Heat conduction mechanism in nanofluids [†]

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Abstract

Nanofluids are produced by dispersing nanoparticles in basefluid. Given its superior thermo-physical properties, nanofluids are gaining increasing attention and are showing promising potential in various applications. Numerous studies have been conducted in the past decade to experimentally and theoretically investigate thermal conductivity. The experimental finding is briefly summarized in this study; however, we do not intend to present a systematic summary of the available references from the literature. The primary objective of this study is to review and summarize the most debated mechanisms for heat conduction in nanofluids, such as the effects of a nanolayer, the Brownian motion of nanoparticles and aggregation, as well as induced convection. Finally, at a low concentration of nanoparticles, nanoconvection is the leading contributor to thermal conductivity enhancement, whereas at a higher concentration, the natural thermal transport along the backbone would aggregate, and the effects of the nanolayer would become significant and become ineligible.

Keywords: Nanofluids; Thermal conductivity; Nanolayer; Aggregation; Nanoconvection

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1. Introduction

Nanofluids, which are suspensions obtained by dispersing nanoparticles in conventional fluids, are the new class of nanotechnology-based heat transfer fluids. Nanofluids generally contain small volumetric quantities (typically less than 5 vol%) and exhibit thermal properties superior to those of host fluids or conventional particle fluid suspensions [1]. Compared with conventional solid-liquid suspensions, nanofluids have a number of unique characteristics, such as a high specific surface area, and therefore, greater heat transfer surface between the particles and the fluid; high dispersion stability with the predominant Brownian motion of particles; reduced pumping power; reduced particle clogging; and adjustable properties, including thermal conductivity and surface wettability, by varying particle concentrations to suit different applications [2]. The Argonne National Laboratory produced metal nanofluids in 2005 to cool down truck engines. Nanoparticles transfer heat better than microparticles in particle-dispersed suspensions, and nanocoolants enhance heat transfer better than the best competing fluid [3]. Recently, Chen et al. [4-6] investigated the heat conduction characteristics of nanofluids and searched for suitable materials that would further enhance heat transfer capabilities. In 2011, the results of the reversible temperature regulation of electrical and thermal conductivity using liquid-solid phase transitions were published in *Nature Communications* [4]. The results indicated that a new way of manipulating both the thermal and electrical conductivity of percolated composite materials, which are tiny flakes of graphite suspended in liquid hexadecane, is to simply change the external conditions, such as the surrounding temperature. Such technique can change electrical conductivity by factors of well over 100 and heat conductivity by more than threefold. This novel idea of using phase change to control the conductivity of nanocomposites suggests that heat switches could exist in the future and that this new system could be used as a fuse to protect electronic circuitry or for storing heat. The significant amount of literature is proof of the promising potential application of heat transfer in nanofluids.

The heat flow in a process can be calculated by $Q = hA\Delta T$. Increasing the heat transfer coefficient h by improving the transport properties of the heat transfer materials has been gaining considerable attention, because ΔT is often limited by process or material constraints and the strategy of maximizing the heat transfer area A cannot be employed in microprocessors and micro-electro-mechanical systems (MEMS) since the area cannot be increased.

Numerous studies have discussed the characteristics of thermal conductivity and heat transfer in nanofluids [7-12]. In addition to major studies on thermophysical properties, nanofluids and binary nanofluids (binary mixtures with added
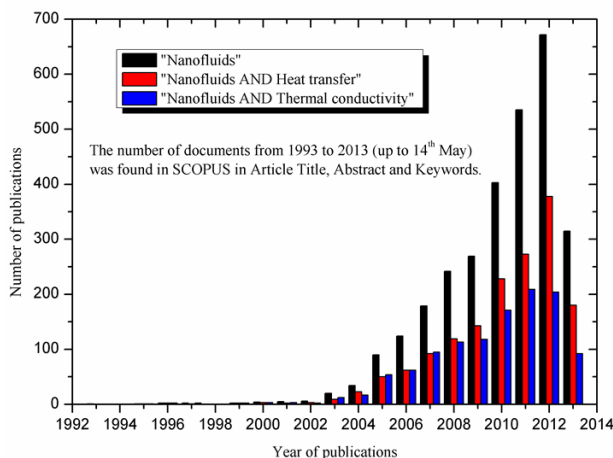
Fig. 1. Nanofluids technology publication rate.

nanoparticles) were recently developed to improve mass transfer performance. Krishnamurthy et al. [13] proposed a faster mass transfer in nanofluids by visualizing a comparison of the dye diffusion properties of both nanofluids and water. Kang et al. [14-18] investigated the heat and mass transfer characteristics in the thermal absorption system with binary nanofluids. The research on nanofluids has significantly grown since the term "nanofluids" was coined. The number of published papers in the Scopus database from 1993 to 2013 (up to 14th May) that are filed under "Nanofluids," "Nanofluids and Heat transfer," and "Nanofluids and Thermal conductivity" are summarized in Fig. 1. The results strongly confirm the intensive interest and activity in research and engineering applications of nanofluids. In fact, the exponential increase in the number of research articles dedicated to this subject shows a noticeable growth as well as the importance of nanofluids technology.

