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# Effective viscosity of nanofluids – A modified Krieger–Dougherty model based on particle size distribution (PSD) analysis



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## ABSTRACT

Nanofluids (colloidal suspensions of nano-sized metallic and non-metallic particles in conventional cooling liquids) are well known for their potential to enhance the thermal transport. Excessive attempts have been made to utilize these nanofluids in heat transfer applications and energy conversion systems, etc. Effective viscosity of nanofluids is a pivotal parameter in determining the flow and heat transfer performance of nanofluids. The prime aim of this work is to develop a new model for effective viscosity of nanofluids. The influences of aggregation and interfacial layer formation have been incorporated into the Krieger–Dougherty (K–D) equation to predict the effective viscosity of nanofluids. This is accomplished by characterizing the clusters based on particle size distribution (PSD) analysis. Furthermore, attention has been paid to showcase the effects of cluster volume fraction, particle diameter and surfactants on effective viscosity of nanofluids. The predicted results are in good agreement with a wide variety of experimental data from literature consisting of different combinations of nanoparticles and basefluid. The accuracy and ease of application of the newly proposed model make it more interesting and useful for practical engineers in design and development of heat transfer systems using nanofluids.

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## 1. Introduction

Recent advances in technology such as miniaturization of devices, elevated power outputs and high operating speeds have made efficient heat transfer as the top priority in many industrial applications. Present day cooling technologies such as increasing the surface area by fins, micro-channel cooling and dual-phase heat transfer techniques have reached their technological limits due to the poor intrinsic thermal conductivity of common coolants. This has led to the search for innovative cooling liquids with superior thermo-physical properties. Recent growth in nanotechnology has led to a new class of coolants referred as 'nanofluids', which has become a hotly debated topic in the domain of heat transfer. Nanofluids are stable suspensions of fine nano-sized metallic/non-metallic particles in traditional coolants. They exhibit higher thermal conductivities than their basefluids and are far more stable than millimeter or micrometer sized solid–liquid suspensions. By virtue of very small size of nanoparticles, nanofluids do not cause erosion, clogging or excessive increase in pumping power. Owing to the above mentioned advantages, nanofluids have proved beneficial in terms of

energy and cost savings in several heat transfer applications [1]. Furthermore, nanofluids find exotic applications in biosciences such as targeted drug delivery and implantable nano-thermal devices [2]. The increase in effective thermal conductivity of nanofluids is also accompanied by corresponding increase in effective viscosity, which is a serious limitation either in terms of elevated pressure drop in forced convective applications or notable fall in fluid motion in case of buoyancy driven flows. Thus, effective viscosity of nanofluids is a crucial property which determines the pumping power and pressure drop, etc. in flow and heat transfer applications. In overall, clear understanding and precise prediction of viscosity of nanofluids are inevitable for determining the thermo-fluidic behavior and design of systems which involve nanofluid flow and heat transfer.

The field of rheology of nanofluids has attracted a considerable volume of research and numerous experimental works on influence of particle geometry, volume fraction and temperature, etc. on effective viscosity of nanofluids have been reported in literature [3–23]. In general, nanofluids show Newtonian behavior except for few exceptions such as that of the non-Newtonian shear thinning behavior reported by Tseng and Lin [8]. These anomalies can be attributed to the higher volume fractions or very small diameter of nanoparticles. The ratio of effective viscosity of the nanofluid to the viscosity of basefluid is known as the effective viscosity ratio and it increases with increase in particle volume fraction. Exponentially

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**Nomenclature**

PSD	particle size distribution
$\mu_{bf}$	viscosity of basefluid [kg/ms]
$\mu_{eff}$	effective viscosity [kg/ms]
$[\eta]$	intrinsic viscosity[kg/ms]
$\phi$	initial particle volume fraction
$\phi_m$	maximum particle volume fraction
$\phi_{cs}$	volume fraction of cluster spheres
$\phi_c$	volume fraction of clusters in nanofluid
$\phi_{ins}$	volume fraction of nanoparticles in a cluster
$\phi_{ecs}$	effective volume fraction of cluster spheres
$d_p$	diameter of primary nanoparticles [m]
$d$	diameter of cluster/aggregate [m]
$d_c$	average diameter of cluster/aggregate [m]
$P$	number percentage of clusters
$n$	number of nanoparticles in a cluster
$s$	maximum packaging density of a cluster
$\omega$	relaxation factor
$\delta$	thickness of interfacial layer [m]
$\beta$	thickness ratio of interfacial layer

high increase in effective viscosity ratios were reported in literature for nanofluids containing very small nanoparticles [23]. This can be attributed to the non-Newtonian behavior of nanofluids containing very small nanoparticles even at low particle volume fractions as observed by Pak and Cho [4]. In general, we can say that the effective viscosity of nanofluids are higher than that of the basefluids and smaller nanoparticles lead to higher increase in effective viscosity of nanofluids. In addition to particle volume fraction and primary nanoparticle diameter, systematic experimental studies on rheology of nanofluids indicate that parameters such as fabrication methods, surfactants, particle clustering, shear rate, sonication and time after sonication are also critical in determining the effective viscosity of nanofluids [24–26]. Recent numerical studies of Sheikholeslami et al. [27–33] reveal that rheological behavior of nanofluids is also influenced by electric and magnetic fields. Adding to the experimental and numerical studies, some theoretical works on prediction of effective viscosity of nanofluids have also been reported in literature. A detailed review of these theoretical and empirical correlations for effective viscosity of nanofluids has been well documented in literature [34–36]. The traditional models which were originally developed for micro and millimeter sized particles suspended in basefluids are poor in predicting the effective viscosity of nanofluids. The conventional models consider only the influence of particle volume fraction, whereas, experimental investigations on viscosity of nanofluids show that the average size of nanoparticles/clusters present in nanofluids is more critical for a precise prediction of effective viscosity [37]. The number of theoretical attempts on predicting the effective viscosity of nanofluids is very limited and there is still a discrepancy prevailing among the experimental data and theoretical predictions of effective viscosity of nanofluids. In this framework, a simple yet comprehensive model to predict the effective viscosity of nanofluids has been proposed in this work. Present model is built by incorporating the effects of aggregation and interfacial layering into Krieger–Dougherty model for effective viscosity of solid–liquid suspensions. The uniqueness and simplicity of the model lies in the fact, that the aggregates in nanofluids are characterized using PSD analysis. The model proposed in this work will be useful and interesting to practical engineers in design of systems involving flow and heat transfer of nanofluids.

