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Modeling of Thermal Conductivity of Nanofluids Considering Aggregation and Interfacial Thermal Resistance

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Abstract

A model with the consideration of particle size, aggregate size and interfacial thermal resistance is developed to predict the thermal conductivity of nanofluids. Interfacial thermal resistance is modeled to have relationship with the equivalent particle size in terms of keeping thermal resistance constant. The shape factor of aggregate is determined by the number of particles in the aggregate. The present model agrees well with the wide accepted experimental data. It concludes that particle size and aggregate size have positive effect on the thermal conductivity enhancement, since the increase of particle size can weaken the effect of interfacial thermal resistance, and the increase of aggregate size can offer fast heat transfer path for adjacent particles and it significantly increases the shape factor of aggregate. The thermal conductivity of nanofluids increases linearly with particle volume fraction and the increase rate differs according to particle size and aggregate size. The inferred values of interfacial thermal resistance are in a reasonable range and fit well with different experimental data. If the particle volume fraction is lower than 0.1% or the particle size is smaller than 10 nm without aggregation, the factors of nano-convection and nanolayer need to be taken into account.

Key words

Nanofluids, Thermal conductivity, Theoretical model, Aggregate size, Interfacial thermal resistance

1. Introduction

The discovery of nanofluids, in which solid nanoparticles disperse in conventional base fluid, has aroused the interests worldwide in the last two decades. Numerous studies showed that, compared with base fluid, nanofluids can dramatically improve thermal conductivity, convective heat transfer and solar energy absorption features.[1-3]

Reports showed that the thermal conductivity, k, of nanofluids depended on factors like particle volume fraction (ϕ), single particle diameter (d), particle morphology, additives, pH value, temperature, nature of the base fluid and particle materials etc.[4] The studies of the effect of particle size on the k of nanofluids concluded conflicting reports. Patel et al.[5] and Cui et al.[6] used experimental method and the molecular dynamics (MD) simulations respectively to find out that the k increased with the reduction of particle diameter. However, Beck et al.[7] measured the k of nanofluids that contain nanoparticles of different diameters and found an enhancement in k as particle size increased. Considering temperature, most studies showed an enhancement in k with the increase of temperature.[5,8,9] Taking the materials of base fluid and nanoparticles into account, most of the studies showed an increase in k with the reduction of k of the base fluid and the increase of k of the nanoparticles.[5,10] The shape of particles and the forming of clusters in nanofluids significantly influenced the k of nanofluids. The benchmark study on alumina nanoparticles and nanorods in PAO showed that the k was higher if the particles have larger aspect ratio (nanorods).1[1] Particles in the nanofluids were prone to form aggregates, and the reason why aggregates can enhance the k of nanofluids was because aggregates created paths of lower thermal resistance among particles and heat could conduct rapidly in the cluster. What is more, aggregates set up percolating structures and the effective volume of aggregates could be much larger than the total volume of particles.[12,13] The factors of pH value and the addition of sufactant on k were also studied. Lee et al.[14] found that since the pH value was far from the isoelectric point, the particle size changed and particles became more stable. Younes et al.[15]

demonstrated that the pH value affected zeta potentials and aggregate size, and surfactant could separate particles to avoid forming clusters and stabilize nanofluids.

The mechanisms of the unusual high thermal conductivity of nanofluids with low particle volume fraction are controversial. Maxwell model was firstly introduced to model the *k* of nanofluids based on the effective medium theory (EMT).[16] Hamilton[17] took particles shape effects into account. The phenomenon that the measured *k* of nanofluids was anomalously greater than theoretical predictions attracted considerable attentions.[1] The existence of an ordered layer of liquid molecules at the solid-liquid interface was experimentally proved, and it could measurably increase the *k* of nanofluids when particle diameter was below 10 nm.[18,19] Brownian motion and the convective heat transfer induced by Brownian motion of nanoparticles were considered as the mechanisms of the enhancement of *k* in nanofluids.[20-22] On the contrary, Gao et al.[23] measured the *k* of nanofluids in both liquid and solid states and figured that the effect of Brownian motion on *k* of nanofluids was much less than that of the clustering formation.

Mathematical models to predict the *k* of nanofluids are built on many investigations. Brownian motion of particles may result in convection-like effects on the nanoscale.[24] Prasher et al.[21] introduced a model considering local convection caused by Brownian movement of particles based on EMT. The factor of convection was given $k_{con} = (1 + A\,\mathrm{Re}^{m}\,\mathrm{Pr}^{0.333}\,\phi)k_f$, where *Re* and *Pr* are Reynolds number and Prantl number respectively, and *A* and *m* are constants determined by experimental results. Yu et al.[19] modified the Maxwell model to include the effect of the ordered nanolayer. With the nanolayer of thickness *h* attaching to the surface of particles, the equivalent particle radius became *r*+*h* and the equivalent particle volume fraction increased. The effect of the ordered layer was rapidly weakened as particle diameter increased and it wore off if particle diameter was larger than 10 nm. Increasing evidence suggests that EMT can estimate thermal conductivity considering the effect of aggregation and interfacial resistance.[10,25] Based on the study of Nan et al.[26] who introduced a methodology to predict the *k* of particulate composites with interfacial