The current interest toward nanofluids is fueled by both fundamental science and applications. On the fundamental side, thermal property measurements on nanofluids are frequently conducted, and many potential mechanisms have been proposed to explain the anomalous enhancement of the thermal conductivity of nanofluids. The experimental data, theoretical models, and mechanisms have been summarized by various researchers [19-21]. On the application side, the enhanced thermal conductivity of nanofluids improves its application in thermal systems [2, 22-25]. For example, with the development of miniaturization, heat removal has become a critical factor for continued progress in the electronic industry, as a result of the increased levels of dissipated power. The search for materials that are good heat conductors has become essential for the design of next-generation integrated circuits and 3D electronics [26]. However, thermal conductivity is not always improved when nanoparticles are added. A number of studies have demonstrated the ineffectiveness of adding nanoparticles and the reduction of thermal conductivity that is dependent on factors such as particle shape, particle species, concentration, and working temperature. [27-29].

Thermal conductivity is of great theoretical and practical value to scientists and engineers, and is therefore the most extensively studied property of nanofluids. In fact, by May 14th 2013, 107 publications on thermal conductivity were found in the Scopus database under "Nanofluids and Review." Thus, we do not intend to present a systematic summary of previous studies in the literature but to summarize the heat conduction mechanism in nanofluids based on our understanding and on previous studies [30-34]. Therefore, the findings on the thermal conductivity of nanofluids are first briefly summarized. Then, we review and discuss the most debated mechanisms, such as the effects of a nanolayer, the Brownian motion of nanoparticles and aggregation, and induced convection.

2. Thermal conductivity measurement

A two-step technique is commonly used to prepare nanofluids. First, dry nanoparticles are produced, and the particles are dispersed in a suitable liquid host. However, given that nanoparticles have a high surface energy, aggregation and clustering are unavoidable and will appear easily. Afterwards, the particles will clog and settle at the bottom of the container. An ultrasonic equipment is generally used to intensively disperse the particles and reduce the agglomeration of nanoparticles [11]. In addition, chemical surfactants are mostly used to improve the stability of nanofluids because the hydrophobic surfaces of nanoparticles are modified to become hydrophilic; a repulsion force among suspended particles is caused by the zeta potential, which is from the surface charge of the particles suspended in the basefluid. The common techniques to measure the thermal conductivity of nanofluids include the transient hot-wire method, steady-state parallel plate method, temperature oscillation method, and 3-ω method [33]. The hot-wire technique is based on a constant heat generation source, which is an infinitely long and thin continuous line that dissipates the heat into an infinite test medium; this technique is known to be a fast and accurate method and has been extensively used [35, 36].

In the past decade, numerous studies have shown that dispersing nanoparticles into a conventional liquid can enhance thermal conductivity. For example, up to 160% enhancement of thermal conductivity was observed in a nanotube volume fraction of only 1% [7]. The experimental data has been summarized by many previous pioneering researchers, such as Fan and Wang [37]. Fig. 2 shows a sample of the summary of typical experimental studies [38-46] on the effective thermal conductivity of the most commonly used nanoparticles-basefluid pairs. Apparently, the particle material might be a main factor affecting the thermal conductivity of nanofluids. Fig. 2 also shows that the thermal conductivity of nanofluids has been reported to increase with the increase in the thermal conductivity of solid particles. However, studies show that particle type may affect the thermal conductivity in other ways. Lee et al. [38] proved that CuO nanofluids had better enhancements in thermal conductivity compared to the Al_2O_3
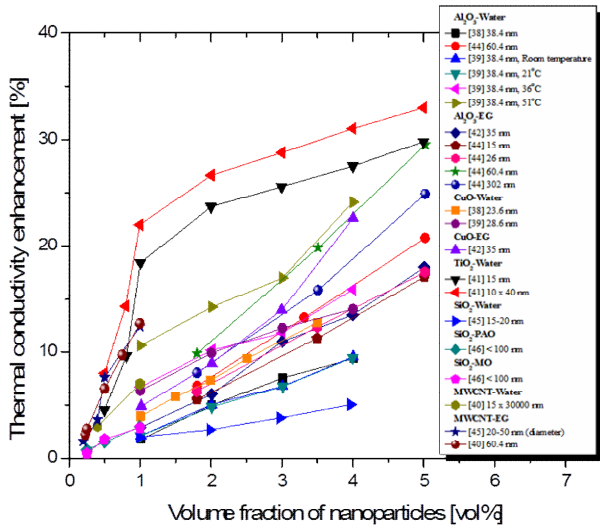
Fig. 2. Summary of experimental thermal conductivity enhancement.

Fig. 3. Thermal conductivity of common solids.

Fig. 4. Thermal conductivity enhancements of nanofluids versus thermal conductivities of basefluids.

nanofluids, although Al_2O_3, as a material, has a higher thermal conductivity than CuO. According to the authors, the key contributor for the anomalous enhancement of heat conduction in nanofluids is the aggregation of nanoparticles, that is, the Al_2O_3 nanoparticles forming relatively larger clusters than the CuO nanoparticles. The analogous trend appeared in our previous experiments as well [30], where the SiO_2 nanofluids obtained a higher thermal conductivity enhancement than that of the Al_2O_3 nanofluids. Obviously, this result indicates that the thermal conductivity of the particle material may not be the dominant parameter that determines the thermal conductivity of nanofluids.