**2. Theoretical background**

Theoretical works on colloidal suspensions is a frequently visited and well documented topic in literature [38–43]. Since the pioneering work of Einstein [44] on analysis of infinitely dilute suspensions of hard solid spheres, several works on rheology of colloidal suspensions were reported in literature. Einstein's works considered the particles to be rigid, uncharged and devoid of any attractive forces. The suspension is very dilute such that the disturbance caused by one particle does not interact with that caused by another particle. In these circumstances, a particle is expected to move with the velocity of the streamline and in line with the particle center as depicted in Fig. 1 (a). The velocity of fluid on the upper side of the particle is higher than that in the bottom side. This difference in velocity leads to a rotational motion of the particle, which is referred as the vorticity of the shear field and leads to the following dispersion viscosity:

$$\frac{\mu_{eff}}{\mu_{bf}} = 1 + [\eta]\phi + O(\phi^2). \quad (1)$$

In the above equation,  $[\eta]$  is the intrinsic viscosity which is typically equal to 2.5 for a suspension of hard spheres of uniform size [37]. This equation is valid when particle volume fraction ( $\phi$ ) is less than or equal to 0.01. Brinkman [45] improved the Einstein's model by accounting for the hydrodynamic interactions and proposed the following relation:

$$\frac{\mu_{eff}}{\mu_{bf}} = \frac{1}{(1 - \phi)^{2.5}}. \quad (2)$$

At higher volume fractions ( $\phi > 0.01$ ), as the disturbance from one particle will interact with that of the neighboring particles, it is essential to consider the hydrodynamic interactions due to Brownian motion among particles. The viscosity in this condition is described by Batchelor [46] as

$$\frac{\mu_{eff}}{\mu_{bf}} = 1 + 2.5\phi + 6.2\phi^2 + O(\phi^3). \quad (3)$$

Above equation of Batchelor [46] is valid for flows which are influenced by single particles and pair-particle micro-structures (Fig. 1 (b)), where the particle volume fraction is usually less than or equal to 0.1. At particle volume fractions greater than 0.1, multi-particle collisions as shown in Fig. 1 (c), begin to dominate and hence higher order terms in the order of 3 have to be taken into account. Krieger and Dougherty [47] proposed a semi-empirical correlation for effective viscosity of solid-liquid suspensions which is valid for full range of particle volume fraction as follows:

$$\frac{\mu_{eff}}{\mu_{bf}} = \left(1 - \frac{\phi}{\phi_m}\right)^{-[\eta]\phi_m}. \quad (4)$$

In Eq. (4),  $\phi_m$  is the maximum particle volume fraction at which the flow can occur. The value of  $\phi_m$  is usually taken as 0.605 [47]. The theoretical correlations discussed above were successful in explaining the effective viscosity of micro/millimeter sized suspensions in the respective volume fraction ranges (see Fig. 1 (a), (b) and (c)). However, experimental studies on effective viscosity of nanofluids reveal that the rheological behavior of nanofluids is more governed by the aggregates; as the nanoparticles in nanofluids mostly exist as aggregates (Fig. 1 (d)) even at very small volume fractions [37,48]. Assuming that the hydrodynamic forces are weak to break the clusters into individual primary nanoparticles but, only aid in formation

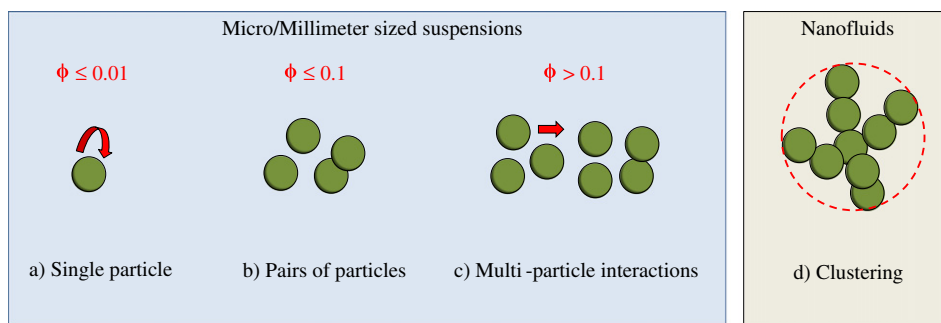


Fig. 1. Interactions of particles in a shear flow.

of clusters, Chen et al. [20] proposed the following correlation for effective viscosity of nanofluids:

$$\frac{\mu_{eff}}{\mu_{bf}} = \left(1 - \frac{\phi_{cs}}{\phi_m}\right)^{[\eta]\phi_m} \quad (5)$$

where,  $\phi_{cs}$  is the volume fraction of clusters which are enclosed in an imaginary sphere which form the flow units. Chen et al. [20] determined  $\phi_{cs}$  based on fractal theory. Similar to Chen et al. [20], recently some equations/correlations have been proposed for effective viscosity of nanofluids. But, these correlations are developed based on certain conditions (such as for a given particle diameter and volume fraction) and are valid only for those conditions. For instance, Chen et al.'s [20,21] model is developed based on the experimental data of TiO<sub>2</sub>-EG nanofluids with primary nanoparticle diameter equal to 25 nm. Maiga et al. [49] proposed a correlation for effective viscosity of nanofluids based on the data reported by Wang et al. [5] for Al<sub>2</sub>O<sub>3</sub>-H<sub>2</sub>O nanofluids. Similarly, Corcione [37] proposed a semi-empirical model which is based on regression analysis of Chevalier et al.'s [22] experimental results for ethanol based silica nanofluids. A comparison of predictions of Chen et al.'s [20] and Maiga et al.'s [49] models with the models based on conventional theories is shown in Fig. 2. It is noted that the models based on conventional theories fail notably for nanofluids. Furthermore, the predictions of models of Chen et al. [20] and Maiga et al. [49] vary significantly with each other. Thus, Fig. 2 clearly reveals the uncertainty prevailing among the models for effective viscosity of nanofluids. This is a clear indication that a reliable model which take into account the size distribution of particles is essential for predicting the effective viscosity of nanofluids.

