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Graphene-based nanofluids: A comprehensive review about rheological behavior and dynamic viscosity

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Abstract: Graphene derivatives are promising nanomaterials for producing nanofluids due to their excellent intrinsic characteristics. Among thermophysical profile, and in addition to thermal properties, relevant property to evaluate the potential of graphene-based nanofluids as efficient and reliable heat transfer fluids is viscosity, and rheological behavior in a wider sense. Therefore, the aim of this review paper is to give a comprehensive overview of the current knowledge and results about the rheological properties of graphene-based nanofluids. After a brief description of the most common methods used for fabricating or extracting graphene derivatives, the main steps of graphene-based nanofluids preparation are introduced. Then, literature results on Newtonian/non-Newtonian behavior as well as variations in apparent dynamic viscosity of suspensions containing graphene derivatives are reviewed, analyzing the effects of shear rate, concentration, base fluids and temperature. Such an analysis is performed distinguishing the different types of graphene derivatives, namely graphene oxide, reduced graphene oxide, pristine graphene, graphene quantum dots, functionalized and doped graphene. Also, the impact of base fluid, temperature, concentration and surfactant on viscosity enhancement of graphene-based nanofluids is graphically and newly presented and discussed. In addition, the current models for viscosity prediction or correlation of those nanofluids are detailed. Finally, challenges and future works are summarized.

Keywords: Graphene derivatives, nanofluids, rheological behavior, dynamic viscosity, concentration and temperature effect, Influence of base fluid and surfactant

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Nomenclature

A, B	Andrade's equation fitting parameters
A-GQD	Amine-treated graphene quantum dots
Ag-rGO	reduced graphene oxides decorated with silver nanoparticles
cGnP	clove-treated graphene nanoplatelets
crGO	controlled reduced graphene oxide
CVD	chemical vapor deposition
DW	distilled water
E, F, G	Vallejo et al.'s fitting parameters.
EG	ethylene glycol
EGGnP	ethylene glycol-functionalized graphene nanoplatelets
[EMIM][DCA]	1-Ethyl-3-methylimidazolium dicyanamide
[EMIM][TFSI]	1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide
[HMIM]BF_4	1-hexyl-3-methylimidazolium tetrafluoroborate ionic liquid
f-GnP	functionalized graphene nanoplatelets
f-GnS	functionalized graphene nanosheets
f-HEG	functionalized graphene (Hummers method followed by exfoliation and reduction by hydrogen gas and then by functionalization using acid)
FLG	few-layer graphene
G	graphene
GA	gum arabic
GAGnP	gallic acid-functionalized graphene nanoplatelets
Gly	glycerol
GMF	graphene oxide(GO)/carbon nanotube(MWCNT)/Fe_3O_4
GnP	graphene nanoplatelets
GnP-COOH	COOH-covalently functionalized graphene nanoplatelets

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GnP/MWCNT	graphene nanoplatelets/multi-walled carbon nanotubes hybrid nanoparticles
GnP-Pt	functionalized graphene nanoplatelets-platinum
GnP-SDBS	SDBS-functionalized graphene nanoplatelets
GnS	graphene nanosheets
GO	graphene oxide
GO/Co_3O_4	graphene oxide decorated with copper oxide
GO-SiO_2	graphene oxide-silica hybrid nanoadditive
GQDs	graphene quantum dots
GtO	graphite Oxide
HOPG	highly ordered pyrolytic graphite
HTFs	heat transfer fluids
MD	molecular dynamics simulation
MLG	multi-layer/thick graphene
MWCNTs	multi-walled carbon nanotubes
NDG	nitrogene-doped graphene
NFs	nanofluids
N_{layer}	number of layers
PEG	poly(ethylene glycol)
PG	propilene glycol
PVA	polyvinyl alcohol
PVP	polyvinylpyrrolidone
rGO	reduced graphene oxide
rGO-Fe_3O_4	reduced graphene oxide-magnetite hybrid nanoadditive
SDBS	sodium dodecylbenzene sulfonate

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SDC	sodium deoxycholate
SDC	sodium deoxycholate
SDS	sodium dodecyl sulfate
SLG	single-layer graphene
SSA	specific surface area
T	temperature (K)
TEA-GnP	triethanolamine-treated graphene nanoplatelets
W	water
W-rGO	red wine reduced graphene oxide
δ_1, δ_3	relaxation (δ_1) and retardation (δ_3) times in Deborah number
μ	dynamic viscosity (Pa·s)
μ_0, D, T_0	Vogel-Fulcher-Tammann (VFT)- fitting coefficients
$\mu_r = \mu_{nf}/\mu_{bf}$	relative viscosity
φ	mass fraction
ϕ	volume fraction
ϕ_m	maximum packing volume fraction

