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# A multi-level homogenization model for thermal conductivity of nanofluids based on particle size distribution (PSD) analysis



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#### ABSTRACT

Nanofluids are engineered suspensions of fine nanoparticles in basefluids. Owing to their enhanced thermal conductivity, nanofluids find applications in many heat transfer and energy conversion systems. Enhanced thermal conductivity of nanofluids is attributed to several mechanisms such as Brownian motion, interfacial layer formation and particle clustering, etc. Many theoretical models have been proposed based on these mechanisms to predict the thermal conductivity of nanofluids. But, still there is an uncertainty in predicting the thermal conductivity of nanofluids. In this work, a simple model to predict the thermal conductivity of nanofluids based on particle size distribution and multi-level homogenization has been proposed. This model considers the effects of Brownian motion, interfacial layer formation and particle clustering. Particle clusters are characterized based on particle size distribution (PSD) analysis and their thermal conductivity is calculated exclusively. The complex nanofluid system is subdivided into smaller systems and a level by level homogenization is carried out to determine the effective thermal conductivity of nanofluids. Present model predictions are compared with experimental results from literature and are found to match well. Contributions of aggregation, Brownian motion and interfacial layer formation are individually exhibited. This model aids to develop a better understanding of the thermal transport in nanofluids and hence, is expected to contribute to several industrial applications.

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# 1. Introduction

Recent trend of miniaturization and development of high power output devices have made efficient heat transfer as the prime requirement of many industries. In recent years, using nanofluids to increase the heat transfer is gaining much more attention among engineers and researchers [1]. Nanofluids are stable suspensions of fine nanosized metallic/non-metallic particles in common cooling liquids. They exhibit higher thermal conductivity than their basefluids and are considered to be the coolants of future. A chance to increase the heat transfer by employing nanofluids have opened the way for a spectrum of promising applications like miniature electronic devices, high power electric devices like transformers and enhanced heat transfer in many other energy conversion systems [1–4].

Since the initiating work of Choi [5] in exhibiting the enhanced thermal conductivity of nanofluids, many experimental works have been carried out to measure the thermal conductivity enhancement in nanofluids. Review of these works show that, classical models like MG model [6] are unable to explain the thermal conductivity enhancement seen in nanofluids. These classical models which are based on Effective

Medium Theory (EMT) substantially underestimate the thermal conductivity of nanofluids [7]. The mechanisms behind the unexplainable increase in thermal conductivity of nanofluids have become a hotly debated topic and various explanations have been proposed to explain the thermal conductivity enhancement in nanofluids [8]. A huge volume of works on theoretical modeling of thermal conductivity of nanofluids are available in literature [4,9–18]. These works on modeling of thermal conductivity of nanofluids suggest that the enhancement can be attributed to nano-scale convection caused by Brownian motion of particles, ordered layering of liquid molecules at the solid-liquid interface and particle clustering. [11–15,18,19].

Effects of Brownian motion of nanoparticles on thermal conductivity of nanofluids have been extensively studied by many researchers. Keblinski et al. [8] showed that, particle-particle collision caused by Brownian motion is too slow (in comparison with thermal diffusion) to cause a significant enhancement in thermal transport of nanofluids. However, Brownian motion can indirectly influence the thermal conductivity of nanofluids by thermal interactions of randomly moving particles with basefluid molecules which results in convection-like effects at nano-scale [10]. An order of magnitude analysis by Prasher et al. [20], proved that the nano-scale convection caused by Brownian motion of nanoparticles is a possible mechanism to explain the thermal conductivity enhancements seen in nanofluids. A recent work by Song et al. [21] showed that

