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ROLE OF NANOSTRUCTURES IN ANOMALOUS THERMAL CONDUCTIVITY OF NANOFLUIDS

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ABSTRACT

Nanofluids have stimulated immense research interest due to their superior thermophysical properties, heat transfer features and potential applications in numerous important fields. Role of nanostructures in heat transfer mechanisms and thermal properties particularly thermal conductivity of nanofluids has been presented and relevant studies are critically reviewed in this study. Research demonstrated that nanofluids exhibit anomalous thermal conductivity (generally higher than their base fluids) which increases with the loading of nanoparticles. Despite of some findings on positive impact of agglomeration or clustering of nanoparticles on thermal conductivity, contrary findings (negative) and argumentations are still widely accepted in the nanofluids research community. Literature results showed that while cluster size increases with concentration of nanoparticles, thermal conductivity of nanofluids decreases with increasing the cluster size. However, it is not yet well-understood how to control the morphology of the clusters of nanoparticles and how do they play role in changing the thermal properties of nanofluids. Furthermore, studies revealed that the primary shape or structures of nanomaterials also influence the properties of nanofluids. Nanofluids containing nanotubes of large aspect ratio exhibit superior thermal conductivity compared to nanofluids having nanoparticles of any other shapes. Nanorods (cylinder)-laden nanofluids showed slightly higher thermal conductivity than that of nanosphere-based nanofluids. Nevertheless, the structures of agglomerated or individual nanoparticles and their nano- or molecular- level activities in the host fluids are mainly responsible for the anomalous thermal conductivity of nanofluids.

INTRODUCTION

Nanofluids are prepared by suspending nanometer-sized structures including particles, rods, ribbon, flakes, and tubes in conventional fluids such as water (W), ethylene glycol (EG), engine oil (EO), and other heat transfer fluids. This is a new class of engineering fluids which showed anomalously enhanced thermophysical and transport properties, as well as potential applications in numerous important fields such as microelectronics, MEMS, microfluidics, transportation, manufacturing, instrumentation, medical, and HVAC systems [1-7]. While micro-sized structures easily agglomerate and sediment quickly in host liquid which hinder their practical applications in thermal managements and other systems, nanoscale structures with orders of magnitude large relative heat transfer surface areas can remain well-dispersed in the base fluids and are suitable for even microsystems or microchannels. Although nanofluids are more stable as compared to suspensions of micro-sized particles, most of the cases nanofluids still lack long-term stability which is very crucial for their practical applications in thermal management or other advanced cooling technologies. Nanoparticles are also prone to agglomerate and to form clusters inside the base fluids. Since stability is the first key requirement for characterizing the properties and features of nanofluids, sonication and surfactant are commonly used to breakdown the agglomerated nanoparticles or clusters for the homogenous dispersions of nanoparticles and better stability of nanofluids. In the absence of appropriate type and quantity of surfactant, nanoparticles in nanofluids can easily agglomerate and settle down at the bottom of the container. Agglomeration and sedimentation of nanoparticles in base fluids depend on various factors such as particles concentration, shape and size as well as properties of base fluids. Nonetheless, any agglomeration or clustering of

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nanoparticles can hinder their numerous applications like microchannel flow or in heat exchangers.

Despite of extensive research interest and efforts devoted on these new fluids, there are controversies in the heat transfer mechanisms and findings of their thermal properties particularly thermal conductivity [2-3,8-9]. However, it is undisputed that nanofluids possess significantly higher thermal features such as thermal conductivity, heat transfer coefficient and boiling critical heat flux compared to their base fluids [2-4, 10-15]. All controversies and anomalies of enhancements of thermal properties of nanofluids are raised mainly due to not correctly identifying or understanding the role of nanostructures in altering the properties of host fluids. Morphologies and nanoscale level activities of dispersed nanostructures (commonly used nanoparticles and tubes) are believed to be responsible for the anomalous properties of nanofluids. After dispersion in base fluids, because of acting interparticle forces, gravity, number density and other factors, nanoparticles change their primary structures and form clusters of ununiformed shapes, sizes and distributions. However, it is not yet well-understood how to control or obtain uniform size and structures of clusters in base fluids for optimizing (enhancing) their thermal properties. In recent years, some research groups [8,16-19] have tried to establish that chain-like (interconnected) clusters of nanoparticles is the major player behind the reported anomalous thermal conductivity of nanofluids. Interestingly some of these groups previously ignored any positive impact of clustering or agglomeration in enhancing the thermal conductivity of nanofluids. On the other hand, most of other researchers demonstrated that besides hindering practical application in flowing systems (e.g. clogging or eroding flow channels), agglomeration or clustering of nanoparticles have negative (decreasing) on thermal conductivity of nanofluids. Thus new controversy on clustering of nanoparticles raised and it is very crucial to study the morphology and structures of nanoparticles in dispersed conditions in order to better understand their role in changing the thermal properties of nanofluids.

