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Thermophysical properties of graphene-based nanofluids

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ABSTRACT

Heat transfer operations are very common in the process industry to transfer a huge amount of thermal energy, i.e., heat, from one fluid to another for different purposes. Many fluids are used as heat transfer fluid (HTF), in which water is the most common HTF due to its high specific heat, availability, and affordability. However, conventional HTFs, including water, have a lower thermal conductivity, which is the most critical thermophysical property, hence decreased heat transfer efficiency. The addition of solid particles of highly thermally conductive material, specifically at nano-size, i.e., nanoparticles NPs, result in nanofluid NF, which has evolved over the last two decades as efficient HTF and have been investigated in a wide range of applications. Among NPs, graphene (Gr) based materials have shown very high potential as NF due to the very high thermal conductivity up to 5,000 W/m.K, hence higher thermal conductivity NF. This work aims to thoroughly discuss the thermophysical properties of Gr-based NFs, including thermal conductivity, heat capacity, density, and viscosity. The discussion focus on the thermophysical properties as it is the ultimate determinator of the heat transfer characteristics of the HTF, such as the convective and the overall heat transfer coefficient as well as the heat transfer capacity of the NF. The discussion expands to the relative enhancement in such thermophysical properties reaching up to a 40% increase in thermal conductivity, as the most critical thermophysical property. The discussion shows that Gr-based NF has a much higher thermal conductivity relative to widely studied metal oxide NF and at much lower content, and lower density and viscosity increase, which is critical for determining the pumping power requirements. Critical challenges facing the application of Gr-based NFs such as cost, stability, increased density and viscosity, and environmental impacts are thoroughly discussed with mitigation recommendations given.

Introduction

Heat transfer operations are very crucial for different energy conversion and transport processes, with heat transfer fluids (HTF) as the energy carrier in different heat exchange processes [1–3]. The most common HTF is water or steam, being available, cheap, and compatible with a wide range of materials and processes [4–6]. Other HTFs are used for specific applications such as ethylene glycol (EG) and its water mixtures and different oils to expand the operating temperature range beyond that of water [7,8]. The main characteristic of evaluating any HTFs for their function and performance is the thermophysical properties, such as thermal conductivity, heat capacity, density, and viscosity [9]. The improved thermophysical properties of HTF enable a highly efficient heat transfer process more specifically for waste heat recovery applications as an evolving industrial energy efficiency effort [4,10–12]. However, the thermal conductivities of common HTF are low, with water as the HTF with the highest thermal conductivity of 0.6 W/m.K, which is far lower compared to that of metals and metal oxides with 17.65 and 39 W/m.K for copper oxide and alumina respectively [13]. Accordingly, it has been proposed to add solid particles of such material to fluids to enhance the thermal conductivity, with special attention to particle sizes of nano-scale, i.e., nanoparticles (NPs) typically below 100 nm in size, hence the term nanofluid (NF) [14,15].

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Table 1
Thermophysical properties of common base fluids and nanoparticles [13,47,53,55–59].

	Thermal conductivity, W/m.K	Specific heat, kJ/kg.K	Density, kg/m^3	Viscosity, 10^{-3} Pa.S
Objective	*Increase*	*Increase*	*Decrease*	*Decrease*
		Common base fluids		
Distilled water (DI)	0.607	4.18	998	0.855
Ethylene glycol-water (1:1 vol.)	0.380	3.28	1,073	3.94
Ethylene glycol (EG)	0.255	2.35	1,111	15.5
Silicone oil (SO)	0.156	1.51	930	11
Engine oil (EO)	0.145	1.88	880	84
		Common nanoparticles materials		
Silver Ag	429	0.234	10,400	-
Copper Cu	398	0.385	8,933	-
Aluminum Al	237	0.877	2,700	-
Magnesia MgO	55	0.955	3,560	-
Alumina Al_2O_3	36–40	0.775	3,970	-
Copper oxide CuO	32.9	0.525	6,500	-
Titania TiO_2	8.4	0.692–0.711	4,000	-
Silica SiO_2	1.38	0.680–0.745	2,220	-
*Graphene**	6–5,000	0.643–2.100	2,000–2,500	-

* Graphene due to the unique properties have a wide range of reported thermophysical properties, as explained in the text.

Nanofluids are formed by simply dispersing NPs of high thermal conductivity material in a base fluid (BF) as a stable suspension [16–18]. A wide range of nanostructures, more specifically NPs have been investigated as additives to a wide range of BFs. This includes metallic NPs such as those of Silver Ag, Copper Cu, and Gold Ag, given the high thermal conductivity of such metals. However, the main challenge was their availability and associated cost [19,20]. Oxides of different elements such as alumina Al_2O_3, silica SiO_2, titania TiO_2, copper oxide CuO, and zinc oxide ZnO were studied as well, given their relatively higher thermal conductivity and higher availability, and lower cost [19,21,22]. However, the improvements in thermophysical properties were not comparable to that of metallic NPs. Conventional carbonaceous materials such as graphite, carbon black, activated carbon were studied as well, given the relatively higher thermal conductivity of almost ten times that of water [23–25]. Among carbon-based material used for NFs, graphene (Gr) have received an extensive research effort due to its outstanding thermal conductivity, which is theoretically in the order of 2,500-5,000 W/m.K, and at least is 6 W/m.K with high surface area and other outstanding properties [26,27]. Accordingly, Gr-based NFs have been investigated for a wide range of applications [28–30].

In the current work, the thermophysical properties of Gr-based NFs are critically discussed, showing the improvements attained for these specific NFs relative to other NFs and conventional HTFs. The focus of the discussion is given to the sole improvement effect of pristine Gr, rather than modified-Gr, to demonstrate its direct improvements on the thermophysical properties of the produced NF. The effect of different parameters such as concentration or loading of Gr as well as its different forms on the thermophysical properties improvements is explored and thoroughly discussed. This is very crucial to better understand the characteristic performance of Gr-based NFs, and helps to better optimize the NF toward the specific application, as well as in comparison to other NFs types. The enhancement of thermophysical properties is generally related to the unique characteristics of Gr material, of extremely high thermal conductivity, which results in outstanding HTF. Additionally, the challenge encountered with the commercial or industrial application of Gr-based NFs both economically and environmentally is discussed with some recommendations to address such challenges.

Nanofluids: preparation, stability, properties, and applications

Nanofluids, as explained earlier, is formed by dispersing NPs in base fluids (BFs). The NPs of many materials have been explored for NFs applications; this includes metals such as Aluminum Al, copper Cu, , gold Au, and silver Ag [31–33]. Oxides such as alumina Al_2O_3, silica SiO_2, titania TiO_2, and zinc oxide ZnO [34–38]. Carbon-based materials such as graphene (Gr), graphene oxide (GrO), carbon nanotube (CNT), and many others [26,30,39,40]. A wide range of BFs have been investigated with almost all feasible NPs; this includes water as the most commonly investigated BF, ethylene glycol (EG), silicone oil, engine oil, and transformer oil [7,41–44]. The thermophysical properties of the formed NFs primarily depend on the thermophysical properties of the respective NPs and BF and the interaction among them.

