Low-Dimensional Nanomaterials for Nanofluids: A Review of Heat Transfer Enhancement

Norhazwani Abd Malek[1,3,*], Siti Ujila Masuri[1], Saidur Rahman[2,4], Che Nor Aiza Jaafar[1], Eris Elianddy Supeni[1], Muhammad Anis Khaliquzzama[1]

Abstract

Nanomaterials can be classified based on its dimension, in which the aspect ratio and surface-to-volume ratio increases as the dimension increases. In nanofluids, suspended nanomaterials' movement can be due to Brownian motion and thermophoresis effect, which causing heat transfer. However, the emergence of nanomaterials with various dimensions has led to more advanced heat transfer mechanisms. The high aspect ratio and surface-to-volume ratio of the nanomaterials is believed to be among the factors in nanofluids' properties enhancement. However, the morphological effect on the heat transfer enhancement in nanofluids is still ambiguous. Hence, this paper aims to explore this significant gap by reviewing the reports that investigate the effect of morphology to the heat transfer enhancement in nanofluids containing low-dimensional nanomaterials and observe the trend and gap. The numerous heat transfer mechanisms in nanofluids are discussed to improve understanding on the phenomena and its methods of study are presented for future reference. This includes the material characterizations since these approaches can provide morphological information, hence heat transfer can be studied. Heat transfer mechanisms associated with the movement of nanoparticle was the most researched mechanism, mostly by experimentations and theoretical predictions. However, there has not been substantial amount of research linking the morphological studies to the heat transfer enhancement in nanofluids. The study on phonon heat transport, nanolayer and nanoclustering, have also been made possible by recent advancements in high performance computing applications such as molecular dynamics simulation and machine learning, offering a more efficient method for exploring novel low-dimensional nanomaterials beyond zero-dimension.

*Corresponding author: Norhazwani Abd Malek (norhazwani@uniten.edu.my)

1 Department of Mechanical and Manufacturing Engineering, Universiti Putra Malaysia, 43400, Serdang, Selangor, Malaysia

2 Research Centre for Nano-materials and Energy Technology (RCNMET), School of Engineering and Technology, Sunway University, Jalan Universiti, 47500 Bandar Sunway, Petaling Jaya, Selangor, Malaysia

3 Department of Mechanical Engineering, College of Engineering, Universiti Tenaga Nasional, 43000, Kajang, Selangor, Malaysia

4 School of Engineering, Lancaster University, Lancaster LA1 4YW, United Kingdom

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List of Abbreviations

AFM	Atomic Force Microscopy	MDS	Molecular Dynamics Simulation
CFD	Computational Fluid Dynamics	MWCNT	multi-walled carbon nanotubes
CNT	carbon nanotubes	NP	nanoparticle
CP	coefficient of pressure	*Nu*	Nusselt number
CRGO	Controlled reduced graphene oxide	OA	oleic acid
Cryo-TEM	Cryo-Transmission Electron Microscopy	PG	propylene gylcol
DEG	Diethylene Glycol	*Pr*	Prandtl number
DLS	Dynamic Light Scattering	*Re*	Reynolds number
DSC	Differential Scanning Calorimetry	SEM	Scanning Electron Microscopy
DTA	Differential Thermal Analysis	STEM	Scanning Transmission Electron
DW	deionized water		Microscopy
EDX	Energy-Dispersive X-Ray Spectroscopy	SWCNT	single-walled carbon nanotubes
EEW	Electrical explosion of wire	TGA	Thermal Gravimetric Analysis
EG	ethylene glycol	TMD	Transition Metal Dichalcogenides
FoM	figure of merit	TEM	Transmission Electron Microscopy
FTIR	Fourier Transform Infrared Spectroscopy	UV-Vis	UV-Vis Spectroscopy
GNP	graphene nanoplatelet	VSM	Vibrating Sample Magnetometry
HRTEM	High-resolution Transmission Electron Microscopy	XRD	X-ray diffraction

Introduction

Nanomaterials offer a wide diversity, making it challenging to categorize them into a single group, hence demanding classification. Nanomaterials can be classified according to their dimensionality, morphology, state, and chemical composition [1]. Dimensionality-based classification can also be related to the aspect ratio (length of the major axis/width of the minor axis) and surface-to-volume ratio (surface area/volume) of the nanomaterials as different dimension of nanomaterials has different aspect ratio and surface-to-volume ratio. This will allow researchers to modify a material property of interest. Nanofluids are the suspension of nanomaterials in any conventional base fluid. When a nanomaterial is suspended in a base fluid, the high surface energy in the nanomaterials causes attractions between the particles, so the particles are subjected to forces from both the base fluid and also the nanomaterial, which causes them to be suspended and moving within the base fluid [2]. Studies reveal that nanofluids are promising heat transfer fluids for many heat transfer applications, such as in solar collectors, heat exchangers, electronic cooling, automobile radiators, thermal storage and refrigeration [3]. In fact, it is also applied in many areas of study, as seen in Figure 1. The results were extracted from the Scopus database by searching ‘‘heat transfer’’ and ‘‘nanofluids’’ in the article title, abstract, and keywords for the year 2018 – 2022 (date of extract: 12 October 2022). Hence, detailed

knowledge of heat transfer mechanisms in nanofluids is vital for those who wish to use nanofluids in their heat transfer applications.

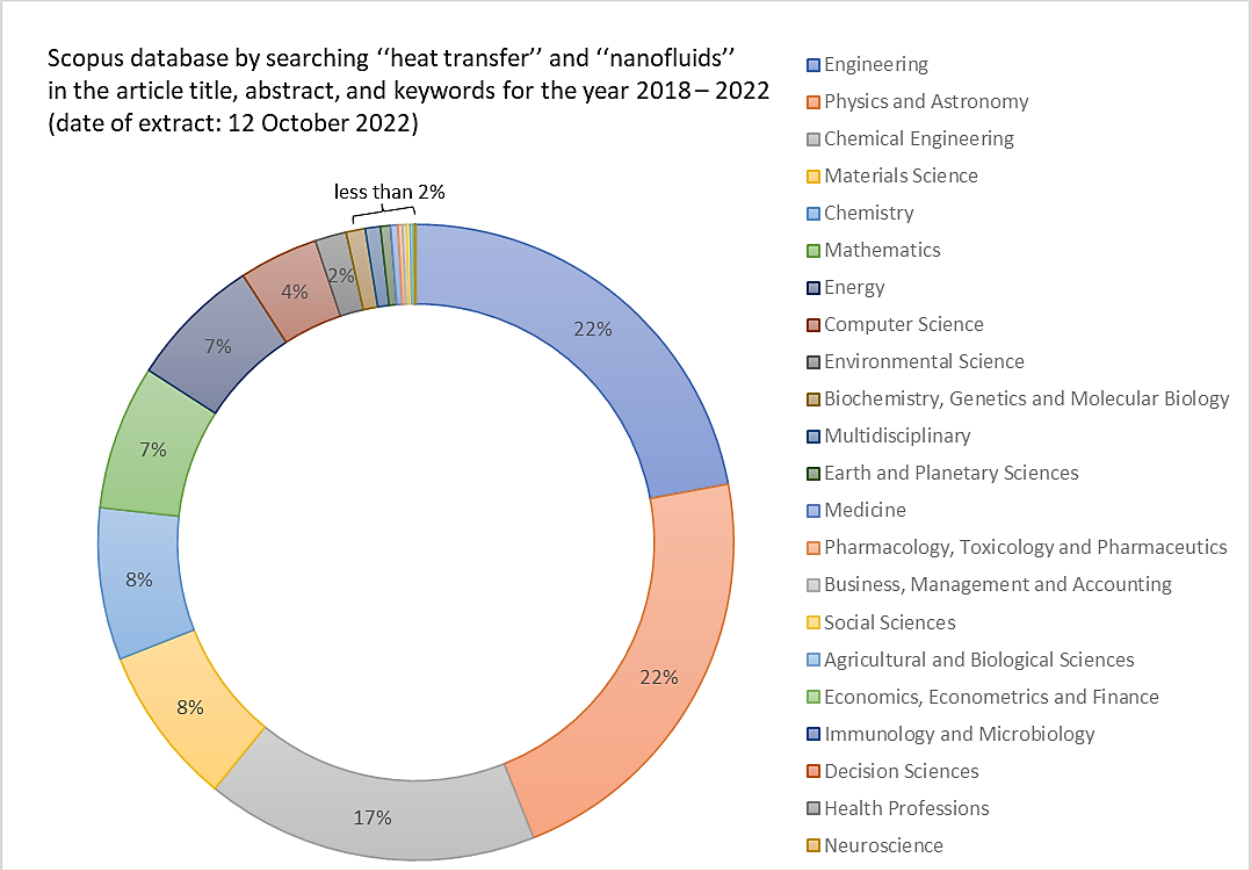


Fig. 1: Scopus database by searching ‘‘heat transfer’’ and ‘‘nanofluids’’ in the article title, abstract, and keywords for the year 2018 – 2022 (date of extract: 12 October 2022)

Based on Pokropivny and Skorokhod, nanomaterials can be classified according to their dimension, in which the same chemical compounds exhibit remarkable differences in characteristics when structured in zero-, one-, two-, and three-dimensional crystal formations [4]. Most prevalently, zero-dimensional nanomaterials are spherical nanoparticles and its size is within the nanoscale (1 – 100 nm) in all dimensions [5]. However, it can also be in the form of other shapes, such as brick, blade, platelet and cylinder [6]. For one-dimensional nanomaterials, one of its dimensions is beyond the nanoscale range, while the remaining two dimensions are within the nanoscale. Nanotubes, nanorods, nanofibers and nanowires are examples of one-dimensional nanomaterials [7]. In recent years, two-dimensional nanomaterials have attracted a lot of attentions and one of the widely studied nanomaterials. This is due to two-dimensional nanomaterials that exhibit a special morphology from which novel properties emerge and are different from other dimensions of its counterparts [8,9]. Typically, two-dimensional nanomaterials exhibit plate-like shapes with two of its dimensions are beyond the nanoscale range, while its thickness is within the nanoscale range. Hence, they can be called as nanofilms, nanosheets, nanolayers, nanoflakes, nanoplates or nanocoatings. Three-dimensional nanomaterials are not restricted to the nanoscale in any dimension. It can contain a bulk powder of

nanoparticles, bundles of nanowires and nanotubes, as well as multi-nanosheets [8]. Zero-, one- and two-dimensional nanomaterials are also called as low-dimensional nanomaterials [10]. Its schematic can be seen in Figure 2.

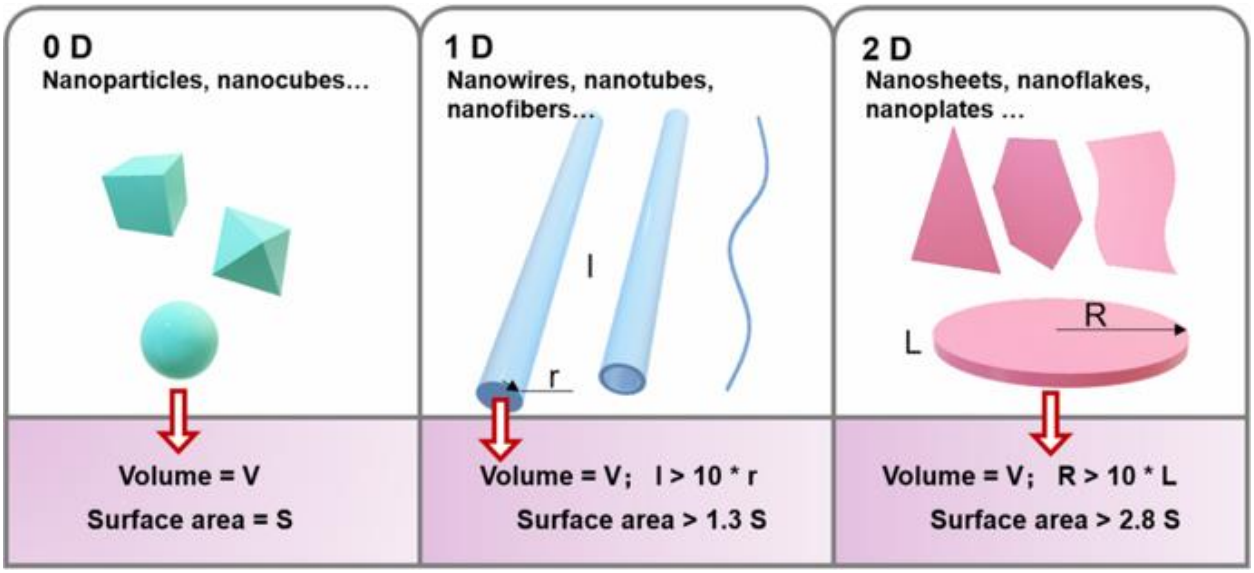


Fig. 2: Schematic of zero- (0D), one- (1D) and two-dimensional (2D) nanomaterials (reproduced from [11]) – Open Access)

The term aspect ratio is commonly used for nanorods [12]. It can also be used for two-dimensional nanomaterials with assumptions that the lateral size is the major axis and the thickness is the minor axis [13]. Meanwhile, the surface-to-volume ratio can be a useful metric in comparing all the nanomaterial dimensions. Generally, the surface-to-volume ratio will be higher for smaller diameter particles of spherical-shaped. But when comparing with other low-dimensional nanomaterials with similar volume as in Figure 3, the surface-to-volume ratio of two-dimensional nanomaterials is the highest, followed by one-dimensional and zero-dimensional nanomaterials. It is to be noted that the calculations assume the shape of the nanomaterials to be spherical for zero-dimension, cylindrical for one-dimension and cubical for two-dimensional, whereas in reality the particles are anisotropy. Based on the calculations, two-dimensional nanomaterials have higher area of interface between the particles and liquids compared to one- and zero-dimensional nanomaterials with similar volume. However, the impact of its higher area of interface to its heat transfer capability is yet to be investigated and compared with zero- and one-dimensional nanomaterials. The implications of this investigation can help in understanding the morphology of the various dimensions of nanomaterials and its potential in heat transfer systems. Moreover, the existence of some two-dimensional nanomaterials in layered structure may offer other exciting prospects to fine-tune a property of interests [14–16].