Furthermore, there is no unanimous agreement among thermal conductivity data from different research groups or laboratories since most results are not easily reproducible. For instance, the thermal conductivity of a nanofluid having 4 vol. % loading of alumina nanoparticles in water measured by three different groups showed different increments ranging from 9% to 30% [20-22]. Also for extremely low volumetric loading (1.3×10^{-4} %) of gold nanoparticles (4 nm) in water, large increase (20%) in conductivity of water was reported by Kumar *et al.* [23], whereas for 0.018 % volumetric loading of the same nanoparticle (4 nm gold) in ethanol, Putnam *et al.* [24] found negligible enhancement (1.3%) of thermal conductivity. It is believed that most of the discriminations of reported data of thermophysical properties of nanofluids are mainly because of the degree of stability of nanofluids, peculiarities of nanoparticle synthesis, purity of nanoparticles, nature of stabilizing agents, and differences in the adopted measurement techniques.

Therefore role of dispersed nanostructures in the heat transfer mechanisms and thermal conductivity of nanofluids are discussed in this study. In addition to our works, findings from relevant studies are also critically reviewed and compared.

STUDIES ON CLUSTERING OF NANOPARTICLES IN NANOFLUIDS

While most of the research efforts have been devoted to study the effect of concentration of nanoparticles and other parameters such as temperature and particle size on thermal conductivity of nanofluids, only a handful effort has been reported on the influence of clustering on thermal conductivity and stability of nanofluids [25-30]. Among very limited efforts, Hong *et al.* [25] reported decreasing of thermal conductivity of nanofluids due to the agglomeration or clusters of the nanoparticles. Another research group [31] also showed that the formation of large and compact clusters at high concentration produced large particle-free regions in host liquid with high thermal resistances which can offset the enhanced thermal conductivity. This hypothesis was also noted by Koblinski *et al.* [32] when they first proposed several potential mechanisms for the anomalously high thermal conductivity of nanofluids. Hwang *et al.* [26] reported that the characteristics of base fluids and suspended nanoparticles have strong influence on the stability of nanofluids which can also improve by adding surfactant. Effect of clustering on the thermal conductivity of CuO dispersed nanofluids was investigated by Karthikeyan *et al.* [27] who found significant enhancement (up to 54 %) of thermal conductivity of CuO water and EG-based nanofluids. They also reported that thermal conductivity actually decreases with elapsed time due to clustering of nanoparticles. This group [33] later studied the influence of aggregation of nanoparticle on thermal conductivity of two nanofluids in stable and unstable conditions. While the enhancement of thermal conductivity of CuO/EG-based nanofluids decreases with elapsed time until reaching an equilibrium value (about 45 minutes in their case), the particle cluster size increases substantially (from nano to micron scales) with elapsed time [33]. Whereas Wu *et al.* [28] experimentally showed that the clustering did not give rise to any distinct difference in the thermal conductivity of nanofluid even at very high volumetric loading (about 23%) of SiO₂ nanoparticles.