Table 1 shows the different thermophysical properties of common NPs and BFs. The table shows clearly the relative lower thermal conductivities of common HTF with 0.607, 0.255, and 0.156 W/m.K for water, EG, and silicone oil, respectively, which will result in poor heat transfer performance. On the other hand, metals such as silver Ag and Copper Cu have thermal conductivities of 429 and 389 W/m.K, respectively. However, these metals are not naturally occurring and require energy-intensive extraction processes, hence higher cost and environmental impacts. Metallic and non-metallic oxides are very abundant, with silica or sand as the most abundant oxide on earth, have reasonably higher thermal conductivity with about 55 W/m.K for Magnesia MgO, down to 1.4 W/m.K for silica SiO_2. Carbonaceous materials such as graphite and many others, with thermal conductivity of about 6 W/m.K, have been widely explored as well due to their high availability and lower cost and being more green chemicals as they can be derived from many wastes such as agriculture waste. Graphene among all carbon-based material has received huge attention due to its extremely high thermal conductivity up to 5,000 W/m.K and other shape-driven characteristics of being a single-layer of carbon atoms, which can impose different structure and surface functionalities on their performance. Additionally, Gr has a higher specific heat capacity relative to all other material present in the table, along with lower density, which both are favored characteristics for NF.

The shape and size of NPs have a significant role as well in determining the thermophysical properties of the result NF as well as the NF synthesis method [42,45–47]. It can be seen that increasing the thermal conductivity of NF, while reducing other thermophysical properties such as specific heat capacity, density, and viscosity is an ultimate goal. Hence the addition of high thermal conductivity NPs to the BF results in enhanced thermal conductivity of the result NF. NF stability is another critical factor that has to be considered, as it affects the ability of long operation of NFs without aggregation or clustering of NPs, which might lead to deposition of NPs and hence deteriorate the functionality of NFs [17,47,48].

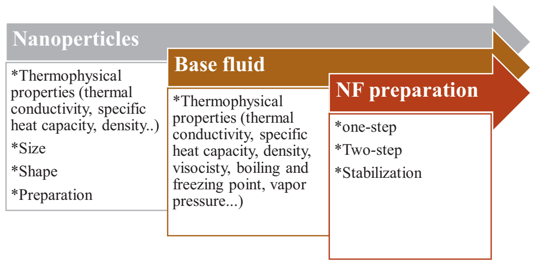

Fig. 1. Factors affecting the properties and performance of nanofluid.

Fig. 1 below shows the scheme of NF from preparation, considering the different factors that affect the properties and performance of NF. The first step is to choose the specific material to be used, as earlier discussed, a wide range of metals, oxides, and carbon-based material are available for such application. The selection of the material depends mainly on their availability and hence cost, as well as their thermophysical properties, more importantly, the thermal conductivity, in addition to specific heat capacity, as it contributes to the heating/cooling capacity of the NF, usually lowering the specific heat capacity of NF relative to the BF, more specifically for water, being the fluid with highest specific heat capacity [49–51]. The density is important as well, which usually increases due to the addition of the dense NPs, hence increases fluid friction and pumping energy requirements. In addition to the thermophysical properties of NPs, other characteristics such as particle shape and size play a crucial role as well, contributing to the final thermophysical properties of the NF. NPs preparation methods will have a direct impact on their characteristics, and more importantly on their stability in the NF, hence careful consideration should be given to the selection of the preparation method [48,52]. The thermophysical properties of NF depend mainly on those of NPs and BF. The thermophysical properties of BF as the continuous phase and the major component of the NF has a huge impact, hence the careful selection of the BF is equally required. The main thermophysical properties such as thermal conductivity, specific heat capacity, density, and viscosity are very important and have a direct impact on the obtained NF thermophysical properties. Additionally, other properties such as boiling point, freezing point, vapor pressure, and many others are very important from operational and safety aspects [15,53,54].

Preparation of nanofluids

Preparation of NF is very crucial as it has a substantial effect on the properties and stability of the results NF; hence careful attention should be given. Generally, there are two approaches for the preparation of NF, either one-step or two-step method [47,54]. In the one-step preparation method, the NPs are produced and readily dispersed in the BF. While in the two-step method, the NPs are separately produced or acquired readily and then dispersed in the BF. The selection of the preparation method depends mainly on the nature of NPs and BF used and their suitability for the chosen method [25,60,61].

One-step preparation of NFs can be achieved by several techniques such as direct evaporation, physical vapor deposition, and chemical vapor deposition [61]. The main advantages of the one-step method are uniform dispersion of NPs in BF, hence enhanced stability and minimized agglomeration of NPs. This is mainly because NPs are directly formed in the BF; therefore, drying, storage, and transportation of NPs are avoided. On the other hand, the one-step method is limited to certain NPs and BF and their specific combination. Additionally, the method results in limited NF quantity and NPs concentration, as well as being costly [62].

Two-step preparation of NFs is much simpler, which includes the dispersion of ready-made NPs in the BF. The NPs are synthesized separately or readily acquired and then added to the BF with violent agitation to ensure proper dispersion and homogeneity of the NF [17,42]. The main advantage of the two-step method is its simplicity as it involves only the addition of NPs to BF without any special chemical or physical conditions. The other advantage is the wide applicability, as a wide range of NPs, almost all types of NPs, can be dispersed in many BFs; hence the wide range of NFs can be produced. However, the two-step method requires very well dispersion of NPs in the BF, which is mainly due to the poor stability of NPs, hence tending to agglomerate and aggregate forming clusters of NPs with increased particle size, which results in less enhanced thermophysical properties, and even to sedimentation [36,63].

Stability of nanofluids

One of the major issues and challenges with NFs is the high tendency of NPs to agglomerate and aggregate. The nature of NPs, their loading or volume fraction (φ), i.e., percentage or concentration of NPs in the BF, along with their interaction with the BF, highly affect the stability of the NF [28]. Fig. 2 below demonstrate NPs as suspended in the BF. Nanoparticles are usually negatively charged, hence attracting positive charge surrounding the particle creating an immobilized fluid layer, hence creating an effective particle volume. Some charge balance or equilibrium is attained among positively and negatively charged species in the NF in order to maintain stability. The increase in the number of particles present in suspension along with charge neutralization results in aggregation of particles to form larger particles, hence agglomerate and eventually results in sedimentation due to the increased size and density of particles relative to the fluid [64,65]. This, in turn, might result in the loss of NPs from the NF and creating some problems in process equipment.

The stability of NF can be accomplished by different approaches. One of the first approaches is the proper preparation and dispersion of NPs in the BF. The strong and violent agitation during the dispersion of NPs in BF in the two-step method ensures good stability of NF, which is usually followed by ultrasonication to augment the dispersibility of NPs [65]. The stability of NF can be measured by the polydispersity index (PDI), with the lower PDI, the higher the stability of the NF. Zeta potential is another more accurate measure of NF stability, as it is a measure of the thickness of the layer surrounding the particle as shown in Fig. 2, with the higher the absolute zeta potential values, the higher the stability of the NF. Generally, a zeta potential < 30 mV indicates limited stability, while 30-60 mV indicates physical stability, and ≥ 60 mV indicates excellent stability [17,64]. Additionally, the measurement of ultraviolet UV absorbance can be used as another measure for the NF stability with the decrease in absorbance intensity as a measure for aggregation and sedimentation of NPs.