Subscripts

nf	nanofluid
bf	base fluid

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1. Background

The rapid growth in population and subsequent increase in worldwide energy consumption requires more efficient transport, conversion and management of thermal energy [1]. Over 70% of consumed energy is currently produced in or through the form of heat [2]. The poor thermal conductivity of conventional heat transfer fluids (HTFs) such as water (W), ethylene glycol (EG) or engine oils is the primary limitation to achieve the essential requirement of increased efficiencies in technologies and industries of our current century [3]. For that reason, one of the most crucial pursuits of thermal investigation and engineering is to develop novel carrier fluids and lubricants with advanced heat transfer capabilities [4]. Recent outcomes in nanotechnology have opened a research avenue in various scientific or technical fields including the use of nanostructures as thermal conductivity enhancers of heat transfer and lubrication materials [5]. Thus, the dispersion of nanomaterials with at least one-dimensional size in the range of 1-100 nm (such as nanoparticles, nano-rods, nano-wires, nanotubes or nano-sheets) and high thermal conductivity can be used to improve the apparent thermal properties of the material used as base fluids. However, as additives stay in the nanometric-size range, this kind of colloids are expected to exhibit superior temporal stability, considerably higher heat transfer performance with better rheological properties, when compared with dispersions of micrometric or macrometric materials [6]. Nanoparticle suspensions based on thermal fluids such as water [7], ethylene glycol [8], thermal oils [9] or commercial coolants [10] are referred as nanofluids [11], while lubricant oil-based suspensions are known as nanolubricants [9,12].

Metallic (Au, Ag, Cu, etc.) [13,14], metal oxide (Al_2O_3, CuO, Fe_3O_4, SiO_2, TiO_2, etc.) [15,16], carbon-based (graphite, carbon nanotubes, graphene, nanodiamonds, etc.) [17] and other non-metallic (carbides: SiC, TiC; nitrades: AlN, Si_3N_4, TiN, etc.) [18,19] nanomaterials have been used to produce nanofluids. Among them, carbon nanostructures have attracted particular interest in several research fields because

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of their remarkable thermal, mechanical and electrical properties [20]. As well as diamond and graphite three-dimensional (3D) carbon allotropes, which have been known since ancient times, carbon-based materials have been developed as zero-dimensional (0D) fullerene, one-dimensional (1D) carbon nanotubes or two-dimensional (2D) graphene [21]. **Figure 1** shows the number of studies on graphene and carbon-based nanofluids published in JCR-indexed (Journal Citation Report) journals in the years of 2010~2019.

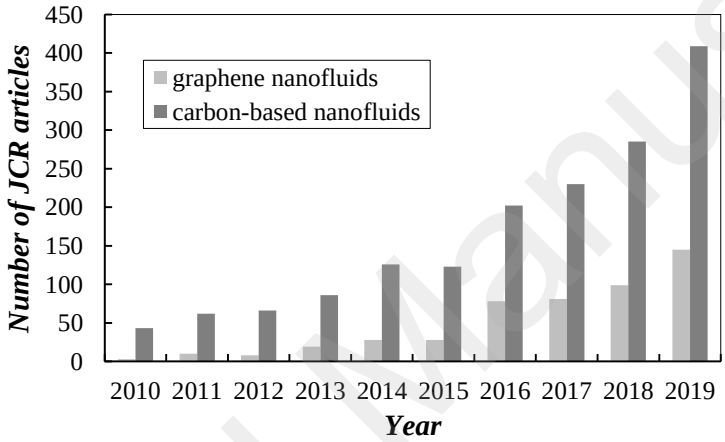

Figure 1. Number of JCR-indexed publications on graphene and carbon-based nanofluids between 2010 and 2019. Results retrieved from Web of Science [22] via the keywords: *i)* "nanofluid" and "graphene"; *ii)* "nanofluid" and ("fullerene", "carbon nanotube", "graphene", "graphite" or "nanodiamond") in the topic of the paper.