thermal resistance, Prasher et al.[12] and Evans et al.[27] built a three-level homogenization model to evaluate the *k* of colloids, assuming that an aggregate was composed of a few linear chains that span the whole cluster and side chains. In the first level, the *k* of aggregate with dead end was calculated by Bruggeman model, then the *k* of the aggregate that includes a backbone was calculated by the model of Nan et al. Finally, the *k* of nanofluids with aggregates was obtained by Maxwell model. Based on the three-level homogenization, Okeke et al.[28] numerically investigated the *k* of nanofluids with the consideration of different factors and found that aggregate size affected thermal conductivity while interfacial thermal resistance did not play the major role. Zhou et al.[29] built a model based on particle size distribution and found that the *k* of nanofluids could be enhanced with clusters in it.

In this paper, the authors build a model to predict the *k* of nanofluids based on the EMT, considering the factor of particle size, aggregate size, particle volume fraction and interfacial thermal resistance. According to this model, the increase of interfacial thermal resistance is equivalent to the decrease of particle size in terms of keeping thermal resistance constant, which further affects the effective particle volume fraction. Aggregate size affects the shape factor and the number of particles in the aggregate. Then the predictions of present model are compared with experimental results in the literature and they are in good agreement. After that, various simulations of the *k* of nanofluids are established to analyze factors such as particle size, aggregate size and particle volume fraction on the *k* of nanofluids. Finally, further discussion on the divergence of present model and some experimental data is made.

2. Model development

The schematic of a particle in nanofluids is shown in Fig. 1. The particle is assumed to be sphere and has a radius of r_2. The thermal resistance in nanofluids contains particle thermal resistance $R_p\,(K/W)$, fluid thermal resistance $R_f\,(K/W)$ and interfacial thermal resistance $R_b\,(Km^2/W)$ at the solid/liquid interface. The boundary conditions at the interface can be expressed as[30]

$$k_p \frac{\partial T_p}{\partial r} = k_f \frac{\partial T_f}{\partial r}, \quad (1)$$

$$T_p - T_f = -k_p R_b \frac{\partial T_p}{\partial r}, \quad (2)$$

where k_p and k_f are particle and base fluid thermal conductivity (W/mK) respectively, T_p and T_f are particle temperature and base fluid at the solid/liquid interface, and *r* refers to radius vector.

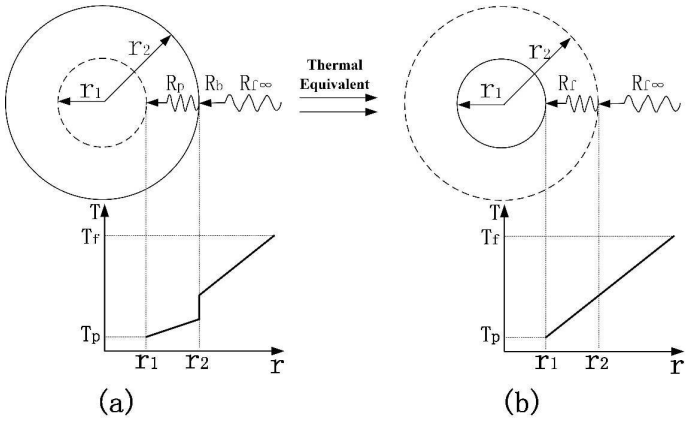


Fig. 1. The schematic of the geometry of a particle. The circle with solid line shows (a) the actual particle and (b) the thermal equivalent particle. The temperature change along the radius direction when assuming there is temperature gradient around a particle is also presented. Nomenclature $R_{f,\infty}$ refers to water thermal resistance around the particle.

Considering that there is temperature gradient around a particle, as shown in Fig. 1, temperature difference exists at the solid/liquid interface due to interfacial thermal resistance. Since temperature difference makes it difficult to model the process of heat conduction, we assume that there is no temperature difference at the solid/liquid interface and particle radius is smaller than r_2, as depicted in Fig. 1(b). We can get certain value of hypothetical particle radius r_1 that make the hypothetical particle thermal resistance (Fig. 1(b)) equal to the original particle thermal resistance (Fig. 1(a)).