The addition of surfactants enhances the stability of NPs and preventing the aggregation of these particles in the NF. Surfactants molecules are attached to the NP surface, causing some surface charge repulsion among particles and keeping the NPs well dispersed. Fig. 3 below shows a schematic of the typical surfactant action on the surface of NPs. The surfactant usually alters the surface nature of the NPs from hydrophilic to hydrophobic and vice versa hence increasing the zeta potential [17,64,67].

Thermophysical properties of nanofluids

The main thermophysical properties of NF is increased thermal conductivity. This is simply due to the dispersion of high thermal conductivity material in particulate form. The mechanisms by which the NPs increase the thermal conductivity of the BF are either due to the Brownian motion, molecular-level layering at the particle/liquid interface, natural heat transfer between liquid and particle, and NPs clustering effect [68]. These mechanisms are illustrated in Fig. 4 below, showing the respective thermal conductivity enhancement, which varies significantly with the NPs and BF type, NF preparation

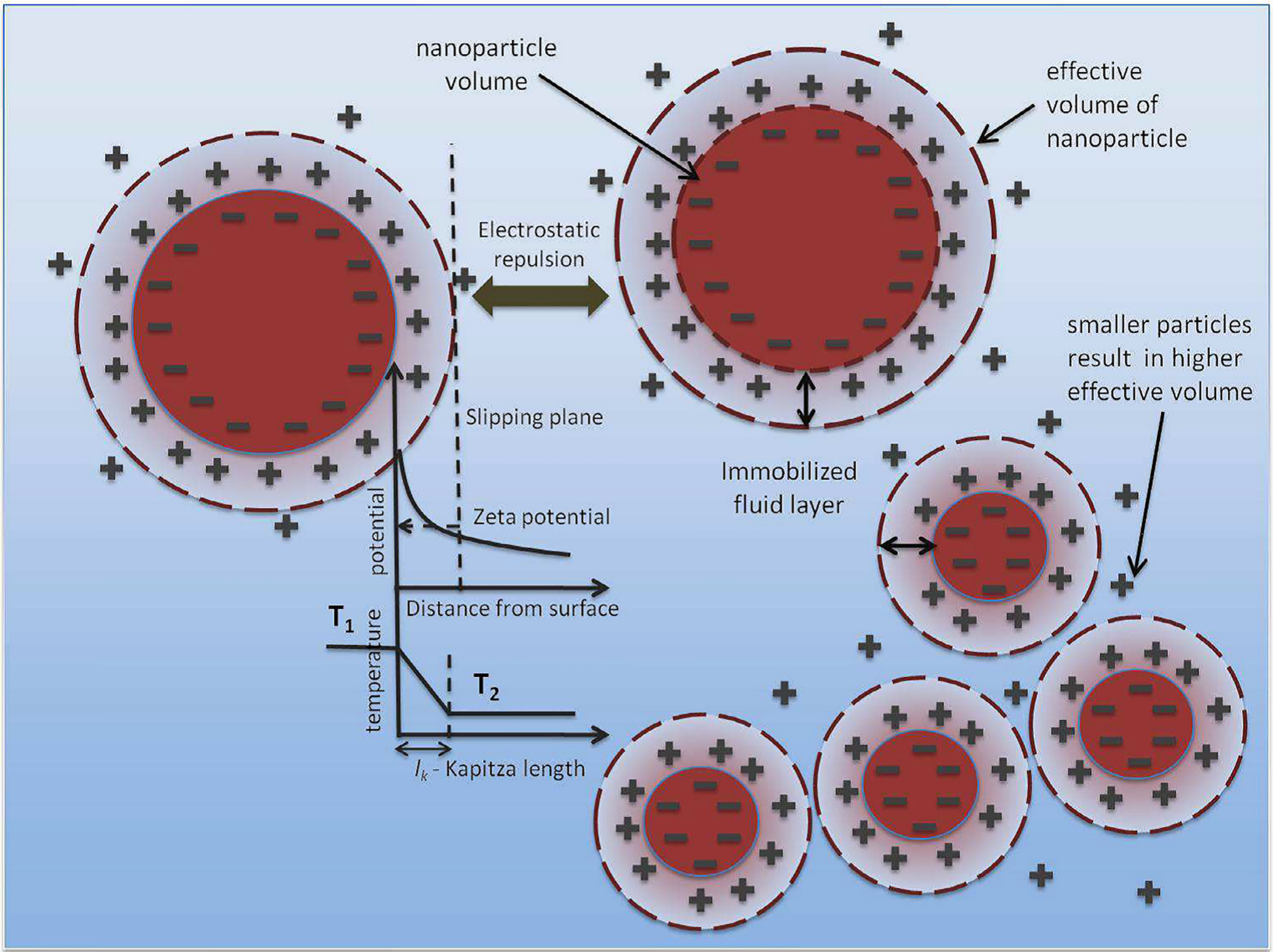

Fig. 2. Interfacial phenomena of particles suspended in fluid [66].

method, and NF stability. Fig. 5 below shows the enhancement of thermal conductivity of a wide range of NFs as reported by Elena V. Timofeeva [66]. The Fig. shows an increase in thermal conductivity of the NF, reaching almost three times that of the BF. The effective thermal conductivity of NF, k_{NF} can be related to the thermal conductivity

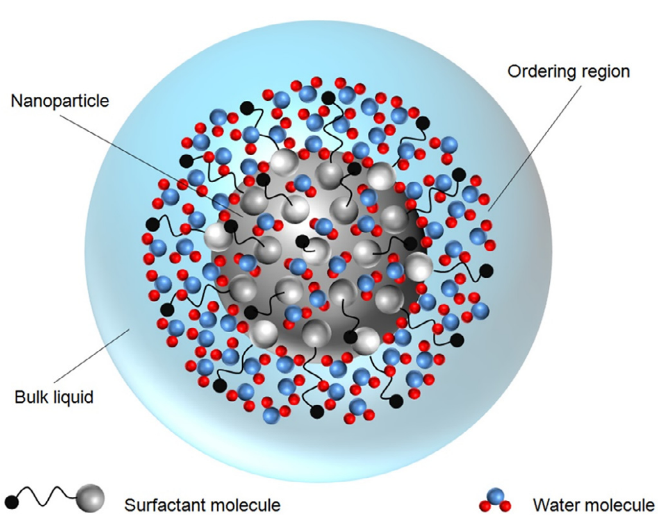

Fig. 3. Surfactant action on the surface of nanoparticles [53].

of the NPs, k_{NP}, thermal conductivity of the BF, k_{BF}, and the volume fraction (φ) by the simple Maxwell equation, Eq. (1); however, several investigations have shown that this equation tends to underestimate the thermal conductivity enhancements, being mainly valid for micro-sized suspension [69–71]. As a result, many correlations have been developed to relate these parameters by modifying the principle Maxwell equation.