Graphene is a one-atom-thick layer of sp^2-bonded carbon atoms arranged in a honeycomb (hexagonal) lattice [23]. Despite fullerenes, graphite, and carbon nanotubes not being produced directly from graphene, the structural shape of these carbon allotropes can be conceived as graphene sheets wrapped into spheres (fullerenes), rolled into cylinders (nanotubes) or forming multiple-sheet stacks hold together by van der Waals bonds (graphite) [24]. Graphene has been reported to exhibit large theoretical specific surface areas (up to 2630 $m^2 \cdot g^{-1}$) [25], thermal conductivities (up to ~3000-5000 $W \cdot m^{-1} \cdot K^{-1}$ in the longitudinal direction) [26], good intrinsic mobility (200,000 $cm^2 \cdot V^{-1} \cdot s^{-1}$) [27] and high optical transmittance (~97.7%) [28]. Single-layer graphene has been reported to exhibit better features than multi-layer, in which the interlayer linkages could substantially reduce thermal conductivity (among

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other properties) [29]. Hence, ideally graphene nanofluids should contain freely suspended one-layer graphene. However, numerous experiments show that below a specific thickness, film-like or sheet-like structures become thermodynamically unstable unless they constitute an inherent part of a 3D system [30]. For that reason, in practice, most graphene suspensions contain few-layer graphene (FLG), multi-layer/thick graphene (MLG) or graphene nanoplatelets (GnP). Although there is not a uniform nomenclature in the specialized literature [31], graphene-stacks with layer numbers from 2 to about 5 are usually categorized as FLG, while graphene layers exceeding 5 and up to 10 (sometimes up to 30) are generally called multi-layer graphene/thick graphene. The term nanoplatelets (used in several commercial nanopowders) usually embraces 2D hexagonal lattice graphene with between 10 and 30 layers [30]. In all cases the transverse sizes of graphene sheet- or flake-like structures can range from several nanometers to the macroscale [30,32]. Within the family of graphene derivatives, graphene quantum dots (GQDs) have recently attracted great interest. GQDs are graphene nanosheets, with less than 100 nm of lateral dimension and less than 10 layers of stacked graphene, take advantage of quantum confinement and edge effect of nano-scale graphene [33].

The addition of nanoparticles to a base fluid usually leads to enhancements in thermal conductivity and heat transfer performance. However, nanoparticle loading may also modify other thermal and physical properties such as dynamic viscosity (μ) [34], density [35], specific heat capacity [36] or surface tension [37]. In particular, dynamic viscosity is related to the fluid resistance to flow and, thus reliable information regarding this transport property is essential to properly assess the thermo-fluidic behavior of nanofluids. A review of the most common techniques used for measuring the dynamic viscosity of nanofluids can be found in Le Ba *et al.* [38]. Flowing skin friction coefficient in laminar and turbulent flow heat transfer strongly depends on dynamic viscosity and thereby pumping power and pressure drop [39]. Besides the influence of dynamic viscosity on the flow status via Reynolds number, μ affects the

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heat transfer characteristics of the nanofluids since this property is also associated with Nusselt number. Additionally, dynamic viscosity may also have a greater impact than thermal conductivity in the diffusion of heat in both bulk and interfacial regions of nanofluids [40]. The study of nanofluid dynamic viscosity requires also analyzing the (non-)dependence of shear rate on this property. Thus, shear rate-shear viscosity flow curves offer information regarding whether the fluid following Newton's linear friction law (Newtonian fluid) or not (non-Newtonian fluid). Rheological tests may also provide an insight on nanoparticle structuring within colloidal suspensions such as nanofluids [41]. The addition of nanoparticles may change (or not) the dynamic viscosity and (non-)Newtonian behavior of nanofluids. As with other thermal and physical properties, modifications in this transport property may rely on some nanofluids preparation parameters such as morphology, type and volume fraction of nanoparticles, type of base fluid, the presence of additives such as surfactants or pH values [42,43].

This article presents an overview on cutting-edge research progress witnessed in the last years regarding the dynamic viscosity and rheological behavior of nanofluids based on graphene derivatives. The paper begins with a brief description of the most common methods used for fabricating or extracting graphene derivatives and an analysis of how authors prepared graphene-based nanofluids in the literature. Previous results on Newtonian/non-Newtonian behavior as well as increases in apparent dynamic viscosity of suspensions containing graphene derivatives are comprehensively reviewed, analyzing the effects that shear rate, concentration and temperature have on the rheological behavior of those nanofluids. Recommendations for future works and conclusions are finally provided.

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2. Synthesis of graphene derivatives and nanofluid preparation

2.1 *Synthesis of graphene derivatives*

Graphene was successfully isolated for the first time in 2004 by Novoselov *et al.* [44] using micromechanical cleavage. Despite the high purity and quality of graphene synthesized by micromechanical cleavage, this method is time-consuming and incapable of mass-scale production [45]. Within the last two decades, much research has been conducted to explore feasible synthesis routes for mass production of graphene [46]. Generally, fabrication methods can be broadly divided into top-down and bottom-up processes. A classification of the most common approaches to produce graphene derivatives is shown in **Figure 2**.