THEORETICAL MODELS AND MECHANISMS

At the early stage of nanofluids research, several heat transfer mechanisms for nanofluids were proposed and analyzed by Wang *et al.* [34] and Koblinski *et al.* [32]. These mechanisms include nanoparticles Brownian motion, interfacial nanolayer at the nanoparticle/fluid interface, nature of heat transport in the nanoparticle, and nanoparticle clustering. Later it was found that the contributions of some of these factors particularly Brownian motion and nature of heat transport in nanoparticles to the enhanced thermal conductivity of nanofluids are not significant. Although interfacial nanolayer and nanoparticles clustering are considered as key factors for the thermal conductivity of nanofluids, there remain controversies about the actual heat transfer mechanisms of nanofluids [8-9]. Considering the effect of interfacial nanolayer, numbers of models [14, 35-40] have been developed for the prediction of thermal conductivity of nanofluids. Despite the fact that the properties and thickness of nanolayer are undetermined, this nanolayer factor is widely accepted and no further discussion on nanolayer and nanolayer-based models will be made here. Instead clustering of nanoparticles in nanofluids has been briefly discussed. It is known that nanoparticles suspended in host fluids prone to form clusters

and thus for any concentration of nanoparticles, some degrees of clusters of nanoparticles exist in nanofluids. However, controversy on the impact of clustering mainly arises from whether clusters have positive or negative impact on the thermal properties and stability of nanofluids.

Considering chain-like clustering of nanoparticles as major factor an effective medium theory (EMT) based model developed by Hashin and Shtrikman (H-S) in 1962 [41] for the magnetic permeability of multiphase materials was first put forward by a research group [8, 16,18] for the prediction of thermal conductivity of nanofluids.

The Hashin and Shtrikman [41] model (bounds) can be expressed as:

$$\frac{k_p + 2k_f + 2\phi(k_p - k_f)}{k_p + 2k_f - \phi(k_p - k_f)} \leq \frac{k_{eff}}{k_f} \leq \frac{3k_f + 2\phi(k_p - k_f)}{3k_p - \phi(k_p - k_f)} \frac{k_p}{k_f} \quad (1)$$

where k_{eff} , k_p , k_f and ϕ are the effective thermal conductivity, thermal conductivity of particle, thermal conductivity of fluid, and volume fraction of particle, respectively. This model has both lower bound and upper bound cases and it assumed that the thermal conductivity of particle is higher than that of the base fluid; otherwise the upper and lower bounds would simply reverse.

The popular Maxwell model [42] has the form:

$$\frac{k_{eff}}{k_f} = \frac{k_p + 2k_f + 2\phi(k_p - k_f)}{k_p + 2k_f - \phi(k_p - k_f)} \quad (2)$$

It can be seen from Eqs. (1) and (2) that the lower bound of H-S model [41] corresponds to the Maxwell model [42].

Keblinski *et al.* [8] used these bounds of H-S model and showed that it is capable of explaining the experimentally observed thermal conductivity data for nanofluids without resorting to new mechanisms. These groups [8, 16, 18] postulated that the lower bound corresponds to a set of well-dispersed nanoparticles in a host fluid matrix while the upper bound corresponds to large pockets of fluid separated by linked or chain-like nanoparticles. It is worth noting that nanofluids with well-dispersed nanoparticles are highly desired for their stability as well as for the practical applications and thus all researchers used sonication and/or surfactant to breakdown the clustering or agglomeration in achieving better dispersions of nanoparticles in nanofluids. Recently Wang *et al.* [19] critical review conduction heat transfer mechanisms in nanofluids. They also presented their own experimental findings which demonstrated that nanoparticle clustering is the key contributor to the enhancement of thermal conductivity of nanofluids [19].

Interestingly, a round-robin exercise [43] on thermal conductivity of nanofluids involving dozens of research groups or organizations worldwide found no anomalous enhancement of thermal conductivity of various nanofluids tested in the exercise. The effective medium theory developed for dispersed particles by Maxwell [42] and its generalized expression of Nan *et al.* [44] were found to be in good agreement with the experimental data, suggesting that no special nanoscale-based model is necessary for nanofluids [43].

In contrast to this nanoparticles clustering mechanism most of the researchers [9, 25-30, 31, 33, 45] demonstrated insignificant or negative impact of clustering of nanoparticles on stability, thermal conductivity and practical application of nanofluids. Considering the fact that clusters and nanoparticles coexist in the base fluids, Xu *et al.* [46] proposed a theoretical model for the estimation of the effective thermal conductivity of nanofluids. Their model showed a decreasing of thermal conductivity with the increasing concentration of clusters.