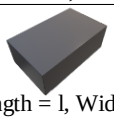
Dimension	r (nm)	l (nm)	w (nm)	t (nm)	Layer	Estimated Surface Area (m^2)	Estimated Volume (m^3)	Estimated Aspect Ratio (m^{-1})	Shape
0D	50					3.14E-14	5.24E-22	6.00E+07	 Radius = r
1D	30	186				4.07E-14	5.26E-22	7.74E+07	 Radius = r, Length = l
2D		250	210	10	1	1.14E-13	5.25E-22	2.18E+08	 Length = l, Width = w, Thickness = t

Fig. 3: Example calculation of surface-to-volume ratio of different low-dimensional nanomaterials

Previously, Ganvir et al. discussed the use of nanofluids in various heat transfer applications such as solar collectors, heat pipes, nuclear reactors, electronic cooling systems, and automobile radiators [17]. The heat transfer characteristics of the nanofluids improved compared to that of the base fluids. However, all cases were zero-dimension nanoparticles, except a few papers describing on carbon nanotubes (CNTs). Similarly, Kaggwa and Carson reviewed recent developments, challenges and future prospects for using nanofluids in heat transfer systems [18]. The paper discussed briefly the potential of suspending nanoparticles in traditional working fluids to enhance heat transfer rates by increasing thermal conductivity and heat transfer coefficients. The theoretical and experimental data of thermophysical properties such as thermal conductivity and viscosity were also presented. Ali and Salam reviewed the previous studies and recent progress in the improvement of heat transfer using nanofluid in the scope of the preparation and stability enhancement of nanofluid, thermophysical and heat transfer characteristics of nanofluid, and different factors affecting thermal conductivity [19]. Baig et al. described the advances in various types of nanomaterials, including fullerenes, carbon nanotubes, graphene, carbon quantum dots, nanodiamonds, carbon nanohorns, nanoporous materials, core-shell nanoparticles, silicene, antimonene, MXenes, metal–organic frameworks, boron nitride, layered double hydroxides, and metal-based nanomaterials [20]. However, the subject was discussed in general as it was too broad and the nanomaterials classification was not discussed in detail. In all the abovementioned review papers, no emphasis was given to the dimensions, shape or size of the nanomaterials.

Narankhishig et al. reviewed the effect of increasing the concentration and size of nanoparticles, as well as the Reynolds number to the convective heat transfer of various nanofluids [21]. Based on their review, the increased in the nanoparticle concentration did not show much effect on the convective heat transfer improvement under laminar and turbulent flow conditions, but the use of smaller-sized nanoparticles enhanced the convective heat transfer. However, the size of some of the reported nanoparticles was greater than the nanoscale, indicating the importance of classification of nanoparticles based on dimensions. In addition to that, increasing the Reynolds number in a turbulent flow region did not enhance the heat transfer coefficient of the nanofluids. Zahmatkesh et al. critically reviewed the effect of nanoparticle shape on the hydrothermal performance of thermal systems utilizing nanofluids [22]. The review indicates that the control of nanoparticle shape is a promising technique for the optimization of heat exchange and the

required pumping power, but no uniform conclusion has been reached for the role of nanoparticle shape on the hydrothermal performance of thermal systems. Paras et al.'s review paper provided a comprehensive guide to produce low-dimensional nanomaterials, namely nanoparticles (zero-dimensional), nanorods, nanowires, and nanobelts (one-dimensional), and atomically thin layered materials (two-dimensional), including their characterization [10]. However, no comparison was done on their heat transfer enhancement. Additionally, the paper highlighted various significant applications of low-dimensional nanomaterials such as photonics, sensors, catalysis, energy storage, diverse coatings, and various bioapplications, but it focused on the possibility of two-dimensional transition metal dichalcogenides (TMD) nanomaterials for sensitive chemical sensing or biosensing only. Recently, Li et al. reviewed the thermal conductivity and heat transfer enhancement of nanofluids with non-spherical nanoparticles only. The nanoparticles were categorized based on three types of geometric particle structure, namely CNTs, nanofibers and nanowire (one-dimensional), nanorods and ellipsoids (two-dimensional) and platelets, blades, bricks, diamonds, polygons (arbitrary shapes) [23]. The paper also discussed the numerical predictions and related heat transfer mechanisms. By incorporating non-spherical nanoparticles into the base fluid, the convective heat transfer coefficient was enhanced, but no significant thermohydraulic performance enhancement for convective heat transfer was obtained, specifically for turbulent flows due to increased pumping power. Despite that, the effect of the surface-to-volume ratio of low-dimensional nanomaterials to the heat transfer enhancement in nanofluids is still unclear and need to be discussed further, especially in the flowing fluid in heat transfer applications.

Motivated by the abovementioned review papers, the aim of the current review is to investigate the effect of morphology to the heat transfer enhancement in nanofluids containing low-dimensional nanomaterials, specifically in the scope of convective heat transfer in internal flow in heat transfer applications. Prior to that, the various mechanisms reported in the literature that contribute to the heat transfer enhancement in nanofluids will be discussed. Additionally, characterization techniques of nanomaterials and nanofluid will also be discussed because the material characterizations can give information about the morphology of the material, hence the heat transfer enhancement can be studied. According to the author's knowledge, this kind of review is the first time reported. The information from this review paper can aid the researchers in making better selection of nanomaterials according to their heat transfer enhancement, particularly for heat transfer applications such as in solar collector and heat exchanger. Nevertheless, the thermohydraulic performance is not covered in great length in this review as to focus and narrow the field.

Heat Transfer Mechanism in Nanofluids

Despite the various potential benefits of nanofluids for heat transfer applications, there remains a need for ongoing research to accurately understand the heat transfer mechanism in nanofluids due to continuous emerging nanomaterials.

Movement/Motion of Nanoparticles

Even though the thermal conductivity enhancement in nanofluid is essential for its advantage over conventional fluids, the more appropriate means of evaluating the heat transfer enhancement comes from the evidence of the performance of nanofluids in convective environment. Moreover, in heat transfer systems that involves flowing fluids, such as heat

exchangers, radiators, or cooling systems, the thermohydraulic performance is desirable to determine the overall efficiency and effectiveness of the heat transfer process. Among other parameters that need to be included are the heat transfer rate, pressure drop, fluid flow rate, and temperature. Nusselt number, *Nu* and heat transfer coefficient can be used as a useful metric for analytical investigations. Nusselt number is the dimensionless parameter which characterizes the ability for heat transfer of a fluid in various flow regime as a result of convection relative to conduction across the same fluid layer. It can be used to compare a real situation, such as heat transfer in a pipe or channel in a small-scale model. In principle, the Nusselt number is not a fluid constant, but is it is largely influenced by the flow's parameters, which are represented by other dimensionless parameters, such as Prandtl number, *Pr* and Reynolds number, *Re*. These dimensionless parameters also rely on some transport and thermophysical properties and the geometry of the pipe/channel. Therefore, to investigate the potential heat transfer of a nanofluid, its thermophysical properties must be known, but to prove the heat transfer enhancement solely using these properties are not enough.

The heat transfer characteristics of nanofluids can be predicted by employing two main approaches, namely single-phase model and the two-phase model. Single-phase model assumes nanofluid as homogeneous and Newtonian with modified thermophysical properties from the base fluid, and there is no slip between the particles and the base fluid. One of the key mechanisms of enhanced thermal conductivity in nanofluids is microconvection due to Brownian motion at the nanoscale levels as suggested by Jang and Choi [24]. Brownian motion is the erratic movement or motion of particles driven on by random thermal fluctuations. Similar to the kinetic theory of particles, Brownian motion is proportional to the temperature. As suggested by Koo and Kleinstreuer [25], collisions between the nanoparticles lead to the heat exchange among the nanoparticles, which then resulting in an increase in the thermal conductivity. Many theoretical and experimental studies on nanofluid heat transfer have been reported in the literature and some new correlations have been proposed as the traditional correlations such as Dittus-Boelter [26], Shah and London's [27] and Gnielinski's [28] cannot predict the average Nusselt number correctly. Among the pioneer works were attributed to Pak and Cho [29] and Xuan and Li [30]. The convective heat transfer behaviors of γ-alumina (γ-Al_2O_3)/water and titanium dioxide (TiO_2)/water nanofluids heated with constant heat flux in a fully developed turbulent flow in circular stainless steel tube were experimentally studied by Pak and Cho [29]. It was reported that, the Nusselt number of the nanofluids increased with increasing volume concentration, as well as the Reynolds number. However, the convective heat transfer coefficient of the nanofluid was 12% smaller than that of water at a volume fraction of 3% under the same average velocity A new Nusselt number correlation was also proposed. Xuan and Li suspended copper (Cu) nanoparticles in water and the heat transfer coefficient of the nanofluid enhanced as compared to that of water under the same Reynolds number [30]. The heat transfer characteristics of the nanofluid also increased with the volume fraction of the nanoparticles. A new Nusselt number correlation that takes into account the microconvection and microdiffusion effects of the suspended nanoparticles was also developed. Despite their limited considerations of the nanofluid properties, Pak and Cho's and Xuan and Li's model serve as useful references to develop other advanced heat transfer models.

The remarkable enhancement in the thermal conductivity of nanofluids in the early years of its discoveries has led to excitement and interest in the potential applications of nanofluids for thermal management and heat transfer

applications and it continues until now. Leela Vinodhan et al. observed a heat transfer enhancement when using CuO/water nanofluid as a coolant in laminar regime through a U-shaped minitube as compared to water [31]. The key factors contributing to the enhancement were the improved thermal conductivity and the particle movement without particle deposition. However, it was also suggested that the existence of an optimum nanoparticle concentration may be attributed to the enhancement, where in this case was 0.05 wt%. Abdolbaqi et al. investigated the convection heat transfer enhancements of BioGlycol/water-based TiO_2 nanofluid in a flat tube with a uniform heat flux under turbulent flow conditions [32]. The heat transfer coefficient and friction factor were determined experimentally and new correlations were obtained for both parameters using non-linear models in ANOVA. However, the Nusselt number was approximately 3% lower than that of the base fluid for volume fraction 2% at 30 °C. The reason for this decrease in the Nusselt number at this specific condition is not explicitly discussed in the paper, but it could be due to the fact that the increase in concentration causes an increase in the viscosity, which could affect the flow behavior and heat transfer performance of the nanofluid. The optimum nanoparticle concentration may also be the reason for such behavior. However, further investigations are required to confirm this hypothesis. Agromayor et al. determined the heat transfer coefficients and the pressure drops of sulfonic acid-functionalized graphene/water nanofluids [33]. The convection coefficients increase with the flow rate and the temperature of the fluid. The increment was obvious for all volume fraction less than 0.79%. The pressure loss increased slowly with the nanoparticle concentration as a consequence of the viscosity increase. New correlations to describe the Nusselt number and the friction factor were introduced.

Single-phase model has been used by Saha and Paul to investigate the flow characteristics of alumina (Al_2O_3)/water and titania (TiO_2)/water nanofluids in a horizontal pipe under constant heat flux numerically [34]. As a result, the heat transfer enhancement is found to be dependent on the concentrations of the nanoparticles and flow regime, but not much difference observed with the size of the nanoparticles. Some new correlations have been proposed using non-linear regression analysis to calculate the average Nusselt number. However, the proposed correlations are not generic as it may not valid for Reynolds numbers below 2000. Ramirez-Tijerina et al. employed single-phase in a mathematical model to investigate the laminar forced convection for the flow of nanofluids in conventional straight tube and straight microtube under constant temperature and constant heat flux conditions [35]. The study varied three different types of base fluids including water, ethylene glycol, and turbine oil with five different types of nanoparticles, namely Al_2O_3, TiO_2, CuO, SiO_2 and ZnO. The results showed that the Nusselt number increased with Reynolds number and considerably enhanced with the increase in the nanoparticle concentrations. New Nusselt number correlation was developed for the flow of nanofluids in conventional straight tube and straight microtube. Additionally, the single-phase model was compared with the single-phase dispersion model, in which the fluid was still considered as a single phase but the properties of the fluid were not uniform, but to be dispersed throughout the domain. The single-phase dispersion model was found to be more accurate than the single-phase model. Khanjari et al. employed conjugate heat transfer in the computational fluid dynamics (CFD) simulation using ANSYS Fluent software to evaluate the effects of Al_2O_3/water nanofluids as coolants in a photovoltaic/thermal (PV/T) collector system [36]. The results were validated and compared with water. It was revealed that increasing the nanoparticle

concentration increased the heat transfer coefficient, as well as the efficiency of the PV/T system. Furthermore, the heat transfer performance of the PV/T system was increased by increasing the pipe inlet fluid velocity.

Despite the simplifications that the single-phase models offer, such as ignoring some interaction forces and assuming that the nanoparticles to be in thermal equilibrium with the base fluid, distributed uniformly within the base fluid and move with the same velocity as the bulk base fluid particles, the results are sometimes limited to certain range. To tackle these discrepancies, the two-phase model or non-homogeneous nanofluid model can be used. Two-phase model considers the nanofluid as a mixture of two distinct phases and takes into account the interactions and dynamics between these two phases. Realizing that the boundary layer effect is one of the important factors influencing the heat transfer characteristics, Buongiorno proposed the concept of nanoparticle slip mechanisms in nanofluids, which refers to the relative motion between nanoparticles and the base fluid [37]. According to his work, the major slip mechanisms in nanofluids were caused by seven transport mechanisms, but Brownian motion and thermophoresis (as illustrated in Figure 4) were more dominant than the others. Thermophoresis causes nanoparticle movement by following a temperature gradient, resulting in a non-uniform distribution of nanoparticle volume fractions. The higher-heat-affected nanoparticles will flow to the lower-heat-affected nanoparticles. The viscosity inside the boundary layer reduced dramatically as a result of this impact, resulting in enhanced heat transfer.

