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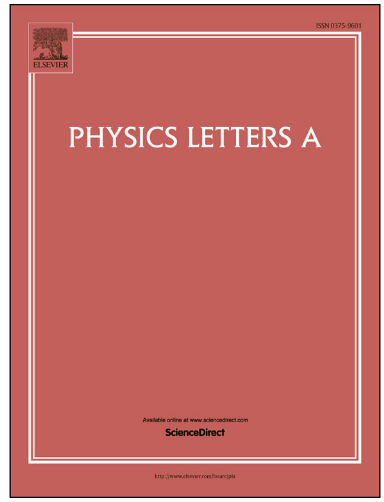

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Fractal analysis of the effect of particle aggregation distribution on thermal conductivity of nanofluids (Revised)

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Note: words/sentences/paragraphs in blue indicate revisions or newly-added material.

Abstract

A theoretical effective thermal conductivity model is derived based on fractal distribution characteristics of nanoparticle aggregation. Considering two different mechanisms of heat conduction including particle aggregation and convention, the model is expressed as a function of the fractal dimension and concentration. In the model, the change of fractal dimension is related to the variation of aggregation shape. The theoretical computations of the developed model provide a good agreement with the experimental results, which may serve as an effective approach for quantitatively estimating the effective thermal conductivity of nanofluids.

Highlights

A thermal conductivity model is derived based on fractal aggregation distribution.

The relationship between aggregation shape and fractal dimension is analyzed.

Predictions of the proposed model show good agreement with experimental data.

Keywords: Thermal conductivity; Fractal; Aggregation distribution; Aggregation shape

1. Introduction

Quantitative estimate of the effective thermal conductivity has attracted substantial attentions since it is one of the most important parameters characterizing the heat transport properties of nanofluids [1-4]. Nanofluids are liquid suspensions that contain nanometer-size particles, with size much smaller than 100 nm, and their thermal conductivity is higher than that of their base liquids [5-8]. In recent years, a great amount of efforts has been exerted to study conductivity characteristic, and significant progress has been made towards the theoretical modeling [9-14] and laboratory experiments [15-19]. In 19th century, Maxwell [20] predicted that the thermal conductivity of mixtures increase by suspending some higher-conductivity substance such as solid particles. Since Maxwell model is only a first-order approximation, it applies only to mixtures with low particle volume fraction and small values of the ratio of thermal conductivity between particle and liquid [21]. Moreover, other traditional models for multiphase systems, such as Wiener approximation [22] and Bruggeman approach [23], fail to illuminate the abnormal enhancement of the effective thermal conductivity for low particle volume fraction in nanofluids.

Several researchers concluded that the major factors of heat conduction mechanisms in nanofluids including particle aggregation [24, 25], particle motion [26-28] and liquid-layering [9, 29]. Particularly, the fact that particle aggregation can enhance the effective thermal conductivity of nanofluids has been confirmed experimentally [30-32]. Wang et al. [33] claimed that particle clustering could prominently affect the enhancement of thermal conductivity of nanofluids. Hamilton and Crosser [34] presented a mixture model to explain heterogeneous two-component systems. In their model, the particle aggregation shape is invariable, which ignores the effect of aggregation shape on the effective thermal conductivity of nanofluids.

After fractal geometry was introduced by Mandelbrot [35], it became a powerful tool for the analysis of physico-geometrical properties and processes, such as electricity conductivity [36, 37], spontaneous capillary imbibition [38, 39], thermal conductivity [40-44] and permeability [45-48]. Several researchers [33, 49-53] also apply fractal geometry to study heat conduction of nanofluids. Wang et al. [33] established an effective thermal conductivity model based on the effective medium approximation and the fractal theory to describe nanoparticle cluster and radial distribution. Xu et al. [50] applied fractal geometry to predict the thermal conductivity in terms of particles sizes distribution and heat convection of nanofluids. Considering the effect of Brownian motion of nanoparticles, Xiao et al. [52] presented a fractal model of thermal conductivity which is expressed as a function of the average diameter of nanoparticles, the nanoparticle concentration, the fractal dimension of nanoparticles and physical properties of fluids.

To the best of our knowledge, there is no full relationship to depict the effective thermal conductivity of nanofluids with fractal clustering distribution in terms of particle aggregation and convection. In the present study, based on modified Hamilton and Crosser model and Xu et al. model, an analytical model considering fractal distribution characteristic of nanoparticle aggregation is derived to estimate the effective thermal conductivity. The validity of the model was confirmed by comparison with the experimental results.