$$\frac{k_{NF}}{k_{BF}}=\frac{k_{NP}+2k_{BF}+2\varphi(k_{NP}-\mathrm{k_{BF}})}{k_{NP}+2k_{BF}-\varphi(\mathrm{k_{NP}}-\mathrm{k_{BF}})} \quad (1)$$

The added NPs results in the decrease of specific heat capacity of BF as such solid particles are common of the lower heat capacity as compared to the BF, with water having the highest heat capacity as shown in Table 1. Similar to thermal conductivity, some general and principle equations can predict the specific heat capacity of NF, $C_{p,NF}$, in relation to the specific heat capacity of the NPs and BF, i.e., $C_{p,NP}$ and $C_{p,BF}$, respectively, and the volume fraction (φ) by the simple Maxwell equation, Eq. (2) or Eq. (3), with the latter involving the density of NF, NPs, and BF, i.e., $\rho_{NF,}$ ρ_{Np}, and ρ_{BF} respectively [53,55,72]. Although these equations are subject to accuracy and applicability issues for specific NPs and BF pairs, it is still can be used to provide primary guide values for the heat capacity.

$$C_{p,NF}=(1-\varphi)C_{p,BF}+\varphi C_{p,NP} \quad (2)$$

$$C_{p,NF}=\left[(1-\varphi)\rho_{BF}C_{p,BF}+\varphi\rho_{NP}C_{p,NP}\right]/\rho_{NF} \quad (3)$$

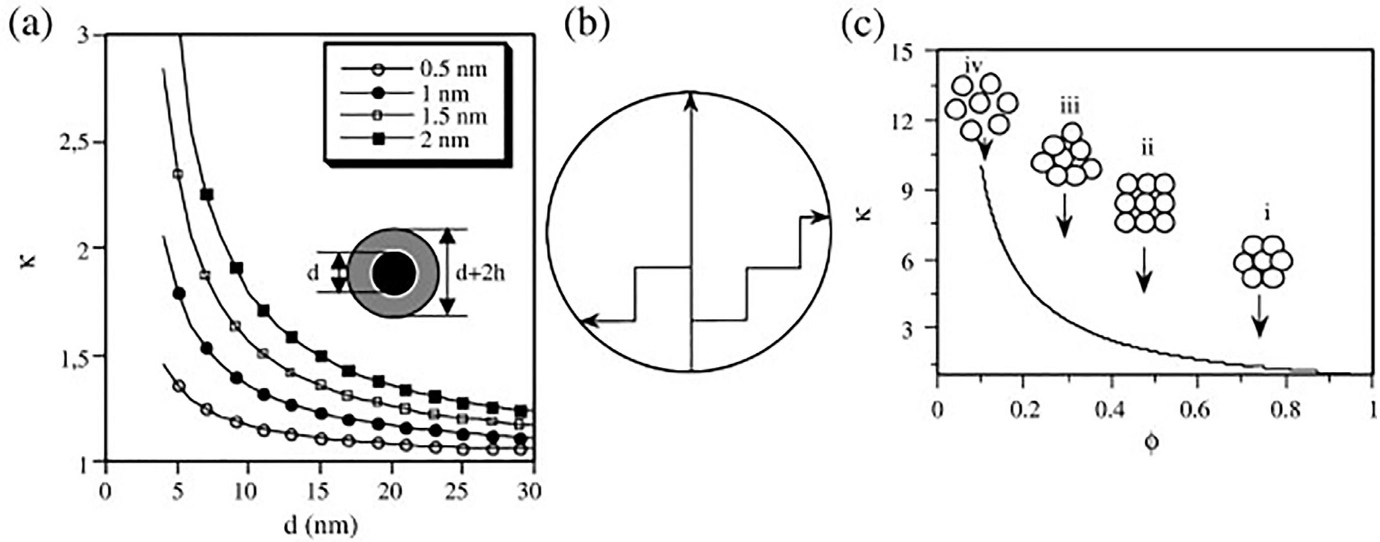

Fig. 4. Thermal conductivity enhancement mechanisms in nanofluids: a) formation of a high thermal conductive layer, b) diffusive and Ballistic and phonon transport, c) Formation of highly thermal conductive clusters [68].

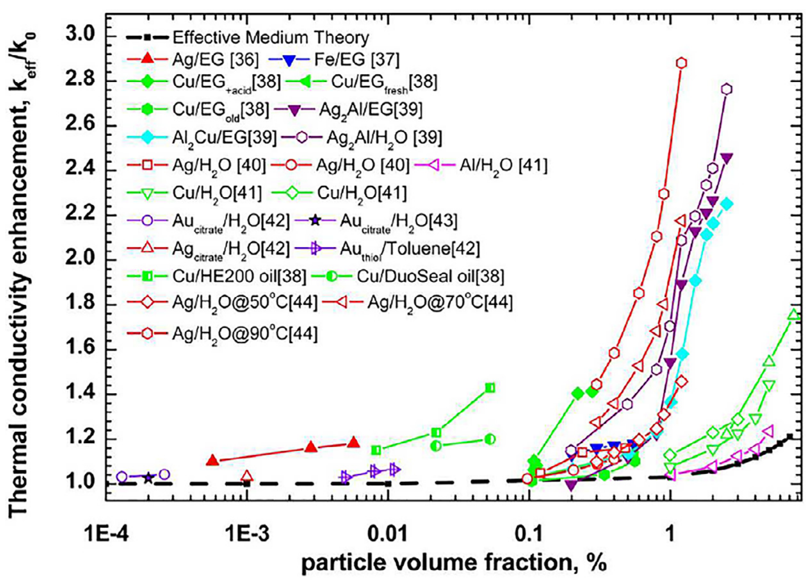

Fig. 5. Thermal conductivity enhancement in different nanofluids [66].

The density and viscosity of NF are typically higher than that of the BF, which is attributed to the typical higher density of NPs, and the fluid flow resistance due to the presence of NPs. This, in turn, is considered one of the disadvantages of NF, as such an increase in density and viscosity will result in higher pressure drop and hence higher pumping power requirement for a specific duty. Simple equations to those of thermal conductivity and specific heat capacity are developed. Eqs. (4) and (5) are usually used to provide rough estimation for the density and viscosity of NFs, i.e., ρ_{NF} and μ_{NF}, respectively, in terms of the densities of NPs and BF, ρ_{NP}, and ρ_{BF} respectively, and viscosity of the BF, μ and the volume fraction (φ) [53,72,73].

$$\rho_{NF} = (1-\varphi)\rho_{BF} + \varphi\rho_{NP} \tag{4}$$

$$\mu_{NF} = \left(1 + 2.5\varphi + 6.25\varphi^2\right)\mu \tag{5}$$

The thermophysical properties of NF play a crucial role in determining the enhancement in heat transfer coefficient (HTC), which can be related to these properties and flow nature using the dimensionless group of number, Reynolds number Re, Prandtl number Pr, and Nusselt number Nu according to Eqs (6) - (9) [74]:

$$Re = \frac{\rho du}{\mu} = \frac{du}{v} \tag{6}$$

$$Pr = \frac{v}{\alpha} = \frac{C_p\mu}{k} \tag{7}$$

$$Nu = \frac{hL}{k} \tag{8}$$

$$Nu = f(Re, Pr) = aRe^n Pr^m \tag{9}$$

Where: ρ = fluid density, d =characteristic diameter, u = flow velocity, μ = fluid dynamic viscosity, v = fluid kinematic viscosity, α = thermal diffusivity, C_p = fluid specific heat capacity, k = fluid thermal conductivity, h = convective heat transfer coefficient, L = characteristic heat transfer length, and a, m, n = correlation coefficients.