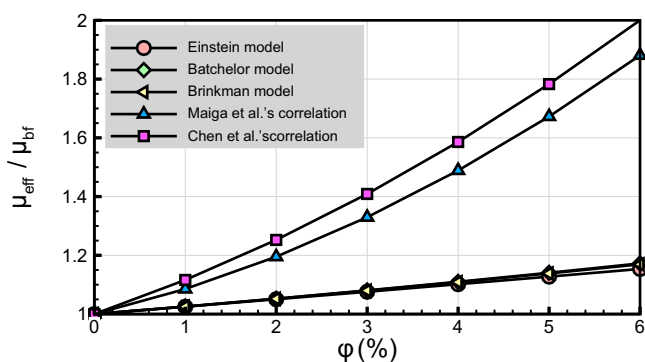


Fig. 2. Comparison of Chen et al.'s [20] and Maiga et al.'s [49] correlations with conventional models of Einstein [44], Batchelor [46] and Brinkman [45].

### 3. Development of new model

Above literature review reveals that the theoretical models proposed specifically for nanofluids are very limited in number. Moreover, those models which are specific for nanofluids are bounded within certain conditions and are dependent on fractal theory to characterize the clusters. According to fractal theory, the cluster morphology or structure is characterized by the average radius of gyration. The average radius of gyration of clusters is a function of fractal and chemical dimensions of the aggregates/clusters. The clusters are considered to be composed of a linear chain known as backbone and side chains called as dead ends. Fractal dimension is a primary parameter that describes the cluster morphology and it is influenced by numerous factors. Hence, fractal theory which is a completely theoretical method is based on numerous assumptions and a precise calculation of average cluster diameter is very difficult [50]. Fractal theory assumes that the clusters in nanofluids are of uniform size [51]. But in practice, the clusters in nanofluids are polydisperse in nature [52]. A suitable choice of cluster diameter is very essential to precisely predict the effective thermophysical properties of nanofluids. Studies of Song et al. [24,25] have clearly showcased the importance of a precise calculation of cluster dimension in predicting the effective properties of nanofluids. To overcome the limitations of fractal theory based models, size and volume fraction of clusters in nanofluids can be determined using particle size distribution (PSD) analysis. PSD of nanofluids can be easily obtained using dynamic light scattering (DLS) experiments and it provides all required information to characterize the clusters [53]. A classical study by Farris [54] also suggests that the PSD of colloidal suspensions is much more dominant in determining the effective viscosity. Recently, a theoretical study by Song et al. [26] revealed that predicting the hydrodynamic and optical properties of nanofluids using PSD is much more precise and exhibit better agreement with experimental data than the fractal theory based methods. In this work, the Krieger–Dougherty model has been modified for nanofluids considering the effects of particle clustering and interfacial layer formation. The cluster volume fraction ( $\phi_{cs}$ ) and average diameter ( $d_c$ ) are calculated using PSD analysis. The change in volume fraction of cluster spheres due to interfacial layer formation is also taken into account. The detailed derivation of the new model is described in the following subsections.

#### 3.1. Characterization of clusters using PSD analysis

A schematic diagram of particle size distribution in a nanofluid is shown in Fig. 3. Particle size distribution (PSD) is an index (means of expression) indicating the sizes of particles and their corresponding proportions (relative particle amount as a percentage where the total amount of particles is 100%) in the suspension to be measured. In nanofluids, the clusters formed are considered to be enclosed by

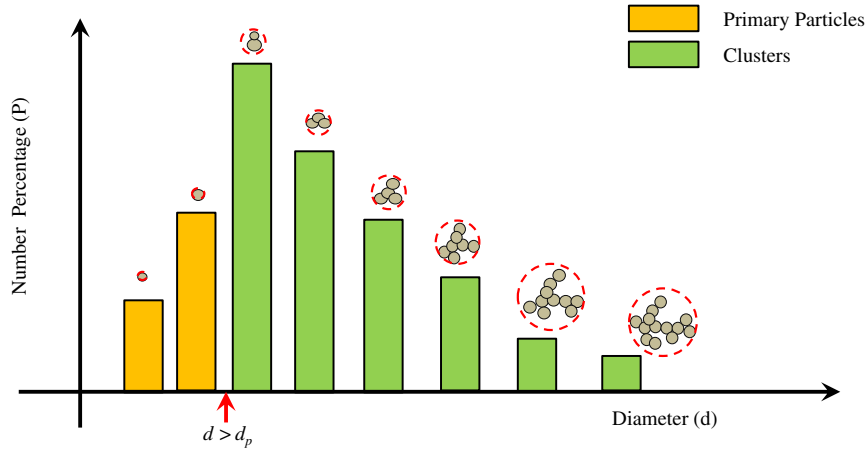


Fig. 3. Schematic diagram of PSD data for nanofluids.

an imaginary sphere with diameter ( $d$ ). In Fig. 3,  $P$  is the number percentage of corresponding particles of diameter ( $d$ ). The cluster size distribution vary in a wide range and hence, the average diameter of the clusters ( $d_c$ ) is calculated from the PSD data. Before that, it is essential to define a cluster. Any particle that is larger than the diameter of primary nanoparticles ( $d_p$ ) is taken as a cluster. The average diameter of the cluster ( $d_c$ ) and the volume fraction of cluster spheres ( $\phi_{cs}$ ) can be calculated from the PSD data as given below [55,56]

$$d_c = \frac{\sum_{d \geq d_p} (P \times d)}{\sum_{d \geq d_p} P} \tag{6}$$

The volume fraction of total clusters in the nanofluid is given by

$$\phi_c = \phi_{cs} \phi_{ins} \tag{7}$$

In the above equation,  $\phi_{cs}$  is the volume fraction of corresponding cluster spheres and  $\phi_{ins}$  represent the volume fraction of primary nanoparticles present in cluster spheres.  $\phi_{cs}$  and  $\phi_{ins}$  are calculated as [56]

