

# A prediction model for the effective thermal conductivity of nanofluids considering agglomeration and the radial distribution function of nanoparticles

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**Abstract** Conventional heat transfer fluids usually have low thermal conductivity, limiting their efficiency in many applications. Many experiments have shown that adding nanosize solid particles to conventional fluids can greatly enhance their thermal conductivity. To explain this anomalous phenomenon, many theoretical investigations have been conducted in recent years. Some of this research has indicated that the particle agglomeration effect that commonly occurs in nanofluids should play an important role in such enhancement of the thermal conductivity, while some have shown that the enhancement of the effective thermal conductivity might be accounted for by the structure of nanofluids, which can be described using the radial distribution function of particles. However, theoretical predictions from these studies are not in very good agreement with experimental results. This paper proposes a prediction model for the effective thermal conductivity of nanofluids, considering both the agglomeration effect and the radial distribution function of nanoparticles. The resulting theoretical predictions for several sets of nanofluids are highly consistent with experimental data.

**Keywords** Nanofluid · Thermal conductivity · Modeling · Agglomeration · Radial distribution function

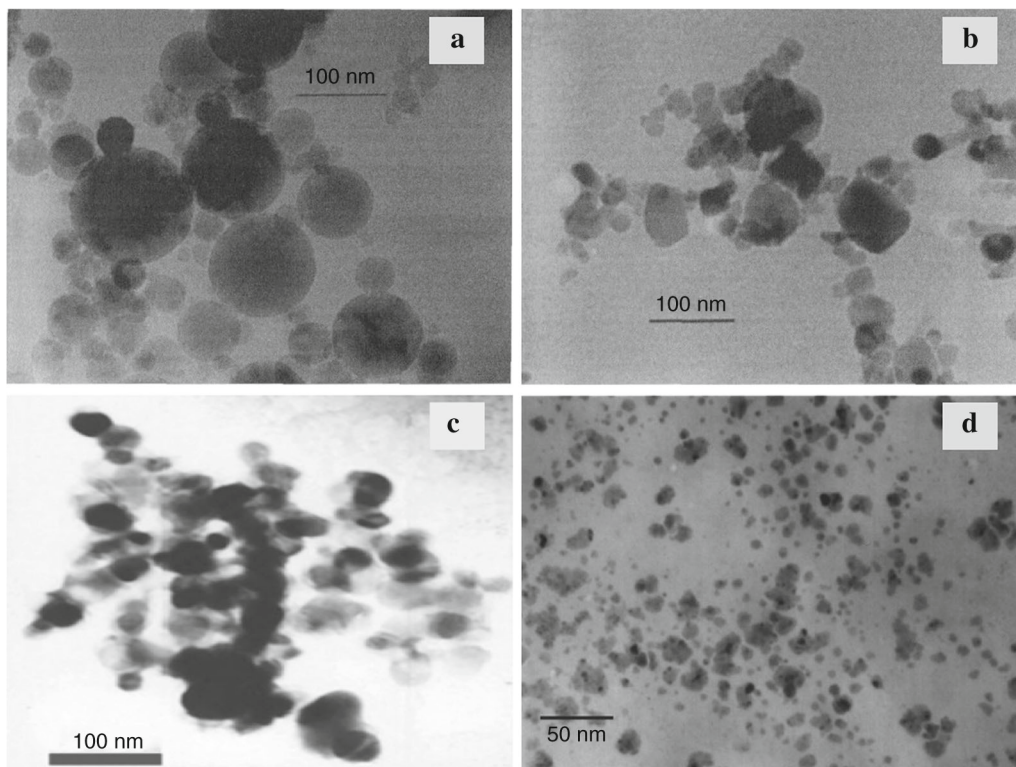
## 1 Introduction

The intrinsically low thermal conductivity of conventional heat transfer fluids is one of the major limitations on development of high-efficiency cooling devices in industrial fields. To achieve development objectives, these fluids must be replaced or their thermal properties improved. On the other hand, it is well known that solid particles exhibit better thermal conductivity compared with conventional heat transfer fluids. However, addition of micro- or millisize solid particles to base fluids, which will enhance the thermal conductivity, also results in anomalous, high flow resistance and severe pipe wear. These negative side-effects have been obstacles to their feasible applications. In 1995, a breakthrough study on enhancing the thermal conductivity of conventional heat transfer fluids was carried out by Choi and Eastman [1], who put forward the concept of, viz. a suspension of nanoparticles dispersed in a conventional fluid, and firstly synthesized  $\text{Al}_2\text{O}_3$ –water nanofluid. Intriguingly, he found that addition of nanoparticles to conventional fluids resulted in anomalously enhanced thermal conductivity. In recent years, many experimental investigations have also been performed on thermal conductivity enhancement of nanofluids [2–8]. For example, Lee et al. [2] measured the thermal conductivity of several kinds of nanofluids, revealing enhancements of about 12.1 % for 3.4 vol.% CuO–water nanofluid and 9.3 % for 4.0 vol.%  $\text{Al}_2\text{O}_3$ –water nanofluid. They also found that the nanoparticles in these systems were agglomerated (Fig. 1a, b), since they did not use an electrostatic repulsion technique during the preparation of nanofluids. Kole and Dey [8] studied the effects of volume fraction and temperature on the enhancement of the thermal conductivity of gear-oil-based nanofluid. Data from transmission electron microscopy (TEM) (Fig. 1c) and dynamic light scattering