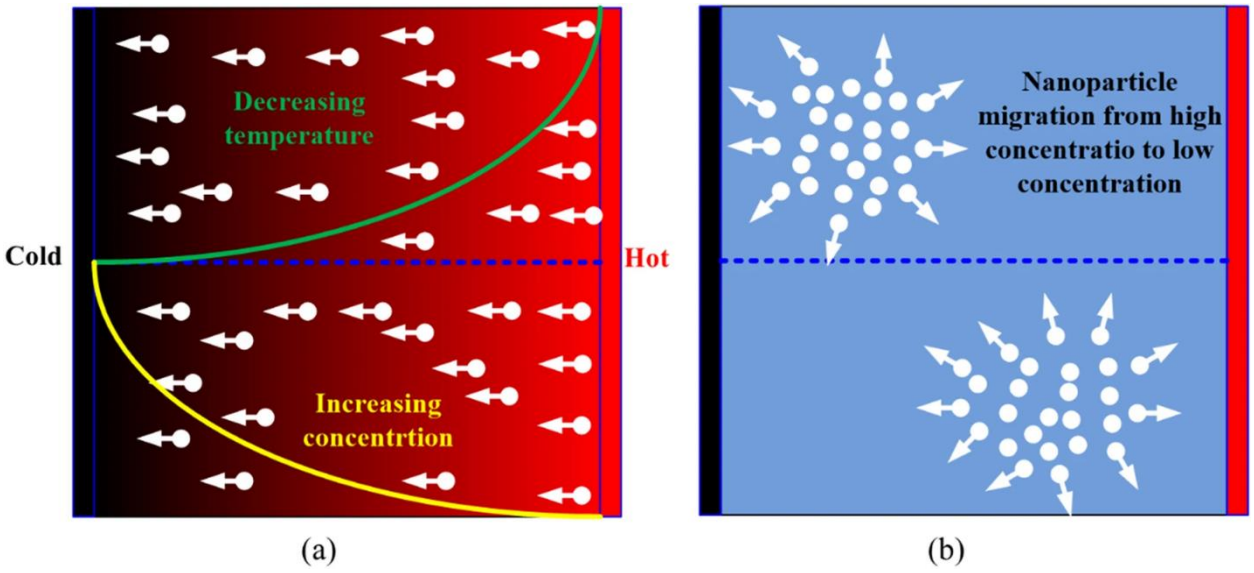


Fig. 4: An illustration of nanoparticle slip motions: (a) thermophoresis; (b) Brownian motion ((reproduced from [38] – Open Access)

There are many other recent papers which deal with the Buongiorno's model and incorporated the effects of Brownian motion and thermophoresis in the energy equation under the boundary layer theorem. Sayyar et al. adopted Bungiorno's model in a numerical study to assess the forced and natural convective heat transfer of laminar developing flow of Al_2O_3/water nanofluid inside a vertical tube [39]. The results showed that the distribution of nanoparticles remained almost uniform except at the region near the hot wall. At this region, the thin boundary layer of nanoparticles reduced drastically, probably due to thermophoresis effect. Meanwhile, the flow behavior of two-dimensional

graphene/water nanofluids with small amount of polyvinylpyrrolidone (PVP) as surfactant in a heated circular copper pipe were comprehensively studied by Demirkir and Erturk by experiment [40]. Surfactant is a kind of coatings to the nanoparticles to maintain the dispersion of nanoparticles in the base fluid [41]. A significant enhancement of the Nusselt number was observed beyond laminar flow regime with a maximum of 36% heat transfer enhancement obtained at a Reynolds number of 3950 compared to that of using water. However, the pressure drop is higher in the transitional region compared to laminar and turbulent region. For laminar flow, the local heat transfer coefficient enhancement merely did not vary with Reynolds number but increased as the nanoparticle mass concentration increased. Considering that the increase is comparable to that of thermal conductivity, Brownian motion thermophoresis are expected to be the reason for the enhancement. Scale analysis were done to compare the effect of Brownian motion and thermophoresis beyond laminar flow regime. As a result, thermophoresis effect is greater than the Brownian motion effect beyond laminar flow regime. Additionally, it was noticed that when particle concentration increased, the onset of the transition from laminar flow shifted to lower Reynolds numbers. For turbulent flow, a significant improvement in the heat transfer coefficient was observed.

Many studies and research have explored and modified Buongiorno's work in several ways, including Yang et al., who modified the Buongiorno's model equations for convective heat transfer in nanofluids by considering the effects of nanoparticle volume fraction distributions on the continuity, momentum, and energy equations [42]. Exact solutions for fully developed laminar flows of nanofluid in channels and tubes were reported. The unusual heat transfer enhancement was identified due to the relatively low volume percentage of nanoparticles close to the wall. As a result, there was a dramatic temperature variation along with elevated velocity close to the wall. Furthermore, it was found that the heat transfer enhancement was higher in the tube compared to that of the channel due to the radial effects. By using Yang's model, Malvandi et al. found that the nanoparticle movement had a significant impact on the flow fields and heat transfer rate [43]. Smaller nanoparticles had a lower thermophoresis strength because they were able to gather at the heated wall and increase the heat transfer rate. On the other hand, larger nanoparticles were depleted at the heated walls, reducing thermal conductivity and restricting a significant increase in heat transfer rate. Goyal and Sharma estimated the forced convective heat transfer characteristics of nanofluids in stainless steel tube by using two different carbon-based nanomaterials, namely multi-walled carbon nanotubes (MWCNTs) and multi-walled carbon nano coils (MWCNCs) [44]. In both cases, the heat transfer coefficient increased as the volume fraction of the carbon nanomaterials increased. The enhancement can be explained by the high thermal conductivity and large aspect ratio of the carbon nanomaterials. It was also more predominant in the entry length region, where the thermal boundary layer was developed. Thus, the inclusion of boundary layer in the analysis is vital.

Alsarraf et al. employed the two-phase mixture model to evaluate the turbulent forced convection of molybdenum disulfide (MoS_2)/water nanofluid flow through a flat plate solar collector tube [6]. According to the paper, the Nusselt number increases with increasing solid volume fraction and mass flow rate. The results were also validated using some other established experimental data. Amiri et al. used two-phase Eulerian-Eulerian model to investigate the heat transfer of CuO nanofluid in microchannel [45]. In comparison to pure water, the heat transfer enhancement was observed in the nanofluid as the Reynolds number and nanoparticle volume concentration increase.

At the same time, pressure drop increased slightly. The heat transfer also increased with the decrease in the nanoparticle diameter. In both papers, the governing mass, momentum and energy equations for both phases were solved using the finite volume method (FVM) in ANSYS Fluent.

The new correlations based on the studies discussed earlier for the heat transfer enhancement of nanofluids in laminar to turbulent flows reported in the abovementioned papers are presented in Table 1. The dimensionless governing parameters for Reynolds and Prandtl numbers used in the correlations are given as follows:

$$Re = \frac{\rho u D}{\mu} \tag{1}$$

$$Pr = \frac{c_p \mu}{k} \tag{2}$$

It is to be noted that the correlations suggested may not be applicable to different flow regimes, as well as they are not generalizable to other base fluids and nanoparticles. Some of the studies do not have new correlations as the accuracy of the results are validated by some well-known correlations correctly. This is primarily due to the specific characteristics and conditions of the of the data being studied, such as non-conventional geometries being used or complex phenomena of novel materials. Traditional correlations are typically developed for common geometries, such as circular tubes. Furthermore, heat transfer in nanofluids goes beyond just Brownian motion and thermophoresis, but it also involves a complex interaction of various mechanisms, such as fluid-particle and particle-particle interactions. As research methodologies, computational approaches, and experimental capabilities evolve, new insights may be discovered and new correlations may be developed to address the limitations and improve the accuracy of the predictions.

Table 1: Traditional correlation and correlations developed from experimental or numerical study of heat transfer enhancement in nanofluids

Author(s) (Year)	(Dimensionality) Nanoparticle (NP)/ base fluid	Size	Volume Fraction	Geometry	Other Conditions	Proposed Correlations
Dittus-Boelter [29]	-	-	-	Circular pipe/tube	$Re \geq 10000$, $Pr = 0.7 – 160$ $L/D \geq 10$	$Nu = 0.023Re^{0.8}Pr^{b}$ (Note: b is 0.4 for heating and 0.3 for cooling)
Shah and London (1978) [27]	-	-	-	Circular pipe/tube	$Re < 2300$	$Nu = \begin{cases} 1.953\left(\frac{1}{L^*_{th,d}}\right)^{\frac{1}{3}} \; for \; L^*_{th,d} \leq 0.03 \\ 4.36 + 0.0722\left(\frac{1}{L^*_{th,d}}\right) \; for \; L^*_{th,d} > 0.03 \end{cases}$ (*$L_{th,d}$ = dimensionless thermal length based on the pipe diameter)
Gnielinski's (2013) [28]	-	-	-	Circular pipe/tube	$Re = 2300 – 10^6$, $Pr = 0.5 – 2000$	$Nu = \frac{(f/8)(Re - 1000)(Pr)}{1 + 12.7\sqrt{\frac{f}{8}}(Pr^{2/3} - 1)}$
Pak and Cho (1998) [29]	(0D) γ-Al_2O_3/water, TiO_2/water	Diameter = 13 nm (γ-Al_2O_3), 27 nm (TiO_2)	0 – 3%	Circular pipe/tube	$Re = 10^4 – 10^5$, $Pr = 6.5 – 12.3$	$Nu_{nf} = 0.021Re_{nf}^{0.8}Pr_{nf}^{0.5}$
Xuan and Li (2003) [30]	(0D) Cu/water	Diameter < 100 nm	0 – 2%	Circular pipe/tube	$Re = 1 \times 10^4 – 2.5 \times 10^4$	$Nu_{nf} = 0.0059(1.0 + 7.6286\phi^{0.6886}Pe_p^{0.002})Re_{nf}^{0.9238}Pr_{nf}^{0.4}$
Leela Vinodhan et al. (2016) [31]	(0D) CuO/water	Diameter = 20 – 30 nm	0 – 0.1	U-shaped minitube	$Re = 322 – 2293$	$Nu_w = 0.1446Re^{0.3735}Pr^{1/3}$
					$Re = 448 – 2001$	$Nu_{0.025\,wt\%} = 0.3403Re^{0.2708}Pr^{1/3}$

					Re = 498 – 2258	$Nu_{0.05\,wt\%} = 0.316Re^{0.2882}Pr^{1/3}$
					Re = 367 – 1993	$Nu_{0.1\,wt\%} = 0.2017Re^{0.3313}Pr^{1/3}$
Abdolbaqi et al. (2017) [32]	(0) TiO_2/BioGlycol + water	Diameter = 50 nm	0 – 2%	Flat tube/duct	*Re* = 3300 – 37400, *Pr* = 5.75 – 13.28	$Nu_{nf} = 0.023Re_{nf}^{0.847}Pr_{nf}^{1/3}(f_r/8)^{0.486}\left(0.1 + \frac{\phi}{100}\right)^{-0.309}$ $f_{nf} = 0.366Re_{nf}^{-2.65}\left(\frac{\rho_{nf}}{\rho_{bf}}\right)^{-0.392}\left(\frac{\mu_{nf}}{\mu_{bf}}\right)^{0.317}$
Agromayor et al. (2016) [33]	(2D) sulfonic acid-functionalized graphene + water	-	0 – 0.75%	Annular tube	*Re* = 5×10^3 – 4×10^4, *Pr* = 4.8 – 10.8 Pr_{nf}/Pr_{wall} = 1.06 – 1.36	$Nu_{nf} = 0.023Re_{nf}^{0.847}Pr_{nf}^{1/3}(f_r/8)^{0.486}\left(0.1 + \frac{\phi}{100}\right)^{-0.309}$
Saha and Paul. (2018) [34]	Al_2O_3/water	Diameter = 10 – 40 nm	0 – 6%	Circular pipe/tube	*Re* = 2300 – 10×10^3 *Pr* = 8.45 – 20.29	$Nu_{nf} = 0.03945Re_{nf}^{0.76746}Pr_{nf}^{0.24025}\left(\frac{d_f}{d_p}\right)^{-0.0004483}$
	TiO_2/water					$Nu_{nf} = 0.03930Re_{nf}^{0.76746}Pr_{nf}^{0.24025}\left(\frac{d_f}{d_p}\right)^{-0.0007074}$
Ramirez-Tijerina et al. (2018) [35]			0 – 10%	Straight tube and microtube	*Re* = 25 – 1500 *Pr* = 6 – 500	$Nu = 0.04381Re^{0.36}Re^{0.42}$

Liquid layering at the base fluid/nanoparticle interface

Liquid layering at the base fluid/nanoparticle interface occurs when the base fluid particles come into contact with the nanoparticles, owing to the intermolecular interactions between the nanoparticle core surface and the base fluid, such as the van der Waals force and the Coulomb force [46]. As seen in Figure 5, this interfacial layer is called as nanolayer and has different thermophysical properties from the bulk base fluid and the nanoparticle [47–50]. If surfactant is added to the nanofluid, it will also act as the interfacial layer [51]. The influence of liquid layering on convective heat transfer in nanofluids is a complex phenomenon that is often not reflected by straightforward correlations. Thus, the fundamental challenge of measuring nanolayer thickness still requires further study.