$$\phi_{cs} = \frac{\sum_{d \geq d_p} (P \times d^3)}{\sum_{d \geq 0} (P \times d^3)} \phi \tag{8}$$

and

$$\phi_{ins} = \frac{d_p^3 n}{d_c^3} \tag{9}$$

In Eq. (9),  $n$  indicates the total number of primary nanoparticles in a single cluster sphere with average diameter ( $d_c$ ) and is estimated as [55]

$$n = \frac{d_c^3}{d_p^3} s \omega \tag{10}$$

The maximum packaging density of the cluster sphere according to Kepler juncture is given as

$$s = \pi / \sqrt{18} \tag{11}$$

In Eq. (10),  $\omega$  is a relaxation factor to indicate the particle packing condition. Average random packing density of a cluster sphere is given by the product of  $s$  and  $\omega$ . Review of related literature indicates that the random packing density of a cluster sphere is approximately 63% [57]. Hence, the relaxation factor  $\omega$  is considered as 0.85, so that the average random packing density is 63%. For later analysis, we introduce a parameter ( $f$ ), which is defined as

$$f = \frac{\sum_{d \geq d_p} (P \times d^3)}{\sum_{d \geq 0} (P \times d^3)} \tag{12}$$

This parameter  $f$  varies from 0 to 1 and indicates the volume fraction of clusters present in the nanofluid system. If  $f = 0$ , all the nanoparticles in nanofluid are completely mono-dispersed with no aggregation and  $f = 1$  indicates that all the nanoparticles have formed clusters and no individual primary nanoparticles are present in the nanofluid.

### 3.2. Calculation of effective volume fraction due to interfacial layer formation

In Eq. (8), the volume fraction of cluster spheres ( $\phi_{cs}$ ) has been calculated using the PSD data. The clusters present in nanofluids consist of closely aggregated nanoparticles into a sphere. It has been experimentally proved that the liquid molecules closer to the solid surface arrange themselves in an orderly fashion at the solid–liquid interface [58]. This ordered layer of liquid molecules behave like a solid and result in increase of effective volume fraction of cluster spheres. Hence, it is essential to calculate the effective volume fraction of cluster spheres ( $\phi_{ecs}$ ) to determine the effective viscosity of nanofluids. The thickness of interfacial layer ( $\delta$ ) is calculated by Hashimoto et al. [59] based on electron density profile as follows:

$$\delta = \sqrt{2\pi} \sigma \tag{13}$$

Here,  $\sigma$  is a parameter which indicates the diffusiveness of interfacial boundary and it varies 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. [58] and molecular dynamic simulations by Xue et al. [60] show that the interfacial layer will be in the order of few atomic distances, which is approximately 1 nm. Hence in this work, the thickness of

**Table 1**

Details of experimental data for effective viscosity of nanofluids from literature, considered for comparison with model predictions.

S. no.	Sample	Details	$\phi$ (%)	$d_{pm}$ (nm)	$\mu_{bf}$ (kg/ms)	Reference
1	S1	CaCO <sub>3</sub> -H <sub>2</sub> O	0.1200	35 nm	0.00089	Zhu et al. [62]
2	S2	ZnO-H <sub>2</sub> O	1.0000	30 nm	0.00089	Jeong et al. [63]
3	S3	TiO <sub>2</sub> -H <sub>2</sub> O	0.0300	21 nm	0.00089	Bobbo et al. [64]
4	S4	Al <sub>2</sub> O <sub>3</sub> -H <sub>2</sub> O	0.5000	50 nm	0.00089	Anoop et al. [65]
5	S5	ZnO-ethylene glycol	1.0000	50 nm	0.01620	Kole et al. [66]
6	S6	ZnO-ethylene glycol	3.0000	50 nm	0.01620	Kole et al. [66]
7	S7	ZnO-ethylene glycol	3.7500	50 nm	0.01620	Kole et al. [66]

interfacial layer is considered to be 1 nm. The effective volume fraction of cluster spheres due to interfacial layer formation is calculated as [61]

$$\phi_{ecs} = \phi_{cs}(1 + \beta)^3 \quad (14)$$

where,  $\phi_{cs}$  is the volume fraction of cluster spheres without interfacial layer formation and  $\beta = 2\delta/d_c$  is the thickness ratio of the interfacial layer to average cluster radius. From the above PSD analysis, the effective volume fraction of cluster spheres ( $\phi_{ecs}$ ) present in the nanofluid can be estimated by Eq. (14). Hence, effective viscosity of nanofluids can be calculated by substituting the expression for effective volume fraction of clusters ( $\phi_{ecs}$ ) in Eq. (5) as follows:

$$\frac{\mu_{eff}}{\mu_{bf}} = \left(1 - \frac{\phi_{ecs}}{\phi_m}\right)^{[\eta]\phi_m} \quad (15)$$

Eq. (15) explains the effective viscosity of nanofluids considering the phenomena of particle clustering and interfacial layer formation seen in nanofluids. The present model is valid though all particle volume fraction ranges as it is built upon the K-D equation (Eq. (4)) and is based on real time PSD data.

#### 4. Results and discussion

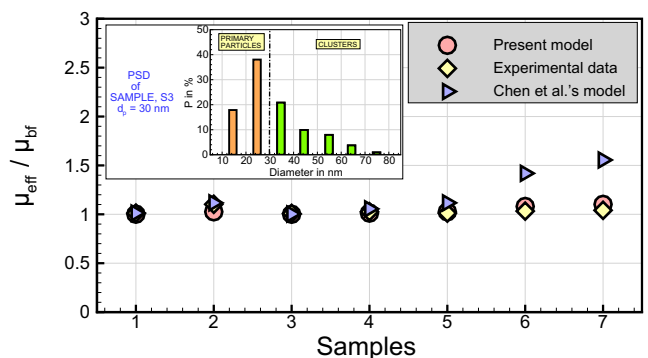
In this work, a new model for effective viscosity of nanofluids has been proposed considering the effects of particle clustering and interfacial layer formation. The clusters in nanofluids are characterized based on PSD analysis. PSD data is used to calculate the average diameter ( $d_c$ ) of the clusters and volume fraction of cluster spheres ( $\phi_{cs}$ ). In addition to that, the effective volume fraction of cluster spheres ( $\phi_{ecs}$ ) due to interfacial layer formation is also taken into account. As the present model is built upon K-D equation which is valid for all ranges of volume fractions [47], this model is also valid at all volume fraction ranges.