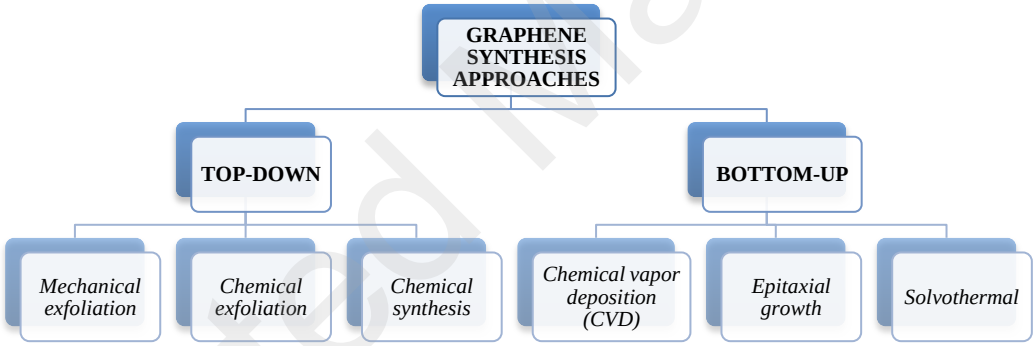

Figure 2. Most common approaches to synthesize graphene derivatives.

In top-down strategies, graphene derivatives are produced by exfoliation or stripping off highly ordered pyrolytic graphite (HOPG) or graphene oxide (GO). As mentioned, mechanical or micromechanical exfoliation was the first established method of extraction of graphene flakes on a substrate. An exfoliation agent (such as adhesive tape) is used to slice down layers from graphite surface [45]. Although mechanical exfoliation is still a significant method for producing superior graphene films (transverse sizes up to 5-10 μm), scalability and reproducibility are still the main limitations [30].

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Chemical exfoliation is another route in which graphene-intercalated compounds (such as alkali metals) are used to increase the interlayer spacing and thereby reduce van der Waals forces between graphite layers [47]. Then, single-layer or few-layer graphene is easily detached from bulk graphite using a mechanical source such as sonication. Chemical reduction of graphite oxide is also one of the most widely extended ways to prepare graphene derivatives in large quantities. Graphite oxide exhibits similar carbon structure to graphite, but the plane of carbon atoms is heavily decorated with oxygen-containing groups such as hydroxyl (-OH), alkoxyl (C-O-C), carboxyl (C=O), carboxylic (-COOH) radicals [48]. Such functional groups increase the interlayer spacing, which, in turn, ease the exfoliation into different layers of graphene oxide (GO).

Brodie [49], Staudenmaier [50] and Hummers [51] were pioneers in the development of chemical routes to efficiently oxidize graphite. Hummers' method, which strongly attacks sp^2 carbons present in the structural plane by sulfuric acid and potassium permanganate and finishes the reaction with hydrogen peroxide, is the most widely spread. Over the years Hummers' method has been modified by optimizing some parameters (reaction time, temperature, reactant nature and dosages), though, keeping the core of the original method [52]. The groups containing polar oxygen atoms bring positive effects in terms of facilitating the suspension of GO in aqueous solvents. However, the acid treatment of graphene oxide can lead to the incorporation of defect sites in the well-arranged carbon network of pristine graphene. This atomic roughness has a negative impact on the thermal conductivity in sheet, among other thermal and physical properties [38,53]. To partially recover the thermal properties of pristine graphene but maintaining the proper dispersibility of graphene oxide in water, the oxygen content of GO is chemically reduced via a controlled thermal, microwave, photo-thermal, photo-chemical, or microbial/bacterial approach to produce reduced graphene oxide (rGO) [30]. A scheme of the different chemical methods to

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oxidize graphite into GO and reduce it into rGO as well as the most common graphene derivatives is presented in **Figure 3**.