2. The fractal thermal conductivity model

2.1. Consideration of size effect of nanoparticles

Hamilton and Crosser [34] used empirical shape factor F to consider the effect of two heterogeneous phases and improved Maxwell equation [20] to calculate the effective thermal conductivity of nanofluid k_s that is induced by stationary nanoparticles in the liquids:

$$k_s = k_f \frac{a+(F-1)-(F-1)(1-\alpha)\phi}{a+(F-1)+(1-\alpha)\phi} \tag{1}$$

and

$$F = \frac{3}{\psi} \tag{2}$$

where $a = k_p / k_f$ (k_p is thermal conductivity of particle and k_f is thermal conductivity of fluid), ψ is defined as the ratio of the surface area A'_p of a sphere to the surface area A_p of the particle whose volume V_p equal to that of the sphere, therefore

$$\psi = \frac{A'_p}{A_p} = \frac{6}{\lambda} \frac{V_p}{A_p} \tag{3}$$

where λ is aggregation size.

However, λ usually has different diameters due to aggregation in nanofluids and thus ψ is not a constant. According to Hamilton and Crosser, $\psi = 1$ for spherical particle and $\psi = 0.5$ for elliptic particle. If substituting λ, V_p and A_p with average particle size $\bar{\lambda}$, average volume $\bar{V}_p$ and average area $\bar{A}_p$, respectively, Eq. (3) can be deduced as

$$\psi = \frac{A'_p}{A_p} = \frac{6}{\bar{\lambda}} \frac{\bar{V}_p}{\bar{A}_p} \tag{4}$$

It has been shown that the size distribution of aggregation in nanofluids follows the fractal power law [33, 49, 50]. Analogous to pores in fractal porous media, the fractal probability density function can be expressed as [50]

$$f(x) = D\lambda_{\min}^{D} \lambda^{-(D+1)} d\lambda \tag{5}$$

The fractal dimension D is determined by [48]

$$\xi = \phi^{\frac{1}{D_E - D}} \quad \text{or} \quad D = D_E - \frac{\ln \phi}{\ln \xi} \tag{6}$$

where $D_E = 3$ for three-dimension space, ϕ is the concentration of nanoparticles and $\xi = \lambda_{\min} / \lambda_{\max}$, where $\lambda_{\max}$ and $\lambda_{\min}$ are the maximum and minimum diameters of nanoparticle cluster, respectively. When the particle cluster has fractal characteristics, its area and volume are $\pi\lambda^2$ and $\pi/6\cdot\lambda^3$, respectively, Eq. (4) can be expressed

$$\psi = \frac{6}{\bar{\lambda}} \frac{\int_{\lambda_{\min}}^{\lambda_{\max}} \frac{\pi}{6}\lambda^3 f(\lambda)d\lambda}{\int_{\lambda_{\min}}^{\lambda_{\max}} \pi\lambda^2 f(\lambda)d\lambda} \quad (7)$$

Combining Eqs. (5), (6) and (7), ψ can be obtained as

$$\psi = \frac{2-D}{3-D}\frac{\lambda_{\min}}{\bar{\lambda}}\frac{\phi^{-1}-1}{\phi^{\zeta_2}-1} \quad (8)$$

where $\zeta_2 = (D-2)/(3-D)$ and $\bar{\lambda}$ can be found from the statistical property of fractal object [50], as

$$\bar{\lambda} \approx \frac{D}{D-1}\lambda_{\min} \quad (9)$$

Inserting Eq. (9) into Eq. (8), the following equation can be obtained

$$\psi = \frac{D-1}{D}\frac{2-D}{3-D}\frac{\phi^{-1}-1}{\phi^{\zeta_2}-1} \quad (10)$$

Therefore, inserting Eq. (10) into Eq. (2) yields

$$F = 3\frac{D}{D-1}\frac{3-D}{2-D}\frac{\phi^{\zeta_2}-1}{\phi^{-1}-1} \quad (11)$$

In Hamilton and Crosser's model, *F* is constant for same shape particles (*F*=6 for ellipse and *F*=3 for sphere). However, it is observed that *F* is the function of fractal dimension and concentration as expressed in Eq. (11), and *F* increase with the increasing of concentration (see figure 1). As shown in figure 1, considering fractal distribution of nanoparticle aggregation, the shape of aggregation gradually grow to chain with the increasing concentration. When *F*<6, most aggregation shapes are circles. Eqs. (1) and (11) are the present fractal models that predict to effective thermal conductivity of nanofluids relating with nanoparticles cluster.