##### 4.1. Validation and analysis of the present model

The new model proposed in this work is validated by comparing the model predictions with experimental results for effective viscosity ratio of nanofluids from literature. The model predictions are compared with a wide variety of data for different nanofluids consisting of different sizes of nanoparticles and with different particle loadings. The details of the experimental data such as the particle diameter, volume fraction, viscosity of the basefluid; taken for comparison are clearly presented in Table 1. The particle size distribution of sample S3 is shown in the inset image of Fig. 4. The comparison reveals that the model predictions are closely matching with the

experimental data for effective viscosity of nanofluids from literature. The number of samples considered for comparison is restricted by the availability of PSD data and effective viscosity measured independently at same instant of time. However, it is to be noted that the samples considered in this study accommodate different particle diameters and volume fractions. Furthermore, predictions of Chen et al.'s model [20] are also presented in Fig. 4. The percentage variation between the present model prediction and the experimental data is found to be less than 8% for all samples. Whereas, the percentage variation between Chen et al.'s model [20] and the experimental data reaches upto 33%. Thus, it is clear that the present model based on PSD analysis is more precise and consistent than the fractal theory based model. It is necessary to mention that the present model is sensitive to the experimental errors involved in the measurement of PSD data of nanofluids. However, as shown in Fig. 2, there is an uncertainty prevailing among the classical models and recent models based on fractal theory for effective viscosity of nanofluids. This uncertainty will be rectified by the present model which is based on real time particle size distribution (PSD) analysis.

It is a well known fact that the effective viscosity of nanofluids is significantly influenced by particle clustering. Fig. 5 shows the influence of volume fraction of cluster spheres present in the nanofluid on effective viscosity calculated by the present model. The parameter  $f$  defined in Section 3.1, indicates the volume fraction of cluster spheres present in the nanofluid. The volume fraction of clusters present in nanofluid is the driving factor which determines the increase in effective viscosity ratio of nanofluids. The value of  $f$  varies from 0 to 1. The percentage of cluster spheres are higher when the value of  $f$  is high.  $f = 0$  indicates that there are no clusters present in the nanofluid and all the nanoparticles are completely mono-dispersed. If  $f = 1$ , all the nanoparticles in the nanofluid have formed clusters. The variation of effective viscosity ratio of nanofluids with the parameter  $f$  varying from 0 to 1 has been



**Fig. 4.** Comparison of predictions of present model and Chen et al.'s model [20] with experimental data from literature.

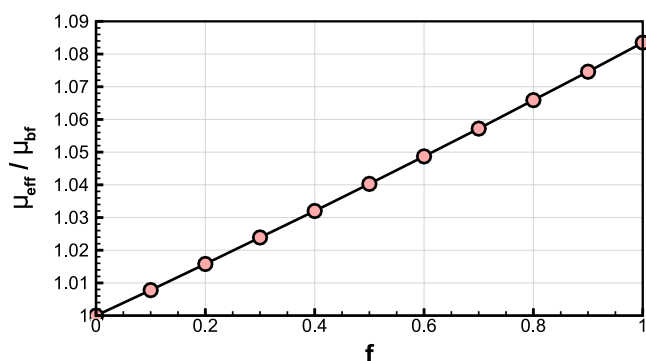


Fig. 5. Effect of volume fraction of clusters on effective viscosity ratio of nanofluids.

pictorially shown in Fig. 5. It can be clearly seen that the effective viscosity ratio of nanofluid increases with an increase in particle clustering which matches with the experimental observations reported in literature [4,67].

In addition to particle clustering, the effective viscosity of nanofluids is also influenced by several other parameters such as nanoparticle diameter and surfactants. Present model is good enough to capture the effects of nanoparticle diameter and surfactants on effective viscosity of nanofluids. The ability of the model to predict the fall in effective viscosity with increase in particle diameter is showcased by comparing the model predictions with experimental data of Pastoriza et al. [18] in Fig. 6. Two samples of CuO–Water nanofluid with 0.5% volume fraction with particle diameters 11 nm and 30 nm, respectively are taken into account. Experimental results revealed a decrease in effective viscosity with increase in particle diameter. The model is also capable of capturing this decrease in effective viscosity with increase in particle diameter with less than 6% deviation from the experimental data.

The effective viscosity of nanofluids is also affected by the type of surfactants used. The model is capable of capturing the effect of surfactants used on effective viscosity, by means of the PSD data. The model predictions are compared with experimental data for effective viscosity ratio of two samples of TiO<sub>2</sub>–H<sub>2</sub>O nanofluid with primary particle diameter 21 nm, prepared by Das et al. [68]. In their study, one sample was stabilized using cetyl trimethyl ammonium bromide (CTAB) and the other was stabilized using acetic acid (AA). Due to the influence of surfactants, both the samples exhibited different effective viscosity ratios. The model is able to predict

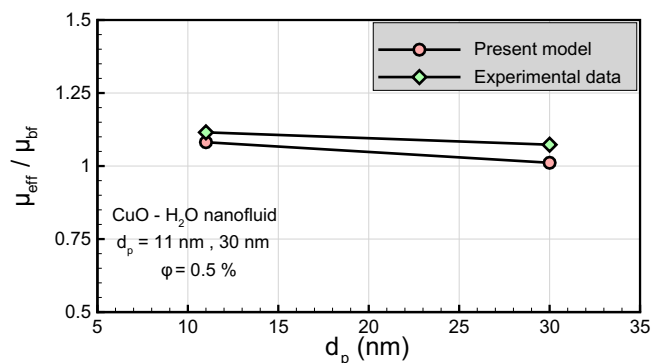


Fig. 6. Effect of primary particle diameter on effective viscosity ratio of nanofluids predicted by the present model compared with experimental data from Pastoriza et al. [18].