Particle shape may be an essential factor affecting the thermal conductivity of nanofluids. Since the first stage of using common nanoparticles such as Al_2O_3, SiO_2, TiO_2, and CuO to produce nanofluids, carbon nanotubes (CNTs), graphite, and graphene are extensively used in many recent research. The thermal conductivities of the commonly used materials are shown in Fig. 3. Balandin [26] summarized the experimental data of the thermal conductivity for single-walled CNTs (SWCNTs) and multi-walled CNTs (MWCNTs). A substantial data scatter in the reported room temperature thermal conductivity value for CNTs ranging from 1100 W mK^{-1} to 7000 W mK^{-1} was found. The commonly quoted values for individual CNTs are 3000 W mk^{-1} for MWCNTs and 3500 W mK^{-1} for SWCNTs at room temperature. These values are above the bulk-graphite limit of 2000 W mK^{-1}. Differences in the strain distribution of the suspended graphene of various temperatures, sizes, and geometries will affect the results; the thermal conductivity of suspended graphene ranged from 1500-5000 W mK^{-1}. Recently, CNTs, graphite, and graphene have been dispersed in conventional liquids; afterwards, the thermal conductivities of the nanocomposites used were observed to have increased with the increase in the loading of nanoparticles [5, 47, 48]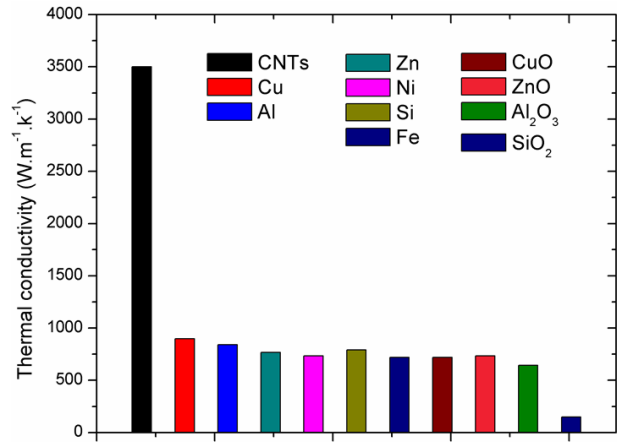. In our previ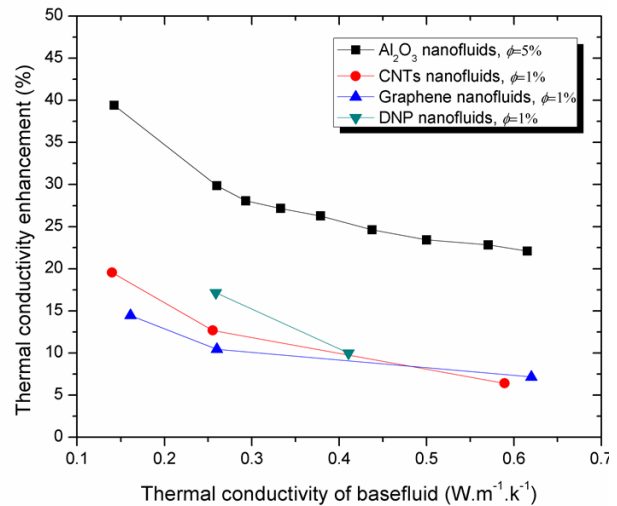ous prediction, the tube-shaped particles were found to possibly have a higher thermal conductivity enhancement of nanofluids [34], which could explain the observed higher measurements for CNTs and graphene.

The commonly used basefluids are deionized (DI) water, ethylene glycol (EG), and pump oil (PO). Fig. 4 shows the thermal conductivity enhancements of nanofluids versus the thermal conductivities of basefluids [49]. The thermal conductivity enhancement obviously increases with a decrease in the thermal conductivity of the basefluid. For example, considering a PO-based Al_2O_3 nanofluid with 5.0% nanoparticle loading, the thermal conductivity enhanced by more than 38%, while the corresponding thermal conductivity enhancement was limited to 22.0% for DI water-based nanofluids [44]. In addition, the thermal conductivity in CNT nanofluids significantly enhanced because the basefluid had a lower thermal conductivity. The viscosity of the basefluid also apparently affects the Brownian motion of nanoparticles, which in turn affects the thermal conductivity of nanofluids [50]. Dul'nev et al. [51] and

Xie et al. [52] have experimentally examined thermal conductivity versus basefluid, and the results were compared with a theoretical analysis made by Hasselman and Johnson in 1987. Contrary to the experimental data, thermal conductivity was found to be nearly independent from the basefluid.

Nanoparticle concentration is a parameter that is extensively investigated in a number of early research; this parameter generally indicates that thermal conductivity increases with an increasing the particle volume fraction, as shown in our previous review literature [33]. However, the nonlinear behavior of enhancement versus particle concentration was reported in a large number of references, which disagrees with conventional theory [20]. Choi et al. [7] demonstrated the fundamental limits of conventional models and interpreted the enhanced thermal conductivity as two consequences of the presence of nanotubes in the liquid: the nature of heat conduction in nanotube suspensions, and an organized structure at the solid-liquid interface. In addition, Choi et al. [7] pointed out that both shape and size of the added nanoparticles played key roles in the nonlinear phenomena.

Particle size, which includes the powder size and aggregate size, is the key parameter for the study of heat conduction in nanofluids. The general trend in the experimental data is that the thermal conductivity of nanofluids increases with a decreasing powder size or increasing aggregate size. However, there were also some contradictory data in numerous literature, for example, Mintsa et al. [53] measured the thermal conductivity of Al_2O_3-water nanofluids with powder sizes of 36 and 47 nm, the enhancements of the thermal conductivity were observed to be similar for the two nanofluids with different particle sizes at room temperature. Murshed et al. [54], and Chon and Kihm [55] also measured thermal conductivity, however, no clear difference in the enhancements could be obtained regardless if the powder sizes ranged from 38.4 to 80 nm. However, the velocity of the Brownian nanoparticles in the suspension would increase with a decrease in the powder size, which leads to aggregation and convection, the two strongly debated heat conduction enhancements in nanofluids. The effect of the particle size on the faster heat conduction was discussed in our previous literature [32, 34]. In fact, there exists a critical particle size for the effective thermal conductivity of nanofluids. Prasher et al. [56] developed the combined conduction-convection model, which includes aggregation. This model demonstrated that the thermal conductivity enhancement initially increases with the decreasing radius of nanoparticles, reaches a peak, and then decreases because of the aggregation effect. This behavior is an effect of the particle size and was also experimentally observed by Xie et al. [44]. The effect of aggregation on heat conduction in nanofluids would be discussed in the following section.

