considering the interfacial thermal resistance (Kapitza resistance), the effective thermal conductivity of dead-end particles based suspension (including the basefluid and dead-end particles) is given by using Maxwell model [\[24\]](#page--1-0)

$$
k_{bf,de} = \frac{[k_{pn}(1+2\alpha)+2k_{bf}] + 2\phi_{pa,de}[k_{pn}(1-\alpha)-k_{bf}]}{[k_{pn}(1+2\alpha)+2k_{bf}] - \phi_{pa,de}[k_{pn}(1-\alpha)-k_{bf}]} \tag{1}
$$

where k_{pn} , k_{bf} and $\phi_{pa,de}$ represent the thermal conductivity of particles with nanolayer, thermal conductivity of basefluid and the volume fraction of dead-end particles in the aggregate, respectively. α = 2 $R_b k_{b} d_p$, R_b and d_p represent the thermal resistance and powder diameter, respectively. The thermal conductivity of particles with nanolayer would be estimated by using Yu and Choi model $[8]$. The effective thermal conductivity of aggregate, k_a , is determined using composite theory for misoriented ellipsoidal particles for the backbone, in a matrix of the non-percolation contributing portion, the following equations are used [\[25\]](#page--1-0)

$$
k_a = k_{bf,de} \frac{3 + \phi_{pa,ba} [2\beta_{11}(1 - L_{11}) + \beta_{33}(1 - L_{33})]}{3 - \phi_{pa,ba} (2\beta_{11}L_{11} + \beta_{33}L_{33})}
$$
(2)

where $\phi_{pa,ba}$ represents the volume fraction of backbone in the aggregate. L_{ii} , $i = 1,3$, are well-known geometrical factors which are dependent upon the particle shape, and it is given by $L_{11} = \frac{p^2}{2(p^2-1)} - \frac{p}{2(p^2-1)^{3/2}} \cosh^{-1} p$, and $L_{33} = 1 - 2L_{11}$. p is the aspect ratio of the ellipsoid, which generally is determined to be the ratio be-tween radius of the aggregate to the radius of the particle [\[20,26\].](#page--1-0) $\beta_{ii}=\frac{k_{ii}^c-k_{bf,de}}{k_{bf,de}+L_{ii}(k_{ii}^c-k_{bf,de})}$, and the equivalent thermal conductivities along the symmetric axis of this ellipsoidal particles are determined by $k_{ii}^c = \frac{k_{pn}}{(1 + iL_{ii}k_{pn}/k_{bf})}$, and $\lambda = \alpha(2 + 1/p)$. When the ellipsoidal inclusions become spheres, $p = 1$, $L_{11} = L_{33} = 1/3$, then Eq. (2) reduce to

$$
k_a = k_{bf,de} \frac{k_{pn}(1+2\alpha) + 2k_{bf,de} + 2\phi_{pa,ba}[k_{pn}(1-\alpha) - k_{bf,de}]}{k_{pn}(1+2\alpha) + 2k_{bf,de} - \phi_{pa,ba}[k_{pn}(1-\alpha) - k_{bf,de}]} \tag{3}
$$

For continuous fiber composites with uniformly distributed long fibers, $1/p = 0$, $L_{11} = 0.5$, $L_{33} = 0$, then the mainly effective thermal conductivity would be expressed

$$
k_a = (1 - \phi_{pa,ba})k_{bf,de} + \phi_{pa,ba}k_{pn}
$$
\n
$$
\tag{4}
$$

Fig. 1. Aggregate of nanoparticles, including the backbone and dead-end particles.

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