The thermal resistance of the sphere between the surfaces of radius r_1 and r_2 that contains interfacial thermal resistance can be expressed as

$$R = \frac{1}{4\pi k_p}\left(\frac{1}{r_1} - \frac{1}{r_2}\right) + \frac{R_b}{4\pi r_2^2}. \quad (3)$$

Assuming that there is no interfacial thermal resistance at the solid/liquid interface and particle radius is r_1, as shown in Fig. 1(b), the space between r_1 and r_2 is filled with liquid, so the thermal resistance between the surfaces of radius r_1 and r_2 becomes

$$R' = \frac{1}{4\pi k_f}\left(\frac{1}{r_1} - \frac{1}{r_2}\right). \quad (4)$$

Then we can get certain value of r_1 that makes the thermal resistance in Fig. 1(a) and Fig. 1(b) be equal. If so, the thermal resistance of the original particle and the hypothetical particle are the same, which is called "thermal equivalent condition". In this condition we can get

$$\frac{R_b}{4\pi r_2^2} + \frac{1}{4\pi k_p}\left(\frac{1}{r_1} - \frac{1}{r_2}\right) = \frac{1}{4\pi k_f}\left(\frac{1}{r_1} - \frac{1}{r_2}\right). \quad (5)$$

Transforming Eq. 5, r_1 is expressed as

$$r_1 = \frac{(k_p - k_f) r_2^2}{(k_p - k_f) r_2 + R_b k_p k_f}. \quad (6)$$

Considering the particle in Fig. 1(b), the effective particle volume fraction can be expressed as

$$\phi_{eff} = \frac{r_1^3}{r_2^3}\phi, \quad (7)$$

where ϕ is the particle volume fraction. Interfacial thermal resistance reduces the equivalent particle radius and the effective particle volume fraction.

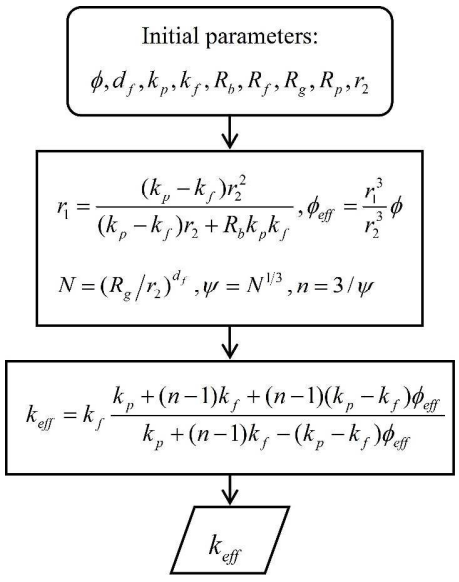
Hamilton[17] developed a model to predict the effective thermal conductivity of suspensions based on EMT. The influence of irregular shapes of particles was

158 considered and the expression of the *k* of nanofluids was given:

159 $$k_{eff} = k_f \frac{k_p + (n-1)k_f + (n-1)(k_p - k_f)\phi_{eff}}{k_p + (n-1)k_f - (k_p - k_f)\phi_{eff}}, \quad (8)$$

160 where k_{eff} is the effective thermal conductivity of nanofluids, *n* is the shape factor
161 given by $n = 3/\psi$ with ψ denoting the sphericity of particles. ψ is defined as the
162 ratio of the surface area of the sphere with the same volume as a particle to the
163 external surface area of a particle. In present model, the effective thermal conductivity
164 of nanofluids can be determined by Eq. 8.

165 Due to Brownian move and the interaction among particles, particles are
166 randomly packed together and form aggregates of fractal structure.[31-33] The number of
167 particles in an aggregate, *N,* is given by $N = (R_g/r)^{d_f}$, where R_g is the aggregate
168 radius of gyration, and d_f is the aggregate fractal dimension. As previously denoted,
169 an aggregate act as an independent unit of particle and its surface area is the same as
170 the surface area of primary particles in the aggregate. So assuming that the primary
171 nanoparticles are spheres with uniform size, we can get $\psi = N^{1/3}$ according to the
172 definition of ψ. The block diagram for the guidance of the model usage is shown in
173 Fig. 2.



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175 Fig. 2. Block diagram of the step of model

3. Model verification

The comparisons between the present model and experimental results in the literature are used to validate the present model. The sizes of particles and aggregates are rarely mentioned in literature, and results vary when using different measurement methods for the same sample because of possible size polydispersity, formation of ordered fluid layers and clustering of particles.[7,34]

Since particle size is defined by the area of the solid/liquid interface that acts as a new phase affecting properties of nanofluids, the specific surface area of the particles measured by Brunauer- Emmett- Teller (BET) can be used to determine average particle size. The average hydrodynamic diameter of species involved in Brownian motion can be estimated by dynamic light scattering (DLS). Since one aggregate involves in Brownian motion as a whole, the average aggregate size can be determined by DLS. In these considerations, the reported particle sizes using BET and DLS are used to determine particle size and aggregate size, and the experimentally measured k of nanofluids are used to validate the present model.