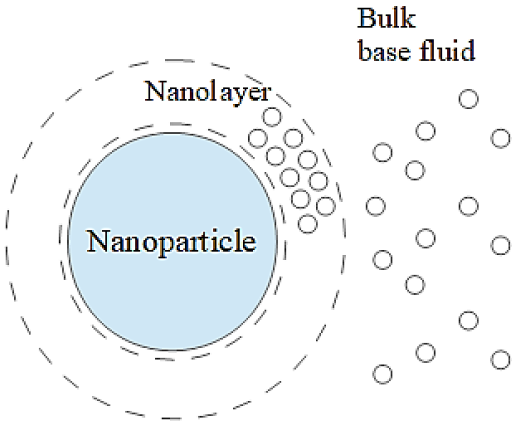


Fig. 5: Liquid layering at the base fluid/nanoparticle interface (redrawn from [46] and [48] – Open Access)

Mamand et al. studied the effect of the liquid nanolayer thickness around Al_2O_3, CuO, ZnO, and SiO_2 in water and ethylene glycol to the thermal conductivity of the nanofluids [52]. The paper also included the effect of the nanoparticles' surface specularity parameter by adjusting the liquid nanolayer thickness by assumption and not by experiment. Despite good agreement with some established data obtained, the accuracy of the results may be affected by this assumption and the effect of other factors, such as particle size distribution, particle concentration, and temperature on the thermal conductivity of nanofluids was also not included. Fan and Zhong predicted the nanolayer thickness in Al_2O_3/water and TiO_2/water nanofluids and proposed a new theoretical model of thermal conductivity that includes Brownian motion and nanolayer [46]. According to the authors, the temperature and nanoparticle concentration influenced the thickness of the nanolayer, by which it was approximately proportional to temperature, and distributed in a power law as the nanoparticle concentration increased. The proposed model may not valid for other types of nanoparticles and base fluids, while the effect of the nanolayer to the heat transfer enhancement remains unpredicted. In other research by Jozwiak et al., the carbon nanotube (CNT) ionanofluids images obtained from cryo-Transmission Electron Microscopy (cryo-TEM) displays the interfacial nanolayers of the base ionic liquid around two different types of CNT, as seen in Figure 6 [13]. The thickness of the nanolayers can be obtained from the cryo-TEM images, but the nanolayer's thermal conductivity remains an issue as it is hardly obtained from experiment.

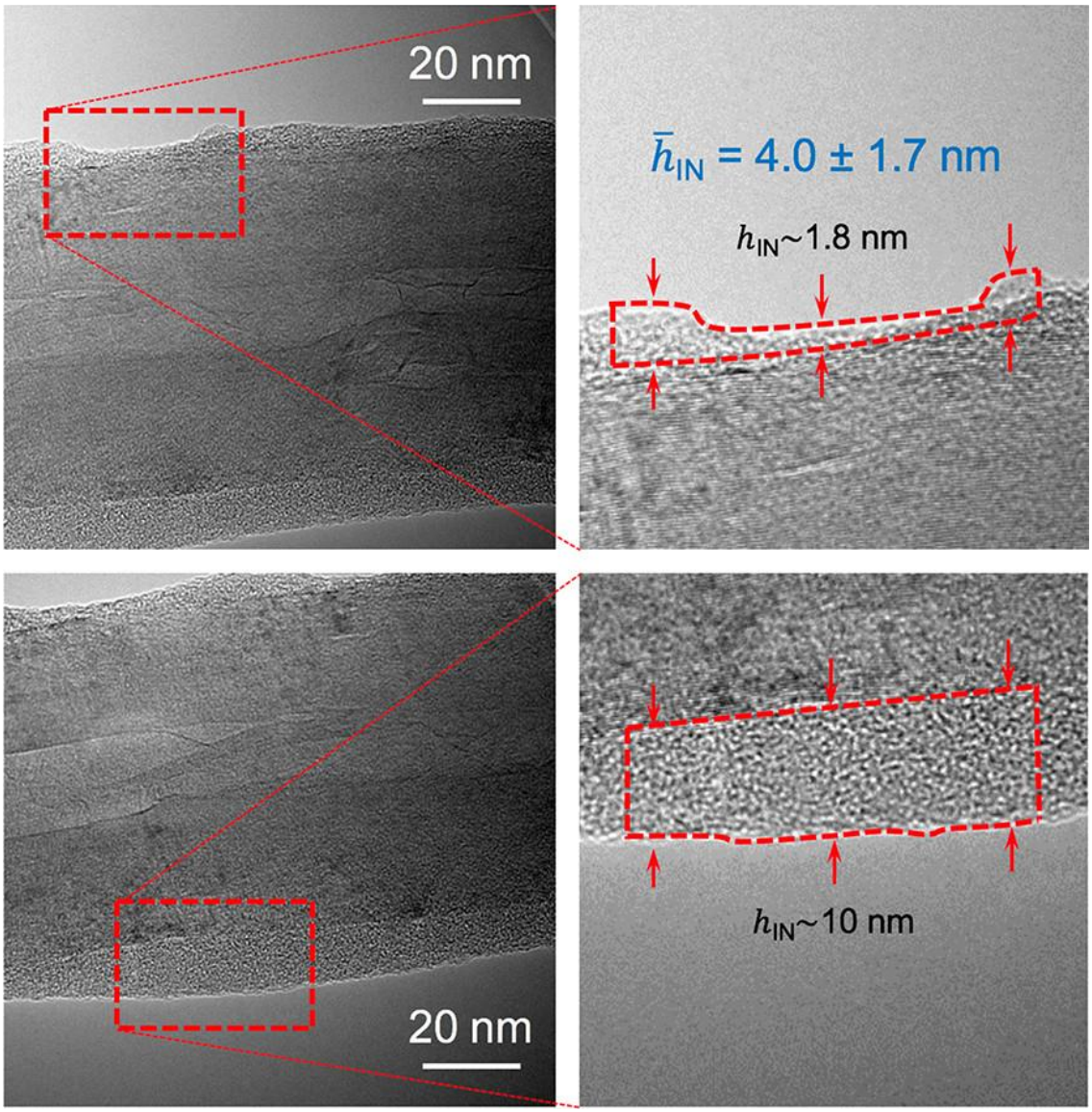


Fig. 6: The interfacial nanolayers of the ionic liquid for two different nanotubes. The indicated areas highlight the interfacial nanolayer of varying thickness (reproduced from [13] – Open Access)

Recent development in high performance computing applications has given light to investigate the heat transfer mechanisms in nanofluids due to the effect of nanolayer numerically. Because the heat transfer process in nanofluids is better explained by using discrete particle models rather than continuum approaches, molecular dynamics simulation (MDS) may be used to explore the characteristics of the nanolayer numerically. MDS describes the atomic mobility within a material, in which interatomic forces cause the atoms to interact and move in certain periods of time. Milanese et al. [53] studied the layering phenomenon as the water molecules surrounded the copper (Cu) and copper oxide (CuO) metal nanoparticles by means of MDS. The software used by the authors is Amsterdam Density Functional (ADF) in Software for Chemistry & Materials (SCM). The numerical findings discovered the two layers of water molecules near the Cu nanoparticle surface, while no substantial layering on the CuO nanoparticle. This might explain the higher thermal conductivity of Cu-based nanofluid compared to CuO-based nanofluid as measured experimentally in their work. Zhai et al. introduced a new parameter that is called as N_δ, which defines as the ratio of

number density, N and its corresponding thickness of interfacial nanolayer, δ to evaluate the relationship between interfacial layer and thermal conductivity of nanofluid [54]. δ value was obtained from MDS. A higher N_δ value indicates a denser distribution of base fluid molecules in the interfacial nanolayer. The highest N_δ and thermal conductivity were obtained in CuO-based nanofluid, followed by Al_2O_3- and Cu-based nanofluids. The contradicted results for Cu-based nanofluid and CuO-based nanofluid compared to Milanese et al.'s [53] might be due to the smaller amount of nanoparticle loading used, which was between 0.5 to 2.0% volume concentration, whereas Milanese et al. was 3%. The size of the CuO nanoparticle for each paper was also different. In addition, no information obtained from the literature on the nanolayer effect in the two-dimensional nanomaterials, possibly due to the ultrathin thickness of most two-dimensional nanomaterials [16,55], hence the prediction is more complex. However, further investigations need to be done to confirm the hypotheses.

Effects of nanoparticle clustering or aggregation

It is undesirable, but some nanoparticles will stick together, creating nanoparticle clustering or aggregation. Brownian motion may influence in the clustering of nanoparticles indirectly. An increase in thermal conductivity can take place if the nanoparticles are not in physical contact but just close enough to allow rapid heat transfer between them. If the clustering is organized, the rate of heat transfer can be increased significantly, as suggested by Prasher et al. [56]. Evans et al. suggested that a linear nanoparticle chain allows heat to be transferred through the nanofluid at a greater rate [57]. However, agglomeration of nanoparticles in the base fluid can occur if the Van der Waals attractions between the particles are strong.

Elsaidy et al. examined the thermal conductivity of aqueous nanofluids containing iron oxide (Fe_3O_4 and γ-Fe_2O_3) nanoparticles in order to study the effect of controlled aggregation of nanoparticles by altering the content of polyethylene glycol (PEG) in the reaction medium [58]. The clusters of iron oxide nanoparticles of different cluster size in the range of 46 – 240 nm diameter were produced as shown in Figure 7. The results showed that the thermal conductivity increased as the cluster size increased with the enhancement between 1 – 5%, while the viscosity was insignificantly higher as compared to water. Despite the thermal conductivity enhancement due to the clusters' existence, the stability of the nanofluids based on the Zeta potential measurements was moderate. Zheng and Wang proposed a prediction model for effective thermal conductivity of nanofluids by considering both the agglomeration effect and the radial distribution function of nanoparticles [59]. The theoretical predictions for several sets of nanofluids showed good agreement with experimental data, indicating the anomalous thermal conductivity due to agglomeration effect and the radial distribution. However, the model assumed one level of agglomeration in the suspension, which may not be true in some cases. Further research is needed in this sense. Moreover, the aforementioned papers did not study the heat transfer enhancement due to clustering or aggregation, but only the thermal conductivity enhancement.

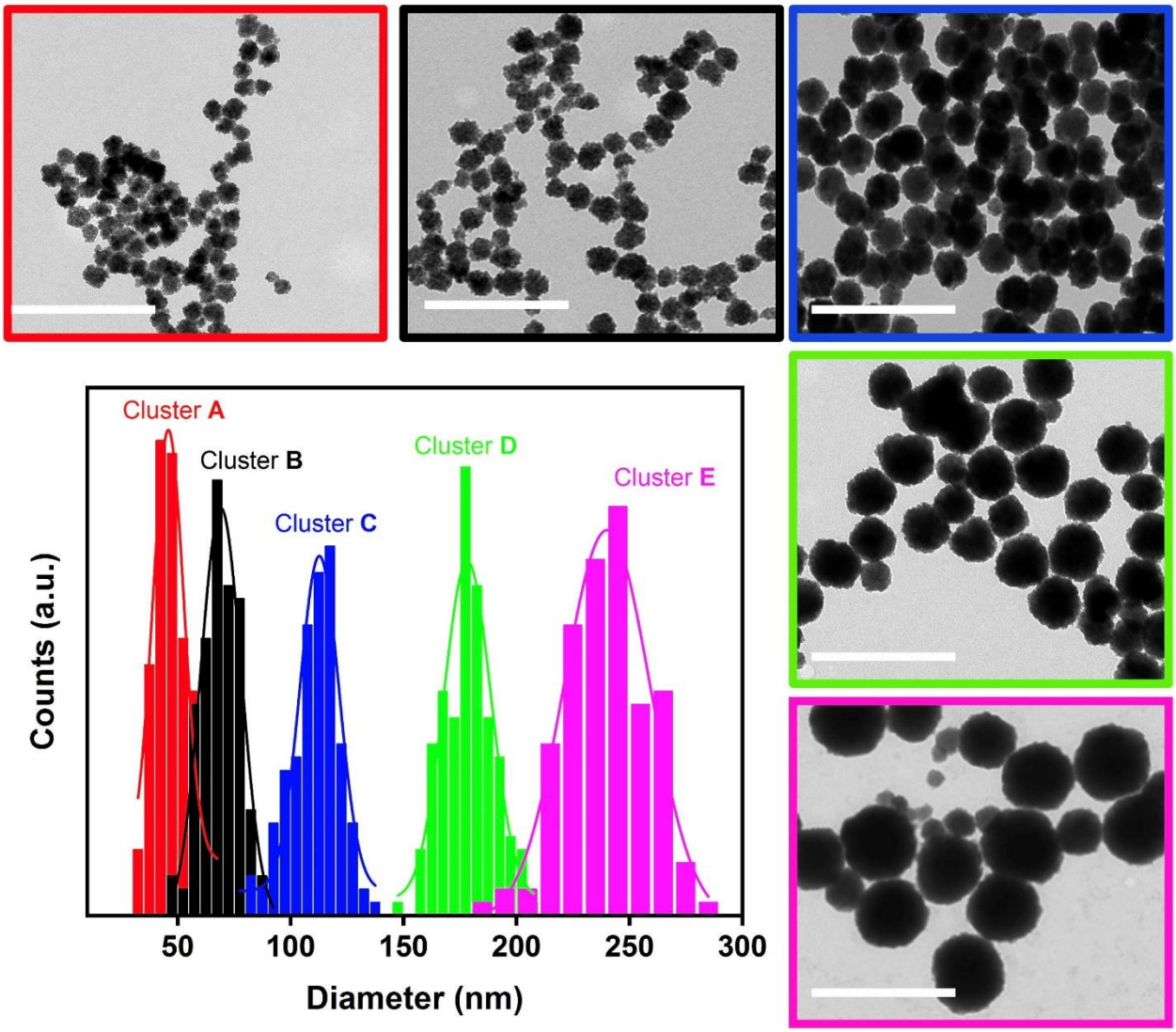


Fig. 7: TEM images (clockwise, scale bar: 500 nm, magnification 10,000x) and particle size distribution analysis of clusters A (framed in red), B (black), C (blue), D (green), and E (magenta) (reproduced from [58] – Open Access)

Nature of heat transport in nanoparticles

The heat transfer by the macroscopic movement of a bulk fluid is known as convection, and the basic theory assumes that heat is transported through diffusion. In crystalline solids, phonons, which are the virtual particles and heat transfer, carry heat by the propagation of lattice vibrations about their equilibrium position. Nevertheless, in nanofluids, as the nanoparticle size falls below the phonon mean-free path, phonons diffusions over the nanoparticle intrinsically invalid and instead proceed ballistically without scattering [60,61]. To put it another way, the nanoparticles are getting free path in between the particles to go through from one place to another because phonons move ballistically without collisions. The diffusive is the bulk base fluid phenomena, but the ballistic is the phenomena in the nanoparticles. During the diffusive transport, heat is carried by numerous phonons scattering activities without macroscopic drift motion, while internal scattering is believed to be absent during the ballistic transport. In both diffusive and ballistic phenomena, the heat transfer occurs.