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the effective property model which considers Brownian motion is much closer with experimental values. Xiao et al. [22] in their model for effective thermal conductivity, considered the effect of Brownian motion by means of fractal geometry. Both the static and dynamic mechanisms were considered and the effects of particle size and volume fraction were successfully explained. Thus, it is evident that Brownian motion is a vital mechanism to consider while modeling thermal conductivity of nanofluids. In solid-liquid suspensions, liquid molecules close to a solid particle arrange themselves in an ordered manner around the solid particle. This ordered layer of liquid molecules behaves like a solid and acts as a thermal bridge between the solid particles and liquid [13]. Even though this layer is only of nanometer thickness, it is expected to play a vital role in thermal transport of nanofluids. As the Specific Surface Area (SSA) of nanoparticles is very large, the effects of interfacial layer are very significant. Yu and Choi [16,17] clearly explained that orderly arrangement of liquid molecules at particle-liquid interface and the corresponding increase in effective volume fraction is a possible mechanism that causes high thermal conductivity enhancements in nanofluids. For instance, an interfacial layer of thickness 2.5 nm is sufficient enough to double the effective volume fraction of the particle with diameter 10 nm [8]. Hence, it is clear that interfacial layer formation at the solid-liquid interface is an important mechanism that influences the thermal transport in nanofluids. In addition to above two mechanisms, nanoparticles suspended in basefluids form clusters that create a low thermal resistance path which can enhance the thermal conductivity [15]. From Kwak and Kim's [23] experiments, it is understood that large enhancements in thermal conductivity are accompanied by significant increase in viscosity, which is a clear indication of particle clustering. Putnam et al. [24], showed that nanofluids with excellent distribution characteristics do not show any anomalous increase in thermal conductivity. Hence, it is understood that particle clustering is also a vital criteria to be taken into account while modeling thermal conductivity of

Prasher et al. [25] and many other researchers [12,15,26] who have considered the effects of particle clustering on thermal

conductivity of nanofluids, characterized the particle clusters based on fractal theory. The cluster structure or morphology is defined by means of average radius of gyration of the clusters and the fractal and chemical dimensions of the aggregates. The clusters are assumed to consist of a linear chain known as the backbone and side chains called as dead ends. The backbone plays a significant role in thermal transport by providing a low resistance path for heat transfer. Xiao et al. [22] employed a fractal method to model the thermal conductivity of nanofluids. Number of particles present in the clusters was calculated using probabilistic theory and fractal dimension. Fractal dimension is a prime factor in fractal theory to characterize the clusters and depends on numerous factors. Hence, a precise calculation using theoretical approach is very difficult [27]. Also, models based on fractal theory consider that the clusters in nanofluids are of uniform size [25]. In reality, cluster size in nanofluids vary in a very wide range [18]. To overcome this limitation, Zhou and Wu [28] calculated the cluster size and volume fraction of clusters based on particle size distribution (PSD) analysis. PSD of nanofluids provide sufficient information to characterize the clusters and is very easy to obtain by Dynamic Light Scattering (DLS) experiments [26]. More recently, Song et al. [29] also showed that hydrodynamic and optical properties of nanofluids predicted using PSD exhibited better agreement with experimental results at all particle volume fractions. It is to be noted that, Zhou and Wu's model [28] consider that the particles aggregate very close to each other and they have the same thermal conductivity as that of the primary particle. Actually, nanoparticle clusters have thermal conductivity less than that of the primary particle due to contact resistance and the basefluid trapped in the interparticle spaces [30].

In this work, a modified model for thermal conductivity of nanofluids based on multi-level homogenization approach of Prasher et al. [25] and PSD analysis of Zhou and Wu [28] is proposed. The thermal conductivity of clusters is calculated exclusively. Effects of nano-scale convection due to Brownian motion and interfacial layer formation are also taken into account. The proposed modified model for thermal conductivity of nanofluids is expected to help in developing better understanding of thermal transport in nanofluids

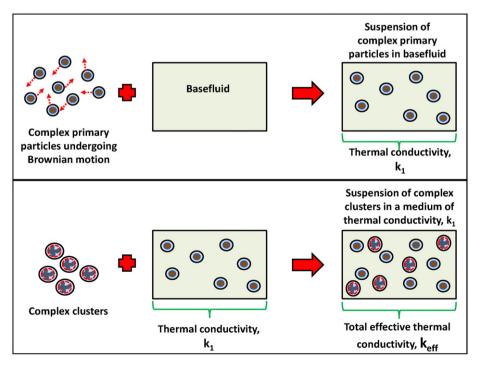


Fig. 1. Schematic representation of the model.

and lead to their applications in many energy efficient heat transfer systems.