The interfacial thermal resistance at the solid/liquid interface has been discussed by researchers. Wilson et al.[35] experimentally estimated $R_b \approx 0.77\times10^{-8}\ Km^2/W$ for pt/water interface and got $R_b \approx 1.61\times10^{-8}\ Km^2/W$ for particle-water interface based on diffuse-mismatch model (DMM). Xue et al.[36] figured that interfacial thermal resistance was strongly dependent on the type of bonding between the solid and the liquid, and nanofluids characterized by weak atomic bonding at the solid- liquid interfaces will exhibit high thermal resistance. According to DMM, the velocity of sound and the heat capacity of the base fluid have influence on the interfacial thermal resistance. Under the assumption that the velocity of sound in ethylene glycol and water were approximately the same, Prasher et al.[21] assumed $R_b \approx 1.21\times10^{-8}\ Km^2/W$ for ethylene glycol based nanofluids. Since the accurate value of R_b for different particle- liquid interface is unavailable, we assume $R_b = 1\times10^{-8}\ Km^2/W$ for particle- water interface, and $R_b = 1.5\times10^{-8}\ Km^2/W$ for

particle- organic solvent interface based on DMM as a first approximation.

The coagulation of primary particles into aggregates, known as "cluster-cluster" aggregation, can be characterized by fractal dimension d_f. Two kinetic regimes, diffusion-limited aggregation (DLA) and reaction-limited aggregation (RLA), have been identified for cluster-cluster aggregation.[31] In DLA, particles collide and combine instantaneously, producing a highly porous, convoluted aggregate, which is similar to the process of aggregation in nanofluids. The value of d_f is around 1.8.[31-33] In RLA, there is a significant repulsive barrier to aggregation and the sticking probability on aggregate-aggregate interaction is less than unity. The value of d_f is around 2.1-2.2.[31] So it is reasonable to assume $d_f = 1.8$ in this paper.

The thermal conductivity of four sets of test nanofluids were measured by over 30 organizations worldwide to resolve the inconsistencies of the reported thermal conductivity of nanofluids.[11] Since the data from most organizations are lied within a narrow band, these data are used to validate the present model. The characteristics of the four sets of samples are listed in Table 1. In the present model, particles are assumed to be sphere, so the samples of alumina nanorods are not used for comparison.

Table 1. Characteristics of the samples for validation

	Particle type	Volume fraction (%)[a]	Particle size (nm)[b]	Aggregate size (nm)[c]	Type Base fluid
1	Alumina	1	10	81.5	PAO+surfactant
2	Alumina	3	10	105.5	PAO+surfactant
3	Gold	0.001	15	15	Water+stabilizer
4	Silica	31.1	22	22	Deionized water
5	Mn-Zn	0.17	7.4	11	Water+stabilizer

[a]The volume fraction of particles are reported by the providers.

[b]The particle sizes are the nominal particle sizes. As gold particles and Silica particles are well dispersed without aggregation, the particle sizes are the same as aggregate

sizes.

[c]The sizes of aggregates are the average sizes of dispersed phase, measured by DLS, or the particle size for the nanofluids without aggregation.

Comparisons of the k enhancement (k_{eff}/k_f) between experimental results and predictions of the present model are depicted in Fig. 3. The k enhancement as a function of fraction dimension is plotted to evaluate the robustness of the model since the value of fraction dimension (d_f) is picked by hand after referring to some references.

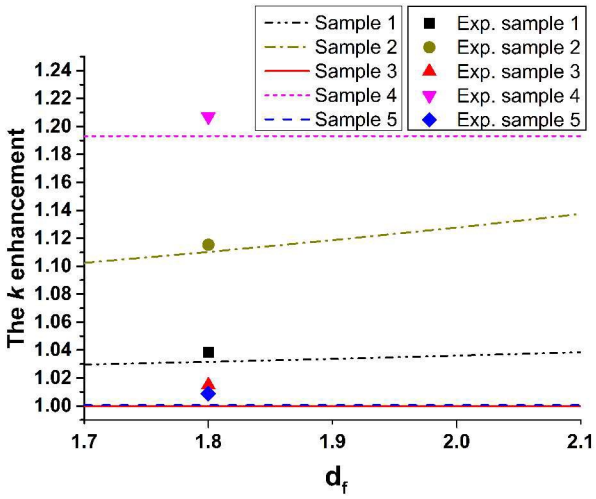

Fig. 3. The k enhancement as a function of fractal dimension. The k enhancement of experimental results from Ref. 11 are also presented.

It shows that the experimental data is consistent with the present model if the fraction dimension is set as 1.8. The influence of the magnitude of the fraction dimension on the k enhancement are minor enough to prove that the agreement is truely convincing. Besides, the volume fraction of gold particles is too low to estimate the k enhancement using present model.

4. Results and discussions

4.1 Effect of particle size on the k enhancement of nanofluids

The predictions of the k enhancement with different particle sizes using present model, three-level homogenization model,[12] and the renovated Maxwell model that considers nanolayer[19] are shown in Fig. 4. The experimental data measured by

Timofeeva et al.[34] are also listed for comparison. They measured the k of water-based α-SiC nanofluids for different particle sizes. The average particle diameters are determined by BET and the average aggregate sizes are measured by DLS. The parameters in the three-level homogenization model are the same as the proposed values in ref. 12. The particle sizes and aggregate sizes used in Fig. 4 are the same as the experimental measurements.