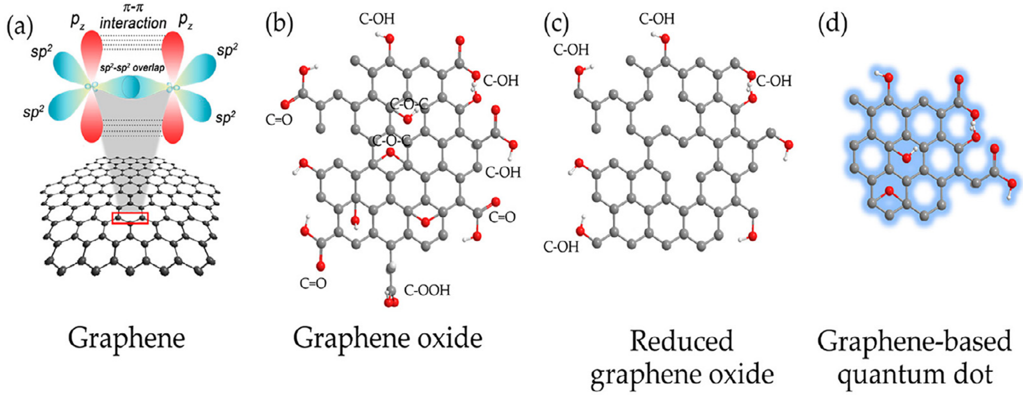

Fig. 6. Schematic structure of a) graphene, b) graphene oxide, c) reduced graphene oxide, and d) graphene-based quantum dots [92].

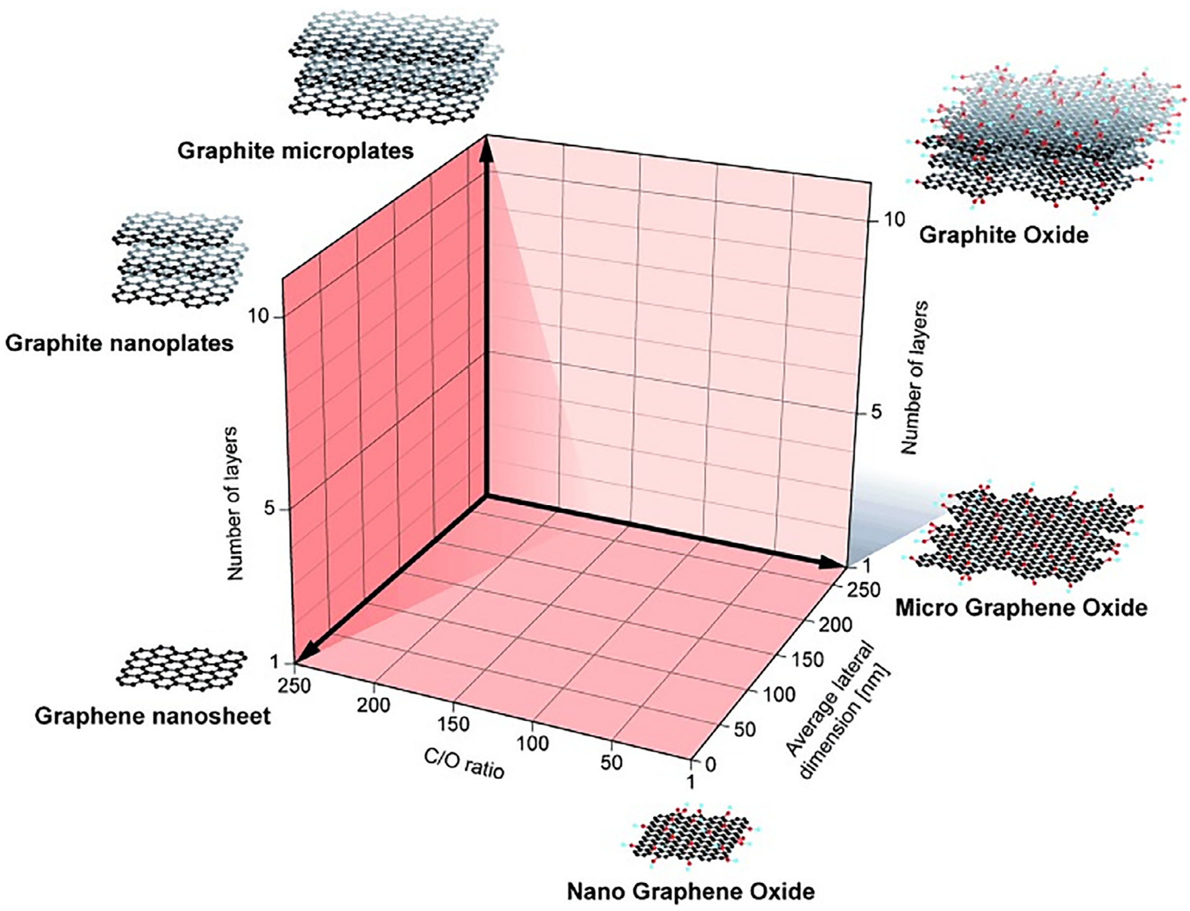

Fig. 7. Classification and forms of graphene and graphene derivatives [93].

Applications of nanofluids

Due to the superior performance of NF relative to the respective BF, it has been widely used as a high-performance heat transfer fluid in a wide range of applications. The main application for NF is in heat transfer in different heat exchanger (HEx) types such as plate heat exchanger [75–77], shell and tube HEx [78], heat pipe [79,80], renewable energy harvesting as in solar collectors and photovoltaic [24,26,81–83], phase change material for energy storage [84], desalination [85,86], fuel additive for combustion improvement [3,12,87,88], and many other applications. NFs have a huge potential for recovery of waste heat to increase the energy efficiency of different processes due to their better thermophysical properties [89]. In all these applications, the utilization of NF has shown a substantial improvement in process performance relative to the use of conventional fluid, resulting in improved efficiency, reduced energy consumption, and lower cost.