# 2. Physical model description

Primary goal of this work is to propose a mathematical model to describe the thermal conductivity enhancement of nanofluids and its relationship with the Brownian motion, interfacial layer formation and particle clustering. A schematic representation of the proposed model is presented in Fig. 1. Nanofluid is a complex system that consists of basefluid, primary nanoparticles and clusters of primary particles. These primary particles and clusters are surrounded by an ordered layer of basefluid molecules at the solid-liquid interface. Primary particles and clusters along with the interfacial layer are termed as complex primary particles and complex clusters, respectively. This complex system is broken into subsystems and then a level by level homogenization is carried out to determine the effective thermal conductivity of the entire nanofluid system. It is to be noted that, nanofluids may contain spherical, cylindrical, planar and spheroidal inclusions. But, this model is built considering spherical inclusions. Initially, only the thermal conductivity of suspension of complex primary particles in basefluid is alone calculated. In this step, these complex primary particles are also considered to undergo Brownian motion. Thus, the thermal conductivity of complex primary particles suspended in basefluids along with the contribution of Brownian motion is calculated as  $k_1$ . Then, the thermal conductivity of clusters along with interfacial layer around them (complex clusters) is calculated exclusively. Finally, it is considered that these complex clusters are suspended in a medium of thermal conductivity,  $k_1$ and the total thermal conductivity of the whole nanofluid system is determined.

# 3. Development of new thermal conductivity model

Detailed derivation of the mathematical model is presented in the following sections.

# 3.1. Characterization of clusters using particle size distribution (PSD) analysis

PSD analysis of nanofluids, clearly indicate that both primary particles and clusters exist together at any instant of time [26]. Hence, the nanofluid is hypothesized as a complex system consisting of both primary nanoparticles and clusters suspended in basefluids. Prior to the homogenization, it is necessary to determine the volume fractions of primary particles  $(\phi_{nm})$  and particle clusters  $(\phi_c)$  using the approach of Zhou and Wu [28]. PSD in nanofluids vary in a wide range [18] and hence, the primary particles and clusters must be clearly defined before calculating  $(\phi_{vm})$  and  $(\phi_c)$ . As a cluster will have at least two primary particles of diameter  $(d_{pm})$  each, anything equal to or larger than the size of two primary particles  $(2d_{pm})$  are considered as clusters and the rest are taken as primary particles. Nanoclusters are assumed to be embedded in an imaginary sphere with diameter (d). Each cluster will be of different size and thus,  $(d_c)$  is defined as the average diameter of clusters. From PSD data, average diameter of clusters can be calculated using the number percentage of clusters (*P*) as follows:

$$d_{c} = \frac{\sum_{d \ge 2d_{pm}} (P \times d)}{\sum_{d \ge 2d_{pm}} P}.$$
 (1)

The volume fraction of clusters present in the nanofluid is given by  $\phi_c = \phi_{cs}\phi_{ins}$ . Where,  $\phi_{cs}$  represent the volume fraction of corresponding cluster spheres and  $\phi_{ins}$  is the volume fraction of primary

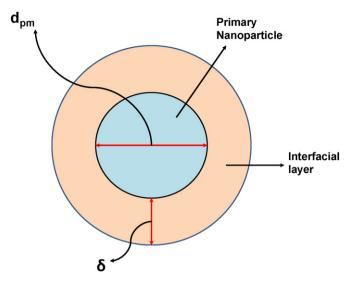


Fig. 2. A complex primary particle.