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**Fig. 1** TEM images of different nanofluids. **a** Al<sub>2</sub>O<sub>3</sub>–water (from Lee et al. [2]). **b** CuO–water (from Lee et al. [2]). **c** CuO–gear oil (from Kole and Dey [8]). **d** Cu–EG (from Eastman et al. [3])

(DLS) measurements confirmed the presence of agglomerated nanoparticles in their prepared nanofluids. Moreover, the thermal conductivity enhancement was about 10.4 % for 2.5 vol.% CuO loading at 30 °C. Intriguingly, the results of Eastman et al. [3] showed that, for a nanofluid consisting of ethylene glycol (EG) and only 0.3 vol.% Cu nanoparticles, the thermal conductivity was measured to be increased by 40 %, while very little agglomeration of Cu particles was observed by TEM (Fig. 1d).

These amazing experimental results motivated researchers to study the heat conduction mechanisms behind the effective thermal conductivity of nanofluids theoretically. Most existing models are based on effective medium theory, in which the size, shape, and volume fraction of particles are typically incorporated as variables, and the interfacial nanolayer at solid–liquid interfaces is also taken into account [9–14]. For instance, Lu and Song [9] developed a theoretical model to second order in volume fraction  $\phi$  to predict the effective thermal conductivity of composites with coated or bonded spherical inclusions. Yu and Choi [10, 11] believed that the several-nanometer-thick interfacial nanolayer at solid–liquid interfaces played an important role in the effective thermal conductivity of nanofluids, proposing two novel models based on the Maxwell model and Hamilton and Crosser model, respectively. Xue and Xu [12] also modified Bruggeman’s model for the effective thermal conductivity of nanofluids by taking the

interfacial layer effect into account. However, like the predictions of classical models (some of which are listed in Table 1) applied for computing the effective thermal conductivity of mixtures including millimeter-size or micrometer-size solid particles, the predictions of their models were lower than experimental results for the thermal conductivity of nanofluids. The weakness of these modified classical models for the thermal conductivity of nanofluids indicates that some mechanisms may have been missed.

Since nanoparticles have high surface energy, they easily agglomerate together when added to a base fluid, as shown in some experimental results [2, 4–8]. Moreover, adjacent particles in agglomerates create percolation paths with lower thermal resistance, which may have a major effect on the effective thermal conductivity [15, 16]. To estimate the contribution of this agglomeration effect to the thermal conductivity enhancement of nanofluids, several prediction models have been proposed. Based on the effective thermal medium approximation and fractal theory, Wang et al. [17] developed a fractal model for nanofluids to take the agglomerates and their radial distribution into account. Their predictions for the effective thermal conductivity of CuO nanofluids fit successfully with their experimental data for  $\phi < 5\%$ . However, this model could not match a wide range of experimental results and not be able to predict the nonlinear behavior of the effective thermal conductivity of

nanofluids. Prasher et al. [18] used a three-level homogenization method, validated by Monte Carlo simulations of heat conduction in fractal aggregates, to investigate the role of agglomeration in the enhancement of the purely conductive phenomenon in nanofluids. Their prediction results showed that the conductivity enhancement strongly depended on the fractal dimension of the agglomerates and their radius of gyration. Feng et al. [19] considered the nanolayer and agglomeration effects of particles and proposed a theoretical model for the thermal conductivity of nanofluids based on a two-dimensional (2D) lattice model, assuming that the formation of agglomerates followed the simple packing principle. Zhou and Wu [20] proposed a theoretical model to predict the thermal conductivity of nanofluids, in which the influence of nanoparticle clusters was considered, by introducing particle size distribution, with an assumption that particles in clusters aggregated with each other closely. Xiao et al. [21] took into account heat convection between nanoparticles and liquids due to Brownian motion of nanoparticles in nanofluids. Their expression for the thermal conductivity of nanofluids was developed based on fractal geometry theory. However, the random distribution and disorder effect of nanoparticles to structure formation in nanofluids contribute to heat conduction. Therefore, these effects should be considered in the prediction model for effective thermal conductivity of nanofluids, by introducing the radial distribution function of nanoparticles [9, 22–24].

Against this background, we propose herein a model for predicting the effective thermal conductivity of nanofluids. The development of the model proceeds as follows: The nanofluid system is first divided into several parts, each of which contains agglomerates at a specific level with a nanolayer and the surrounding base fluid; We treat each agglomerate and its surrounded nanolayer as an equilibrium agglomerate with equilibrium nanolayer. To describe the effect of the presence of agglomerates on the effective thermal conductivity in each equilibrium part, a radial distribution function is introduced. Thus, the effective thermal conductivity of the whole system can be evaluated by combining each part. In this way, the agglomeration effect and the radial distribution function of the nanoparticles are taken into account. Finally, the predictions of the present model for several sets of nanofluids are also compared with experimental results, as well as with other theoretical models.