Significantly enhanced thermal conductivity of nanofluids has been reported in a number of experimental results, as described in several review literature [10, 20, 37, 57]. Lee et al. [20] summarized previous thermal conductivity data and proposed advanced features of thermal conductivity in nanofluids,

such as the anomalously high enhancement at low nanoparticle concentration, the nonlinear relationship between enhancement and concentration, and the temperature-/size- and pH-dependent thermal conductivity enhancements. The specific parameters that would affect the enhancement were reviewed as well [19]; these parameters are the following: (1) nanoparticle material, shape, size, and concentration; and (2) basefluid, temperature, and pH. However, most of the available thermal conductivity data from different contributors do not agree well with one another because of various experimental conditions and measurement deviations. Regarding nanofluids, as the controversial heat transfer fluids [58], dramatic increases in thermal conductivity with small nanoparticle loadings have been reported. Accordingly, the mechanism of thermal conductivity enhancement is a strongly debated topic. Thus, theoretical investigation on the enhanced thermal conductivity of nanofluids becomes highly important to understand and to improve the applied heat transfer enhancement.

3. Heat conduction mechanism and model

There are numerous explanations through numerical models and visualizations to elucidate the mechanism of heat conduction in nanofluids [37]. Clearly, the most debated theoretical explanations are the effect of the liquid molecule nanolayer, the aggregation induced by the Brownian nanoparticles, and the formative nanoconvection. In addition, the effect of interfacial thermal resistance was included. Moreover, these above mentioned debating theories were developed based on classical effective medium theory (EMT).

Two common methods are used in EMT to treat the effective transport coefficient of the mixture and composites: Maxwell-Garnett's self-consistent approximation (MG model) [59] and the Bruggeman approach [60]. The MG model fits well for diluted and randomly distributed components included in a homogenous host medium, where the suspended particles are isolated and no interactions exist among them. The Bruggeman approach with mean field approach is applied to discuss the interactions among randomly distributed particles. The Bruggeman model has no limitations on the concentration of spherical particles. For low particle concentration suspension, the predictions of thermal conductivity from the Bruggeman model and the MG model are identical. However, for high particle concentration suspension or a particle percolation situation, the discrepancy between the two models increases and the MG model fails to predict the experimental data, whereas the Bruggeman model fits this situation well [61]. The MG model and Bruggeman model are expressed as follows in Eqs. (1) and (2), respectively.

$$\frac{k_{nf}}{k_{bf}} = \frac{3\varphi_p k_p + (1-\varphi_p)(k_p + 2k_{bf})}{3\varphi_p k_{bf} + (1-\varphi_p)(k_p + 2k_{bf})} \tag{1}$$

$$\varphi_p \left(\frac{k_p - k_{nf}}{k_p + 2k_{bf}} \right) + (1-\varphi_p)\left(\frac{k_{bf} - k_{nf}}{k_{bf} + 2k_{nf}} \right) = 0 \tag{2}$$
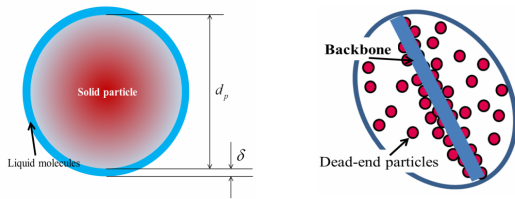
(a) Solid particle with nanolayer   (b) Aggregate of nanoparticles

Fig. 5. Schematic diagram of nanolayer and aggregate (ellipsoid).

where k means the thermal conductivity; p, bf, and nf represent the nanoparticle, basefluid, and nanofluids, respectively; and φ_p is the volume fraction of the nanoparticles in the fluid. Hamilton and Crosser [62] extended the Maxwell model to account for the effect of the shape of the solid particles, in addition to the thermal conductivities of the solid and liquid phases and particle volume fraction. The effective thermal conductivity ratio of nanofluids is determined by

$$k_{eff} = \frac{k_p + (n-1)k_{bf} - (n-1)\varphi_p(k_{bf} - k_p)}{k_p + (n-1)k_{bf} + \varphi_p(k_{bf} - k_p)} k_{bf} \quad (3)$$

where n is the empirical shape factor, which is defined as $n = 3/\psi$, where ψ is the sphericity. Sphericity is the ratio of the surface area of a sphere, with a volume equal to the ratio of the particle to the surface area of the particle. Therefore, n = 3 for a sphere, and in that case the Hamilton and Crosser model becomes identical to the Maxwell model. Lee et al. [20], and Fan and Wang [37] summarized more references on the EMT model in detail. Although classical EMT-based models can predict the thermal conductivity of suspensions with particles sizes of a micrometer or larger, these models fail to predict most thermal conductivity data for nanofluids. Nevertheless, these models are utilized frequently because of their simplicity in the study of nanofluids, and to compare between theoretical and experimental findings.