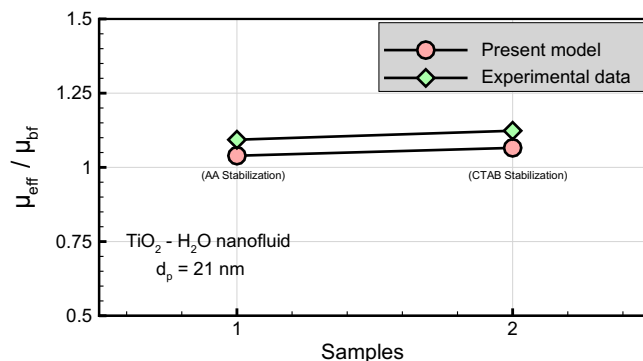


Fig. 7. Effect of surfactant on effective viscosity ratio of nanofluids predicted by the present model compared with experimental data from Das et al. [68].

the change in effective viscosity due to different stabilizing agents with a deviation of less than 8% from the experimental data (refer Fig. 7). Furthermore, the effective viscosity ratio of nanofluids is also affected by several other parameters such as temperature, physicochemical properties of constituents, sonication time and time after sonication. Present model characterizes the cluster size and volume fraction based on PSD data which is dependent on all above mentioned factors. Thus, we believe that the present model will be capable of predicting the effects of all above factors based on the PSD data obtained experimentally by dynamic light scattering (DLS) experiments.

## 5. Conclusions

A simple yet comprehensive model has been proposed to explain the effective viscosity of nanofluids based on particle size distribution (PSD) analysis. The model considers the effects of aggregation and interfacial layer formation on effective viscosity of nanofluids. The model is built upon the Krieger–Dougherty (K–D) model for effective viscosity of solid–liquid suspensions. The influence of aggregation and interfacial layer formation has been incorporated into the K–D model to make it applicable for nanofluids. The model predictions are closely matching with the experimental data for effective viscosity of nanofluids taken from literature. The model is also able to capture the effects of nanoparticle diameter and stabilizing agents on effective viscosity of nanofluids. By means of PSD data, the model is also capable of addressing the effects of several parameters such as particle size, temperature, surfactants, pH, physicochemical properties of nanoparticles and basefluids, sonication time and time after sonication. Hence, the model will be useful in explaining the contradictions in effective viscosity data of nanofluids available in literature. The PSD data obtained experimentally will provide sufficient information to characterize the clusters in nanofluids and is more practical than the characterization based on fractal theory. Thus, the present model will be interesting and useful to practical engineers in design and development of heat transfer and energy conversion systems using nanofluids.