## 2 Models of effective thermal conductivity

### 2.1 Earlier models

Many classical models have been developed to calculate the effective thermal conductivity of solid–liquid suspensions. However, these classical models apply to dilute suspensions

containing milli- or microparticles. To extend them to predict the effective thermal conductivity of nanofluids, various researchers have proposed modified models derived from classical models. Some of these are listed in Table 1.

#### 2.1.1 Present model

The TEM images in Fig. 1a–c show nanoparticle agglomerates of different sizes. This random distribution of multisize agglomerates (or multilevel agglomerates) makes a great contribution to the enhancement of the heat conduction in nanofluids. Therefore, we introduce an equivalent agglomeration method [28] to take the agglomeration effect into account in the thermal conductivity calculation. A schematic of the method is shown in Fig. 2. The main idea is that the multilevel agglomerates consist of two parts: pure solid-phase particle material and liquid phase bounded in pores. For an  $i$ th-level agglomerate, the contained solid-phase particles are regarded as an equivalent solid particle with the same volume, while the liquid phase, including the liquid in the gaps and the nanolayer on the surface of the agglomerate, is considered as an equivalent interfacial nanolayer on the surface of the equivalent solid particle. The equivalent nanolayer thickness  $t_i$  satisfies the equation:  $t_i = R_{pe}^i - R_p^i$ , where  $R_{pe}^i$  and  $R_p^i$  are the radius of the equivalent agglomeration particle  $i$  and equivalent solid particle  $i$ , respectively. Moreover, each primary single particle with its interfacial nanolayer is treated as a zero-level agglomerate for convenience, thus  $R_p^0$  represents the radius of such a single solid particle.

Since the random distribution of each level of agglomerates makes a different partial contribution to the enhancement of the heat conduction in the nanofluid, the agglomeration ratio  $\lambda_i$  of the  $i$ th-level agglomerates is introduced to weigh their contribution to the total thermal conductivity enhancement. The agglomeration ratio  $\lambda_i$  can be calculated as  $\lambda_i = N_c^i / N$ , where  $N_c^i$  and  $N$  are the number of agglomerates  $i$  and the total number of particles (single particles and agglomerates), which can both be measured approximately from TEM images. Moreover,  $\lambda_i$  should satisfy  $\sum_{i=0}^n \lambda_i = 1$ ,  $i = 0, 1, 2, \dots, n$ .

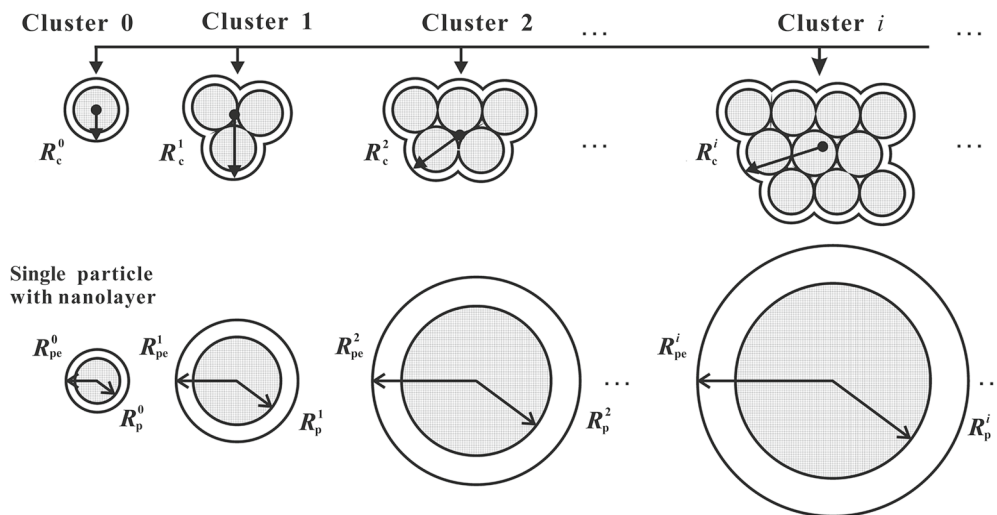
To calculate the effective thermal conductivity, we assume that the contribution of each level of agglomerates to the effective thermal conductivity of the nanofluid complies with the principle of linear weighted superposition. Thus, we define  $k_e^i$  as the effective thermal conductivity of a nanofluid in the presence of only  $i$ th-level agglomerates, which can be expressed as [9]

$$\frac{k_e}{k_f} = 1 + 3\theta_1\phi_e^i + \frac{3\theta_1^2(\phi_e^i)^2}{1 - \theta_1\phi_e^i} + \frac{M^*(\phi_e^i)^2}{1 - \theta_1\phi_e^i}, i = 0, 1, \dots, n. \quad (1)$$