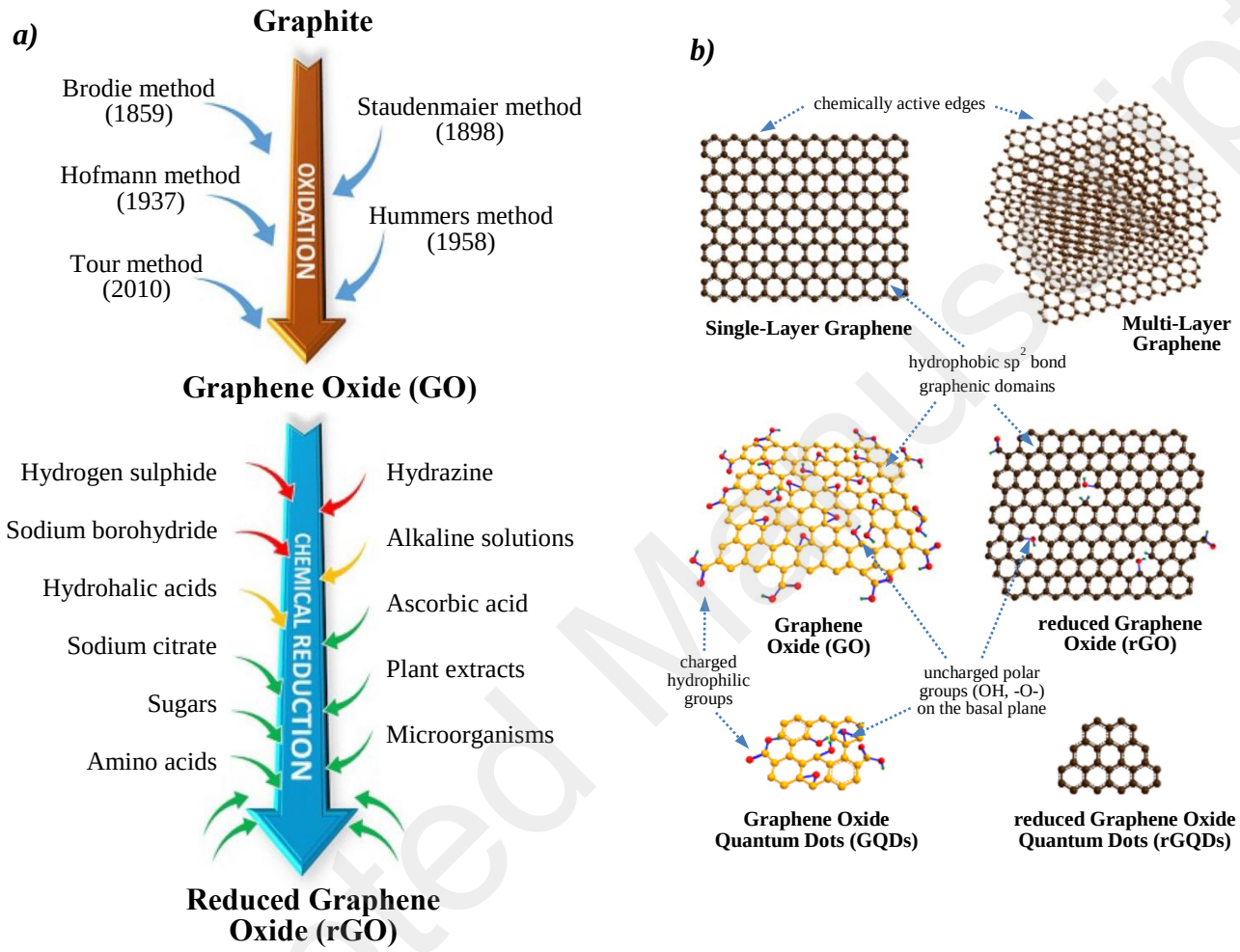

Figure 3. *(a)* Scheme of different chemical methods to oxidize graphite into GO and reduce it into rGO [54] and *(b)* Common forms of graphene derivatives [55].

In bottom-up approaches, graphene sheets are fabricated by building up the nanomaterials via an atomic or molecular arrangement of carbon [45]. Chemical vapor deposition (CVD) is a relatively cheap and accessible technique in which a carbon-based substrate is exposed to thermally decomposed precursors in a gaseous state. Thermal- and plasma-enhanced are the most commonly used CVD techniques [45]. Epitaxial thermal synthesis is another well-known synthesis route which allows the growth of an

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epitaxial graphene film on the surface of a single-crystalline subtract [56]. Solvothermal synthesis has also been used to produce single-layer graphene film. There, graphene layers are yield by the solvothermal process in the rapid pyrolysis of sodium ethoxide, using ethanol and sodium as the carbon precursors [57].

Table 1 summarizes described synthesis methods and mechanisms. Several other ways including microwave synthesis, electrochemical exfoliation or unzipping multi-walled carbon nanotubes have also been reported in the literature [30,56,57].