particles present in cluster spheres.  $\phi_{cs}$  and  $\phi_{ins}$  are estimated estimated as [28]

$$\phi_{cs} = \frac{\sum_{d \ge 2d_{pm}} \left(P \times d^3\right)}{\sum_{d \ge 0} \left(P \times d^3\right)} \phi_p \text{ and } \phi_{ins} = \frac{d_{pm}^3 n}{d_c^3}.$$
 (2)

In Eq. (2),  $\phi_p$  is the total initial volume fraction of nanoparticles including the primary particles and clusters. For the convenience of calculation, a parameter (f) is defined as

$$f = \frac{\sum\limits_{d \ge 2d_{pm}} \left(P \times d^3\right)}{\sum\limits_{d \ge 0} \left(P \times d^3\right)}.$$
 (3)

This parameter f varies from 0 to 1 and indicates the volume fraction of clusters present in the nanofluid system. Again, in Eq. (2) n is the number of nanoparticles in a cluster sphere with equivalent diameter  $(d_c)$  and is given by

$$n = \frac{d_c^3}{d_{pm}^3} s\omega. (4)$$

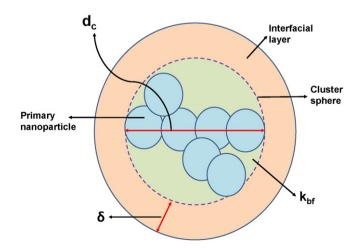


Fig. 3. Graphical representation of a complex cluster.

 Table 1

 Nanoparticles, basefluids, initial particle volume fraction and primary particle diameter used in the nanofluid samples taken for comparison [35].

S. no.	Sample	Details	Initial particle volume fraction, $\phi_p$	Particle diameter, $d_{pm}$
1	S1	Fe <sub>3</sub> O <sub>4</sub> -kerosene	0.0171	8 nm
2	S2	Ag-hexadecane	0.0116	7 nm
3	S3	Fe <sub>3</sub> O <sub>4</sub> -hexadecane	0.0164	8 nm
4	S4	Fe <sub>3</sub> O <sub>4</sub> -water	0.0102	8 nm
5	S5	CuO-Eg	0.0018	10 nm
6	S6	CuO-Eg	0.0131	10 nm

According to Kepler conjecture [31],  $s=\pi/\sqrt{18}$  is the maximum packing density of cluster sphere and  $\omega$  is a relaxation factor which indicates the particle packing condition. The average packing density is given by the product of s and  $\omega$ . Review of related literature show that, the random packing density in cluster spheres is approximately 63% [32]. Thus, the relaxation factor  $\omega$  is taken as 0.85, which gives the average packing density as 62.9%, which is very close the random packing density. Finally, the volume fraction of primary particles  $(\phi_{pm})$  is given by  $\phi_{pm} = \phi_p - \phi_c$ .

# 3.2. First level of homogenization

It is experimentally proved that the liquid molecules arrange themselves into an ordered layer at the solid-liquid interface [33]. The primary particles along with the interfacial layer are considered as complex primary particles. A pictorial representation of a complex primary particle is shown in Fig. 2.

This interfacial layer acts as a thermal bridge between the particle and liquid with its thermal conductivity  $(k_l)$  varying between the thermal conductivity of the primary particle  $(k_{pm})$  and that of the basefluid  $(k_{bf})$ . Hashimoto et al. [34] proposed a relation for the thickness of interfacial layer  $(\delta)$  based on electron density profile at the interface as follows:

$$\delta = \sqrt{2\pi}\sigma. \tag{5}$$

 $\sigma$  is a parameter which indicates the diffusiveness of interfacial boundary and its value ranges from 0.2 to 0.8 nm. If  $\sigma$  = 0.4 nm, the interfacial layer thickness is 1 nm. Experimental results of Yu et al. [33] and molecular dynamic simulations by Xue et al. [14] show that, the interfacial layer will be in the order of few atomic distances, which is approximately 1 nm. Hence in the present model, the thickness of the interfacial layer is considered to be 1 nm.