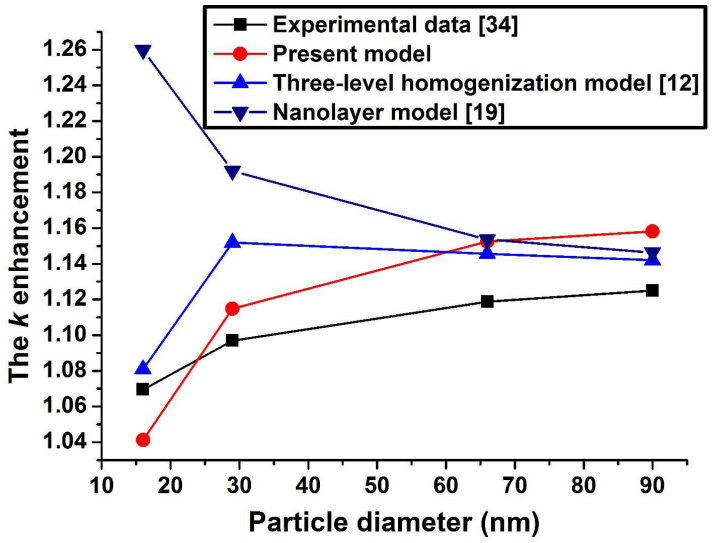

Fig. 4. Dependence of the k enhancement on the average particle diameter in 4.1 vol% water based SiC nanofluids. The experimental data in ref. 34 and predictions obtained by the present model, the three-level homogenization model, and the renovated Maxwell model that considers nanolayer are presented.

Fig. 4. indicates that both the present model and the experimental results show the increase of k enhancement with the rise of average particle sizes. The reason why particle size has positive effect on k enhancement is that as the average particle size increases, the total surface area of the solid/liquid interface decreases geometrically, thus weakening the effect of interfacial thermal resistance(according to Eq. 3). The predictions of this model (k_{cal}) fit well with the experimental results ($k_{\exp}$) with the deviations ($(k_{cal}-k_{\exp})/k_{\exp}\times 100\%$) within $\pm 3\%$.

For the three-level homogenization model, although the increase of particle size can weaken the effect of interfacial thermal resistance and enhance the k of nanofluids, the aggregate size has greater impact on the k of nanofluids. It also shows that

compared with the three-level homogenization model, the present model can better predict the k of nanofluids. According to the renovated Maxwell model, the nanolayer has significant impact on small particles. However, with the increase of particle sizes, the *k* enhancement decreases and the renovated Maxwell equation reduces to the original Maxwell equation, because the impact of nanolayer becomes smaller.

4.2 Effect of aggregate size on the *k* enhancement

Studying the k enhancement with absolute value of aggregate size is meaningless because particle sizes vary. In this consideration, the authors investigate the effect of relative aggregate size, defining as the ratio of aggregate diameter to particle diameter (D_g/d), on the *k* enhancement. The relative aggregate size reflects the amount of particles of an aggregate and its aggregate shape factor.

The *k* enhancement as the function of D_g/d for water based alumina nanofluids is shown in Fig. 5. The particle volume fraction is set as 2% and the particle diameters are kept constant for each line. The *k* of 2 vol% water based alumina nanofluids reported by Beck et al.[7] are also presented to make a comparison. The particle diameters were from BET measurement and the aggregate sizes were from DLS.

Fig. 5. shows that the *k* enhancement rises rapidly as D_g/d increases, and the growth rate decreases as aggregate size becomes larger. If the aggregate size is fixed, *k* enhancement increases as particle size becomes larger. The estimates of the present model show a good agreement with the experimental data, with deviations within 4.67%.

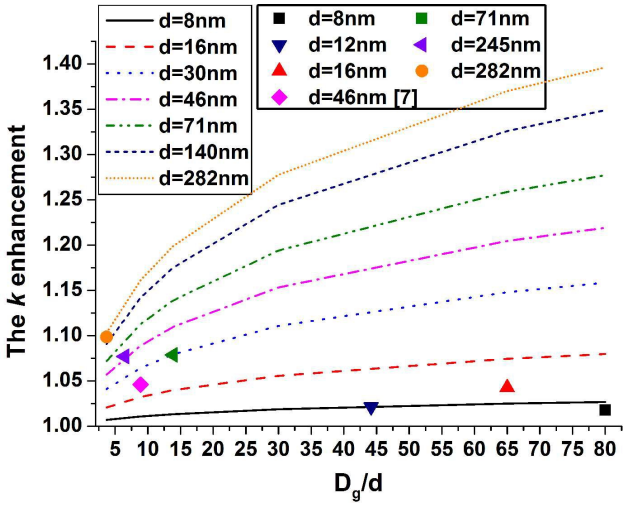

Fig. 5 Influences of relative aggregate size (D_g/d) on the *k* enhancement of 2 vol% water based alumina nanofluids. Experimental data from ref. 7 are also presented.