Insert here Table 1

2.2 *Preparation of nanofluids based on graphene derivatives*

Nanofluids can be produced using one-step or two-step methods. One-step approaches yield nanofluids directly through chemical techniques. In two-step strategies, graphene derivatives are first produced separately in powder form (using one of the synthesis routes above described) and then dispersed in the base fluid by means of mechanical stirring, ultrasonication, homogenization or other mixing techniques along with possible surfactants/dispersants [58]. One and two-step methods have their own benefits and limitations, the selection definition of the most appropriate nanofluid preparation route depends upon the production scale, nanoparticle concentration or the functional groups/dispersants required to achieve stable dispersions, among other design parameters [29].

Unfortunately, it is well-known that strong π-π interactions present in pristine graphene make this material hydrophobic in nature. Thus, graphene sheets readily agglomerate and settle in the presence of common solvents and particularly in water, the most widely used thermal medium [59]. Over the last years, authors have worked on effective techniques to disperse graphene-derivatives in common solvents, including thermal fluids and lubricants [60,61].

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Surfactants can be added to the base fluid to reduce the surface tension and ease the dispersibility of nanoparticles [37]. When it comes to nanofluids based on graphene derivatives, stable dispersions were obtained using ionic or non-ionic surfactants as dispersants such as: sodium dodecyl sulfate (SDS), sodium dodecylbenzene sulfonate (SDBS) [62,63], cetyltrimethylammonium bromide (CTAB) [62], polyvinylpyrrolidone (PVP) [64], gum arabic (GA) [62], sodium deoxycholate (SDC) [65–67], 4-(1,1,3,3-tetramethylbutyl) phenyl-polyethylene glycol (Triton X-100) [68–70], oleylamine [71] or Tween 80 [72]. However, in certain applications (see for instance the case of some lubricants) the use of surfactants/dispersants may not be appropriate [30]. Additionally, large amounts of surfactant can considerably increase the viscosity of the resulting nanofluid. As an example, Amiri *et al.* [73] reported that rises in viscosity of graphene aqueous nanofluids stabilized using SDBS doubled the values of covalent-funtionalized nanofluids using equivalent nanoparticle concentration.

Chemical functionalization [74] of graphene helps to improve the dispersibility of this material by inserting additional chemical species/moieties between its basal planes. There exist two ways to incorporate functional groups on the surface of graphene nanosheets: by covalent or non-covalent chemical routes. The former approaches (based on covalent functionalization) chemically create covalent bonds on graphene surface by converting sp^2 of carbon networks into sp^3 orbitals [75], while non-covalent functionalization attaches polymer species to the surface of graphene via multiple π-π stacking, H-bonding, and hydrophobic interactions [76]. Doubtless, the most common functionalization way is the introduction of oxygen-containing groups on the nanosheet surface, which takes place during graphite oxidation to produce graphene oxide [77]. As it was already mentioned, the incorporation of hydroxyl, alkoxy, carboxyl or carboxylic polar oxygen groups facilitates the dispersibility of graphene oxide (GO) in aqueous solvents [78]. Thus, Paredes *et al.* [79] reported the good stability of GO suspensions in water and other organic solvents. However, such functional groups also bring a negative

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downside effect in terms of substantial reductions in the thermal conductivity of the sheets. Hence, a controlled acid treatment followed by an appropriate reduction of graphene oxide are key-issues to produce graphene derivatives that allow preparing nanofluids with enhanced thermal properties that can become the next generation of heat transfer liquids.

In the following sections, we review and describe the results, mainly experimental, concerning the rheological behavior and dynamic viscosity of graphene-based nanofluids distinguishing the different types of graphene derivatives. Then, the effect of the base fluid, concentration in graphene derivatives and temperature on viscosity enhancement is presented and discussed, before to introduce future works and conclusions

3. Rheological behavior and dynamic viscosity of graphene-based nanofluids

3.1 Graphene Oxide (GO) nanofluids

Several studies on rheology properties of graphene oxide (GO) based nanofluids have been performed as explained in the following. Besides, the type of treatment, nanoparticles dimensions and concentration range, temperature, and key results are gathered in **Table 2**. Ranjbarzadeh *et al.* [80] investigated water-based nanofluids containing 0.025-0.1 vol% concentrations of graphene oxide with hydroxyl (-OH) and carboxyl (-COOH) functional groups. Dynamic viscosity increase at the maximum GO loading was ~78%, which led to a rise in friction factor of ~16% (in both cases in comparison to water). Nazari *et al.* [81] worked on surfactant-free aqueous nanofluids loaded with low GO-concentrations (0.25-1.5 g/L) as possible heat transfer media for pulsating heat pipes. Their viscosity measurements at 298 K showed that the relative viscosity (μ_r) of nanofluids increases from 1.041 at 0.25 g/L to 1.248 at 1.5 g/L. Graphene oxide-distilled water dispersions were studied also by Esfahani and Languri [82] who worked on two concentrations (0.01 and 0.1 wt%) at temperatures of 298 and 313 K. The results showed that the