3.1 Nanolayer of liquid molecules

Liquid molecules near a solid surface are known to form layered structures called nanolayer. The nanolayer structure was recently introduced by Keblinski et al. [63], and Yu and Choi [64] as the first static mechanism to explain the enhanced thermal conductivity of nanofluids. Fig. 5(a) shows the schematic diagram of the basic concept of nanolayers. However, the fact that there is no experimental data regarding the thickness (δ) and thermal conductivity of these nanolayers is an important drawback of the proposed mechanisms. Nanolayers have been considered as a thermal bridge between a solid particle and a bulk liquid to improve the thermal transport in a suspension. The nanolayer around the particle is assumed to be more ordered than that of the bulk liquid, and the thermal conductivity of the ordered nanolayer k_{layer} is higher than

that of the bulk liquid [64].

Accounting for the concept of the nanolayer, Yu and Choi [64] modified the Maxwell model to predict the effective thermal conductivity of nanofluids. Two essential features of the model of Yu and Choi are 1) the consideration for a new nanoscale structure that has not been considered in classical EMT models, and 2) a new assumption has been made that the solid-like nanolayer acts as a thermal bridge between a solid nanoparticle and a bulk liquid in nanofluids, an idea contrary to that in nanocomposite solids. Finally, the modified MG model was determined, and the equivalent thermal conductivity of the nanolayer-based particle k_{pn} was presented as

$$\frac{k_{pn}}{k_p} = \frac{(1+\beta)^3(1+2\gamma) + 2(1-\gamma)}{(1+\beta)^3(1+2\gamma) - (1-\gamma)}\gamma \quad (4)$$

where the ratio of the nanolayer thickness to the original particle radius (d_p) is defined as $\beta = 2\delta/d_p$; and $\delta = \sqrt{2\pi}\sigma$, where σ is a parameter that characterizes the diffusion of the interfacial boundary whose typical value is within 0.2 to 0.8 nm [65]. The thermal conductivity of the nanolayer is defined as $k_{layer} = \gamma k_p$. In 2004, Yu and Choi [66] extended the above mentioned model, which is the nanoscale structural model for spherical nanoparticles, to nonspherical particles, and the Hamilton-Crosser model was also modified. The nanolayer is expressed as a confocal ellipsoid with a solid particle. In addition, Leong et al. [67] developed a new nanolayer-based model, which includes the effect of the nanoparticle size, nanolayer thickness, and volume fraction. Xie et al. [68] derived a new expression for calculating the effective thermal conductivity of nanofluids considering a linear thermal conductivity distribution across the interfacial nanolayer. Murshed et al. [54] proposed two models for the effective thermal conductivity of nanofluids containing spherical and cylindrical nanoparticles by considering the effect of the interfacial layer at the solid-liquid interface. In our previous study [32, 34], the nanolayer of liquid molecules was indicated to have insignificant effects on the thermal conductivity enhancement of nanofluids at a low concentration of nanoparticles of ~0.1 vol%, whereas at a higher concentration, the prediction demonstrates that the nanolayer would play a key role for the enhancement. This indication of the nanolayer may be induced by the liquid molecules being more orderly near the solid-liquid interface [69]. Thus, such organized liquid layers were speculated to have acted as a bridge to generate more effective thermal transport across the interface [7]. Another possibility proposed was that if the separation of the two particles is so small that only the organized liquid layer is in between, such solid-like liquid layer may facilitate the ballistic phonons initiated in one particle to persist in the liquid and reach the nearby particle, consequently increasing thermal conductivity. Moreover, the liquid layer itself was speculated to have a better thermal transport ability than the bulk liquid because of the ordered molecular structure of the layer [63].

Table 1. Summary of the models on Brownian motion-induced effects.