In the first level of homogenization, the thermal conductivity of complex primary particles  $(k_{cpm})$  is calculated using the approach of Yu and Choi [16] as

$$k_{cpm} = \frac{\left[2(1-\gamma) + (1+\beta)^3 (1+2\gamma)\right] \gamma}{-(1-\gamma) + (1+\beta)^3 (1+2\gamma)} k_{pm}.$$
 (6)

Here,  $\gamma = k_l/k_{pm}$  is the ratio of the thermal conductivity of interfacial layer  $(k_l)$  to the thermal conductivity of primary particle  $(k_{pm})$ .  $\beta = 2\delta/$ 

 $d_{pm}$  is the thickness ratio of the interfacial layer to particle radius. Thermal conductivity of the interfacial layer  $(k_l)$  around a primary particle is given by Ren et al. [19] as

$$k_{l} = \frac{k_{bf}M^{2}}{(M-\beta)\ln(1+M) + \beta M}.$$
(7)

In Eq. (7),

$$M = \varepsilon_{pm}(1+\beta) - 1 \tag{8}$$

where,  $\varepsilon_{pm} = k_{pm}/k_{bf}$  is the ratio of thermal conductivity of the primary particle  $(k_{pm})$  to the thermal conductivity of the basefluid  $(k_{bf})$ . It is to be noted that, by considering the interfacial layer formation there will be a corresponding change in volume fraction. The volume fraction of complex primary particles is calculated as [16]

$$\phi_{cpm} = \phi_p (1+\beta)^3. \tag{9}$$

# 3.3. Second level of homogenization

In the second level of homogenization, complex primary particles are considered to be suspended in basefluid. These complex primary particles exhibit Brownian motion by virtue of their small size [8]. Random motion of these complex particles cause a nano-scale convection in the basefluid which enhances the thermal transport in the nanofluids [10]. A time-scale analysis by Prasher et al. [20] proved that, nano-scale convection caused by Brownian motion of particles is capable of producing an enhancement in thermal conductivity of nanofluids. Hence, the thermal conductivity of suspension of complex primary particles in basefluid by taking into account the nano-scale convection caused by Brownian motion is calculated using MG model [6] as

$$k_{1} = \frac{k_{cpm} + 2k_{f} + 2(k_{cpm} - k_{f})(1 + \beta)^{3}\phi_{cpm}}{k_{cpm} + 2k_{f} - (k_{cpm} - k_{f})(1 + \beta)^{3}\phi_{cpm}}k_{f}.$$
(10)

The effect of nano-scale convection caused by Brownian motion is included by using the approach of Prasher et al. [20].  $k_f$  used in Eq.

**Table 2**Thermophysical properties of nanoparticles and basefluids considered in this study.

S. no.	Materials	Thermal conductivity in [W m <sup>-1</sup> K <sup>-1</sup> ]	Density in [kg m <sup>-3</sup> ]	Dynamic viscosity in [kg m <sup>-1</sup> S <sup>-1</sup> ]
Nanoparticle	rs			
1	$Fe_3O_4$	7.2 [36]	5,170 [37]	=
2	Ag	429.0 [36]	10,500 [37]	-
3	CuO	13.5 [36]	6,315 [37]	-
Basefluids				
4	Water (de-ionized)	0.600 [36]	997.10 [37]	0.000890 [38]
5	Hexadecane	0.140 [36]	773.30 [37]	0.003044 [39]
6	Kerosene	0.150 [36]	799.00 [37]	0.001640 [38]
7	Ethylene glycol	0.254 [36]	1,113.00 [37]	0.016200 [38]

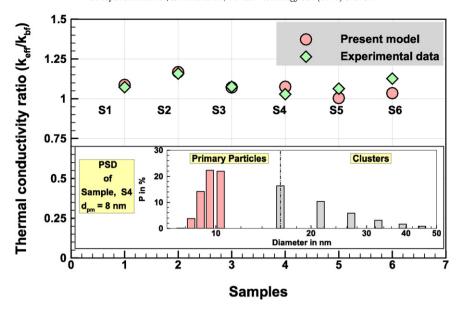


Fig. 4. Comparison of present model with experimental data from literature [35]. Inset image shows the particle size distribution of sample, S4.