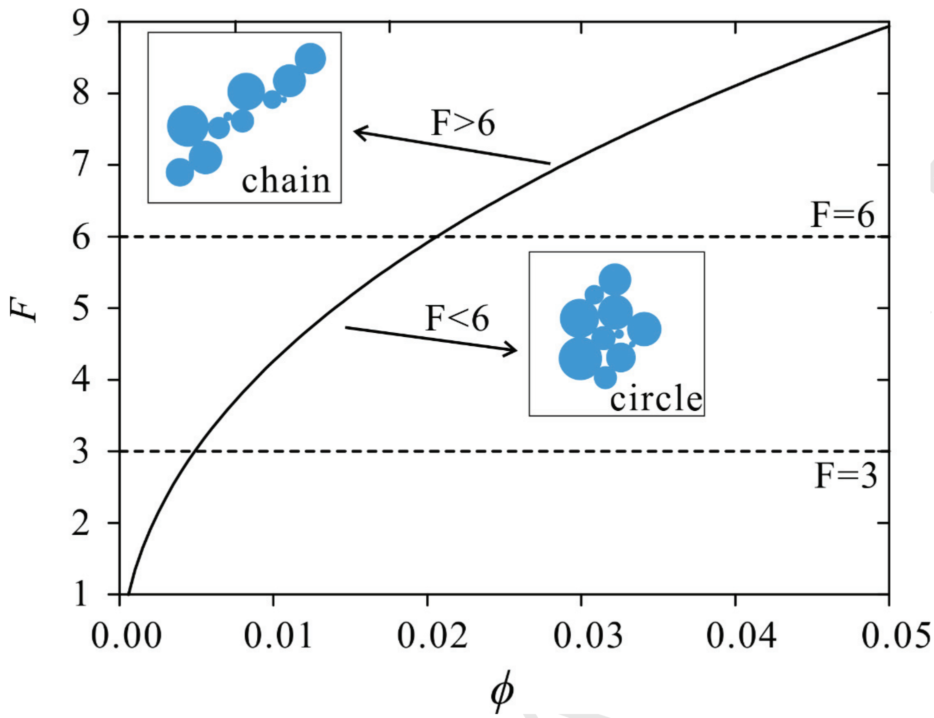

Figure 1. Relationship between F and concentration ϕ in Eq. (11). The dashed line for F=3 and F=6 [34] representing respectively sphere and ellipse for suspended aggregation.

2.2. Consideration of convention effect of nanoparticles

Heat convection due to the Brownian motion of nanoparticles could enhance heat transfer in nanofluids. While most convention models are based on an assumption that suspended aggregation in nanofluids have uniform diameter. Xu et al. [50] theoretically analyzed thermal conductivity k_c for heat convection by using the fractal geometry for different sizes of nanoparticle cluster, which can be expressed as

$$k_c = c\frac{k_f \cdot Nu \cdot d_f}{\Pr}\frac{D(2-D)}{(1-D)^2}\frac{\left(\xi^{1-D}-1\right)^2}{\xi^{2-D}-1}\frac{1}{\overline{\lambda}} \qquad (12)$$

where c is an empirical constant, Nu is the Nusselt number for liquid flowing around a sphere, Pr is the Prandtl number for fluids and d_f is diameter of liquid molecule. Combining Eq. (6) and Eq. (12), the following can be obtained

$$k_c = c\frac{k_f \cdot Nu \cdot d_f}{\Pr}\frac{(2-D)D}{(1-D)^2}\frac{\left(\phi^{\zeta_1}-1\right)^2}{\phi^{\zeta_2}-1}\frac{1}{\bar{\lambda}} \quad (13)$$

where $\zeta_1=(D-1)/(3-D)$. The thermal conductivity for heat convection k_c can express as a complex function of the Prandtl number Pr, the average diameter of aggregation $\bar{\lambda}$, the diameter of molecule of fluids d_f, the concentration ϕ, the Nusselt number Nu and the fractal dimension D. Next section, the model will be simplified and combine Eq. (1) to form a new effective thermal conductivity model with particle aggregation and convection.