Reference	Expression	Remark
Wang et al. (2003) [61]	$$\frac{k_{nf}}{k_{bf}} = \frac{(1-\varphi)+3\varphi\int_0^\infty \frac{k_{cl}(a_{cl})n(a_{cl})}{\left[k_{cl}(a_{cl})+2k_{bf}\right]}da_{cl}}{(1-\varphi)+3\varphi\int_0^\infty \frac{k_{bf}(a_{cl})n(a_{cl})}{\left[k_{cl}(a_{cl})+2k_{bf}\right]}da_{cl}}$$	Modified Bruggeman model with aggregation effect
Xuan et al. (2003) [50]	$$\frac{k_{nf}}{k_{bf}} = \frac{k_p+2k_{bf}+2(k_p-k_{bf})\varphi}{k_p+2k_{bf}-(k_p-k_{bf})\varphi} + \frac{\rho_p\varphi C_{v,p}}{2k_{bf}}\sqrt{\frac{k_bT}{3\pi a_{cl}\mu}}$$	EMT approach with aggregation and Brownian Motion effect
Prasher et al. (2006) [56]	$$\frac{k_{nf}}{k_{bf}} = (1+A\,\mathrm{Re}^m\,\mathrm{Pr}^{0.333}\,\varphi)\frac{k_{ag}+2k_{bf}+2(k_{ag}-k_{bf})\varphi_{ag}}{k_{ag}+2k_{bf}-(k_{ag}-k_{bf})\varphi_{ag}}$$	EMT approach based on the aggregation kinetics and Brownian Motion effect
Feng et al. (2007) [76]	$$\frac{k_{nf}}{k_{bf}} = (1-\varphi_e)\frac{k_{pe}+2k_{bf}+2(k_{pe}-k_{bf})(1+\eta)^3\varphi}{k_{pe}+2k_{bf}-(k_{pe}-k_{bf})(1+\eta)^3\varphi}$$ $$+\varphi_e\left[\left(1-\frac{3}{2}\varphi_e\right)+\frac{3\varphi_e}{\eta}\left(\frac{1}{\eta}\ln\frac{a_p+\delta}{(1-\eta)(a_p+\delta)}-1\right)\right]$$	EMT approach with nanolayer and aggregation effect
Jang and Choi (2004) [70]	$$\frac{k_{nf}}{k_{bf}} = 1+\varphi\left(\frac{\beta k_p}{k_{bf}}-1\right)+C_1\varphi\frac{d_{bf}}{d_p}\mathrm{Re}_{d_p}^2\,\mathrm{Pr}$$	Brownian motion-induced nanoconvection
Koo and Kleinstreuer (2004) [71]	$$\frac{k_{nf}}{k_{bf}} = \frac{k_p+2k_{bf}+2(k_p-k_{bf})\varphi}{k_p+2k_{bf}-(k_p-k_{bf})\varphi} + 50000\frac{\beta\varphi\rho_p C_{v,p}}{k_{bf}}\sqrt{\frac{k_BT}{\rho_p d_p}}f(T,\varphi,etc.)$$	Brownian motion-induced nanoconvection with EMT
Kumar et al. (2004) [73]	$$\frac{k_{nf}}{k_{bf}} = 1+C\frac{\varphi a_{bf}}{k_{bf}(1-\varphi)a_p}\cdot\frac{2k_BT}{\pi\mu d_p^2}$$	Brownian motion with kinetic theory
Prasher et al. (2005) [72]	$$\frac{k_{nf}}{k_{bf}} = (1+A\,\mathrm{Re}^m\,\mathrm{Pr}^{0.333}\,\varphi)\frac{k_p+2k_{bf}+2(k_p-k_{bf})\varphi}{k_p+2k_{bf}-(k_p-k_{bf})\varphi}$$	Brownian motion-induced nanoconvection with EMT considering interaction of nanoparticles
Yang (2008) [77]	$$\frac{k_{nf}}{k_{bf}} = \frac{k_p+2k_{bf}+2(k_p-k_{bf})\varphi}{k_p+2k_{bf}-(k_p-k_{bf})\varphi} + \frac{157.5\varphi C_f u_p^2\tau}{k_{bf}}$$	Brownian motion-induced nanoconvection approximation with Maxwell model

3.2 Brownian motion

A suspension of nanosized particles is different from that of micro- or millimeter-sized particles given that the latter is static and the former is dynamic because nanoparticles are constantly in random motion even if the bulk fluid is stationary. Hence, a fundamental difference in the mechanisms of the heat transport in nanofluids is expected because of the dynamic effects in nanofluids. Given that the particles suspended in the liquid are very small, the Brownian movement of the particles is quite possible [70-74]. Kumar et al. [73] developed a moving particle model from the Stokes-Einstein formula to explain the temperature effect. The root-mean-square velocity (ν) of a Brownian particle can be defined as [71]

$$\nu = \sqrt{\frac{18\kappa T}{\pi\rho d_p^3}} \tag{5}$$

where κ, T, and ρ represent the Boltzmann constant, temperature, and density, respectively. Some early discussions proposed that the Brownian motion of nanoparticles is the straightforward mechanism to explain the thermal conductivity enhancement of nanofluids. However, the thermal diffusivity of the basefluid is usually two or more orders larger than the particle diffusivity [35]. Therefore, such a Brownian diffusive motion of nanoparticles has a negligible effect on the thermal conductivity enhancement of nanofluids [72, 75]. Generally, there are two kinds of Brownian motion in nanofluids [20]: collision among Brownian nanoparticles and convection induced by Brownian nanoparticles. In addition, the Brownian motion of nanoparticles will lead to another key dynamic phenomenon, that is, the aggregation of nanoparticles. Recently, aggregation and convection induced by Brownian motion are extensively discussed to explain the enhanced thermal conductivity of nanofluids as essential factors. Table 1 shows the summary of models considering the

aggregation and convection induced by the Brownian motion of nanoparticles [50, 56, 61, 70-73, 76, 77].

3.3 Aggregation of nanoparticles

Aggregation or clustering is an inherent property of nanoparticles whether they are in liquid or powder form, resulting from van der Waals forces. Recently, several aggregation models have been developed to investigate the enhancement mechanisms. Keblinski et al. [63] conceptualized the clustering of nanoparticles as a mechanism of the enhanced k of nanofluids, assuming that clustered nanoparticles provide local percolation-like paths for rapid heat transport and increase the effective nanoparticle volume fraction. Prasher et al. [78] studied the effect of aggregation on thermal conductivity enhancement by analyzing the aggregation kinetics of nanoscale colloidal solutions combined with the physics of thermal transport. Prasher et al. [78] also reported that the aggregation time constant decreases rapidly with a decrease in the nanoparticle size, and the thermal conductivity enhancement increases with an increase in the level of aggregation, which levels off after the optimum level of aggregation is reached. Gao et al. [79] experimentally investigated the enhancement mechanism through thermal conductivity measurements and structural analysis for the same materials in both liquid and solid states, and the idea that clustering holds the key contribution for the enhancement was strongly suggested. Guided by this insight, Wang et al. [6] used graphite flakes as additives and developed stable graphite suspensions in water and oil to discuss the heat conduction mechanisms in nanofluids. The thermal percolation phenomenon was observed and explained based on combined optical and AC impedance spectroscopy studies. Evans et al. [80] revealed that clustering could result in a fast transport of heat along relatively large distances because heat could be conducted much faster by solid particles. Evans et al. [80] also showed that the effective thermal conductivity increased with an increase in the cluster size. Feng et al. [76] found that clustering improved thermal conductivity enhancement more significantly in nanofluids with smaller nanoparticles because of the major contribution of the van der Waals force. Prasher et al. [78] discussed the aggregation of nanoparticles in nanofluids and concluded that the aggregated nanoparticles would improve the heat conduction around the percolation path. At this point, notably, the excessive clustering of nanoparticles may result in sedimentation, which adversely affects thermal conductivity.