(10) is the conductivity of basefluid including the contribution of nanoscale convection due to Brownian motion and is given as

$$k_f = k_{bf} \left( 1 + ARe^m Pr^{0.333} \phi_{cpm} \right) \tag{11}$$

where,  $A = 4 \times 10^4$  and m = 2.5 are empirical constants which are determined by considering the interaction of convection currents caused by Brownian motion of multiple particles [20].

$$Re = \frac{1}{\nu} \sqrt{\frac{18K_B T}{\pi \rho_{pm} (d_{pm} + 2\delta)}}.$$
 (12)

Re is the Reynolds number based on root-mean-square velocity of Brownian particle and Pr is the Prandtl number of the basefluid.  $\nu$  is the kinematic viscosity of the basefluid.  $K_B$  is the Boltzmann constant and T is the temperature.

# 3.4. Third level of homogenization

Nanofluid system consists of clusters which are aggregates of primary particles attached with each other and basefluid trapped in the interparticle spaces. Hence, Bruggeman model [9] which is more suitable at high volume fractions of highly conductive particles is employed to calculate the thermal conductivity of clusters  $(k_c)$  as follows:

$$\frac{(1-\phi_{ins})(k_{bf}-k_c)}{k_{bf}+2k_c} + \frac{\phi_{ins}(k_{pm}-k_c)}{k_{pm}+2k_c} = 0.$$
 (13)

Similar to the primary particles, clusters will also have an interfacial layer around them. These cluster spheres surrounded by interfacial layer are together referred as complex cluster spheres. A graphical representation of this complex cluster is shown in Fig. 3. In the third level of homogenization, thermal conductivity of the complex cluster spheres ( $k_{cc}$ ) using the same approach used for calculating the thermal conductivity of complex primary particles is given by the following equation.

$$k_{cc} = \frac{\left[2(1-\alpha) + (1+\xi)^{3}(1+2\alpha)\right]\alpha}{-(1-\alpha) + (1+\xi)^{3}(1+2\alpha)}k_{a}.$$
(14)

Here,  $\alpha = k_{lc}/k_c$  is the ratio of thermal conductivity of the interfacial layer around a cluster sphere  $(k_{lc})$  to the thermal conductivity of a

cluster  $(k_c)$ .  $\xi=2\delta/d_c$  is the thickness ratio of interfacial layer to cluster radius. Thermal conductivity of interfacial layer around a cluster  $(k_{lc})$  can be estimated as

$$k_{lc} = \frac{k_{bf}Q^2}{(Q - \xi) \ln(1 + Q) + \xi Q}.$$
 (15)

In the above equation

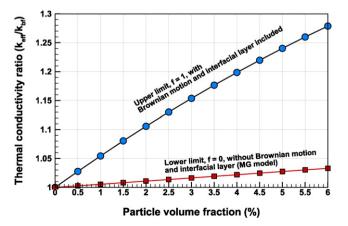
$$Q = \varepsilon_c(1+\xi) - 1. \tag{16}$$

where,  $\varepsilon_c = k_c/k_{bf}$  is the ratio of thermal conductivity of cluster to the thermal conductivity of basefluid.

# 3.5. Fourth level of homogenization

In the final level of homogenization, the complex cluster spheres are suspended in a medium of thermal conductivity ( $k_1$ ). Using MG model [6], the total effective thermal conductivity of the nanofluid system is calculated as

$$k_{eff} = \frac{k_{cc} + 2k_1 + 2\phi_{cc}(k_{cc} - k_1)}{k_{cc} + 2k_1 - \phi_{cc}(k_{cc} - k_1)} k_1$$
(17)



**Fig. 5.** Upper limit (f=1) and lower limit (f=0) calculated for Alumina-Water nanofluid,  $d_{pm}=10$  nm and  $d_c/d_{pm}=4$  at 293 K using present model.