The reason why the aggregate size has positive effect on the k enhancement is that the number of particles in an aggregate increases as aggregate size becomes larger, which significantly increases the shape factor of aggregate and enhances the *k* of nanofluids according to Eq. 8.

4.3 Effect of particle volume fraction on the *k* enhancement

The predictions of *k* enhancement as a function of particle volume fraction for a series of alumina nanofluids are depicted in Fig. 6. The sizes of particles and aggregates used in the present model are the same as those measured by Timofeeva et al.[10] In their experiment, the *k* of water and ethylene glycol based alumina nanofluids were measured and aggregate size distribution were determined by DLS. Although the intensity-weighted distributions of aggregate sizes have two peaks, the volume fractions of bigger aggregates are much less than that of the smaller ones. Therefore, the sizes of smaller aggregate are considered as the aggregate sizes, which are 88, 120, and 40 nm, and the nominal particle diameters are 11, 20, and 40 nm, respectively.

It can be seen from Fig.6 that the *k* enhancement of nanofluids increases linearly with the rise of particle volume fraction, which is consistent with the experimental data. For ethylene glycol based nanofluids (Fig. 6(a)), when volume fraction is fixed, the highest enhancement is observed in nanofluids with particle size of 20 nm, the second highest for 11 nm particles and the lowest for 40 nm particles. The

experimental data are within the range of predictions and show that all particle sizes present the same trend and this factor less influence on the *k* enhancement. The reason why nanofluids with the largest particle size (d=40 nm) shows the lowest k enhancement is that particles (d=40 nm) are well dispersed in the base fluid without aggregation and cannot form rapid thermal conduction path among themselves. By contrast, for water based nanofluids (Fig. 6(b)), experimental data show that nanofluids with particle size of 40 nm shows the highest k enhancement, which is not consistent with the present model. The maximum deviations between the experimental data and present model are within $\pm 8.5\%$ and the reason might be the variation of particle size distribution and the uncertainty of interfacial thermal resistance.

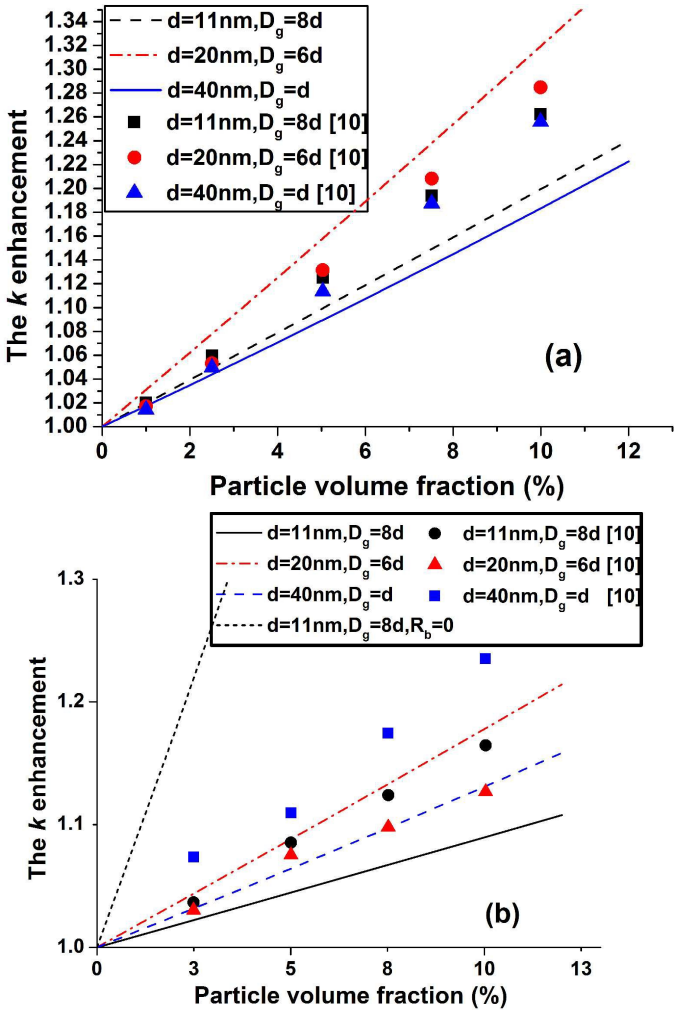

Fig. 6. The *k* enhancement as a function of particle volume fraction. The lines are the

present model predictions of *k* enhancement for nanofluids with 11, 20, and 40 nm nominal size alumina particles in (a) ethylene glycol and (b) water. The aggregate sizes are four, six and one time（s） of the particle size respectively. The dots are the experimental data from ref. 10.