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viscosity of 0.01 wt% of GO was in a similar range to water while the viscosity of 0.1 wt% GO was 60% higher than distilled water at 298 K. In addition, they obtained a Newtonian behavior for 0.01wt% GO loading, and a non-Newtonian pseudo-plastic behavior in the case of 0.1 wt% concentration, at shear rates lower than 20 s^{-1}. Anin Vincely and Natarajan [83] investigated the thermal performance of deionized water-based nanofluids, where 0.005 to 0.05 wt% mass loadings of GO were dispersed, in a solar flat plate collector. Compared to the base fluid, 0.05 wt.% dispersion exhibited an increase in dynamic viscosity of 10.4% and 20.2% at 323 K and 303 K, respectively. Results showed also a non-Newtonian behavior for 0.01, 0.02 and 0.05% weight concentrations at shear rates less than 2000 s^{-1}. Graphene oxide was dispersed also in deionized water + ethylene glycol (60:40%) by Ijam *et al.* [84], who found a non-Newtonian shear-thinning behavior at shear rates less than 100 s^{-1} and 300 s^{-1} for 0.05 wt% and 0.1 wt% nanofluid concentrations, respectively. Also, the viscosity of the dispersion containing 0.1 wt% of GO increased by 35% compared to the base fluid at 293 K and decreased by 48% when temperature increased from 293 to 333 K. Hadadian *et al.* [85] and Wang *et al.* [86] focused on the study of the GO/ethylene glycol nanofluids. First, in the work of Hadadian *et al.* [85], the average crystalline size of GO was 20 nm, and they did the experiments for 0.001-0.005 wt% nanofluid concentrations in the temperature range from 293 to 232 K. As a result, a shear-thinning behavior was obtained at low shear rates and a Newtonian one at high shear rates. Additionally, they found that the relative viscosity of 0.005% mass content of GO at 293 K under a shear rate of 27.5 s^{-1} was equal to 3.4, and μ_r increased from 1.52 at 0.001 wt% to 2.34 at 0.005 wt%, at 323 K and under a shear rate of 67.5 s^{-1}. On the other hand, Wang *et al.* [86] worked with GO nanoparticles that have a large dimension between 1 and 5 μm, and a thickness range 0.8-1.2 nm. These authors prepared nanofluids with different weight concentrations from 0.5 to 2.5 wt%, and measured their rheological properties at 293 K. A low viscosity increase was obtained (from 21.4 of based-EG to 29.5 mPa.s of dispersion loaded with 2 wt% GO), and

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flow curve results showed a Newtonian behavior for 0.5-1 wt% loading and a shear-thinning behavior at low shear rates when the loading increased to 1.5 wt%. Water, ethylene glycol and ethylene glycol+water (20:80, 40:60, 60:40 weight ratios) were used as base fluids by Syam Sundar *et al.* [87] to prepare nanofluids loaded with 0.05 to 0.2 vol% concentrations of GO/Co_3O_4 hybrid nanoparticles. Viscosity was investigated between 293 and 303 K and this property was observed to decrease with rising temperature and increase with nanoparticle loading. However, rises in viscosity depended on base fluid. Thus, an enhancement in viscosity of water-based nanofluid by 1.70-times and ethylene glycol-based nanofluid by 1.42-times, in both cases at 0.2% volume concentration and a temperature of 333 K. Similar increases in viscosity (μ_r ~1.31-1.36) were observed when 0.2 vol% dispersions were compared with their corresponding base fluid at 303 K. The viscosity ratios of 60EG:40W-based GO/Co_3O_4 hybrid nanofluids reported by Syam Sundar *et al.* [87] were in parallel with the enhancements reported by Kole and Dey [88] for 70EG:30W-based GO nanofluids. Yao *et al.* [89] worked on an epoxy resin-based magnetic nanofluid, composed of graphene oxide (GO)/carbon nanotube (MWCNTs)/Fe_3O_4 (GMF) as core and polyether amine as shell, which was fabricated via a combination of ultrasonic-assisted chemical co-precipitation process and post-modification. The objective was to improve the mechanical and thermal performance of the epoxy resin. Polymer grafted on the surface of GMF core did not only prevent nanoparticle aggregation but, according to authors, the presence of polymer chains also resulted in a larger molecular space, which limited friction between polymer chains producing lubrication and reducing nanofluid viscosity. The authors found that the viscosity decreases from 3.26 Pa.s to 0.81 Pa.s with the increase of temperature from 303 to 353 K. Additionally, oscillatory rheological tests showed that GMF dispersions exhibited a liquid-like behavior in the temperature range from 293 to 353 K. A 0.03 mg/mL concentration of graphene oxide (GO) was used as lubricant additive in an SAE 10W40 engine oil by Mishra *et al.* [90], who showed from their rheological measurements a Newtonian