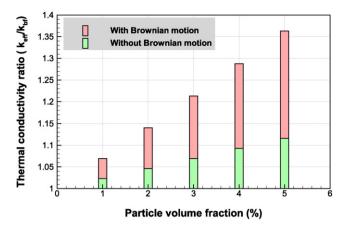


Fig. 6. Contribution of Brownian motion.

where,  $\phi_{cc}$  is the effective volume fraction of complex clusters which is given as  $\phi_{cc} = \phi(1+\xi)^3$ .

# 4. Results and discussion

In the present model, effective thermal conductivity of nanofluids  $(k_{eff})$  is determined using a multi-level homogenization approach considering the effects of Brownian motion, interfacial layer formation and particle clustering. The nanofluid is considered as a complex system which has both primary nanoparticles and particle clusters suspended in the basefluid. The volume fraction and the size of particle clusters are estimated based on particle size distribution (PSD) analysis. The primary particles and the clusters along with the interfacial layer around them are taken as complex primary particles and complex clusters, respectively. Primarily in the first level of homogenization, thermal conductivity of complex primary particles  $(k_{cpm})$  is calculated. In the second level of homogenization, the complex primary particles are alone considered to be suspended in the basefluid. Hence in the second level of homogenization, the thermal conductivity of suspension of complex primary particles in basefluid  $(k_1)$  including the effects of nano-scale convection caused by Brownian motion is determined. In the third level of homogenization, the thermal conductivity of complex cluster spheres ( $k_{cc}$ ) is calculated. Finally, the complex cluster spheres are considered to be suspended in a medium of thermal conductivity

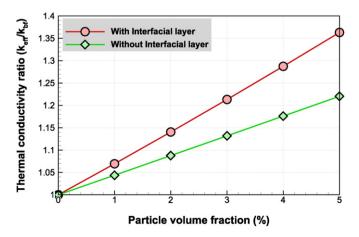


Fig. 7. Contribution of interfacial layer.

 $(k_1)$  to determine the total effective thermal conductivity of nanofluids  $(k_{\it eff})$ .

Proposed model is validated by comparing with the thermal conductivity data for six different samples of nanofluids, available in literature. Philip and Shima [35] have presented the PSD and  $k_{eff}$  measured at same instant of time for six different samples of nanofluids. The details of the samples and thermo-physical properties of the nanoparticles and basefluids taken for comparison are presented in Tables 1 and 2, respectively. The particle size distribution (PSD) of sample, S4 is shown in the inset image of Fig. 4. The comparison between the experimental data and the corresponding model prediction are clearly shown in Fig. 4. It is seen that the model predictions closely match with the experimental data for all the samples. The percentage difference difference between the model prediction and the experimental values range from 0.7% to 8.8%, which is acceptable in theoretical studies. In overall, a convincing agreement is obtained for all the six samples considered for comparison. Only six samples of nanofluids are used for comparison due to the limited availability of data for thermal conductivity and PSD measured independently, at same instant of time.

It can be understood that, f which determines the volume fraction of clusters present in the nanofluid and  $d_c/d_{pm}$  that shows the increased size of the cluster in comparison with the primary particle are the two prime factors which indicate the cluster characteristics and thus, affect the effective thermal conductivity of the nanofluid. Higher values of f indicate larger volume fraction of clusters in the nanofluid and hence, leading to higher enhancements in thermal conductivity. The value of f varies from 0 to 1. f=0, indicate that the particles are completely mono-dispersed and there are no clusters and when f=1, there will be no primary particles in the nanofluid. With the value of f = 1, the model gives the upper limit for any considered nanofluid. The upper and lower limits of the model calculated for an Alumina-Water nanofluid with  $k_{bf} = 0.6 \text{ W m}^{-1}\text{K}^{-1}$ ,  $k_{pm} = 40 \text{ W/mK}$ ,  $d_{pm} = 10 \text{ nm}$  and  $d_c/d_{pm} = 4$  at 293 K are presented in Fig. 5. The upper limit is calculated using f = 1; while, the lower limit which is equal to the MG model is calculated by neglecting the effects of interfacial layer ( $\delta = 0$ ), Brownian motion (Re = 0) and particle clustering (f = 0). When the interfacial layer thickness ( $\delta$ ), Brownian Reynolds number (Re) and the parameter (f) which determines the volume fraction of clusters are taken as zero the model is reduced to the basic MG model. It can be seen that, the thermal conductivity enhancement increases with increase in the volume fraction of clusters.