The uncertainty of interfacial thermal resistance has a great influence on the k enhancement, as shown in Fig. 6(b). When interfacial thermal resistance is neglected, the *k* enhancement is much larger than the prediction with $R_b = 1\times10^{-8}\ Km^2/W$ for water based nanofluids of fixed volume fraction. Although it is hard to obtain the exact value of interfacial thermal resistance, the comparisons between the present model and the experimental data[7,10,11,34] show that the inferred values of interfacial thermal resistance are in a reasonable range.

4.4 Further discussion **about the divergence of the present model and some experimental results**

The relationship between the *k* of nanofluids and the particle volume fraction differs in different experimental results. Although the present model analyzes the factors of particle size, aggregate size and interfacial thermal resistance on the divergence of *k* of nanofluids, some phenomenon still need to be discussed further.

First, the anomalous *k* enhancement at a low particle volume fraction, less than 0.1 vol%, can not be predicted by present model. The measured *k* enhancement (k_{eff}/k_f) of 0.001vol% water based Gold nanofluids is 1.015, as shown in Fig. 3, while the present model predicts almost no *k* enhancement. Pang[50] found non-linear enhancement at low concentration and the *k* of nanofluids was much larger than the prediction when using EMT. The *k* enhancement at low concentration may dominantly contribute to nano-convection and can be predicted by the model built by Pang et al.[37] and Prasher et al.[21]

Second, some nanofluids with a small amount of aggregation and particles whose diameter is lower than 10 nm show larger *k* enhancement than the predictions of the present model. Eastman et al.[38] found that *k* enhancement of ethylene glycol based Cu nanofluids was up to 1.4 when particle volume fraction was 0.3 %. In this study, a

one-step production procedure was used and the particle diameter was less than 10 nm with very little aggregate. Philip et al.[4] reported that the *k* enhancement of Fe_3O_4 nanofluids was larger than the predictions of the present model. The particle diameter was 8 nm and there was little aggregate. The reason why nanofluids without aggregate but with well dispersed particles whose diameter was less than 10 nm show larger *k* enhancement might result from the forming of the solid-like nanolayer at the solid/liquid interface. The solid-like liquid layer of thickness *h* around particles is more ordered than that of the base fluid and the *k* of layer is larger than that of base fluids. The effective particle volume fraction would be calculated as $\phi_{eff} = \phi(1+h/r)^3$, much larger than the primary particle volume fraction.

Third, the present model could not predict the *k* of nanofluids if the shape of particles is not sphere. The *k* of alumina nanorods nanofluids is larger than the *k* of alumina particle at the same particle volume fraction.[11] Philip et al.[4] concluded that all thermal conductivity studies in Carbon nanotube (CNT) nanofluids showed *k* enhancement is inconsistent with the predictions of EMT. The reason might be that the shape factor of nanorods and CNT are larger than that of the sphere particles. Lamas et al.[39] concluded several correlations to predict the *k* of CNT nanofluids and presented critical analysis on these models. However, it is still necessary to conduct further studies about the influence of the particle shape on the *k* enhancement.

5. Conclusions

In the present work, a model for predicting the thermal conductivity of nanofluids is built considering particle size, aggregate size and interfacial thermal resistance. In the present model, the existence of interfacial thermal resistance is considered, and aggregate acts as an independent unit of particle. The shape factor of the aggregate is determined based on the number of particles in the aggregate. The *k* of nanofluids is obtained by using EMT-based Hamilton model. Based on analysis on the factors that influence the *k* enhancement, it is concluded that particle size and aggregate size have positive effect on the *k* enhancement. The increase of particle size can weaken the effect of interfacial thermal resistance and enhance the *k* of nanofluids because the

382 total surface area of the solid/liquid interface decreases as particle size increases. As
383 aggregate size becomes larger, the shape factor of aggregate significantly increases
384 and the *k* of nanofluids will also be enhanced. The *k* of nanofluids increases linearly
385 with particle volume fraction, and the increase rates vary according to particle size
386 and aggregate size. This can explain the divergence of the experimental results. Since
387 present model fits different experimental data well, the inferred values of interfacial
388 thermal resistance are in a reasonable range. Considering the case of nanofluids with
389 particle volume fraction lower than 0.1% and particle size smaller than 10 nm without
390 aggregation, the factors of nano-convection and nanolayer need to be taken into
391 account.