**Table 1** Earlier theoretical models of effective thermal conductivity of solid–liquid suspensions or nanofluids

Model	Expression	Remarks
<i>Classical models</i>		
Maxwell [25]	$\frac{k_e}{k_f} = \frac{k_p + 2k_f + 2\phi(k_p - k_f)}{k_p + 2k_f - \phi(k_p - k_f)}$	Spherical particles are considered
Hamilton and Crosser [26]	$\frac{k_e}{k_f} = \frac{k_e + (n-1)k_f + (n-1)\phi(k_p - k_f)}{k_e + (n-1)k_f - \phi(k_p - k_f)}$	Spherical and nonspherical particles are considered: $n = 3$ for spheres, $n = 6$ for cylinders
Jeffrey [22]	$\frac{k_e}{k_f} = 1 + 3\beta\phi + 3\beta\phi^2 \left( \beta + \sum_{p=6}^{\infty} \frac{B_p - 3A_p}{(p-3)2^{p-3}} \right)$	Considering Dirac step function as pair distribution function to describe pair interaction of randomly dispersed particles
<i>Modified models</i>		
Yu and Choi [10]	$\frac{k_e}{k_f} = \frac{k_{pe} + 2k_f + 2(1+\delta)^3\phi(k_{pe} - k_f)}{k_{pe} + 2k_f - (1+\delta)^3\phi(k_{pe} - k_f)}$	A modified Maxwell model taking the interfacial nanolayer effect into account
Prasher et al. [18]	$\frac{k_e}{k_f} = \frac{k_a + 2k_f + 2\phi_a(k_a - k_f)}{k_a + 2k_f - \phi_a(k_a - k_f)}$	A model using effective medium theory and considering the fractal clusters in nanofluids; $k_a$ and $\phi_a$ are the thermal conductivity and effective volume fraction of agglomerates, respectively
Murshed et al. [27]	$\frac{k_e}{k_f} = \{ (k_p - k_l) \phi k_l [2\gamma_1^3 - \gamma^3 + 1] + (k_p + 2k_l) \gamma_1^3 [\phi\gamma^3(k_l - k_f) + k_f] \} \times \{ \gamma_1^3(k_p + 2k_l) - (k_p - k_l) \times \phi k_f [\gamma_1^3 + \gamma^3 - 1] \}^{-1}$	A modified Maxwell model taking the interfacial nanolayer effect into account; the nanolayer thickness satisfies the Hashimoto equation: $t = \sqrt{2\pi}\delta$

Subscripts “e,” “f,” “p,” and “l” stand for nanofluid, base fluid, particles, and nanolayer, respectively

**Fig. 2** Scheme of equivalent agglomeration method [28]

In Eq. (1),  $\phi_e^i$  is the effective volume fraction of agglomerate  $i$  with its combined nanolayer. The second term on the right-hand side, first obtained by Maxwell, accounts for the presence of rigid spheres without interactions; the third term is a correlation to the first-order coefficient due to the mutual impenetrability of the spheres. The fourth term takes into account the thermal interactions (i.e., how the temperature field around the first spherical particle is affected by the presence of the second), as well as the structural distribution

of pairs of particles (i.e., how the presence of one particles affects the location of the other), where the coefficient  $M^*$  can be expressed as

$$M^* = 3\theta_1^2 \int_2^\infty \left[ \sum_{p=6}^\infty (B_p - 3A_p) \left( \frac{R_p^i}{r} \right)^{p-2} \right] \times g \left( \frac{r}{R_p^i} \right) d \left( \frac{r}{R_p^i} \right), \quad (2)$$



with

$$\theta_1 = \frac{\left[ \left( 1 + \frac{t_i}{R_p^i} \right)^3 - \frac{\beta_{pl}}{\beta_n} \right] \beta_{lf}}{\left( 1 + \frac{t_i}{R_p^i} \right)^3 + 2\beta_{lf}\beta_{pl}}, \quad (3)$$

$$\beta_{xy} = \frac{k_x - k_y}{k_x + 2k_y}, \quad (4)$$

and

$$\phi_e^i = \lambda_i \left( 1 + \frac{t_i}{R_p^i} \right)^3 \phi = \lambda_i (1 + \delta_i)^3 \phi. \quad (5)$$

In Eqs. (2)–(5),  $\phi$  and  $\phi_e^i$  are the volume fraction of all particles without their nanolayers and the effective volume fraction of agglomerate  $i$  with its combined nanolayer, respectively;  $A_p$  and  $B_p$  are the coefficients of each term in the summation, which are expressed in detail in Ref. [22];  $r$  is the distance between the center of each pair of particles in the system;  $\theta_1$  is the dimensionless polarizability, characterizing the interfacial nanolayer effect;  $\beta_{xy}$  is a convenient measure of the disparity between  $k_x$  and  $k_y$  [22, 24];  $k_x$  and  $k_y$  refer to  $k_p$ ,  $k_f$ , and  $k_l$ , which are the thermal conductivity of solid-phase particle, base fluid, and interfacial nanolayer, respectively.  $g(r/R_p^i)$  is a radial distribution function to describe the structure of suspensions of identical rigid spherical particles within a liquid, first proposed by Jeffrey [22]. Employing the simplified statistics of a homogeneous structure, he assumed that the radial distribution function should satisfy the conditions of low-density limits and temperature continuity at liquid–solid interfaces, taking the following form:  $g \rightarrow H(r - 2R_{pe}^i)$  (where  $H$  is a step function). However, a step function cannot represent the structure of an equilibrium rigid sphere fluid at any finite density. Thus, an alternative radial distribution function is introduced in the present model, and can be accurately evaluated using the procedure proposed by Verlet and Weis [29].