Because the nanoparticle size is much less or comparable to the phonon mean free path, the classical Fourier's law is inappropriate to be used in nanofluids to predict the heat transfer in the nanoscale regime. In the ballistic thermal transport, the heat transport is controlled by the phonon-boundary interactions and the size-dependent effective thermal conductivity, due to the increase in the Knudsen number, *Kn* (the ratio of the phonon mean-free path to the size of the nanoparticle) [62]. The heat conductivity of nanofluids can be influenced by a wide range of factors, making it impossible to represent all of them in a single model. Another computational fluid dynamics (CFD) method which has gained more attention in recent years for its advantages over finite element, finite volume and finite difference methods is the lattice Boltzmann method (LBM). Several transport methodologies are generally derived from the Boltzmann transport equation (BTE) to characterize the nanoscale heat transport, such as the ballistic-diffusive equation (BDE) and the Cattaneo-Vernotte (C-V) model [62]. To better study the thermal transport in two-dimensional nanomaterials, interfacial heat conduction investigation is essential.

Carrillo-Berdugo et al. used molecular dynamics simulation (MDS) approach and concluded that phonon propagation along the nanotube longitudinal axis of one-dimensional SWCNT nanofluid increased the heat conduction through the base fluid, hence improving the nanofluid's thermal conductivity and heat transfer [63]. The study was based on the experimental data of one-dimensional SWCNT suspended in water and mixed with Triton X-100 additive. In a research performed by Lee et al., a significant hydrodynamic phonon transport in graphene provides a new perspective beyond diffusive and ballistic transport, depicting the thermal transport phenomenon in two-dimensional nanomaterials which is different from zero- and one-dimensional nanomaterials [64]. Hydrodynamic transport contains many phonon scattering actions. The drift motion of phonons in the hydrodynamic regime developed two fascinating hydrodynamic transport phenomena, i.e. phonon Poiseuille flow and second sound, which are analogous to Poiseuille flow and ordinary sound in a fluid, respectively. In line with the findings, Cepelloti et al. [65] investigated on the phonon transport and heat conductivity of some two-dimensional nanomaterials using density functional perturbation theory (DFPT) and an exact, variational solution of the BTE. A wave-like heat diffusion with second sound were observed at ambient temperature and higher, especially in graphene, boron nitride, and graphane. Recently, Mortazavi et al. [66] found a more stable, efficient, and convenient solution for the investigation of dynamical stability and the phonon transport of low-symmetry and porous two-dimensional nanomaterials using machine-learning interatomic potentials as an alternative to DFPT simulations. The unique layered nature of two-dimensional nanomaterials also provides new opportunities to achieve novel phonon properties by stacking along the thickness direction [7].

Effect of Nanomaterial Morphology to Thermal Conductivity and Heat Transfer Enhancement in Nanofluids

In addition to thermal conductivity improvement, another key reason behind heat transfer enhancement in nanofluids is due to their increased aspect ratio or surface-to-volume ratio. Tahmooressi et al. [12] studied the effect of nanoparticles size and aspect ratio on the thermal conductivity of water-based nanofluids numerically using lattice

Boltzmann method and validated the results with some established data, such as from Hamilton–Crosser model [67], Timofeeva et al. [68] and Murshed et al. [69]. It has been demonstrated that raising the aspect ratio of nanoparticles can enhance the effective thermal conductivity of nanofluids due to good agreement with the previous data. However, the size of nanoparticles is not significant based on their statistical analysis. Nevertheless, this research was limited to zero-dimensional alumina (Al_2O_3) and titania (TiO_2) nanoparticles in spherical- and rod-shaped only and the heat transfer enhancement was not predicted. Despite the complexity, the surface-to-volume ratio of the nanoparticle is a suggested to be a more suitable parameter that should be studied as all the solid/liquid interfacial interactions depend on this ratio.

Studies have shown that nanomaterials with cylindrical or elongated shape show exceptional thermal conductivity increase as reported by Jozwiak et al. [13]. These authors discovered the exceptional thermal conductivity increase in one-dimensional carbon nanotube (CNT) ionanofluids compared to other nanomaterials considered. The study compared various carbon nanomaterials with distinct dimensions, namely zero-dimensional fullerenes, one-dimensional single-walled carbon nanotubes (SWCNT) and multi-walled carbon nanotubes (MWCNT), two-dimensional graphene, and three-dimensional graphite, which were suspended in the ionic liquid of 1-ethyl-3-methylimidazolium thiocyanate [EMIM][SCN]. The two-dimensional graphene ionanofluid showed a slight increase in the thermal conductivity, while the zero- and three-dimensional nanomaterials used in the study did not show any enhancement in the thermal conductivity. The higher thermal conductivity observed in the one-dimensional SWCNT than other nanomaterials studied was due the higher aspect ratio of the SWCNT than other nanomaterials. Further research needs to be conducted to compare the heat transfer enhancement between distinct dimensions of nanomaterial.

By considering convective heat transfer caused by the Brownian motion, Shukla et al. developed a new analytical model for effective thermal conductivity of the nanofluids containing spherical Al_2O_3 and TiO_2 nanoparticles suspended in water or ethylene glycol (EG) [70]. The model is also applicable to non-spherical nanoparticles by assuming volume-equivalent particle diameter, dp_{eq}.

$$d_{p,eq} = \left(\frac{6V_{non,sphere}}{\pi}\right)^{\frac{1}{3}} \tag{3}$$

The accuracy of the model may be limited by the quality of the data used for comparison as the agreement was solely with Hamilton-Crosser model [67], whereas no experimental data was compared due to inconsistency in the experimental data. Besides, simplification was given in the shape of the non-spherical nanoparticles as cylinder and plate. According to the Hamilton-Crosser model for non-spherical particles, a more significant increase in the enhancement of the effective thermal conductivity may be achieved using non-spherical nanoparticles with a greater surface-to-volume ratio.

In the paper reported by Alsarraf et al., four different nanoparticle shapes, namely blade, brick, platelet, and cylinder of MoS_2 nanoparticles suspended in water were investigated [6]. The results showed that the brick-shaped nanoparticles have the highest Nusselt number and the lowest entropy production compared to other shapes, while the blade-shaped nanoparticles have the lowest Nusselt number and the highest entropy production compared to other

shapes. A recent study by Huang et al. can serve as a good reference in investigating the effect of two-dimensional nanomaterial's shape to the thermal conductivity and heat transfer enhancement in nanofluids [71]. The paper reported on the development of a novel model that incorporates the impacts of graphene's length and thickness, interface layer, and anisotropic properties in order to predict the thermal conductivity of graphene nanofluids. Anisotropy refers to the directional dependence of the material's properties. For graphene nanosheets, it indicates that the thermal conductivity is different in different direction. In addition, the model included the possibility of thermal conduction path absence considering the effect of different orientation in graphene nanosheets. The thermal conductivity predicted by the model was correlated well with the temperature, concentration, and length of nanoparticles, but deviated slightly with thickness. When the graphene sheets are aligned parallel to the direction of heat flow, a higher thermal conductivity is observed, but if they are aligned perpendicular to it, a lower thermal conductivity is noticed. This is due to the fact that heat can move along graphene sheets rather than across them more readily. Therefore, for similar shaped nanomaterials, the anisotropic characteristics study should be taken into consideration as it affects their effective thermal conductivity.

As the study of emerging nanomaterials are still progressing, it is important to include the morphological and heat transfer enhancement study. Some of the abovementioned studies did not report all these information due to lack in characterization technique and different aims of study.

Other Method of Study related to Heat Transfer Enhancement of Nanofluids

The methodology for the study of nanofluid heat transfer enhancement involves several steps. Firstly, the selection of the base fluids and nanomaterials from available options is vital. Once the base fluids and nanomaterials are selected, they must be prepared thoroughly. Preparation of nanofluids can be achieved via two different methods, which are one-step method and two-step method as explained by Sezer et al. [72] and Said et al. [73]. The preparation process is crucial as it affects the stability and properties of the nanofluid. The stability of the nanofluid is imperative to ensure that it does not lose its thermal properties over time and remains effective during the heat transfer process. Hence, material characterization must be performed to investigate the stability. Additionally, characterization of nanomaterials and nanofluids can aid in distinguishing between different dimensions of nanomaterials as the microscopic structure, composition and thermophysical properties can be obtained. All these information is useful for the prediction of heat transfer enhancement in nanomaterials and nanofluids.

Characterization of nanomaterials and nanofluids

Nanomaterials and nanofluids characterizations are found to be important in developing scalable nanomaterials and nanofluids production. However, because of the multidisciplinary nature of nanoscience and nanotechnology, not every research team has easy access to a wide variety of characterization tools. In this regard, it is indeed useful to understand the limits and strengths of various approaches in order to determine whether, in certain circumstances, using only one or two of them is sufficient for obtaining reliable data when investigating a given parameter. While several material characterization methods have been used for decades, new methods and techniques are constantly emerging. A comprehensive description of numerous distinct characterization techniques for nanomaterials in relation

to the parameter studied can be found in the report prepared by Mourdikudis et al. [74]. In general, material characterizations can be divided into three categories; namely by using microscopy, spectroscopy and macroscopic testing techniques.

(a) Microscopy Techniques

Microscopy techniques can be used to obtain the morphology or estimated size and shape of nanomaterials. The dimensional classification of nanomaterials includes morphology as a key component. In comparison to bulk materials, the characteristics and behavior of nanomaterials are far more varied and advanced. Thus, size and shape are two of the basic and most important parameters investigated in the characterization of nanomaterials of different dimensions [74]. The frequent approaches for analyzing nanomaterial size and shape are by using transmission electron microscopy (TEM), scanning electron microscopy (SEM), high-resolution transmission electron microscopy (HRTEM), or field emission scanning electron microscopy (FESEM), as shown in Table 2. These approaches provide both the direct images of the nanomaterial and accurate estimation of its homogeneity.

Table 2: Common microscopy techniques to obtain the estimated size and shape of nanomaterials

Author(s) (Year)	**Microscopy technique**	**Type of Nanomaterials**	**Information obtained**
Ahmadi et al. (2016) [75]	FESEM	Graphene nanoplatelets (GNP)	- Image and average particle size of GNP (38.01 – 72.28 nm)
Zhang et al. (2017) [76]	TEM, HRTEM	Controlled reduced graphene oxide (CRGO) nanosheets	- Image and thickness of 4.3 nm of ultrathin two-dimensional graphene sheet.
Nikkam et al. (2017) [77]	SEM, TEM	Cu nanoparticles	- Image of spherical Cu nanoparticles using both SEM and TEM. - Average particle size of Cu nanoparticles using both SEM (20 – 40 nm) and TEM (~20 nm).
Kumar et al. (2018) [78]	SEM, TEM	ZnO nanoparticles	- Image of spherical ZnO nanoparticles using SEM. - Average particle size of ZnO nanoparticles (25 – 30 nm) using TEM.
Casanova et al. (2019) [79]	SEM, HRTEM	Carbon nanotubes (CNTs)	- Images of variations of microstructural of CNTs as a function of temperature
Kang et al. (2019) [80]	FESEM, TEM	Single and multilayer Ti_3C_2 MXene nanosheet	- Image of shape and average particle size of single and multilayer Ti_3C_2 MXene nanosheet

Liu et al. (2020) [81]	FESEM	GNP, graphene (Gr), Al_2O_3 and SiO_2	- Image of shape and average particle size of the metal oxides with different dimensions (shown in Figure 8).
Elsaidy et al. (2021) [58]	TEM	Controlled Fe_3O_4/γ-Fe_2O_3 nanoparticles clusters	- Image of clusters of Fe_3O_4/γ-Fe_2O_3. - Average particle size of clusters of Fe_3O_4/γ-Fe_2O_3.

Apparently, the morphology changes with respect to temperature as reported by Casanova et al. using SEM and HRTEM images [79]. The images can be found in Figure 9 (a) – (e) and 10 (a) – (d), respectively. In their research, the best carbon nanotubes (CNTs) were found to be produced at temperatures ranging between 800 and 850°C, while outside this range, iron impurities, more graphitic carbon, and additional structural defects may be present. With the distinct diameter obtained at different temperature, the aspect ratio will be varied.