Nonetheless, there still remain controversy and debate on heat transfer mechanisms of nanofluids. Thus more systematic and rigorous studies are to be performed to resolve this key issue of nanofluids.

TECHNIQUES TO STUDY MORPHOLOGY OF NANOPARTICLES IN NANOFLUIDS

A Transmission Electron Microscope (TEM) is commonly employed to characterize the dispersion, clustering and morphology of nanoparticles in base fluids, which are considered to be important factors influencing the heat transfer performance of nanofluids. Most of the cases the size of the nanoparticle is found to be larger than those specified by the supplier or primary particle size. This is because of the large particle density, huge number of particles, inter-particle attraction, and clustering of nanoparticles. The particle or cluster size is also measured by using Particle Size Analyzer which adopts the dynamic light scattering principle. Other electron or atomic microscopes (SEM and AFM) are also used to determine the morphology and agglomeration of dispersed nanomaterials inside the base-fluids. The crystal structures of nanoparticles are also determined using XRD analysis.

In order to obtain stable nanofluids containing well-dispersed nanoparticles, almost all researchers used sonication and/or dispersant in their nanofluids. The improved dispersion (i.e. smaller clustering) of nanoparticles due to the addition of surfactant and sonication can also be evidenced from those microscopic photographs.

DISCUSSION OF RESULTS

Effect of Shape and Loading of Nanoparticles on Thermal Conductivity

Although extensive studies have been conducted on the nanoparticle concentration dependence of thermal conductivity, except carbon nanotubes only a handful of investigations have been reported nanofluids containing other nonspherical-shaped (cylinder, flakes, ribbon etc) nanoparticles. This is mainly due to the availability of limited shapes of nanoparticles as well as difficulty in controlling the shape during synthesizing nanoparticles. Among nonspherical nanoparticles only cylindrical shape nanoparticles (also term as nanorods) are used in some studies. Here thermal conductivities of nanofluids having nanoparticles of same materials but different shape are compared and analyzed. For carbon nanotubes (CNT), single wall and multi-wall nanotubes are considered for comparison.

Xie *et al.* [47] was the first to measure the thermal conductivity of nanofluids having both spherical (average diameter of 26 nm) and cylindrical (average diameter of 600 nm) shapes SiC nanoparticles. Although the average diameter of cylindrical SiC nanoparticle was about 23 times larger than that of the spherical SiC nanoparticles, nanofluids with cylindrical SiC had higher thermal conductivity than that of

nanofluids having spherical SiC. The effect of nanoparticle shape on the thermal conductivity of nanofluids containing spherical and cylindrical TiO_2 nanoparticles in water and ethylene glycol was also investigated by the lead author [12, 15]. Figure 1 demonstrates the effect of particle shape on thermal conductivity of different nanofluids. Nanofluids having cylindrical-shape nanoparticles showed larger thermal conductivity as compared to that of nanofluids containing spherical-shape nanoparticles of same materials (Figure 1).

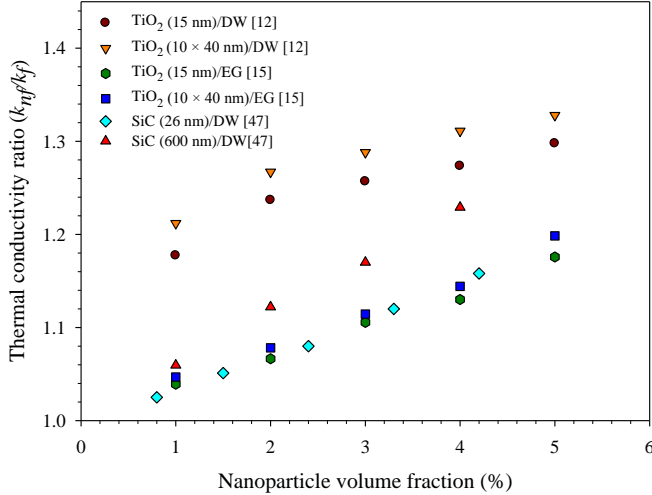


Figure 1. Effect of nanoparticle concentration and shape on the thermal conductivity of different nanofluids.