It is worth mentioning that the thermal conductivity of the interfacial nanolayer  $k_l$  should be a nonlinear function of  $\delta$  (ranging from 0 to  $\delta_i$ ). If not, it would tend to an infinite value as the nanolayer became ultrathin [30, 31] ( $t \rightarrow 0$ ). Moreover, it decreases from  $k_p$  to  $k_f$  as  $\delta$  changes from 0 to  $\delta_i$ , because the interfacial nanolayer represents an intermediate state between the bulk solid and liquid phase. Following the analysis above, the thermal conductivity of nanolayer  $k_l$  should become [28, 32, 33]

$$k_l(\delta) = e^{-c_i \delta}, \quad (6)$$

where  $c_i = \ln(k_p/k_f)/\delta_i$  is related to the ratio  $\delta_i = t_i/R_p^i$ . We then use the integral average of Eq. (6) to calculate

the effective thermal conductivity of nanolayer  $k_l$ , with the form

$$k_l = \frac{1}{\delta_i} \int_0^{\delta_i} k_l(\delta) d\delta. \quad (7)$$

Now, the effective thermal conductivity of the nanofluid, considering the agglomeration effect, can be expressed as

$$k_e = \sum_{i=0}^n \lambda_i k_e^i, \quad (8)$$

and the effective thermal conductivity of the nanofluid can be predicted by substituting Eqs. (1)–(5) and Eq. (7) into Eq. (8). According to experimental studies [2, 8], we posit that there exists one level of agglomeration in a dilute suspension, for simplicity, thus Eq. (8) becomes

$$k_e = \lambda_0 k_e^0 + \lambda_1 k_e^1. \quad (9)$$

For the computation using just one-level agglomeration, the radius of the equivalent solid particle  $R_p^1$  can be replaced by the radius of the first-level agglomerate  $R_c^1$ , since  $R_c^1$  can be measured by TEM and the volume of liquid gaps in a one-level agglomerate are small compared with the solid-phase particles. Also, the agglomeration ratio of the one-level agglomerate  $\lambda_1$  can also be estimated by TEM.

Through its consideration of the structural distribution of pairs of particles, the accuracy of our model is up to order  $\phi^2$ . Moreover, the present model can also be used to predict the effective thermal conductivity of nanofluids without agglomerates. For such cases, one lets the agglomeration ratio  $\lambda_1$  be zero, then Eq. (9) degenerates to the model of Lu and Song with an exponential distribution of thermal conductivity in the nanolayer.

### 3 Results and discussion

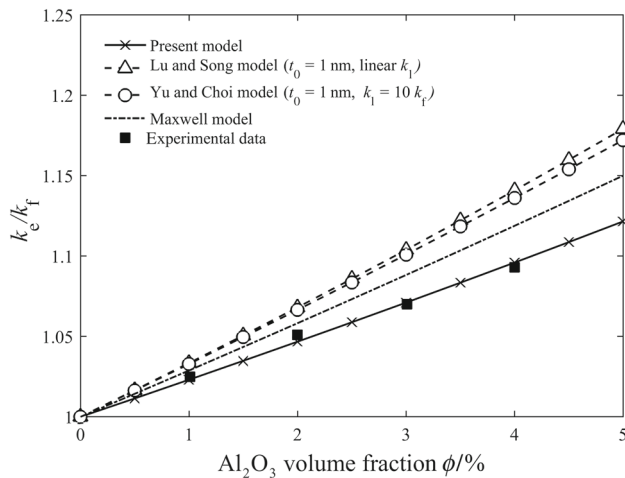
The present model was validated by comparing its prediction results with several sets of experimental data, as well as results calculated using other theoretical models for nanofluids. A linear distribution and a constant value of thermal conductivity in the nanolayer are employed in the calculations using the models of Lu and Song and of Yu and Choi, respectively.

Table 2 presents the values of the parameters  $\lambda_i$ ,  $\delta_i$ , and  $t_i$  ( $i = 0, 1$ ) used in our model according to the experimental data from Lee et al. [2], Kole and Dey [8], and Eastman et al. [4]. The agglomeration ratio of the one-level agglomerate  $\lambda_1$  was taken as 19%, 14%, 26%, 0% for  $\text{Al}_2\text{O}_3$ –water,  $\text{CuO}$ –water,  $\text{CuO}$ –gear oil, and  $\text{Cu}$ –EG, respectively.