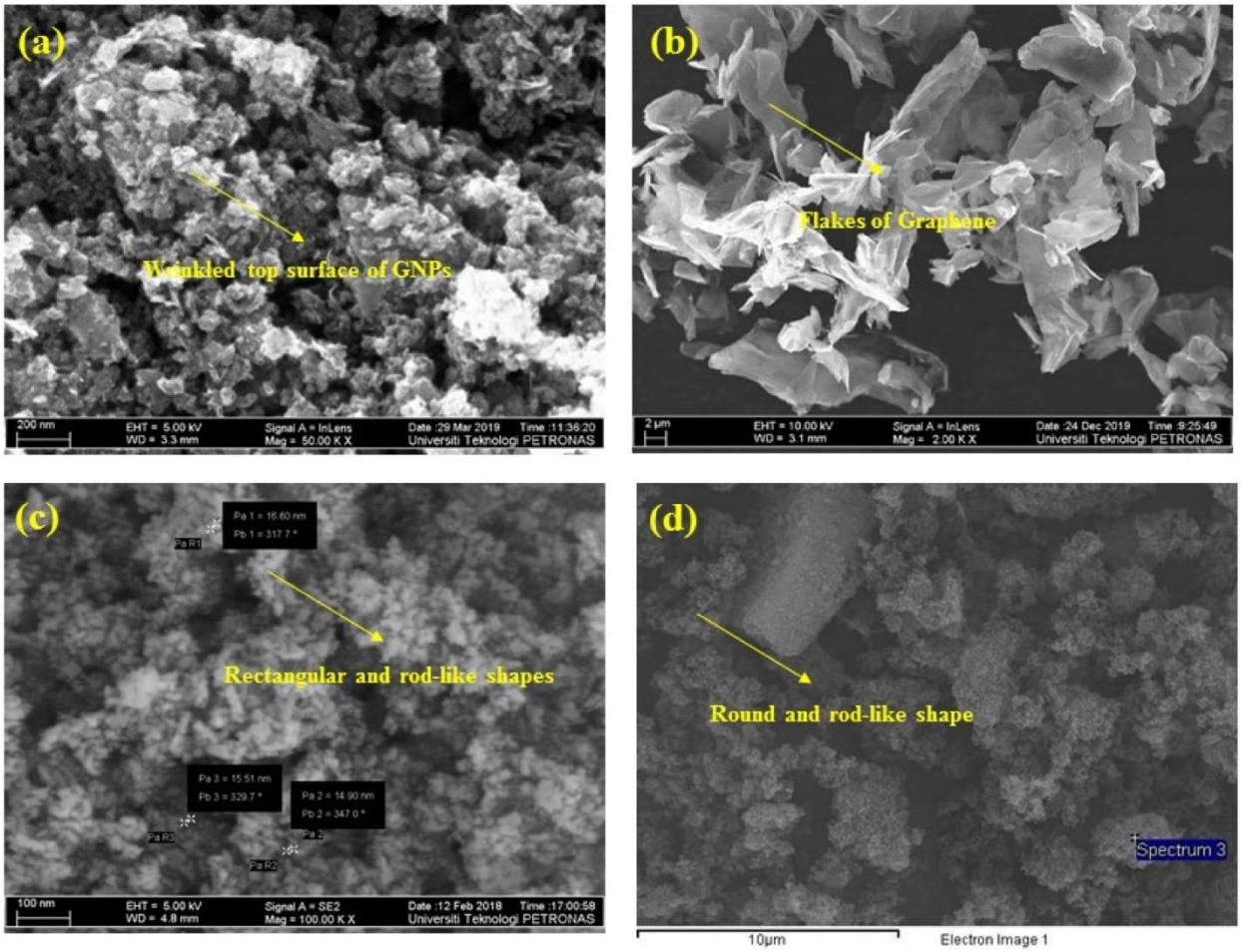


Fig. 8: FESEM images of shape and average particle size of (a) GNPs, (b) Gr, (c) Al_2O_3 and (d) SiO_2 (reproduced from [81] – Open Access)

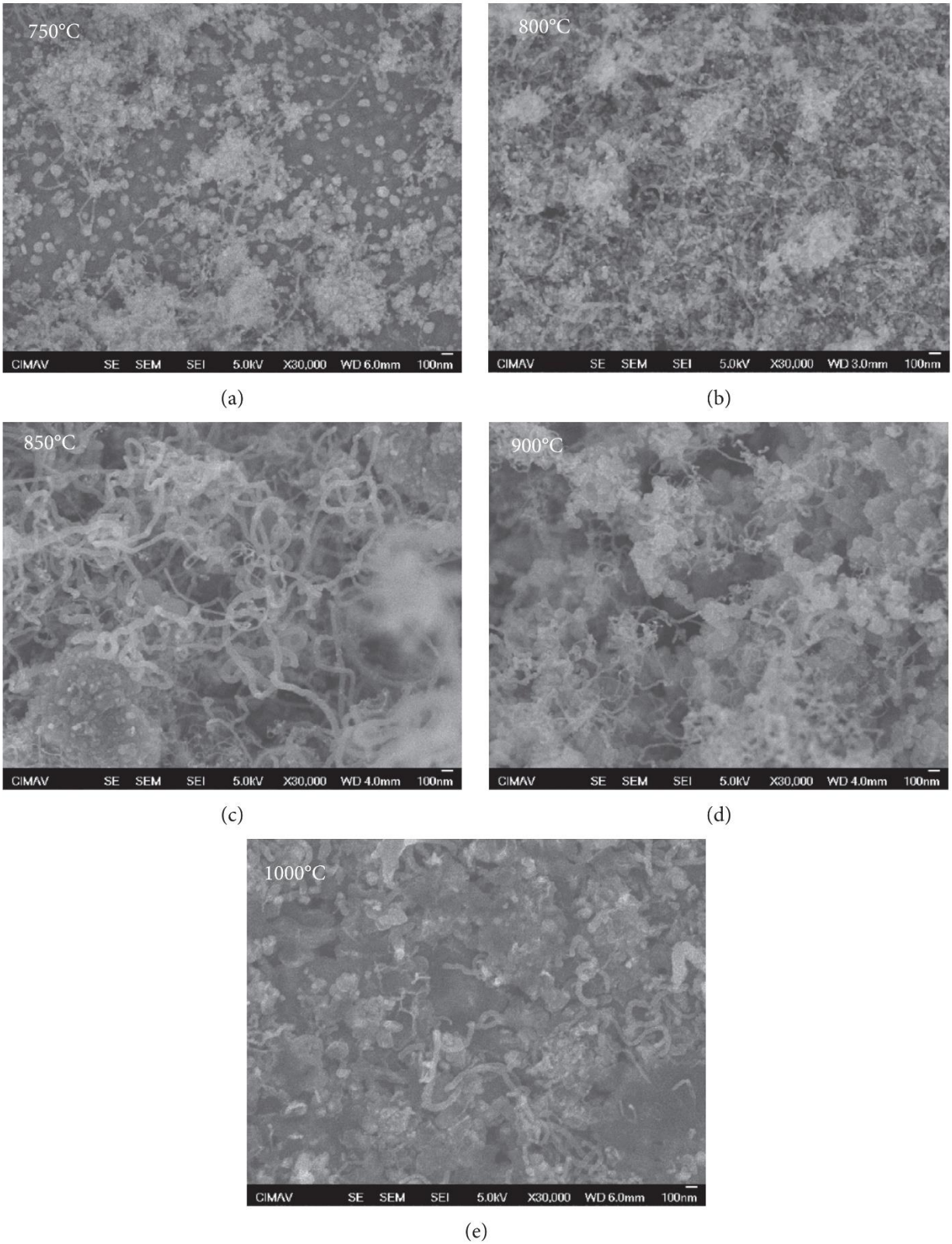


Fig. 9: SEM images of variations of microstructural of carbon nanotubes (CNTs) as a function of temperature (reproduced from [79] – Open Access)

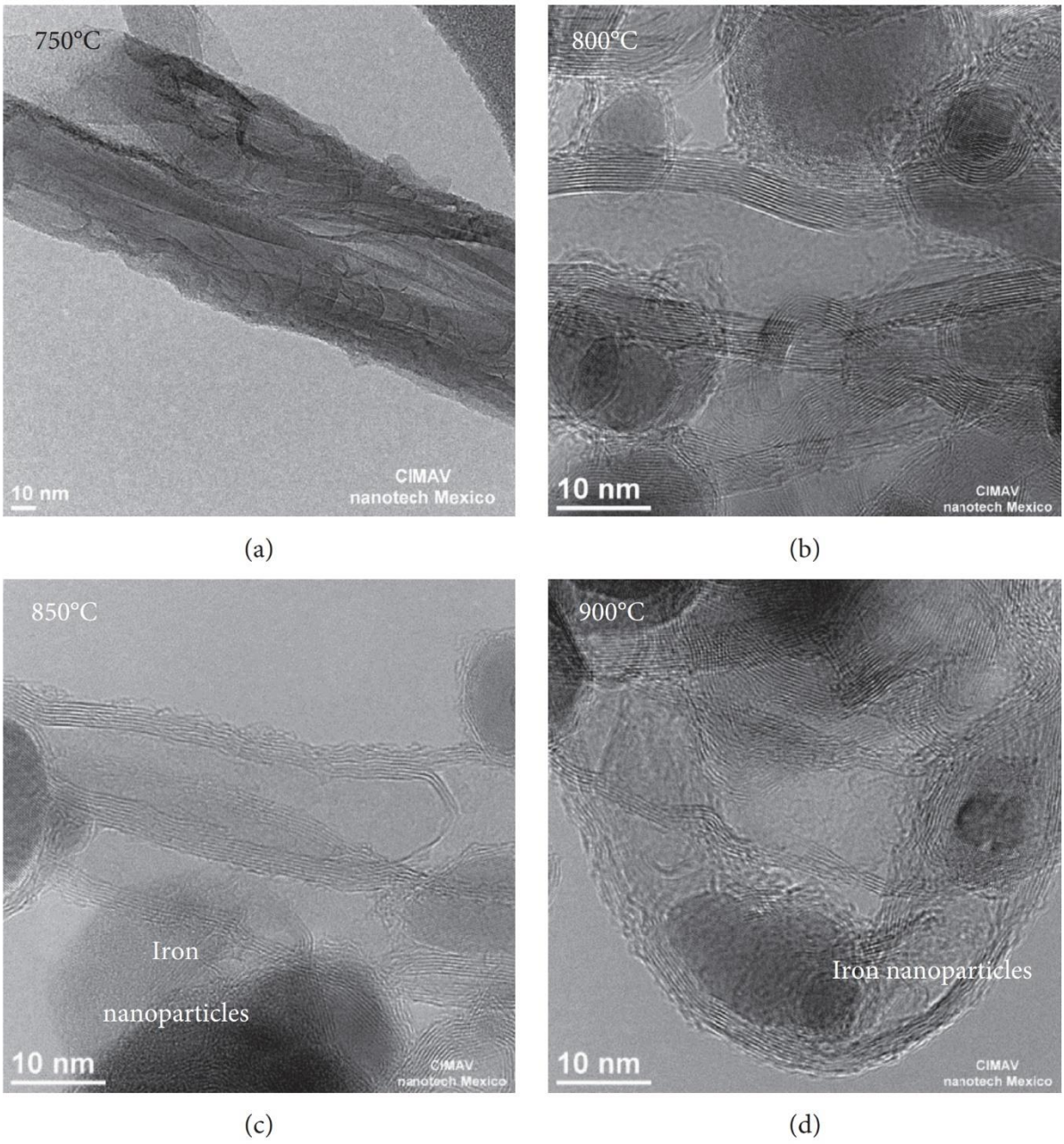


Fig. 10: HRTEM images of variations of microstructural of carbon nanotubes (CNTs) as a function of temperature (reproduced from [79] – Open Access)

Despite the variety of characterization methods, only a few researchers have published about the characterization images of nanomaterials suspended in nanofluids, compared to many reports on the characterization images of nanomaterials before suspending them in nanofluids. Suganthi et al. characterized the dried zero-dimensional ZnO/water nanofluid using SEM [50]. The appearance of the dried nanofluid was not much different from the ZnO nanoparticle with slightly planar in shape compared to the ZnO nanoparticle as the nanoparticle undergone a few processes during the preparation of the nanofluid, including ultrasonication process. Abdolbaqi et al. used TEM to compare the images of TiO_2 nanoparticles before and after the suspension in

BioGlycol/water mixture to produce nanofluid [32]. It was observed that the nanoparticles dispersed well in the base fluid and the distribution of nanoparticles remains the same. Aberoumand et al. utilized both SEM and TEM techniques to obtain the image of uniformity of silver dispersion and average particle size of silver in water [82]. Hydrogen bonds connect the silver nanoparticles to the water molecules. Due to the silver nanoparticles' negative charge and the water molecules around them, a highly stable solution has been created without the need for a surfactant or stabilizing agent. Hence, high temperatures can be tolerated by the silver nanofluid without causing structural changes.

Meanwhile, recent advances in the microscopy techniques reveal a more versatile and powerful technique for studying samples size and shape at nanoscale, especially for two-dimensional nanomaterials. The technique is by using atomic force microscopy (AFM) [9,80,83–90]. It does not only provide image in two-dimensional and three-dimensional topography, but also various types of surface measurements. An example of an AFM image of a two-dimensional single Ti_3C_2 MXene nanosheet obtained from a report published by Kang et al. [80] is depicted in Figure 11. In the study reported by Vallejo et el., different images of GNP morphology with multiple groups of stacked graphene layers in the height range of 2.2 nm to 5.1 nm were seen through AFM analyses [83]. The thickness of each layer and heights per graphene plate between 2 nm and 6 nm were also obtained.

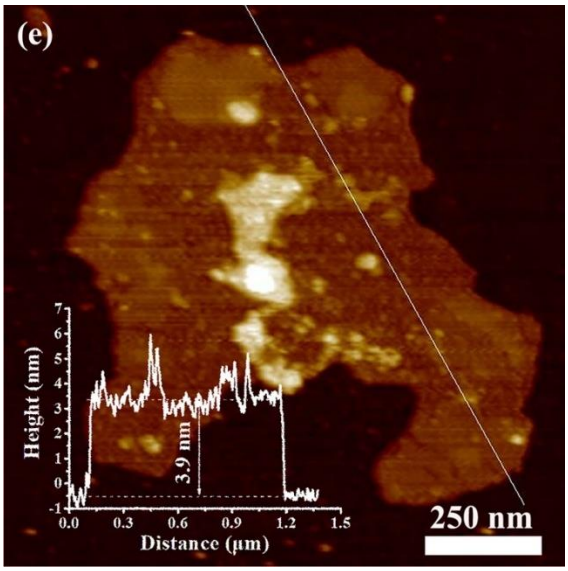


Fig. 11: The AFM image of a two-dimensional single Ti_3C_2 MXene nanosheet (reproduced from [80] – Open Access)

Since nanomaterials are usually used in various applications associated with heat, Kawamoto et al. reported on a novel method to visualize heat conduction pathways with nanoscale spatial resolution using scanning transmission electron microscopy (STEM) and a nano-thermocouple assembled in a TEM [91]. The method is called as STEM-based thermal analytical microscopy (STAM). In their work, the detailed information of heat conduction in the nano-fillers embedded in a composite material and/or between them was visualized in the form of two-dimensional thermal distribution maps as seen in Figure 12. This method is also beneficial to measure a TEM specimen's steady-state thermal conductivity and visualize the heat distribution in any nanostructured object

up to ~20 nm. The challenge in using this method is, the sample must be prepared using focused ion beam (FIB) carefully, as the tendency for sample surface damage is high. Additionally, further research needs to be done to analyze the transient thermal conduction process in addition to the quantitative thermal conductivity measurements based on the Fourier's law.