Graphene-based nanofluids: Preparation and thermophysical properties

Graphene (Gr) has evolved as an emerging material since its early isolation in 2004, although it was theoretically expected as back to the mid-18th century, which was crowned by Nobel Prize in Physics in 2010 [90]. Since its early preparation and isolation, graphene and common Gr-derivatives such as graphene oxide (GrO) and reduced graphene oxide (rGrO) have been investigated for a wide range of applications; almost all applications require unique material. The main driver for such huge attention was mainly due to its outstanding chemical, mechanical, thermal, physical, optical, and electrical properties [30,91]. Gr is simply a two-dimensional 2D allotrope of carbon in which carbon atoms are ordered in one layer with a thickness of one carbon atom. Fig. 6 below shows the structure of Gr, GrO, rGrO, and Gr quantum dots, while Fig. 7 shows the different forms of Gr derivatives. Gr is usually produced by isolating one-layer thickness from graphite as the parent material, which is simply the three-dimensional 3D allotrope of carbon. Gr can be produced by chemical vapor deposition, mechanical, chemical, and electrochemical exfoliation, epitaxial growth on SiC, and many other recent methods [90,92].

Preparation of graphene-based nanofluids

Preparation of Gr-based NF follows the same classification of NF preparation either through one-step method or two-step methods as discussed earlier in 2.1. However, most of the literature work on Gr-based NF have been found to utilize the two-step method [27,30,40,94]. The two-step methods have two main advantages when considered for Gr-based NF. The first advantage is the possibility of acquiring commercially readily available Gr with given specification and known characterization, hence reduce the time required for exhaustive Gr preparation methods, which to be followed with needed advanced characterization techniques. The second advantage is the possibility of carrying out much modification to the graphene, following on-site preparation or external acquiring, which aims to enhance some of its properties. Nitrogen-doped Gr (ND-Gr) has been widely used for NF, hence further enhancing the thermophysical properties of the NF [95–97], silver-decorated [98,99], core/shell copper-Gr [100], and many other metallic and non-metallic species.

However, with the wide application of the two-step NF preparation method special attention has been given to ensure the stability of the NF. In this regard, the unique properties of Gr of low bulk density of about 0.20-0.40 g/ml, and true density of 2.0-2.25 g/ml, which is typical to that of carbon/graphite plays an important role, which is close to that of water relative to other common NPs as shown in Table 1 [56,58]. The sedimentation velocity of suspended particles in the fluid can be determined using Stokes's equation, Eq. (10) below in terms of the particle radius r, fluid density ρ_F, fluid viscosity μ, particle density ρ_P, and acceleration due to gravity g [27,94]. In the case of Gr-based NF, this sedimentation velocity is much less due to the lower density difference between Gr and base fluid, which helps to maintain the stability of the NF.

$$V_{sed} = \left[r^2(\rho_{NP} - \rho_{BF})g\right]/9\mu \tag{10}$$

Thermal Conductivity

The utmost property of NF is the higher thermal conductivity, with high thermal conductive NPs added specifically to increase the thermal conductivity of BF. The enhancement in thermal conductivity can be easily evaluated by obtaining the ratio of the thermal conductivity of the NF to that of the BF, i.e., k_{NF}/k_{BF}, which should be greater than unity, and the higher the ratio, the better the NF. The simplest relation for evaluating the k_{NF}/k_{BF}, is given in Eq. 1; however, experimental data deviate much from this simple equation due to the inherent assumptions of ideal spherical particles, uniform single particle size, and absence of mutual interaction among NPs and BF and NPs themselves, which is not quite accurate [46,53]. Several correlations have been developed to express the enhancement in thermal conductivity, which are validated versus specific experimental data [101]. These correlations are considering particle size, specific surface area, shape, and many other properties of NPs, with the works of Yang et al. [102], Angayarkanni and Philip [16], and Akilu et al. [13] provide a good compilation of these correlations.

Fig. 8 below shows the thermal conductivity ratio for different forms of Gr and some typical metallic oxides in common BF. The figure demonstrates the outstanding thermal conductivity improvement upon the addition of Gr, showing that much less volume fraction can result in a substantial increase in thermal conductivity. The significant rise in thermal conductivity of Gr-based NF is attributed to the high thermal conductivity of Gr, which is claimed to be up to 3,000 W/m.K (parallel to the Gr surface) to a minimum of 6 W/m.K (perpendicular to Gr surface) [56], with former value reported to be

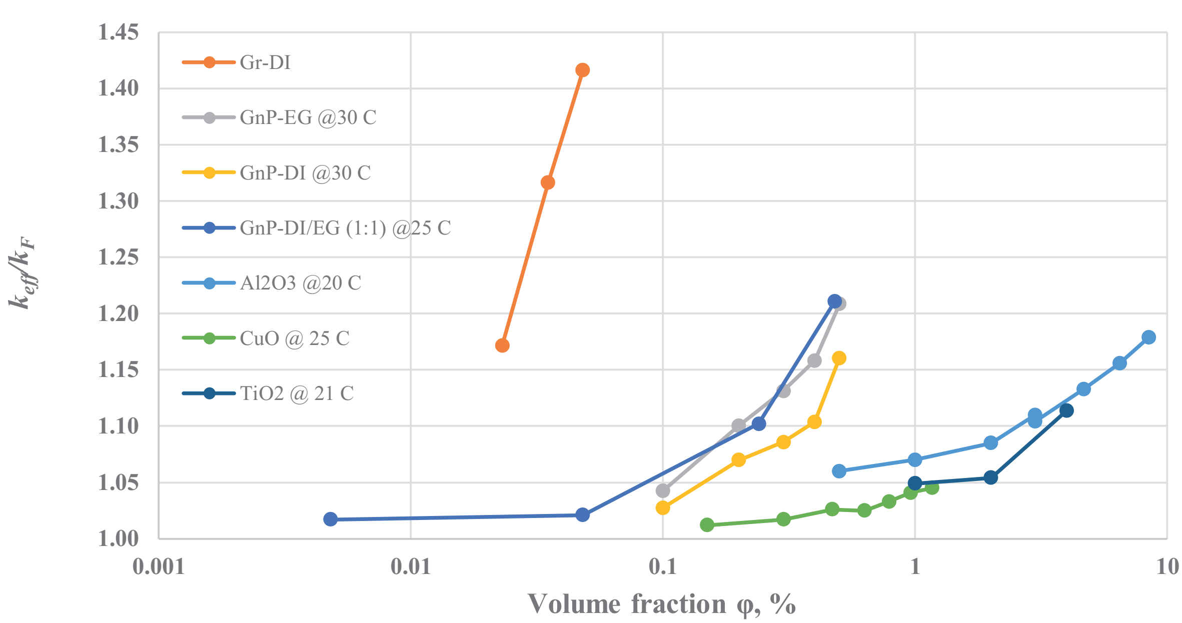

Fig. 8. Thermal conductivity enhancement of graphene-based nanofluids relative to other nanofluids (data obtained from [34,58,103–106]). Gr = graphene, GnP = graphene nanoplatelets, DI = deionized water, and EG = ethylene glycol.

Table 2
Enhancement in thermal conductivity of graphene-based nanofluids.