The predictions of the present model for the  $\text{Al}_2\text{O}_3$ –water nanofluid are in good agreement with the experimental data

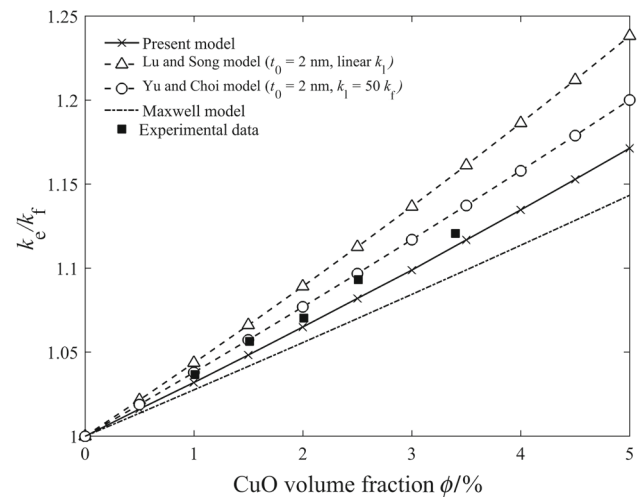
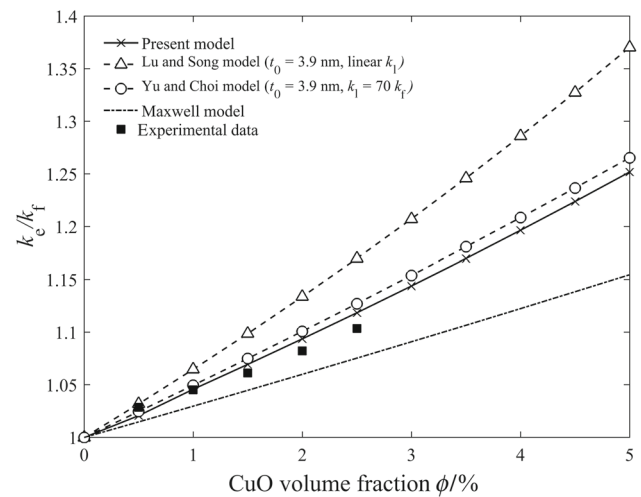
**Table 2** Values of parameters used in our model

Type of nanofluid	$\lambda_1$ (%)	$\delta_0$	$\delta_1$	$t_0$ (nm)	$t_1$ (nm)
Al <sub>2</sub> O <sub>3</sub> –water [2]	19	0.05	0.10	0.96	7.00
CuO–water [2]	14	0.17	0.15	2.00	6.75
CuO–gear oil [8]	26	0.185	0.13	3.90	9.10
Cu–EG [3]	0	1.00	—	3.00	—

**Fig. 3** Thermal conductivity variation for Al<sub>2</sub>O<sub>3</sub>–water nanofluid [2]

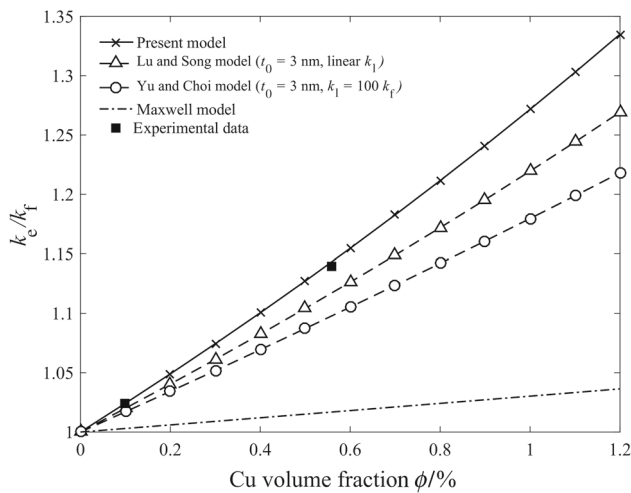
for the Al<sub>2</sub>O<sub>3</sub>–water nanofluid synthesized by Lee et al. [2], as shown in Fig. 3. The average equivalent radius of one-level agglomerates used in the present calculation is about  $R_c^1 \approx 70$  nm based on the scale bar in the TEM images (Fig. 1a), and the nanolayer thickness of the original particle and equivalent agglomeration particle are  $t_0 = 0.96$  nm and  $t_1 = 7.00$  nm, respectively. The value of  $t_0$  is close to the results of molecular dynamics (MD) simulation ( $\approx 1$  nm) carried out by Xue et al. [34] and direct observations using the X-ray reflection technique, suggesting that the nanolayer thickness should be several times the water molecule diameter [35]. Meanwhile, the calculation results of Maxwell model, Lu and Song model with  $t_0 = 1$  nm and Yu and Choi model with  $t_0 = 1$  nm, show higher deviations from experimental data. Comparison between the predictions of the present model and those obtained using the model of Lu and Song indicates that the agglomerates of nanoparticles would make a negative contribution to the effective thermal conductivity of nanofluids.

For the CuO–water nanofluid, the predictions of the present and other models are shown in Fig. 4, in comparison with the experimental data obtained by Lee et al. The values of the nanolayer thickness used in the present calculations are chosen as  $t_0 = 2.00$  nm and  $t_1 = 6.75$  nm, while the average equivalent radius of one-level agglomerates was  $R_c^1 \approx 45$  nm, based on the scale bar in the

**Fig. 4** Thermal conductivity variation for CuO–water nanofluid [2]**Fig. 5** Thermal conductivity variation for CuO–gear oil nanofluid [8]

TEM image (Fig. 1b). Meanwhile, the same value of  $t_0$  is adopted for the predictions using the model of Lu and Song, with a linear distribution of  $k_1$ , as well as the predictions obtained using the model of Yu and Choi, with a constant  $k_1 = 50k_f$ . As expected, good agreement is found between the predictions of the present model and the experimental data.