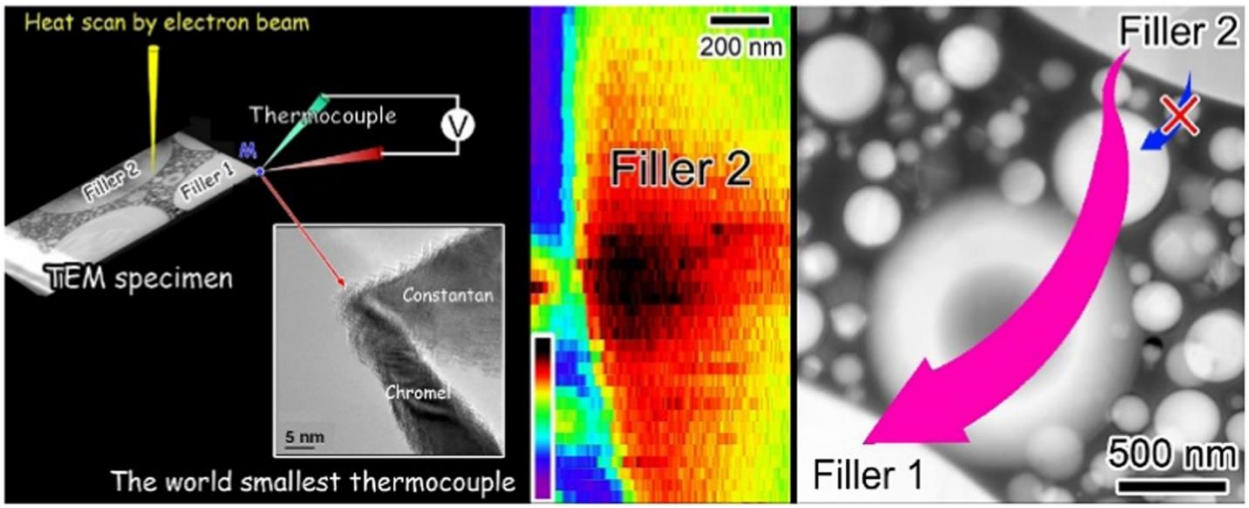


Fig. 12: Visual of heat conduction in the fillers embedded in a composite material and/or between them (reproduced from [91] – Open Access)

(b) Spectroscopy Techniques

Spectroscopy techniques are frequently combined with several microscopy techniques to give a comprehensive overview of the nanomaterial. While microscopy techniques are used to provide the image of the nanomaterial, spectroscopy techniques provide a lot of information about the nanomaterials, such as the chemical composition, composition variation, crystal structure and photoelectric properties of the nanomaterial. Some common examples of spectroscopy techniques include energy dispersive X-ray (EDX) spectroscopy, Fourier transform infrared spectrum (FTIR) spectroscopy, ultraviolet visible (UV-Vis) spectroscopy, Raman spectroscopy, dynamic light scattering (DLS) and electrophoretic light scattering (ELS). Examples of some spectroscopy response from a report published by Kang et al. [80] are shown in Figure 13 (a) – (c).

For nanofluids, DLS can be used to measure the hydrodynamic diameter, which is the inorganic core of the nanomaterials along with the base fluid layers surrounding it [78], while ELS can be used to measure the electrophoretic mobility of nanomaterials in dispersion. The particle mobility is often converted to Zeta potential value. Modern equipment sometimes integrates DLS and ELS into one device, allowing for the measurement of both particle size and Zeta potential. Manikandan and Rajan [92] and Leela Vinodhan et al. [31] reported that there was a difference between the hydrodynamic diameter of the nanoparticle suspended in the base fluid determined by using DLS and the nanoparticle diameter determined using SEM. This is attributed to the liquid layers that surround the nanoparticles, which are taken into consideration when the hydrodynamic diameter is measured using DLS. The hydrodynamic diameter offers information on the nanoparticle core, any surfactant,

and the base fluid layer adhering to the nanoparticle as it moves under Brownian motion. Some limitations of DLS are; it generally assumes spherical shaped particles and lacks the resolution to detect small aggregates, hence the measurements on the one-dimensional- and two-dimensional-based nanofluids might be inaccurate [74].

The Zeta potential value and UV-Vis spectroscopy characterization can also be used as the indication for nanofluids' stability. Stability is the property in which the particles do not aggregate at a significant rate. One of the main challenges of using nanofluids in heat transfer applications is its stability at high temperature and in the long term [93]. Jin and Jing suggested that a stable suspension requires a minimum absolute Zeta potential of 30 mV, and that a value of 20 mV or below indicates inadequate stability [94]. A stable nanofluid is vital and its usage in heat transfer applications must be investigated thoroughly to ensure less maintenance and replacement of the nanofluid in the system. Depending on the structure, aggregated nanoparticles may have a negative impact on the thermal conductivity and convection factor of nanofluids, both of which are crucial for enhancing heat transfer in nanofluids. This statement is supported by the research done by Zhou et al. [95], where the thermal conductivity was shown to increase when the structure of nanoparticle aggregates switched from compact spherical to chain-like structure. Anushree and Philip investigated the stability of three water-based metal oxide (α-Al_2O_3, TiO_2 and γ-Al_2O_3) nanofluids with different particle concentration using UV–Vis spectroscopy, dynamic light scattering (DLS), zeta potential, phase contrast microscopic and visual observation [96]. The observed zeta potential value and its time independent variation indicates sample stability and electrostatically stabilized, which causes a sufficiently large potential barrier that prevents the particles from aggregating. The time dependent absorbance changes in α-Al_2O_3 and TiO_2 nanofluids were very high as compared to γ-Al_2O_3 nanofluids. The result was also confirmed by the phase contrast optical microscopy images, as well as the visual observation which showed a clear phase separation in α-Al_2O_3 and TiO_2 nanofluid, especially at higher particle concentration. Choudhary et al. [97] investigated the behavior of zinc oxide/ethylene glycol:deionized water (50:50) nanofluid for 25 days and its influence on thermal characteristics of flat plate solar collector at the different parametric condition. The Zeta potential value dropped by 51.16% on the 25th day for 1.4 vol% ZnO nanoparticle.

(c) Macroscopic Testing Techniques

It is important to note that the morphology of nanomaterials may be affected by environmental factors such as temperature. Therefore, investigating the thermal change and stability of nanomaterials is necessary to fully comprehend their behavior during heating. Macroscopic testing techniques can be used to obtain some thermophysical properties of the nanofluids and perform thermal analysis as to investigate the thermal change and stability of the nanofluids. Among the common macroscopic testing techniques include differential thermal analysis (DTA), Dielectric thermal analysis (DEA, DETA), thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC).

TGA is performed to measure the sample mass as a function of temperature or time. In the TGA, the materials characterization is done through the analysis of characteristic decomposition patterns based on mass loss which is given by the shape of the TG curve, as shown in Figure 13 (d). From the TG curve, the decomposition temperature is known. A heat transfer fluid with high thermal stability will degrade less than a fluid with low

thermal stability, hence will deliver a longer fluid life than a less stable fluid. Thermal fluid degradation occurs when enough heat is applied to the fluid to cause the breaking of the molecular bonds, which results in a change in the fluid's physical properties.

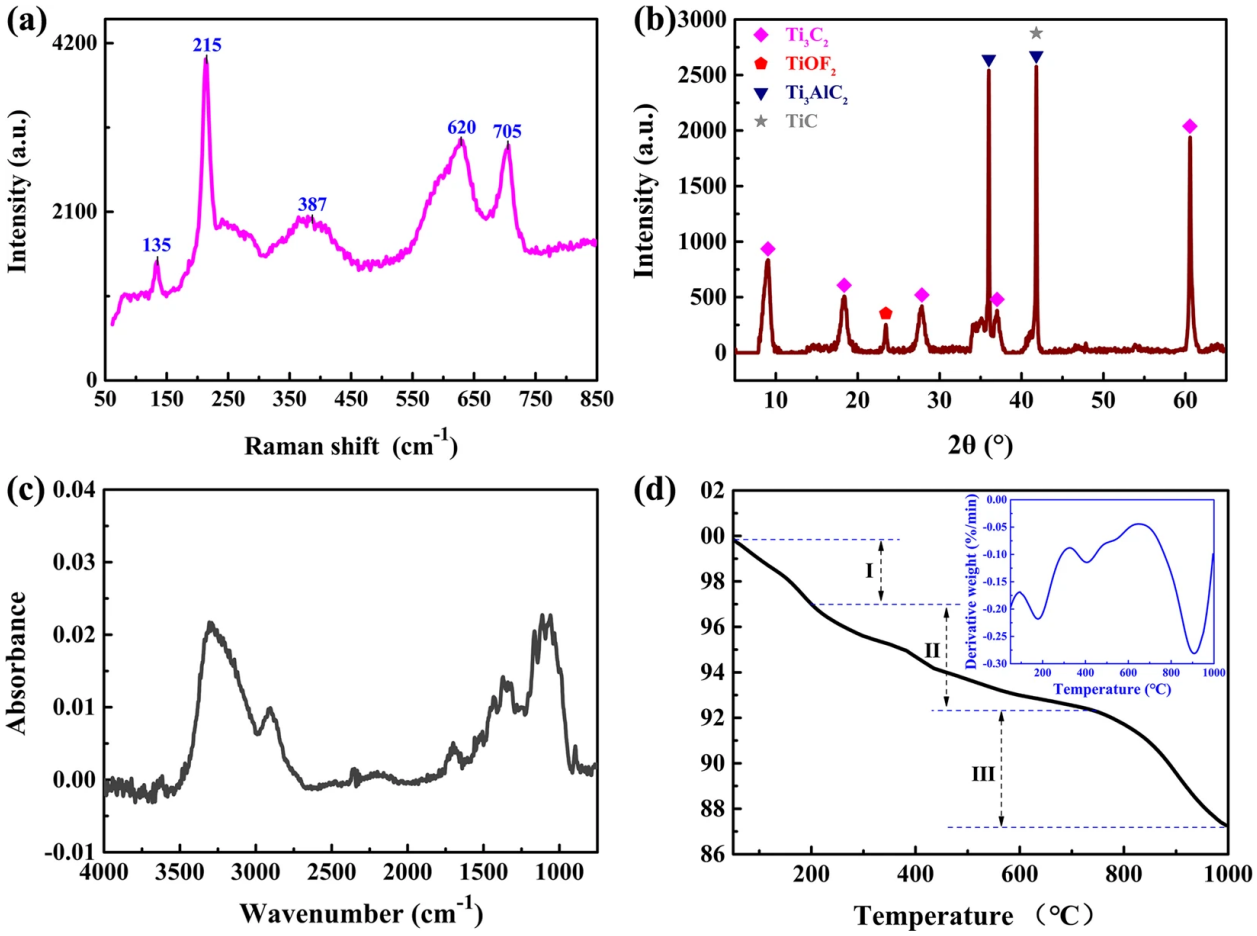


Fig. 13: (a) Raman spectrum, (b) XRD pattern, (c) FTIR spectrum, and (d) Thermal gravimetric analysis (TGA) curve of Ti_3C_2 MXene. The differential thermal gravimetric (DTG) curve of Ti_3C_2 MXene is presented in the inset of (d). (reproduce from [80] – Open Access)

Other Method of Study on Heat Transfer Enhancement in Nanofluids

Alternatively, Sundar et al. used different figure of merit to evaluate the thermal performance of nanodiamond/propylene glycol and water mixture in turbulent flow, rather than using Nusselt number [98]. A dimensional number called the Mouromsteff number, *Mo* that combines the effects of the primary properties of heat transfer fluids such as density, thermal conductivity, specific heat, and viscosity was used in the figure of merit.

$$Mo = \frac{\rho^{0.8} k^{0.67} c_p^{0.33}}{\mu^{0.47}} \quad (4)$$

The figure of merit is obtained by the relative Mouromsteff number of the nanofluid to the base fluid. The relevance of the Mouromtseff number comes from the fact that the liquid coolant with the highest Mouromtseff number will have the highest rate of heat transfer for flow over or through a given geometry at a given velocity. It should be

highlighted, though, that the Mouromtseff number does not account for all heat transfer mechanisms. In a recent work by Upadhyay et al. [99], they proposed the selection of heat transfer fluid for the heating and cooling applications based on solar thermal system by means of another figure of merit, *FoM* as indicated in (5):

$$FoM = \frac{Nu}{CP} \tag{5}$$

CP is the coefficient of pressure, which is defined as the ratio of pressure drop to the dynamic pressure in the piping system. An analysis of the dimensional parameters using the thermophysical properties of pure fluid and nanofluid revealed that the relative performance of a heat transfer fluid was dependent on the temperature and changed as the temperature changed. A novel separation approach was proposed to filter the effect of nanoparticles on the Nusselt number correlation for nanofluids, leading to a generalized form of Nusselt number correlation for nanofluids for a number of base fluids. However, the equation is limited to the temperature range between 373 to 573 K and applicable for some nanoparticles and base fluids only. Since there is no common agreement in the magnitude of the heat transfer enhancement of nanofluids, hence, more investigations are needed in this sense.