Results on concentration-dependent thermal conductivity of CNT-nanofluids from selected studies are presented in Figure 2 which shows that the significant enhancement of thermal conductivity of nanofluids with CNT concentration is obvious and base fluids as well CNT walls also play role in changing the thermal conductivity of nanofluids. It is found that nanofluids containing multi-wall CNT (MWCNT) have slightly larger thermal conductivity than that of single wall CNT (SWCNT)-nanofluids.

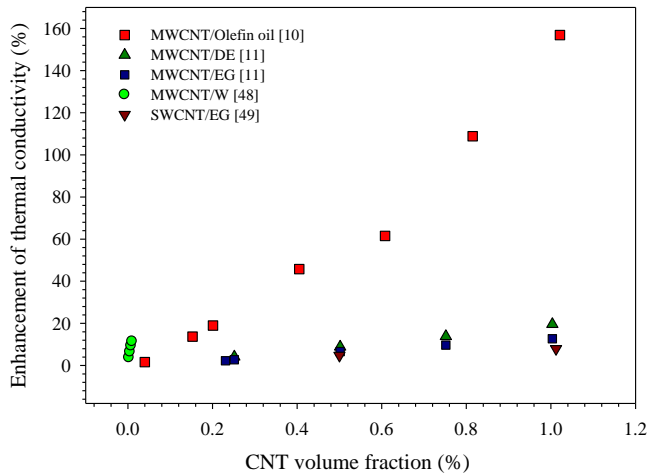


Figure 2. Concentration-dependent thermal conductivity of CNT-nanofluids with different base fluids.

A summary of representative results on thermal conductivity of various nanofluids containing different shapes and types of nanoparticles is also presented in Table 1. It also contains maximum enhancement of thermal conductivity at corresponding concentration of dispersed nanoparticles or tubes at room temperature. Although nonspherical nanoparticles particularly cylindrical nanoparticles showed larger thermal conductivity only a few studies used nonspherical nanoparticles. It is noted that most of the studies listed in Table 1 applied surfactants for better dispersion of nanoparticles and stability of nanofluids. Table 1 demonstrates that although there are some differences in the thermal conductivity enhancement results, nanofluids possess substantially higher thermal conductivity compared to their base fluids.

Effect of Clustering on Thermal Conductivity

The formation of clusters in nanofluids is almost unavoidable except for very low concentration (e.g., <0.05 vol. %) and size (e.g., <10 nm) of nanoparticles. The clustering of nanoparticles is generally known to have negative impact on the stability and the cluster or agglomeration size increases with loading of nanoparticles. Recently we investigated the effect of surfactant and clustering of nanoparticles on the thermal conductivity of aqueous nanofluids [30]. The TEM photographs depicted in Figure 3 clearly show that the cluster size increases with increasing the concentration of TiO_2 nanoparticle [30]. Such increase in cluster size is anticipated due to large number density of loaded nanoparticles. As mentioned before large-sized clusters can create large particle-free regions of higher thermal resistance in the base fluids resulting in lowering the enhancement of the thermal conductivity. However, as can be evidenced from Figure 3 that shape and size of the clusters or agglomerates are not uniform and mostly random. Zhu *et al.* [31] also reported that, the cluster size of Fe_3O_4 nanoparticles increases with increasing volume fraction of nanoparticles. Figure 4 demonstrates that the thermal conductivity of TiO_2 /deionized water (DW)-based nanofluids decreases with increasing cluster size [30]. This is in agreement with the findings of most of the related studies in the literature. For instance, Hong *et al.* [25] reported that the reduction of the thermal conductivity of nanofluids is directly related to the agglomeration or clusters of the nanoparticles. Karthikeyan *et al.* [27] showed that thermal conductivity of CuO /water-based nanofluid actually decreases with elapsed time due to clustering of nanoparticles. Shima *et al.* [33] also showed that the clustering of CuO nanoparticles in ethylene glycol increased with elapsed time which resulted in decreasing the enhancement of the thermal conductivity of this nanofluid.