The predictions for the CuO–gear oil nanofluid are illustrated in Fig. 5, using values for the nanolayer thickness of  $t_0 = 3.90$  nm and  $t_1 = 9.10$  nm, and the average equivalent radius of one-level agglomerates is  $R_c^1 \approx 70$  nm based on the scale bar in the TEM image (Fig. 1c). By considering the agglomeration effect to reach order up to  $\phi^2$ , the present predictions show better agreement with experimental data [4] compared with the other models. In addition, Figs. 4 and 5 reveal that the nanolayer thickness of CuO nanoparticles in water is much thinner than that in gear oil,



**Fig. 6** Thermal conductivity variation for Cu-EG nanofluid [3]

indicating that the nanolayer thickness depends on the type of base fluid, as also reported in several previous investigations [30,36,37].

In the case of the Cu-EG nanofluid, the predictions of the present model are shown in Fig. 6, in comparison with the results of other models and experimental data [3]. The method used by Eastman et al. to synthesize their nanofluid resulted in little or no agglomerates, as shown in the TEM image (Fig. 1d). Therefore, we chose an agglomeration ratio for one-level agglomerates of  $\lambda_1 = 0\%$  in our calculations. The good consistency between the effective thermal conductivity predictions and the experimental data and the results of other models results from the contribution of order  $\phi^2$  and the nonlinear distribution of thermal conductivity  $k_1$ . In addition, this also indicates that the chosen values of  $\delta_i$  presented in Table 2 are reasonable. Another impressive finding is that the ratio  $\delta_i$  becomes relatively large if  $R_c^1$  is small enough, both with and without agglomerates.

## 4 Conclusions

We present a prediction model for the effective thermal conductivity of nanofluids, using the equivalent agglomeration method to consider the agglomeration effect. With the help of the radial distribution function of the nanoparticles in the nanofluid, the present model is up to order  $\phi^2$  in the volume fraction, comparable to most other theoretical models. The highly consistent predictions of the present method indicate that its application for predicting the thermal conductivity of nanofluids, with or without agglomerates, is more appropriate than other models, based on comparisons with experimental datasets. However, validation of the chosen values, including the agglomeration ratio and

nanolayer thickness, must be further investigated. Moreover, other mechanisms for thermal conductivity enhancement, e.g., based on temperature, pH value, and even viscosity, might also be considered in future studies.

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## References

- Choi, S.U.S., Eastman, J.A.: Enhancing Thermal Conductivity of Fluids with Nanoparticles. In: International mechanical engineering congress and exhibition, San Francisco, Nov. 12–17 (1995)
- Lee, S., Choi, S.U.S., Li, S., et al.: Measuring thermal conductivity of fluids containing oxide nanoparticles. *ASME J. Heat Transf.* **121**, 280–9 (1999)
- Eastman, J.A., Choi, S.U.S., Li, S., et al.: Anomalous increased effective thermal conductivities of ethylene glycol-based nanofluids containing copper nanoparticles. *Appl. Phys. Lett.* **78**, 718–720 (2001)
- Das, S.K., Putra, N., Thiesen, P., et al.: Temperature dependence of thermal conductivity enhancement for nanofluids. *ASME J. Heat Transf.* **125**, 567–574 (2003)
- Kang, H.U., Kim, S.H., Oh, J.M.: Estimation of thermal conductivity of nanofluid using experimental effective particle volume. *Exp. Heat Transf.* **19**, 181–191 (2006)
- Karhikeyan, N.R., Philip, J., Raj, B.: Effect of clustering on the thermal conductivity of nanofluids. *Mater. Chem. Phys.* **109**, 50–55 (2008)
- Mintsa, H.A., Roy, G., Nguyen, C.T., et al.: New temperature dependent thermal conductivity data for water-based nanofluids. *Int. J. Therm. Sci.* **48**, 363–371 (2009)
- Kole, M., Dey, T.K.: Enhanced thermophysical properties of copper nanoparticles dispersed in gear oil. *Appl. Therm. Eng.* **56**, 45–53 (2013)
- Lu, S.Y., Song, J.L.: Effective conductivity of composites with spherical inclusions: effect of coating and detachment. *J. Appl. Phys.* **79**, 609–618 (1996)
- Yu, W., Choi, S.U.S.: The role of interfacial layers in the enhanced thermal conductivity of nanofluids: a renovated Maxwell model. *J. Nano. Res.* **5**, 167–171 (2003)
- Yu, W., Choi, S.U.S.: The role of interfacial layers in the enhanced thermal conductivity of nanofluids: a renovated Hamilton–Crosser model. *J. Nano. Res.* **6**, 355–361 (2014)
- Xue, Q., Xu, W.M.: A model of thermal conductivity of nanofluids with interfacial shells. *Mater. Chem. Phys.* **90**, 298–301 (2005)
- Rizvi, I.H., Jain, A., Ghosh, S.K., et al.: Mathematical modelling of thermal conductivity for nanofluid considering interfacial nanolayer. *Heat Mass Transf.* **49**, 595–600 (2013)
- Jiang, H., Xu, Q., Huang, C., et al.: The role of interfacial nanolayer in the enhanced thermal conductivity of carbon nanotube-based nanofluids. *Appl. Phys. A* **118**, 197–205 (2015)
- Buongiorno, J.: Convective transport in nanofluids. *J. Heat Transf.* **128**, 240–250 (2006)
- Kebllinski, P., Prasher, R., Eapen, J.: Thermal conductance of nanofluids: Is the controversy over? *J. Nano. Res.* **10**, 1089–1097 (2008)
- Wang, B.X., Zhou, L.P., Peng, X.F.: A fractal model for predicting the effective thermal conductivity of liquid with suspension of nanoparticles. *Int. J. Heat Mass Transf.* **46**, 2665–2672 (2003)