As a matter of fact, the experimental investigations require careful calibration and control of experimental variables and the theoretical prediction of the effective thermal conductivity of nanofluids is unclear and not generic. Therefore, another option to predict effective thermal conductivity and heat transfer enhancement of nanofluids and generalize the correlation is by doing comparative study. One of the methods is by using dimensional analysis, while a more sophisticated method is by using soft computing modeling or machine learning approaches. Hassani et al. predicted the thermal conductivity of nanofluids using dimensional analysis based on Vaschy-Buckingham theorem [100]. The developed model has wider application capability compared to some theoretical models. It was found that the reduction in particle size, increase in the volume fraction of the nanoparticles and temperature resulted in the rise of the nanofluids thermal conductivity. Zendehboudi and Saidur constructed and tested a Multilayer Perceptron-Artificial Neural Network (MLP-ANN) approach using the data set that contains 993 experimental samples on 26 different types of nanofluids [101]. The most influential physical properties of nanofluids, such as the nanoparticle volume fraction, nanoparticle diameter, thermal conductivity of base fluid, temperature, and thermal conductivity of nanoparticle were considered as the input variables. For validation of the MLP-ANN model, comparisons with other well-known artificial intelligence methods, such as Hybrid-Adaptive Neuro Fuzzy Inference System (Hybrid-ANFIS), Genetic Algorithm-Least Square Support Vector Machine (GA-LSSVM), and Group Method of Data Handling (GMDH) were carried out. The predictions made by the MLP-ANN are similar to the experimental data compared to those of other approaches. Despite that, most of the nanofluids studied by Hassani et al. [100] and Zendehboudi and Saidur [101] were zero-dimensional-based nanofluids except for a few nanofluids using one-dimensional CNT and MWCNT. None of them were two-dimensional-based nanofluids.

A summary of the method of Study related to heat transfer enhancement of nanofluids discussed earlier is shown in Table 3 and in pictorial form in Figure 14.

Table 3: Possible Reason(s) of Thermal Conductivity/Heat Transfer Enhancement in Nanomaterial/Nanofluids

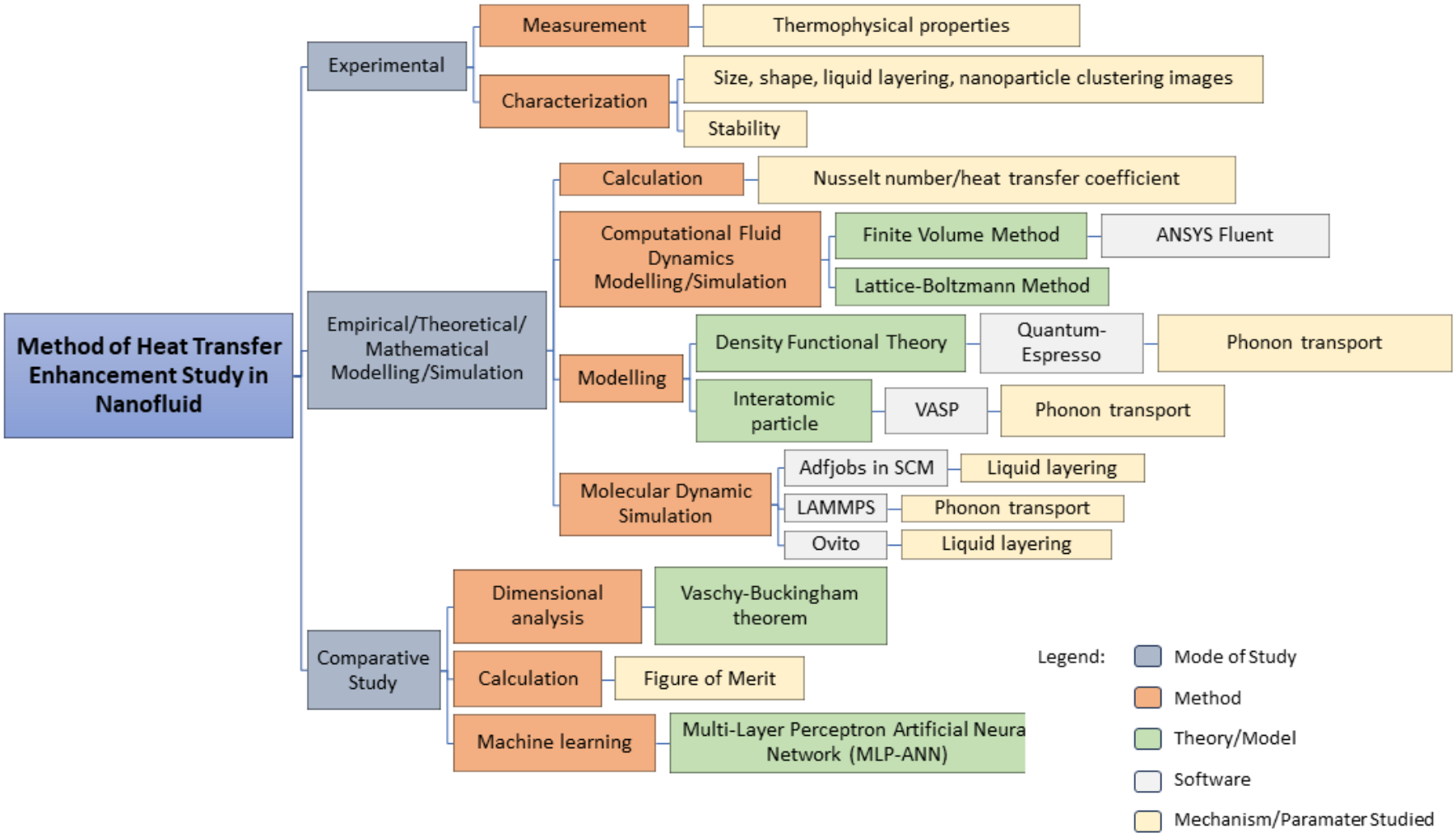
Author(s) (Year)	**(Dimensionality) Nanoparticle (NP)/ base fluid**	**Method of Study (theory/ equation/ software)**	**Thermal Conductivity Enhancement**	**Heat Transfer Enhancement**	**Reason(s) of Thermal Conductivity/ Heat Transfer Enhancement**
Lee et al. (2015) [64]	(2D) Graphene	Theoretical models simulation (DFPT using Quantum-Espresso package)	-	Yes	Hydrodynamic phonon transport
Hassani et al. (2015) [100]	(0D) TiO_2, Al_2O_3, Al, Cu, Fe, Al_2O_3, SiO_2/methanol, TiO_2, Al_2O3, CuO, Al_2O_3/radiator coolant, Al_2O_3/R141b, Al, Cu/Therminol 66, (1D) MWCNTs/EG, MWCNTs/water CNTs/Engine Oil	Comparative study using dimensional analysis (based on Vaschy-Buckingham theorem)	Yes	-	- Nanolayering of the liquid at the liquid/nanoparticle interface. - Brownian motion of the nanoparticles - Effects of nanoparticle clustering
Cepelloti et al. (2015) [65]	(2D) Graphene, Boron Nitride, Molybdenum Disulphide, Functionalized Derivatives Graphane, Fluorographene	Theoretical models simulation (DFPT using Quantum-Espresso package)	Yes	Yes	Phonon transport
Milanese et al. (2016) [53]	(0D) Cu/water, CuO/water	Experiment and simulation (MDS using Adfjobs in SCM software)	Yes	-	Liquid layering: Better thermal conductivity enhancement in Cu
Ahmadi et al. (2016) [75]	(2D) Graphene nanoplatelet (GNP)	Experiment and theoretical study	Yes	Yes	Thermal conductivity enhancement in GNP (not discussed in detail)
Leela Vinodhan et al. (2016) [31]	(0D) CuO/water	Experiment	Yes	Yes	- Thermal conductivity enhancement. - Nanoparticle movement due to Brownian motion

Sundar et al. (2016) [98]	(0D) Nanodiamond/water	Experiment and empirical study (Figure of Merit based on Mouromtseff number)	Yes	-	Microconvection due to Brownian motion
Manikandan and Rajan (2017) [92]	(0D) Sand/propylene glycol (PG) + water (30:70)	Experiment and empirical study (heat transfer coefficient)	Yes	Yes (heat transfer coefficient)	Microconvection due to Brownian motion
Zhang et al. (2017) [76]	(2D) Controlled reduced graphene oxide (CRGO)/water	Experiment	Yes	-	- High thermal property of graphene sheets. - High surface area of graphene, which provides more contact between particles and helps electrons and phonons to conduct heat. - Brownian motion at high temperature
Nikkam et al. (2017) [77]	(0D) Cu/EG, Cu/diethylene glycol (DEG)	Experiment and empirical study	Yes	-	Brownian motion (higher thermal conductivity enhancement using EG)
Abdolbaqi et al. (2017) [32]	(0D) TiO_2/BioGlycol + water (20:80)	Experiment and empirical study (Nusselt number)	Yes	Yes	Nanoparticle clustering/ aggregation
Saha and Paul (2018) [34]	(0D) Al_2O_3/water, TiO_2/water	Computational Fluid Dynamic (CFD) Simulation (Finite volume solver Fluent 6.3.26)	-	Yes	Brownian motion
Vallejo et al. (2018) [83]	(2D) Functionalized GNP/PG + water (30:70)	Experiment and empirical study (Mouromtseff number)	Yes	-	Large surface area of 2D nanomaterial

Zendehboudi and Saidur (2019) [101]	(0D) Al_2O_3/water, Al_2O_3/EG, Al_2O_3/ Oil, CuO/water, CuO/EG, CuO/Oil, CuO/monoethylene glycol (MEG), CuO/Paraffin, Cu/water, Cu/EG, Cu/Oil, Al/water, Al/EG, Al/Oil, Al/engine oil, ZnO/water, ZnO/EG, TiO_2/water, TiO_2/EG, Ag/water, $Mg(OH)_2$/EG, SiO_2/water, SiO_2/TH66, (1D) MWCNT/water, MWCNT/R113, MWCNT/EG	Comparative study using Multilayer Perceptron-Artificial Neural Network (MLP-ANN)	Yes	-	Not discuss in detail
Carrillo-Berdugo et al. (2019) [63]	(1D) SWCNT/water	Experiment and molecular dynamics simulations (Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS))	Yes	Yes	- Phonon heat transport through nanotube longitudinal axis. - Liquid layering
Fan and Zhong (2020) [46]	(0D) Al_2O_3/water, TiO_2/water	Experiment and theoretical study	Yes	-	- Interfacial nanolayer (Interfacial nanolayer increases with fluid temperature but decreases with particle volume fraction) - Brownian effect.
Jozwiak et al. (2020) [13]	(0D) Fullerenes/ionic liquid, (1D) CNT/ionic liquid, (2D) graphene/ionic liquid, (3D) graphite/ionic liquid	Experiment and characterization	Yes	-	Interfacial nanolayer in 1D CNT. Thermal conductivity enhancement is the highest in 1D CNT, moderate in 2D graphene and the lowest in others
Choudhary et al. (2020) [97]	(0D) ZnO/EG:DW (50:50)	Experiment	Yes	-	Brownian motion

Mortazavi et al. (2020) [66]	(2D) Graphene, pentagraphene, haeckelite, phagraphene, graphyne	Machine learning interatomic particle (Vienna Ab initio Simulation Package (VASP))	-	Yes	Second sound in phonon transport
Mamand et al. (2021) [52]	(0D) CuO/water, Al_2O_3/water, SiO_2/water, ZnO/EG	Empirical study (based on Prasher's model)	Yes	-	Nanolayer
Tahmooressi et al. (2021) [12]	(0D) TiO_2/water, Al_2O_3/water	Simulation (Lattice Boltzmann method)	Yes	-	Increasing the nanoparticles aspect ratio
Demirkir and Erturk (2021) [40]	(2D) Graphene/water	Experiment and empirical study (Nusselt number)	Yes	Yes	Brownian motion and thermophoresis
Elsaidy et al. (2021) [58]	(0D) Fe_3O_4/EG, γ-Fe_2O_3/EG	Experiment and characterization	Yes	-	Nanoparticle clustering/aggregation
Zhai et al. (2021) [54]	(0D) Cu/water, CuO/water, Al_2O_3/water	Simulation (N_d using MDS in Ovito software)	Yes	-	Nanolayer
Upadhyay et al. (2021) [99]	(0D) CuO/water, TiO_2/water, Cu/water, (1D) CNT/water, (2D) GNP/water	Comparative study (Figure of Merit = Nu/CP)	-	Yes	Brownian motion and thermophoresis, nanoparticle clustering

Figure 14: Method of study related to heat transfer enhancement of nanofluids



Conclusions and Recommendations

With the rise of nanotechnology, there has been a growing interest in developing materials at the nanoscale with different dimensions to enhance thermal conductivity and heat transfer. Nanofluids have been suggested to be used in many heat transfer applications due to its superior properties compared to conventional fluids. Despite the potential benefits of nanofluids for enhanced heat transfer applications, there remains a need for ongoing research to accurately understand the impact of nanomaterials of different dimensions on the heat transfer properties of nanofluids. Hence, the mechanism of heat transfer of nanofluids have to be understood and the contributing parameters have to be identified to understand the heat transfer enhancement in nanofluids. Material characterizations and thermophysical properties measurements can help in providing the information on the parameters involved in the heat transfer enhancements in nanofluids. The heat transfer enhancement in nanofluids can arise from the thermal conductivity enhancement, movement or motions of nanoparticles due to Brownian motion and thermophoresis, liquid layering, effect of nanoparticle clustering and ballistic phonon transport. The heat transfer enhancement of nanofluids can be studied using a variety of approaches. The majority of the authors opted for experimentation and empirical calculation and focused on improving the thermal conductivity. On the other hand, it should be noted that recent developments in high performance computer applications utilizing more sophisticated methods such as molecular dynamics simulations and machine learning have also made it feasible to study not only Brownian motion effect, but also phonon heat transfer, nanolayers, and nanomaterials clustering. Despite this, little study has been done relating the morphological study with the enhancement of heat transfer in nanofluids. Compared to zero -dimensional nanomaterials, certain low-dimensional nanomaterials have elongated and layered structures, which have a greater aspect ratio and surface-to-volume ratio. These features may present intriguing new opportunities to modify an important property and enhance the heat transfer. In a nutshell, this is a significant gap that has to be explored.

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