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## References

- [1] D. Wen, G. Lin, S. Vafaei, K. Zhang, Review of nanofluids for heat transfer applications, *Particuology* 7 (2) (2009) 141–150.
- [2] R.S. Shawgo, A.C.R. Grayson, Y. Li, M.J. Cima, Biomems for drug delivery, *Curr. Opin. Solid State Mater. Sci.* 6 (4) (2002) 329–334.
- [3] H. Masuda, A. Ebata, K. Teramae, Alteration of thermal conductivity and viscosity of liquid by dispersing ultra-fine particles. dispersion of  $\text{Al}_2\text{O}_3$ ,  $\text{SiO}_2$  and  $\text{TiO}_2$  ultra-fine particles, *NETSU BUSSEI* 07 (04) (1993) 227–233.
- [4] B.C. Pak, Y.I. Cho, Hydrodynamic and heat transfer study of dispersed fluids with submicron metallic oxide particles, *Experimental Heat Transfer an International Journal* 11 (2) (1998) 151–170.
- [5] X. Wang, X. Xu, S.U.S. Choi, Thermal conductivity of nanoparticle–fluid mixture, *J. Thermophys. Heat Transf.* 13 (4) (1999) 474–480.
- [6] S.K. Das, N. Putra, W. Roetzel, Pool boiling characteristics of nano-fluids, *Int. J. Heat Mass Transf.* 46 (5) (2003) 851–862.
- [7] N. Putra, W. Roetzel, S.K. Das, Natural convection of nano-fluids, *Heat Mass Transf.* 39 (8–9) (2003) 775–784.
- [8] W.J. Tseng, K.-C. Lin, Rheology and colloidal structure of aqueous  $\text{TiO}_2$  nanoparticle suspensions, *Mater. Sci. Eng. A* 355 (1) (2003) 186–192.
- [9] C. Nguyen, F. Desgranges, G. Roy, N. Galanis, T. Maré, S. Boucher, H.A. Mintsa, Temperature and particle-size dependent viscosity data for water-based nanofluids-hysteresis phenomenon, *Int. J. Heat Fluid Flow* 28 (6) (2007) 1492–1506.
- [10] C. Nguyen, F. Desgranges, N. Galanis, G. Roy, T. Maré, S. Boucher, H.A. Mintsa, Viscosity data for  $\text{Al}_2\text{O}_3$ –water nanofluid hysteresis: is heat transfer enhancement using nanofluids reliable? *Int. J. Therm. Sci.* 47 (2) (2008) 103–111.
- [11] Y. He, Y. Jin, H. Chen, Y. Ding, D. Cang, H. Lu, Heat transfer and flow behaviour of aqueous suspensions of  $\text{TiO}_2$  nanoparticles (nanofluids) flowing upward through a vertical pipe, *Int. J. Heat Mass Transf.* 50 (11) (2007) 2272–2281.
- [12] J.-H. Lee, K.S. Hwang, S.P. Jang, B.H. Lee, J.H. Kim, S.U. Choi, C.J. Choi, Effective viscosities and thermal conductivities of aqueous nanofluids containing low volume concentrations of  $\text{Al}_2\text{O}_3$  nanoparticles, *Int. J. Heat Mass Transf.* 51 (11) (2008) 2651–2656.
- [13] S. Murshed, K. Leong, C. Yang, Investigations of thermal conductivity and viscosity of nanofluids, *Int. J. Therm. Sci.* 47 (5) (2008) 560–568.
- [14] S.S. Murshed, S.-H. Tan, N.-T. Nguyen, Temperature dependence of interfacial properties and viscosity of nanofluids for droplet-based microfluidics, *J. Phys. D: Appl. Phys.* 41 (8) (2008) 085502.
- [15] T.X. Phuoc, M. Massoudi, Experimental observations of the effects of shear rates and particle concentration on the viscosity of  $\text{Fe}_2\text{O}_3$ -deionized water nanofluids, *Int. J. Therm. Sci.* 48 (7) (2009) 1294–1301.
- [16] W. Duangthongsuk, S. Wongwises, Measurement of temperature-dependent thermal conductivity and viscosity of  $\text{TiO}_2$ –water nanofluids, *Exp. Thermal Fluid Sci.* 33 (4) (2009) 706–714.
- [17] Z. Jia-Fei, L. Zhong-Yang, N. Ming-Jiang, C. Ke-Fa, Dependence of nanofluid viscosity on particle size and pH value, *Chin. Phys. Lett.* 26 (6) (2009) 066202.
- [18] M. Pastoriza-Gallego, C. Casanova, R. Páramo, B. Barbés, J. Legido, M. Piñeiro, A study on stability and thermophysical properties (density and viscosity) of  $\text{Al}_2\text{O}_3$  in water nanofluid, *J. Appl. Phys.* 106 (6) (2009) 064301.
- [19] R. Prasher, D. Song, J. Wang, P. Phelan, Measurements of nanofluid viscosity and its implications for thermal applications, *Appl. Phys. Lett.* 89 (13) (2006) 133108.
- [20] H. Chen, Y. Ding, C. Tan, Rheological behaviour of nanofluids, *New J. Phys.* 9 (10) (2007) 367.
- [21] H. Chen, Y. Ding, Y. He, C. Tan, Rheological behaviour of ethylene glycol based titania nanofluids, *Chem. Phys. Lett.* 444 (4) (2007) 333–337.
- [22] J. Chevalier, O. Tillement, F. Ayela, Rheological properties of nanofluids flowing through microchannels, *Appl. Phys. Lett.* 91 (23) (2007) 3103.
- [23] P.K. Namburu, D.P. Kulkarni, D. Misra, D.K. Das, Viscosity of copper oxide nanoparticles dispersed in ethylene glycol and water mixture, *Exp. Thermal Fluid Sci.* 32 (2) (2007) 397–402.
- [24] D. Song, J. Zhou, Y. Wang, D. Jing, Choice of appropriate aggregation radius for the descriptions of different properties of the nanofluids, *Appl. Therm. Eng.* 103 (2016) 92–101.
- [25] D. Song, Y. Wang, D. Jing, J. Geng, Investigation and prediction of optical properties of alumina nanofluids with different aggregation properties, *Int. J. Heat Mass Transf.* 96 (2016) 430–437.
- [26] D. Song, D. Jing, J. Geng, Y. Ren, A modified aggregation based model for the accurate prediction of particle distribution and viscosity in magnetic nanofluids, *Powder Technol.* 283 (2015) 561–569.
- [27] M. Sheikholeslami, M. Rashidi, T. Hayat, D. Ganji, Free convection of magnetic nanofluid considering MFD viscosity effect, *J. Mol. Liq.* 218 (2016) 393–399.
- [28] M. Sheikholeslami, K. Vajravelu, M.M. Rashidi, Forced convection heat transfer in a semi annulus under the influence of a variable magnetic field, *Int. J. Heat Mass Transf.* 92 (2016) 339–348.
- [29] M. Sheikholeslami, R. Ellahi, Three dimensional mesoscopic simulation of magnetic field effect on natural convection of nanofluid, *Int. J. Heat Mass Transf.* 89 (2015) 799–808.
- [30] M. Sheikholeslami, M. Rashidi, D. Ganji, Numerical investigation of magnetic nanofluid forced convective heat transfer in existence of variable magnetic field using two phase model, *J. Mol. Liq.* 212 (2015) 117–126.
- [31] M. Sheikholeslami, A.J. Chamkha, Flow and convective heat transfer of a ferro-nanofluid in a double-sided lid-driven cavity with a wavy wall in the presence of a variable magnetic field, *Numerical Heat Transfer, Part A: Applications* 69 (10) (2016) 1186–1200.