For the effect of aggregation on thermal conductivity enhancement, the shape and size of aggregates could be two essential factors. To further understand the effect of aggregation on the fluid properties, thermal conductivity is modeled using fractal theory in the Ref. [81]. Therefore, the thermal conductivities of aggregates were evaluated by separating them into two components, the percolation contributing backbone and the non-percolation contributing dead-end particles. Fig. 5(b) shows the schematic diagram of the aggregation of

nanoparticles in the suspension. Considering the interfacial thermal resistance (Kapitza resistance), the effective thermal conductivity of dead-end particles-based suspension (including the basefluid and dead-end particles) is presented by using the Maxwell model [82]

$$k_{bf,de} = \frac{\left[k_{pn}(1+2\alpha)+2k_{bf}\right]+2\varphi_{pa,de}\left[k_{pn}(1-\alpha)-k_{bf}\right]}{\left[k_{pn}(1+2\alpha)+2k_{bf}\right]-\varphi_{pa,de}\left[k_{pn}(1-\alpha)-k_{bf}\right]}$$

(6)

where $\varphi_{pa,de}$ represents the volume fraction of the dead-end particles in the aggregate. $\alpha = 2R_b k_{bf} / d_p$ and R_b represent the interfacial thermal resistance between the solid and bulk liquid. The effective thermal conductivity of aggregate k_b is determined using composite theory for misoriented ellipsoidal particles for the backbone in a matrix of the non-percolation contributing portion, where the following equation is used [81].

$$k_a = k_{bf,de} \frac{3+\varphi_{pa,ba}\left[2\beta_{11}(1-L_{11})+\beta_{33}(1-L_{33})\right]}{3-\varphi_{pa,ba}(2\beta_{11}L_{11}+\beta_{33}L_{33})}$$

(7)

where $\varphi_{pa,ba}$ represents the volume fraction of the backbone in the aggregate. L_{ii}, $i = 1,3$ are well-known geometrical factors that are dependent on the particle shape, and β is the defined dimensionless variation. When the ellipsoidal inclusions become spheres, $L_{11} = L_{33} = 1/3$, then Eq. (7) reduces to

$$k_a = k_{bf,de} \frac{k_{pn}(1+2\alpha)+2k_{bf,de}+2\varphi_{pa,ba}\left[k_{pn}(1-\alpha)-k_{bf,de}\right]}{k_{pn}(1+2\alpha)+2k_{bf,de}-\varphi_{pa,ba}\left[k_{pn}(1-\alpha)-k_{bf,de}\right]}.$$

For continuous fiber composites with uniformly distributed long fibers, $L_{11} = 0.5$, $L_{33} = 0$, then the main effective thermal conductivity would be expressed as $k_a = (1-\varphi_{pa,ba})k_{bf,de} + \varphi_{pa,ba}k_{pn}$. For laminate composites with a matrix containing parallel flat plate inclusions, $L_{11} = 0$, $L_{33} = 1$, the mainly effective thermal conductivity would also be estimated with the expression of that for fiber inclusions [81].

In addition, a thermal boundary resistance (Kapitza resistance) exists at solid-solid and solid-liquid interfaces even if these interfaces are in perfect contact [83]. The interfacial thermal resistance poses a barrier to heat flow that might inhibit the benefit of adding highly conductive fillers. Generally, thermal resistance is obtained through experimental measurement. For water, R_b is assumed to be 0.77×10^{-8} m^{2}K/W [84]. The mechanism for R_b between liquid and solid is still unclear. A simple debye model for interfacial thermal resistance yields is: $R_b = 4/(\rho C \upsilon \eta)$ [85], where C, υ, and η represent the specific heat, debye velocity of the matrix, and the average probability of the transmission of the phonon across the interface into the particles. The mechanisms for the additional re-

sistance at the interfaces are mismatched in the characteristics of the heat carriers on the two sides [86]. In the case of dielectric-dielectric interfaces, the mismatch in the atomic potential and mass leads to a reflection of phonons. The transmission of energy modes across interfaces, however, depends on the details of interfacial structures. Up until now, there are no good model existing that can accurately predict thermal interfacial resistance.

3.4 Nanoconvection

The Brownian motion of particles may result in the convection of the surrounding basefluid, thus, enhancing the thermal conductivity [70, 72, 82]. Jang and Choi [70] developed a theoretical Brownian motion-based model that takes into account nanoconvection. Since then, nanoconvection, as a key contributor, is commonly used to estimate the thermal conductivity of nanofluids. Prasher et al. [72, 78] considered the effect of the convection of the liquid near the particles as a result of their Brownian movement; the Reynolds number and the effective thermal conductivity of the semi-infinite medium were given as

$$k_{nc} = A\varphi \operatorname{Re}^m \operatorname{Pr}^{0.333} k_{bf} \tag{8}$$