2.3. The present fractal thermal conductivity model

In this paper, we assume that the enhancement of thermal conductivity of nanofluids may be caused by aggregation distribution in the liquids and Brownian motion of clustering. Thus, the total dimensionless effective thermal conductivity k_e of nanofluids based on Eqs. (1), (11) and (12) can be written as

$$k_e = \frac{k_s + k_c}{k_f} = \frac{a+(F-1)-(F-1)(1-\alpha)\phi}{a+(F-1)+(1-\alpha)\phi} + c\frac{Nu \cdot d_f}{\Pr}\frac{(2-D)D}{(1-D)^2}\frac{\left(\phi^{\zeta_1}-1\right)^2}{\phi^{\zeta_2}-1}\frac{1}{\bar{\lambda}} \quad (14)$$

Xu et al. [50] found that the values of c is 85.0 both for the Al_2O_3 nanoparticles and for the CuO nanoparticles added in the deionized water, and c equates to 280.0 for the ethylene glycol. The value of c is approximate to be $\bar{\lambda}/d_f$, then Eqs. (14) can be deduced to

$$k_e = \frac{a+(F-1)-(F-1)(1-\alpha)\phi}{a+(F-1)+(1-\alpha)\phi} + \frac{Nu}{\Pr}\frac{(2-D)D}{(1-D)^2}\frac{\left(\phi^{\zeta_1}-1\right)^2}{\phi^{\zeta_2}-1} \quad (15)$$

Eqs. (11) and (15) indicate that the total dimensionless effective thermal conductivity k_e varies with the concentration and fractal dimension for nanoparticle aggregation. In the present model, $Nu \approx 2$ and $\Pr \approx 6.0$ for water at room temperature [50]. Once the concentration ϕ and the fractal dimension D are given/measured, the effective thermal conductivity can be calculated according to Eq. (15).

3. Results and discussion

To our knowledge, the fractal dimension has never been accurately measured to describe thermal conductivity for whole nanofluids. In the following, we therefore evaluate our proposed models (Eqs. (11) and (15)) by fitting experimental measurements, and discuss the relationship between fractal dimension and aggregation shape.

Wang et al. [33] measured the SiO_2/ethanol nanofluids and obtained the fractal dimension equals to 1.57 for nanoparticles when ϕ is about 6.5%. Their model predicted effective thermal conductivity of CuO/water nanofluids could reflect the variation of concentration ϕ qualitatively. The result indicates that the local fractal characteristic represents whole fractal behavior of particles suspensions.

Figure 2 and figure 3 display the present model predictions with the available experimental data. Here the fractal dimension can be obtained by the nonlinear regression method based on Mean Squared Error (MSE) to estimate fitting results. The obtained fractal dimension is 1.572 from fitting to the nanofluids of CuO/water, is very close to the measured fractal dimension, 1.57, by Wang et al [33], which demonstrates the validity of the present model.

Table 1 show that good agreement is found between the predictions of proposed model and experiment results (lower MSE). Figure 2 also clearly indicates that the thermal conductivity of nanofluids increases with the increment of nanoparticles' concentration. It is notable that our proposed model fits better to k_e in the range of 1.1-

1.3 when $0<\phi<0.05$, so the model have not always fitted to lower k_e, such as Al_2O_3/water [54].

In Eq. (11), F is always less than 6 when fractal dimension is larger than a particular value (the value is 1.2 in our model), such as $D=1.693$ for TiO_2/water in table 1. It indicates that the shape of aggregations are near circle, and the increased speed of k_e becomes gradually slow with the increasing concentration. For nanofluids of Al_2Cu/water, the fractal dimension D is approximately 1, which resulting to F>6 and $k_e=2.28$ in smaller concentration (ϕ=0.018). In this situation, aggregation shapes are seem to be behaved as chain and thus play a major role in enhancing heat conduction of nanofluids. Generally, smaller fractal dimension of nanofluids would produce more aggregations of chain shape and enhance heat energy transfer. However, to demonstrate the relationship between fractal dimension and F, more experiments and numerical modeling are needed.