Apart from particle aggregation, thickness of the interfacial layer ( $\delta$ ) and Brownian Reynolds number (Re) also affect the effective thermal conductivity of the nanofluid system. The contribution of nano-scale convection caused by Brownian motion to the thermal conductivity enhancement of nanofluids is shown in Fig. 6. Contribution of Brownian motion is showcased using an Alumina-Water nanofluid with  $k_{bf} =$ 0.6 W/mK,  $k_{pm} = 40$  W/mK,  $d_{pm} = 10$  nm and  $d_c/d_{pm} = 4$  at 293 K. The effect of Brownian motion is neglected by assuming the Brownian Reynolds number as 0. It is seen that, the thermal conductivity of nanofluid is enhanced by the inclusion of effect of Brownian motion at all volume fractions. It is observed that at higher volume fractions, the contribution of nano-scale convection caused by Brownian motion is more significant. Similarly, the effect of interfacial layer for the same Alumina-Water nanofluid is studied by using  $\delta = 1$  nm and  $\delta = 0$  nm (see Fig. 7). The thermal conductivity of the nanofluid is higher when the effect of interfacial layer is taken into account. This proves the role of interfacial layer in thermal transport of nanofluids.

The model considers the effects of particle size and temperature by means of Brownian motion. It is also to be noted that the PSD of nanofluid is dependent on the particle size, temperature, physio-chemical properties of particles and basefluids, and time [35]. The PSD of nanofluids vary depending on all the above factors. Thus, the model will be capable of explaining the effects of all the above factors based on the PSD obtained experimentally by Dynamic Light Scattering (DLS) experiments.

# 5. Conclusions

A simple comprehensive model has been proposed to explain the thermal conductivity of the nanofluid considering the effects of nanoscale convection caused by Brownian motion, interfacial layer and particle aggregation. The model is built based on the particle size distribution and a multilevel homogenization approach. The main advantage of the model is its ease and precise characterization of particle clusters using particle size distribution. The particle size distribution which can be easily measured experimentally will give a clear data about the clusters in nanofluids which is more precise and real time when compared to the characterization by fractal theory. In addition to that, the model considers the mechanisms of nano-scale convection caused by Brownian motion and interfacial layer formation. The model suggests that the effective thermal conductivity increases with increase in volume fraction of clusters. Excellent agreement has been obtained between the model predictions and experimental data from literature. Roles of Brownian motion and interfacial layer in enhancing the thermal transport of nanofluids have been explicitly showcased. The model takes into account the influence of particle size, temperature, physiochemical properties of particles and basefluids and time. Hence, it will be helpful in explaining the contradictions in thermal conductivity data of nanofluids available in literature. Thus, this model is expected to aid in better understanding of the thermal transport in nanofluids and lead to practical application of nanofluids in many industrial applications.

#### Nomenclature

#### **Notations**

N Number of primary particles in a cluster

T Temperature in [K] P Number percentage

K Thermal conductivity in [Wm<sup>-1</sup>K<sup>-1</sup>]

KB Boltzmann constant

## Greek symbols

 $\phi$  Volume fraction  $\rho$  Density in [kgm<sup>-3</sup>]

δ Thickness of the interfacial layer v Kinematic viscosity in [m<sup>2</sup>S<sup>-1</sup>]

# Subscripts

eff Effective property of nanofluid

c Clusters
l Interfacial layer
cc Complex clusters
pm Primary nanoparticles

bf Basefluid

cpm Complex primary particles

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