where A and m are constants determined from experiments, where A should be independent of the fluid type, whereas m will depend on the fluid type. Pr is the Prandtl number, and the Reynolds number is determined by $\operatorname{Re} = 1/\nu \sqrt{18\kappa T / \pi\rho d_p}$, where ν represents the kinematic viscosity. The evaluated thermal conductivity enhancement could reach several percent for a 10 nm alumina particle suspended in water and ethylene glycol [72]. Based on this model, effective thermal conductivity of nanofluids via nanoconvection has been predicted, and the results showed that nanoconvection would be the dominative contributor for the observed enhancement at a low concentration of ~0.1 vol% [34]. While increasing the concentration of nanoparticles, the dynamic contribution of nanoconvection would be reduced, especially for fiber inclusion. To theoretically study the effect of nanoconvection, the distributions of the static and dynamic contributions would be evaluated at a higher concentration of up to 5 vol% by using the combined model, which is expressed in Eq. (6) in the Ref. [34]. The result is shown in Fig. 6. The result indicates that analogous trends of the contribution proportion and a variation in the fiber and sphere inclusions are obtained; by contrast, that for ellipsoidal inclusion, the static contribution increases, but the dynamic contribution decreases with an increase in the nanoparticle loading. For the fiber shaped aggregate-based nanofluids, the dynamic contribution would gradually decrease, in which the 50% proportion is obtained at a concentration of 1.5 vol%. However, the estimation of thermal conductivity enhancement in Ref. [87] also showed an insignificant contribution of either translational- or rotational-
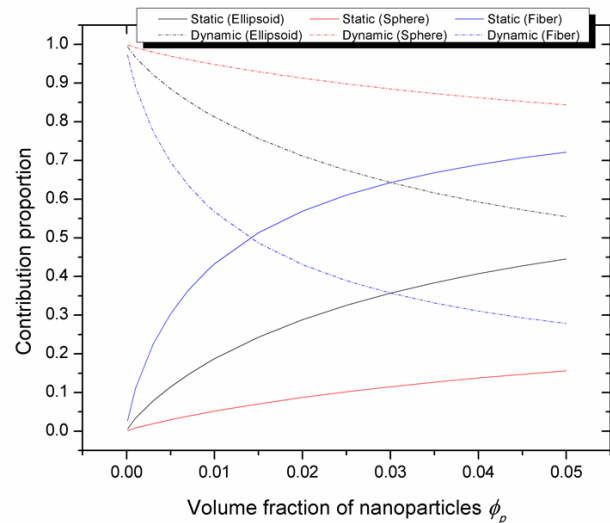

Fig. 6. Contribution proportion on thermal conductivity enhancement.

Brownian-motion-induced convection because of the small Peclet number. Up to now, there are controversies in existing experiments and simulations; some studies support the role of the particles in the Brownian motion [39, 44, 53, 88], while others disqualify the effect of the particles [63, 88, 89]. However, in the convection induced by the Brownian movement of nanoparticles, which is reported to be one of the most debated reasons for thermal conductivity enhancement, the mathematical model includes some experimentally determined constants. Thus, to improve the theory on nanoconvection in nanofluids, more studies are required to understand the fundamental mechanism and the empirical formula.

The molecular dynamics simulation by Volz et al. [90] demonstrated that a near-field radiation such as the Coulomb interaction, was shown to offer an increased thermal conductance between two adjacent particles based on a fluctuating dipole model. With the distance between two particles decreasing to less than the particle diameter, the multipolar contributions can lead to a stronger heat transfer enhancement, which is found to be two to three orders of magnitude more efficient than the enhancement for two contacting particles. However, the surface electrons in polar nanoparticles were argued to play a minor role in the heat-conduction enhancement because the separation distance among particles is comparable with or smaller than the electron wavelength even in nanofluids with a low concentration of particles [91]. Wang et al. [6] suggested that near-field radiation could not be used to explain the experimental thermal conductivity enhancement. Even though experiments have demonstrated that near-field radiation can be significantly higher than the maximum radiation exchange described by blackbodies, the radiation effect is always much smaller than heat conduction through a medium [92]. Several other mechanisms reported that explanations of the enhancement were summarized in a number of review literature [6, 20, 37, 75].

4. Conclusions

This offers an overview of the recent research and development on heat conduction in nanofluids, with emphasis on thermal conductivity. Numerous experimental studies showed that dispersing nanoparticles into conventional liquids can enhance the thermal conductivity of nanofluids. Based on a number of research literature, the significant enhancement of thermal conductivity of nanofluids was found to be dependent on many factors, such as the nanoparticle material, nanoparticle concentration, particle size and shape, basefluid, temperature, and chemical additives. The main explanations contributing to thermal conductivity enhancement include the nanolayer of liquid molecules, Brownian motion and the induced aggregation, and nanoconvection. At a low concentration of nanoparticles, nanoconvection is also suggested to be the leading effect for thermal conductivity enhancement, whereas at a higher concentration, the natural thermal transport along the backbone in aggregate and the effect of the nanolayer will increase their contributions, which could not be neglected especially for the fiber-shaped aggregates. For further studies, the authors shared some recommendations. The nonlinear phenomenon of thermal conductivity enhancement versus nanoparticle concentration, which is reported to be related to the shape and size of the nanoparticle aggregates, was observed. More experiments are required to confirm this interesting phenomenon and explain it in theory. To enhance the thermal conduction in nanofluids, the aggregation of nanoparticles is a key contributor. However, looking for ways of controlling the shape and size of aggregates become crucial and complicated. Thus, more experiments under visualization are required to examine the anomalous enhancements in the heat transfer of nanofluids and the generation of aggregates, which are helpful to clearly understand the fluid dynamic and thermal performance of nanofluids.

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