Table 1. Data for calculating the total dimensionless effective thermal conductivity

	k_p (W/m/K)	k_f (W/m/K)	$\bar{\lambda}$ (nm)	D	MSE (%)
CuO/water [33]	32.9	0.613	50.0	1.572	3.70
Al_2Cu/water [55]	418.7	0.613	30.0	1.011	0.00
TiO_2/water [24]	8.5	0.613	15.0	1.693	1.92

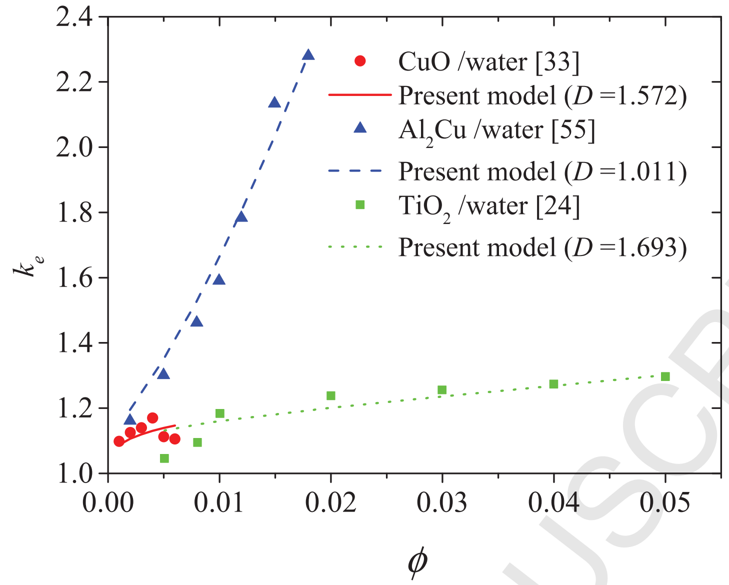

Fig. 2. Comparison between the total dimensionless effective thermal conductivity k_e from fractal model and experimental data in different concentration ϕ.

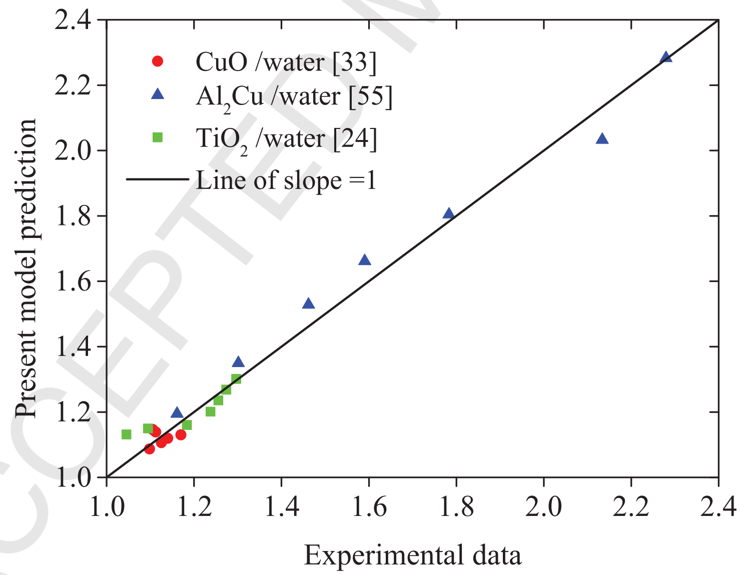

Fig. 3. A comparison of the experimental data with the present model predictions.

4. Conclusions

In this paper, an analytical expression to calculate the thermal conductivity in nanofluids with different space distribution of aggregation is derived base on fractal geometry. The model, which takes into account F in Hamilton and Crosser, is a function of fractal dimension of nanoparticle aggregation and concentration in nanofluids. The effective thermal conductivity calculated based on the developed model provides a good agreement with the experimental results, which validates the validity of the model. The concentration-dependent total dimensionless effective thermal conductivity of three kinds of nanofluids were analyzed. Results show that the fractal dimension may influence the variation of aggregation shape, and more experiment analyses are needed to further quantitatively estimate the influence.

The present study only focus on the effect of particle aggregation and convention for heat conduction mechanism. In the future, more aggregation patterns of nanofluids will be tested and the model will be improved to consider the effect of liquid-layering.

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Figure captions

Fig. 1. Relationship between F and concentration ϕ in Eq. (11). The dashed line for F=3 and F=6 [34] representing respectively sphere and ellipse for suspended aggregation.

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