Gr material	Base fluid	k_{NF}/k_{BF}	Remarks	Ref.
3D-Gr	EG	1.149	At φ = 0.1% wt. and 25 °C.	Bing et al. [103]
Gr	DI	1.416	At φ = 0.1% wt. and 25 °C	Ghozatloo et al. [58]
GnPGnP	EG	1.208	At φ = 0.5% vol. and 35 °C	Selvam et al. [104]
	DIDI	1.160		
		1.287	At φ = 0.1% wt. and 60 °C relative to 1.205 at 20 °C for 500 m^2/g GnP.	Iranmanesh et al. [101]
		1.307	At φ = 0.1% wt. and 60 °C relative to 1.234 at 20 °C for 750 m^2/g GnP.	
Gr	EG-DI (1:1 vol.)	1.092	At φ = 0.2% wt. and 25 °C.	Martin et al. [107]
GrnP		1.211	At φ = 1% wt. and 25 °C and 750 m^2/g.	Wang et al. [105]

*Gr = graphene, GrnP = graphene nanoplatelets, EG = ethylene glycol, DI = deionized water.

down to 2,500 W/m.K [58] and up to 5,000 W/m.K [57]. The Figure also shows that the thermal conductivity increase depends on the BF as well, as in the case of Gr nanoplatelets (GnP) when added to ethylene glycol (EG) and water (DI), which results in a higher increase in the case of the former. Table 2 shows some of the recently reported enhancement in thermal conductivity of Gr-based NF.

Specific heat capacity

The specific heat capacity is simply the amount of energy required to increase the temperature of a unit mass of matter by unit temperature and expressed J/g.K in the metric system of units. The specific heat of NF as typical HTF plays a crucial role as it determines the heating/cooling capacity of the fluid per unit decrease/increase in temperature, which is simply the product of fluid flowrate and specific heat capacity in J/k, with the higher the heat capacity, the better the HTF is. Accordingly, the higher the specific heat capacity, the higher the heat capacity, hence requires less HTF flow for a certain heating/cooling duty, which also decreases the pumping power requirements.

Unfortunately, the addition of NPs to BF results in the decrease of the specific heat capacity of the result NF. As given in Table 1, the specific heat capacity of BF is much higher than that of NPs, with water and ethylene glycol have about 4.18 and 2.35 J/g.K, respectively. While graphene has a value range of 0.643-2.10 J/g.K, which is one of the highest specific heat capacity among NPs, but still lower than that of the BF [53,108]. The specific heat capacity can be related to the specific heat capacities of the NPs and BF along with the volume fraction in simple correlation as given in Eq. (2) and (3). Liu et al. have reported the drop of specific heat capacity of Gr-water NF from 3.915 J/g.K for pure water at 20 °C to 3.875, 3.834, and 3.768 J/g.K for 0.2, 0.4, and 0.8 wt.%, respectively representing a drop up to 3.75% [109].

Density

The density of NF is higher than that of the BF; this is simply due to the higher density of the solid NPs added, as shown in Table 1 and according to the simple relation of Eq. (4). However, in the case of Gr, a clear distinction can be made, given the fact that Gr is porous in nature; hence it has true density and bulk density. The true density of Gr is close to that of graphite of 2.00-2.50 g/ml, while the bulk density of porous Gr structures is much less in the range of 0.2-0.4 g/ml [56,58]. The increase in density, along with the viscosity of NF, has a significant effect on increasing pressure drop and required pumping power. The density of Gr/DI NF has increased from 0.9805 g/ml for pure water to 0.9934 g/ml at 0.8 wt.%, equivalent to an increase of about 1.3% [109].

The increase in density of NF due to the higher density of NPs has another critical role in the stability of NF, with the higher density difference between the BF and NPs, i.e., higher NPs density, the higher settling or sedimentation velocity of the NPs, according to Eq. (10) [110–112]. Hence the lower NF stability, with loss of NPs in the heat transfer loop over time, which result in degradation of thermal performance. In this regard, the density of graphene seems to be of the closest value to the density of most BF, hence the higher stability relative to other NPs types, which is an additional advantage of Gr-based NF relative to other types of NFs.

Viscosity

Viscosity simply represents the resistance of the fluid to flow due to inter-layer or fluid/surface interaction. Similar to density, the viscosity has two detrimental effects on pressure drop and pumping power requirements. The presence of NPs in the BF, i.e., forming the NF, result in increasing the friction at the fluid/surface interface due to NPs/surface collisions and other inter-layer resistance and interfacial forces. These interfacial resistances result in increasing the viscosity of the NF relative to the BF. The simple correlation expressing the viscosity of NF is presented in Eq. (5); however, more representative models for a wide range of NF have been developed and validated as summarized in the works of Yang et al. [102], Angayarkanni and Philip [16], and Akilu et al. [13] provide a good compilation of these correlations. Table 3 and Fig. 9 below shows the increase in viscosity of NF relative to that of BF, i.e., μ_{NF}/μ_{BF}, which is always higher than unity. Table 3 shows an increase in the thermal conductivity of Gr-based NF up to 50% over φ range of 0.1-0.8%. The table also shows that graphene nanoplatlets GrnP results in a higher increase in the viscosity relative to Gr itself. Fig. 9 shows that a much higher increase in the viscosity of the NF is observed at a lower φ for Gr-based NFs reaching about 40-50% in some cases at about φ of 0.1%, which is much higher than that encountered with oxides NF at even higher φ of 1%. Similar to thermal conductivity, the increase in Gr-based NF viscosity is much higher than other NF. However, the increase in viscosity is much higher compared to that of the thermal conductivity.

Challenges and recommendations of graphene-based nanofluids

Gr-based NFs have shown very high potential as an efficient HTF due to the significant increase in thermal conductivity relative to the BF. However, there are some challenges associated with the wide

Table 3
Viscosity increase of graphene-based nanofluids.

Gr material	Base fluid	μ_{NF}/μ_{BF}	Remarks	Ref.
Gr	DI	1.12	At φ = 0.1% wt. and 25 °C	Ghozatloo et al. [58]
		1.506	At φ = 0.8% wt. and 25 °C	Changhui Liu et al. [109]
GrnP		1.380	At φ = 0.1% wt. at 20 °C for 500 m^2/g GnP.	Iranmanesh et al. [101]
		1.440	At φ = 0.1% wt. and 20 °C for 750 m^2/g GnP.	
Gr	EG-DI (1:1 vol.)	1.096	At φ = 0.2% wt. and 25 °C.	Martin et al. [107]
GrnP		1.45	At φ = 0.8% wt. and 25 °C and 750 m^2/g.	Wang et al. [105]

*Gr = graphene, GrnP = graphene nanoplatelets, EG = ethylene glycol, DI = deionized water.