- [32] M. Sheikholeslami, A.J. Chamkha, Electrohydrodynamic free convection heat transfer of a nanofluid in a semi-annulus enclosure with a sinusoidal wall, *Numerical Heat Transfer, Part A: Applications* 69 (7) (2016) 781–793.
- [33] M. Sheikholeslami, D. Ganji, Influence of magnetic field on  $\text{CuO}$ – $\text{H}_2\text{O}$  nanofluid flow considering Marangoni boundary layer, *Int. J. Hydrog. Energy* (2016).
- [34] I. Mahbubul, R. Saidur, M. Amalina, Latest developments on the viscosity of nanofluids, *Int. J. Heat Mass Transf.* 55 (4) (2012) 874–885.
- [35] P.C. Mishra, S. Mukherjee, S.K. Nayak, A. Panda, A brief review on viscosity of nanofluids, *International Nano Letters* 4 (4) (2014) 109–120.
- [36] J.P. Meyer, S.A. Adio, M. Sharifpur, P.N. Nwosu, The viscosity of nanofluids: a review of the theoretical, empirical, and numerical models, *Heat Transfer Eng.* 37 (5) (2016) 387–421.
- [37] M. Corcione, A semi-empirical model for predicting the effective dynamic viscosity of nanoparticle suspensions, *Heat Transfer Eng.* 33 (7) (2012) 575–583.
- [38] W. Russel, D. Saville, W. Schowalter, *Colloidal dispersions* (Cambridge Monographs on Mechanics and Applied Mathematics), 1989.
- [39] T. Chow, Viscosities of concentrated dispersions, *Phys. Rev. E* 48 (3) (1993) 1977.
- [40] C.J. Petrie, The rheology of fibre suspensions, *J. Non-Newtonian Fluid Mech.* 87 (2) (1999) 369–402.
- [41] J.W. Goodwin, R.W. Hughes, *Rheology for Chemists: An Introduction*, Royal Society of Chemistry, 2008.
- [42] R.G. Larson, The rheology of dilute solutions of flexible polymers: progress and problems, *J. Rheol.* 49 (1) (2005) 1–70.
- [43] I.M. Abdulagatov, N.D. Azizov, Experimental study of the effect of temperature, pressure and concentration on the viscosity of aqueous NaBr solutions, *J. Solut. Chem.* 35 (5) (2006) 705–738.
- [44] A. Einstein, Berichtigung zu meiner arbeit:eine neue bestimmung der moleküldimensionen (correction of my work: a new determination of the molecular dimensions), *Ann. Phys.* 339 (3) (1911) 591–592.
- [45] H. Brinkman, The viscosity of concentrated suspensions and solutions, *J. Chem. Phys.* 20 (4) (1952) 571–571.
- [46] G. Batchelor, The effect of Brownian motion on the bulk stress in a suspension of spherical particles, *J. Fluid Mech.* 83 (01) (1977) 97–117.
- [47] I.M. Krieger, T.J. Dougherty, A mechanism for non-Newtonian flow in suspensions of rigid spheres, *Trans. Soc. Rheol.* 3 (1) (1959) 137–152.
- [48] D. Song, M. Hatami, Y. Wang, D. Jing, Y. Yang, Prediction of hydrodynamic and optical properties of  $\text{TiO}_2$ /water suspension considering particle size distribution, *Int. J. Heat Mass Transf.* 92 (2016) 864.
- [49] S.E.B. Maiga, C.T. Nguyen, N. Galanis, G. Roy, Heat transfer behaviours of nanofluids in a uniformly heated tube, *Superlattice. Microst.* 35 (3) (2004) 543–557.
- [50] C. Oh, C.M. Sorensen, The effect of overlap between monomers on the determination of fractal cluster morphology, *J. Colloid Interface Sci.* 193 (1) (1997) 17–25.
- [51] R. Prasher, W. Evans, P. Meakin, J. Fish, P.E. Phelan, P. Keblinski, Effect of aggregation on thermal conduction in colloidal nanofluids, *Appl. Phys. Lett.* 89 (14) (2006) 143119.
- [52] Y. Xuan, Q. Li, W. Hu, Aggregation structure and thermal conductivity of nanofluids, *AIChE Journal* 49 (4) (2003) 1038–1043.
- [53] P.E. Gharagozloo, K.E. Goodson, Aggregate fractal dimensions and thermal conduction in nanofluids, *J. Appl. Phys.* 108 (7) (2010) 074309.
- [54] R.J. Farris, Prediction of the viscosity of multimodal suspensions from unimodal viscosity data, *Trans. Soc. Rheol.* 12 (2) (1968) 281–301.
- [55] R. Deepak Selvakumar, S. Dhinakaran, A multi-level homogenization model for thermal conductivity of nanofluids based on particle size distribution (PSD) analysis, *Powder Technol.* 301 (2016) 310–317.
- [56] D. Zhou, H. Wu, A thermal conductivity model of nanofluids based on particle size distribution analysis, *Appl. Phys. Lett.* 105 (8) (2014) 083117.
- [57] G.D. Scott, D.M. Kilgour, the density of random close packing of spheres, *J. Phys. D: Appl. Phys.* 2 (6) (1969) 863.
- [58] C.-J. Yu, A.G. Richter, A. Datta, M.K. Durbin, P. Dutta, Molecular layering in a liquid on a solid substrate: an X-ray reflectivity study, *Phys. B Condens. Matter* 283 (1) (2000) 27–31.
- [59] T. Hashimoto, M. Fujimura, H. Kawai, Domain-boundary structure of styrene-isoprene block copolymer films cast from solutions. 5. Molecular-weight dependence of spherical microdomains, *Macromolecules* 13 (6) (1980) 1660–1669.
- [60] L. Xue, P. Keblinski, S.R. Phillpot, S.U.S. Choi, J.A. Eastman, Effect of liquid layering at the liquid–solid interface on thermal transport, *Int. J. Heat Mass Transf.* 47 (19) (2004) 4277–4284.
- [61] W. Yu, S.U.S. Choi, The role of interfacial layers in the enhanced thermal conductivity of nanofluids: a renovated maxwell model, *J. Nanopart. Res.* 5 (1–2) (2003) 167–171.
- [62] H. Zhu, C. Li, D. Wu, C. Zhang, Y. Yin, Preparation, characterization, viscosity and thermal conductivity of  $\text{CaCO}_3$  aqueous nanofluids, *Sci. China Technol. Sci.* 53 (2) (2010) 360–368.
- [63] J. Jeong, C. Li, Y. Kwon, J. Lee, S.H. Kim, R. Yun, Particle shape effect on the viscosity and thermal conductivity of  $\text{ZnO}$  nanofluids, *Int. J. Refrig.* 36 (8) (2013) 2233–2241.
- [64] S. Bobbo, L. Fedele, A. Benetti, L. Colla, M. Fabrizio, C. Pagura, S. Barison, Viscosity of water based SWCNH and  $\text{TiO}_2$  nanofluids, *Exp. Thermal Fluid Sci.* 36 (2012) 65–71.

- [65] K. Anoop, S. Kabelac, T. Sundararajan, S.K. Das, Rheological and flow characteristics of nanofluids: influence of electroviscous effects and particle agglomeration, *J. Appl. Phys.* 106 (3) (2009) 034909.
- [66] M. Kole, T. Dey, Thermophysical and pool boiling characteristics of ZnO–ethylene glycol nanofluids, *Int. J. Therm. Sci.* 62 (2012) 61–70.
- [67] W.J. Tseng, C.-N. Chen, Effect of polymeric dispersant on rheological behavior of nickel–terpineol suspensions, *Mater. Sci. Eng. A* 347 (1) (2003) 145–153.
- [68] P.K. Das, A.K. Mallik, R. Ganguly, A.K. Santra, Synthesis and characterization of TiO<sub>2</sub>–water nanofluids with different surfactants, *Int. Commun. Heat Mass Transfer* 75 (2016) 341–348.