18. Prasher, R., Evans, W., Meakin, P., et al.: Effect of aggregation on thermal conduction in colloidal nanofluids. *Appl. Phys. Lett.* **89**, 143119 (2006)
19. Feng, Y., Yu, B., Xu, P., et al.: The effective thermal conductivity of nanofluids based on the nanolayer and the aggregation of nanoparticles. *J. Phys. D: Appl. Phys.* **40**, 3164 (2007)
20. Zhou, D., Wu, H.: A thermal conductivity model of nanofluids based on particle size distribution analysis. *Appl. Phys. Lett.* **105**, 083117 (2014)
21. Xiao, B., Yang, Y., Chen, L.: Developing a novel form of thermal conductivity of nanofluids with Brownian motion effect by means of fractal geometry. *Powder Technol.* **239**, 409–414 (2013)
22. Jeffrey, D.J.: Conduction through a random suspension of spheres. *Proc. R. Soc. Lond. A: Math. Phys. Eng. Sci.* **335**, 355–367 (1973)
23. Chiew, Y.C., Glandt, E.D.: The effect of structure on the conductivity of a dispersion. *J. Colloid Interface Sci.* **94**, 90–104 (1983)
24. Chiew, Y.C., Glandt, E.D.: Effective conductivity of dispersions: the effect of resistance at the particle surfaces. *Chem. Eng. Sci.* **42**, 2677–2685 (1987)
25. Maxwell, J.C.: *Electricity and Magnetism*. Clarendon Press, Oxford (1873)
26. Hamilton, R.L., Crosser, O.K.: Thermal conductivity of heterogeneous two-component systems. *Ind. Eng. Chem. Fund.* **1**, 187–191 (1962)
27. Murshed, S.M.S., Leong, K.C., Yang, C.: Investigations of thermal conductivity and viscosity of nanofluids. *Int. J. Therm. Sci.* **47**, 560–568 (2008)
28. Sui, J., Zheng, L.C., Zhang, X.X., et al.: A novel equivalent agglomeration model for heat conduction enhancement in nanofluids. *Sci. Rep.* **6**, 19560 (2016)
29. Verlet, L., Weis, J.J.: Equilibrium theory of simple liquids. *Phys. Rev. A* **5**, 939–952 (1972)
30. Liang, Z., Tsai, H.L.: Thermal conductivity of interfacial layers in nanofluids. *Phys. Rev. E* **83**, 041602 (2011)
31. Tso, C.Y., Fu, S.C., Chao, C.Y.: A semi-analytical model for the thermal conductivity of nanofluids and determination of the nanolayer thickness. *Int. J. Heat Mass Transf.* **70**, 202–214 (2014)
32. Xie, H., Fujii, M., Zhang, X.: Effect of interfacial nanolayer on the effective thermal conductivity of nanoparticle-fluid mixture. *Int. J. Heat Mass Transf.* **48**, 2926–2932 (2005)
33. Sohrabi, N., Masoumi, N., Behzadmehr, A., et al.: A simple analytical model for calculating the effective thermal conductivity of nanofluids. *Heat Transf. Asian Res.* **39**, 141–150 (2010)
34. Xue, L., Keblinski, P., Phillpot, S.R., et al.: Effect of liquid layering at the liquid–solid interface on thermal transport. *Int. J. Heat Mass Transf.* **47**, 4277–4284 (2004)
35. Yu, C.J., Richter, A.G., Datta, A., et al.: Molecular layering in a liquid on a solid substrate: an X-ray reflectivity study. *Phys. B: Cond. Mater.* **283**, 27–31 (2000)
36. Xue, Q.Z.: Model for effective thermal conductivity of nanofluids. *Phys. Lett. A* **307**, 313–317 (2003)
37. Firlar, E., Çınar, S., Kashyap, S., et al.: Direct visualization of the hydration layer on alumina nanoparticles with the fluid cell STEM in situ. *Sci. Rep.* **5**, 9830 (2015)