Nevertheless, the structures of agglomerated or individual nanoparticles and their nanoscale or molecular level activities in the host fluids are mainly responsible for the observed anomalous thermal conductivity of nanofluids. It is important to ensure that the nanofluids have smaller clusters or interconnected large number of small clusters of nanoparticles in order for their better thermal performance.

Table 1. Summary of thermal conductivity (k) results of nanofluids containing different shapes and types of nanoparticles or tubes in various base fluids.

References	Nanofluids: NP (Size)/BF	Nanoparticle shape/wall	Measurement method	Main findings (Maximum increase in k)
Xie <i>et al.</i> [47]	SiC (26 nm)/DW	Spherical	THWM	At 4.2 vol. %, k of DW increased 16%.
	SiC (600 nm)/DW	Cylindrical		At 4vol. %, k of DW increased 22.9%.
Murshed <i>et al.</i> [12]	TiO ₂ (15 nm)/DW	Spherical	THWM	k of DW increased 29.7% at 5 vol. %.
	TiO ₂ (10×40 nm)/DW	Cylindrical		k of DW increased 32.8% at 5 vol. %.
Murshed [15]	TiO ₂ (15 nm)/EG	Spherical	TDHWM	At 5 vol. %, k of EG increased 17.6%.
	TiO ₂ (10×40 nm)/EG	Cylindrical		At 5 vol. %, k of EG increased 19.9%.
Chen <i>et al.</i> [50]	CNT (15nm×30μm)/EG	Multi-wall	THWM	k of EG increased 17.5% at 1 vol. %.
Assael <i>et al.</i> [51]	CNT (80nm×50μm) /W	Multi-wall	THWM	At 0.6 vol. %, k of W increased 38%.
Amrollahi <i>et al.</i> [49]	CNT (1-4 nm ×NA)/EG	Single wall	Steady method	At 2.5 vol. % of CNT, k of EG increased 20%.
Aravind <i>et al.</i> [52]	CNT (NA×1-3μm)/W/EG	Multi-wall	Lambda Instruments (THWM)	At 0.03 vol. % of CNT, k of water and EG increased up to 33% and 40%, respectively.
Nanda <i>et al.</i> [53]	CNT(1-1.4nm×1.4μm) /PAO	Single wall	Transient planar source technique	At 1.1 vol. % of CNT, k of PAO increased 12%.
Yan <i>et al.</i> [54]	ZnO (140×700 nm)/DW	Cylindrical	KD2 Pro (THWM)	k of DW increased 35% at 0.0022 vol.% of this nanorod.
Neogy and Raychaudhuri [55]	ZnO (10 nm)/Ethanol	Spherical	3ω technique (THWM)	k of this nanofluid increased 4.2% over the base fluid.