References

1. S. U. S. Choi, Z. G. Zhang, W. Yu, F. E. Lockwood, E. A. Grulke, *Appl. Phys. Lett.*, 2001, 79, 2252-2254.
2. S. S. J. Aravinda and S. Ramaprabhu, *RSC Adv.*, 2013, 3, 4199-4206.
3. Y. Xuan, H. Duan and Q. Li, *RSC Adv.*, 2014, 4, 16206-16213.
4. J. Philip and P. D. Shima, *Adv. Colloid Interface Sci*., 2012, 183-184, 30-45.
5. H. E. Patel, T. Sundararajan and S. K. Das, *J. Nanopart. Res.*, 2010, 12, 1015-1031.
6. W. Z. Cui, Z. J. Shen, J. G Yang, S. H. Wu and M. L. Bai, *RSC Adv.*, 2014, 4, 55580-55589.
7. M. P. Beck , Y. H. Yuan, P. Warrier and A. S. Teja, *J. Nanopart. Res.*, 2009, 11, 1129-1136.
8. P. M. Sudeep, J. Taha-Tijerina, P. M. Ajayan, T. N. Narayananc and M. R. Anantharaman, *RSC Adv.*, 2014, 4, 24887-24892.
9. S. K. Das, N. Putra, P. Thiesen and W. Roetzel, *J. Heat Trans*., 2003, 125, 567-574.
10. E. V. Timofeeva, A. N. Gavrilov, et al, *Phys. Rev. E.*, 2007, 76: 061203.
11. J. Buongiorno, et al, J. *Appl. Phys.*, 2009, 106, 094312.
12. R. Prasher, W. Evans, P. Meakin, J. Fish, P. Phelan and P. Keblinski, *Appl. Phys. Lett.*, 2006, 89, 143119.
13. P. Keblinski, S. R. Phillpot, S. U. S. Choi and J. A. Eastman, *Int. J. Heat Mass Trans.*, 2002, 45, 855-863.
14. D. Lee, J. W. Kim and B. G. Kim, *J. Phys. Chem. B.*, 2006, 110, 4323-4328.
15. H. Younes, G. Christensen, X. Luan, H. Hong and P. Smith, *J. Appl. Phys.*, 2012, 111, 064308.
16. J. C. Maxwell, Clarendon Press, Oxford, UK, 1873.
17. R. L. Hamilton and O. K. Crosser, *Ind. Eng. Chem. Fund.*, 1962, 1, 187-191.
18. C. J. Yu, A. G. Richter, A. Datta, M. K. Durbin and P. Dutta, *Phys. B*., 2000, 283, 27-31.
19. W. Yu and S. U. S. Choi, *J. Nanopart. Res.*, 2003, 5, 167-171.
20. R. Prasher, P. Bhattacharya and P. E. Phelan, *Phys. Rev. Lett.*, 2005, 94, 025901.
21. R. Prasher, P. Bhattacharya and P. E. Phelan, *J. Heat Transf.*, 2006, 128, 588-595.

22. S. Mehta, K. P. Chauhan and S. Kanagaraj, *J. Nanopart. Res.*, 2011, 13, 2791-2798.

23. J. W. Gao, R. T. Zheng, H. Ohtani, D. S. Zhu, G. Chen, *Nano lett.*, 2009, 9, 4128-4132.

24. S. P. Jang and S. U. S. Choi, Appl. *Phys. Lett.*, 2004, 84, 4316-4318.

25. R. Prasher, P. E. Phelan and P. Bhattacharya, *Nano Lett.*, 2006, 6, 1529-1534.

26. C. W. Nan, R. Birringer, D. R. Clarke and H. Gleiter, *J. Appl. Phys.*, 1997, 81, 6692.

27. W. Evans, R. Prasher, J Fish, et al. *Int. J. Heat Mass Transf.*, 2008, 51, 1431-1438.

28. G. Okeke, S. Witharana, S. J. Antony, Y. Ding, *J. Nanopart. Res.*, 2011, 13, 6365-6375.

29. D. Q. Zhou and H. Y. Wu, *Appl. Phys. Lett*., 2014, 105, 083117.

30. D. P. H. Hasselman and L. F. Johnson, *J. Comp. Materials*, 1987, 21, 508-515.

31. T. D. Waite, J. K. Cleaver, J. K. Beattiez, J Colloid Interface Sci., 2001, 241, 333-339.

32. B. X. Wang, L. P. Zhou, X. F. Peng, Int J Heat Mass Transf., 2003, 46, 2665-2672.

33. P. Meakin, Reviews Geophysics, 1991, 29, 317-354.

34. E. V. Timofeeva1, D. S. Smith, et al., *Nanotechnology*, 2010, 21, 215703.

35. O. M. Wilson, X. Hu, D. G. Cahill, P. V. Braun, *Phys. Review B.*, 2002, 66, 224301.

36. L. Xue, P. Keblinski, S. R. Phillpot, S. U. S. Choi, and J. A. Eastman, J.Chem. Phys., 2003, 118, 337-339.

37. C. Pang C, J. Y. Jung, Y. T. Kang, *Int. J. Heat Mass Transf.*, 2014, 72, 392-399.

38. J. A. Eastman, S. U. S. Choi, S. Li, W. Yu, and L. J. Thompson, Appl. Phys. Lett. 2001, 78, 718-720.

39. B. Lamas, B. Abreu, A. Fonseca, N. Martins and M. Oliveira, Int. J. Ther. Sci., 2014, 78, 65-76.

Interfacial thermal resistance is modeled to have relationship with the equivalent particle size in terms of keeping thermal resistance constant.