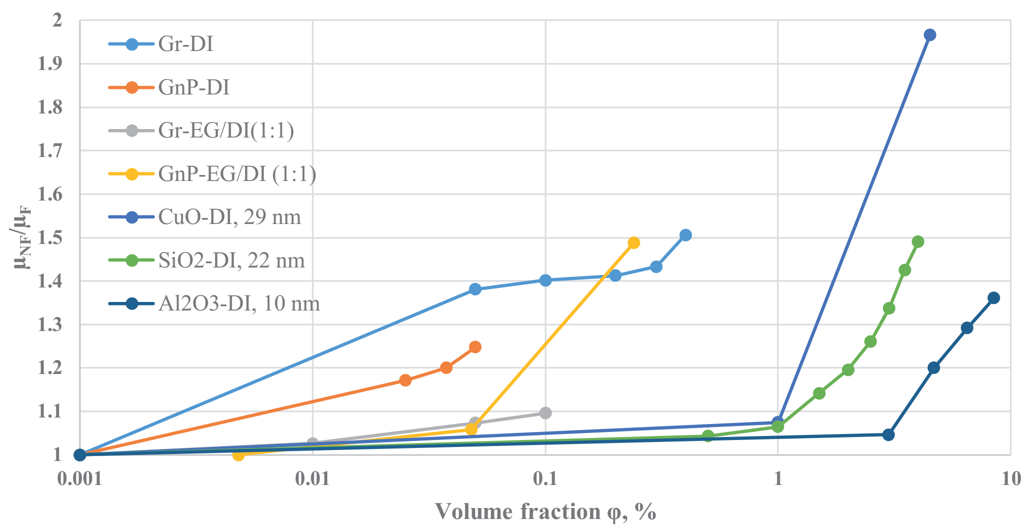

Fig. 9. The relative increase in viscosity of nanofluid relative to base fluid for graphene-based nanofluids and some other nanofluids (data obtained from [34,58,101,105–107,113]. Gr = graphene, GnP = graphene nanoplatelets, EG = ethylene glycol, DI = deionized water.

application of Gr-Based NF. These challenges can be summarized as follow:

Ø High cost,
Ø Stability and dispersion,
Ø Increased density and viscosity
Ø Environmental impacts of Gr NPs

The major challenge to widely utilize Gr-based NF is the high cost associated with Gr and Gr derivatives. Although of the relatively cheap and abundant precursors for synthesizing Gr, i.e., graphite, the synthesis methods for Gr are very advanced and exhaustive in terms of chemicals used, energy consumed, and procedure followed to ensure obtaining the right 2D structure and defect-free Gr with high-quality control measures during synthesis [111,114–116]. Following the synthesis of the Gr complex and delicate purification and downstream processing are required to ensure the recovery of the synthesized Gr at high yield and purity. In addition, the characterization of the Gr and Gr derivatives requires advanced analytical techniques, including different scanning and transmission electron microscopy (SEM and TEM respectively) to confirm the structure and purity of the product. Accordingly, a careful benefit-cost analysis has to be made for the application of Gr-based NF to accurately balance the gained improvement in thermophysical properties versus the higher cost associated with such NF.

The second major challenge facing Gr-based NF, like any other typical NF, is to maintain the stability of NF and avoid NPs aggregation and agglomeration, which lead to sedimentation of accumulation of such NPs in process equipment [25,65,117]. Gr is hydrophobic in nature, and hence particles tend to aggregate rather than to disperse in water as the most common HTF. Accordingly, surface modification with hydrophilic groups was used as an approach to improve its stability [111,112,118,119]. The addition of surfactant helps to alter the surface charge and nature, changing it from hydrophobic to hydrophilic in the case of Gr-based NF, as shown in Fig. 3 [28,48,71,117].

Gr-based NF has a significantly enhanced thermal conductivity, which is due to the high thermal conductivity of Gr. However, the Gr-based NF also has a higher density and viscosity, as discussed earlier relative to other NFs. The increased density and viscosity results in a higher pressure drop of the fluid along with higher pumping power requirements, which is a critical trade-off. Accordingly, it is very crucial to optimize the volume fraction φ of Gr in Gr-based NF to maximize the gain in thermal conductivity while minimizing the increase in density and viscosity. This will also help in decreasing the cost associated with Gr-based NF.

The last challenge with Gr-based NF is its environmental impacts (EI). The EI has become an essential pillar of evaluating and assessing the eco-friendly and sustainable nature of a wide range of processes [120–123]. NF can affect the environment harshly due to the presence of NPs; this is in addition to the BF in case of loss of containment or disposal at the end of duty. Water as BF does not have many EIs, and most of the result EIs are due to the added NPs, which include metals and metal oxides [83,102,124]. NF use is currently under development; hence it requires careful assessment and evaluation of EI. Gr as a carbon allotrope can be approximated to graphite; which will have less EI as compared to other materials, however, due to the nanosize, it can act differently in natural systems. Accordingly, a specific and detailed environmental impact assessment for the use of Gr-based NF is required for each particular application.

Given the high potential and promising enhancement in the different thermophysical properties encountered with Gr-based NF, some recommendations for further study and future work can be provided to expand its application and overcome the faced challenges. These recommendations can be summarized as follow:

- Ø Although of the theoretically very high thermal conductivity of Gr, the enhancement in thermal conductivity of Gr-based NF was up to 50% only, which has to be further studied. Some modeling and theoretical studies can better interpret such behavior, hence help to obtain higher thermal conductivity.
- Ø It was found that the visocity of Gr-based NF is higher compared to other metallic and oxides NFs, which might result in increased fluid flow friction and pumping energy requirements. Hence a careful study of the friction and associated pressure drop, along with pumping energy requirements is required to better evaluate the effectiveness of utilized Gr-based NF measuring the enhanced thermal conductivity versus the increased friction and energy requirements.
- Ø Incorporation of metals, more specifically copper Cu due to its availability and low cost along with high thermal conductivity, into graphene might evolve as a tool to further enhance the thermophysical properties of Gr-based NF.
- Ø Although the technical feasibility of using Gr-based NF, the economical feasibility has to be carefully assessed and evaluated, hence an optimum technical and economical feasibility range can be identified.

Conclusions

Nanofluids (NF) have been evolved recently as highly efficient heat transfer fluid with outstanding thermophysical properties. A wide range of nanostructures, specifically nanoparticles NPs have been added to mostly used fluids to enhance their thermophysical properties. Among materials with high potential as efficient NF is graphene Gr, due to its tremendous thermal conductivity of about 5,000 W/m.K. Accordingly, the Gr-based NFs have been explored and investigated for a wide range of applications with almost all types of fluids. Gr-based NFs have shown much higher thermal conductivity at much lower concentration relative to high thermal conductivity metals and metal oxides such as Ag, Cu, Al_2O_3, and SiO_2, showing up to 40-50% increase at 0.1 wt.%. However, Gr-based NF has a lower specific heat capacity, higher density, and viscosity, which is common for typical NFs. Specifically, the viscosity of Gr-based NFs was found to be higher relative to other types of NF and at lower concentrations. Accordingly, the volume fraction φ has to be well-optimized to gain higher thermal conductivity at affordable density and viscosity increase to minimize pressure drop and pumping power requirements. Some of the challenges facing the wide application of Gr-based NF, including high cost, stability, and environmental impacts, have been discussed, which require careful investigation to promote the use of Gr-based NFs. Most of these challenges are associated with Gr preparation methods, which are currently costly, material, and energy exhaustive, hence advancement in Gr manufacture might relief many technical and economic constraints for the applications of Gr-based NFs.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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