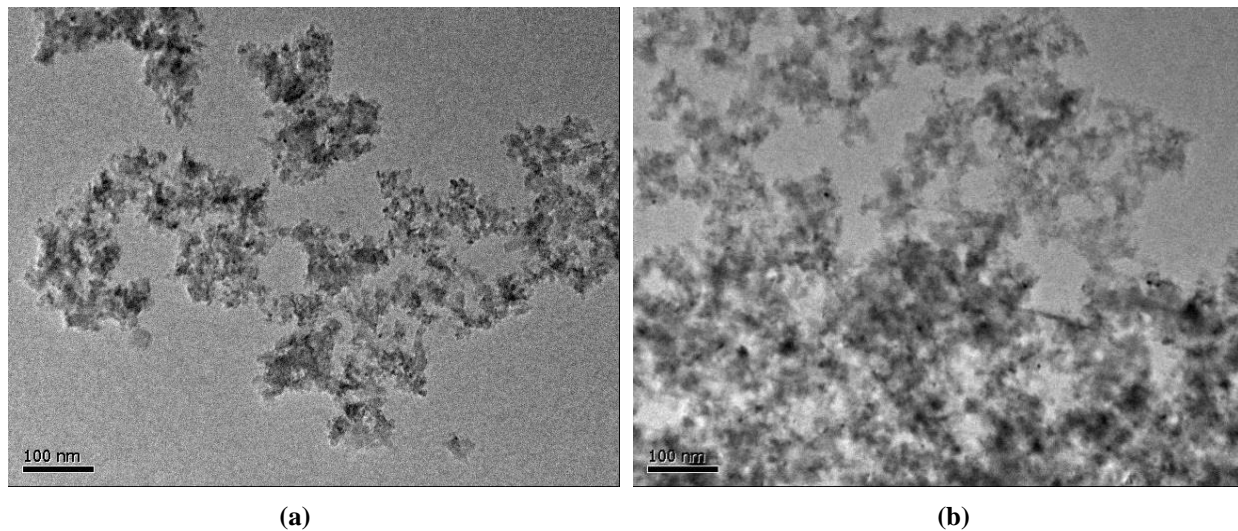


Figure 3. TEM photographs of clusters of nanoparticles at two concentrations: (a) 0.5 vol. % and (b) 1 vol. % of TiO₂ (15 nm) in deionized water.

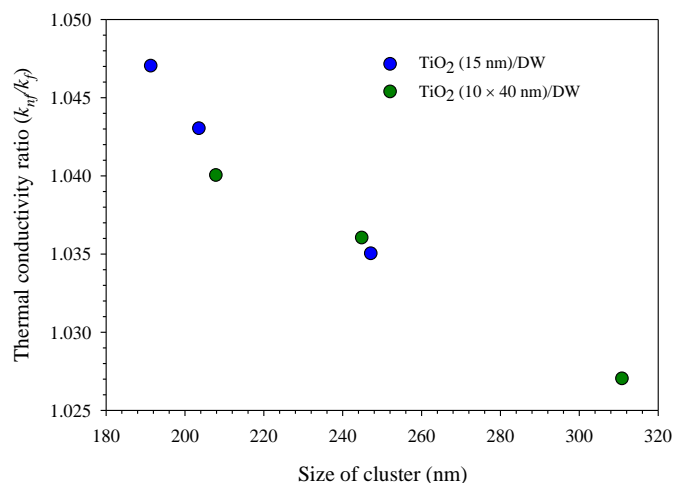


Figure 4. Effect of cluster size on the thermal conductivity of TiO₂ nanofluids.

CONCLUSIONS

Role of nanostructures in heat transfer mechanisms and anomalous thermal conductivity of nanofluids are demonstrated in this study. Results from relevant studies are also critically reviewed and compared. Heat transfer mechanisms related to nanoparticles clustering or agglomerations are discussed. Effect of concentration and shape of nanoparticles on the thermal conductivity of nanofluids are also analyzed.

It is found that regardless of inconsistencies in experimental data of the studies in the literature, nanofluids exhibit substantially higher thermal conductivity compared to their base fluids and it further increases with the concentration of nanoparticles. Despite of some studies on positive impact of clusters of nanoparticles on thermal conductivity, contrary (negative) findings and argumentation are still widely accepted. However, it is not yet understood how to have good control on the size and structures of clusters of nanoparticles and how do they influence the thermal properties of nanofluids. Furthermore, studies revealed that the primary shape or structures of nanoparticles also play role in changing the thermal conductivity of nanofluids. While nanofluids containing nanotubes of large aspect ratio are found to exhibit superior properties compared to nanofluids having any other shape of nanoparticles, nanorods (cylinder)-laden nanofluids also showed slightly larger increase in thermal conductivity than nanosphere-based nanofluids.

Results demonstrated that while the cluster size increases with increasing nanoparticles loading, the thermal conductivity of nanofluids actually decreases with increasing the cluster size.

Finally, the morphology and structures of individual and agglomerated nanoparticles and their activities can modify the thermophysical properties of the base fluids, opening the opportunity to optimize the various heat transfer performances of nanofluids.

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NOMENCLATURE

k	thermal conductivity (W/m·K)
ϕ	particle volume fraction
<i>Subscripts</i>	
f	fluid
eff	effective
nf	nanofluid
p	particle
<i>Abbreviations</i>	
DW	deionized water
BF	base fluid
CNT	carbon nanotube
DE	decene
EG	ethylene glycol
EO	engine oil
NA	not available
NP	nanoparticles
PAO	(poly)-alpha olefins
THWM	transient hot wire method
W	water

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