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behavior for the neat SAE10W40 and the GO/SAE10W40 blend. Ranjbarzadeh *et al.* [91] investigated the rheological properties of graphene oxide silicon oxide (GO-SiO_2)/water nanofluids in the concentration range 0.5-1 vol% at temperatures between 293 and 303 K. These nanofluids were found Newtonian at shear rates from 110-245 s^{-1}. In addition, a maximum increase in viscosity of water from 1.059 mPa.s to 2.421 mPa.s for 1 wt% nanofluid concentration was obtained at 293. The results also showed that the relative viscosity rises with temperature and nanofluid concentration. Authors also performed additional viscosity measurements for graphene oxide (GO)/water nanofluids at equivalent nanoparticle mass concentrations and observed that the presence of silicon oxide reduced the viscosity of (GO-SiO_2)/water hybrid nanofluids in comparison to GO/water dispersions. Such reductions were attributed to the higher specific surface area of GO (in comparison to SiO_2), which leads to stronger nanoparticle-base fluid interactions. Cabaleiro *et al.* [92] prepared graphene oxide (GO)/water nanofluids in the volume concentration range 0.0005-0.1% and investigated their rheological properties at 293 and 303 K. The results showed that the nanofluids with concentrations lower than 0.01 vol% were Newtonian, and above this concentration GO-suspensions become non-Newtonian. In addition, maximum increases by 100-130 % were obtained for the non-Newtonian nanofluids, and the larger increase was observed for the 0.1 vol% concentration at 303 K. The results of relative viscosity showed an increase by about 130% in the considered concentration range, and there are no significant influence of temperature on the relative viscosity. Esfahani *et al.* [93] studied graphene oxide nanofluids based on deionized water. Rheological measurements were done for different concentrations between 0.01 and 0.5 wt% at the temperature range 298-303 K. Results showed a decreasing in the viscosity with the rise of temperature and an increasing with the increase of nanoparticle content. This increase was more pronounced in the case of the highest concentration 0.5 wt%. Thus, at 298 K and shear rate of 100 s^{-1}, the viscosity rises by 38% and 130% for 0.01 and 0.5 wt%, respectively. In addition, the rheological

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behavior of all samples was studied at shear rates between 10 and 100 s^{-1}, and a non-Newtonian behavior was observed for higher concentrations at lower shear rates. Cabaleiro *et al.* [94] investigated sulfonic acid-functionalized graphene oxide nanoplatelets dispersed in ethylene glycol:water mixture 10:90 wt% following two step method. Mass concentrations between 0.1 and 0.5% were prepared and tested at temperatures in the range 283.15-343.15 K. The measurements showed that all samples behave as Newtonian fluids. Their dynamic viscosity was obtained to increase with the temperature decrease and concentration increase. Maximum increase in the viscosity by 12.6% was found for the highest concentration at 343.15 K. Vallejo *et al.* [95] studied also sulfonic acid-functionalized graphene oxide nanoplatelets nanofluids based on propylene glycol/water mixtures (PG:W) at 10:90 and 30:70 weight fractions. Two mass concentrations 0.25% and 0.50% of nanofluids were prepared with PG:W 10:90 wt% and between 0.25% and 1% with PG:W 30:70 wt%. The rheological properties of all those samples were measured at the temperature range 278.15-323.15 K and at shear rates between 1 and 100 s^{-1}. The flow behavior of all samples at studied temperatures showed that the nanofluids are Newtonian. The authors found that PG:W 30:70 wt% based nanofluids are more viscous than PG:W 10:90 wt% based nanofluids by around 123 and 106% for the same mass nanoadditive concentrations at 283.15 and 293.15 K, respectively. Increasing temperature decreased the viscosity by 68 and 80% for PG:W 10:90 wt% and PG:W 30:70 wt% base fluids, respectively. Maximum viscosity increases by 58 and 99% for the PG:W 10:90 wt% base fluid and 39 and 76% for the PG:W 30:70 wt% base fluid were found for the 0.25 wt% and 0.50 wt% nanofluids, respectively.

Insert here Table 2

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3.2 Reduced graphene oxide (rGO)-based nanofluids

Rheological properties of reduced graphene oxide-based nanofluids were investigated also by some researchers as explained below. As in the previous section, **Table 3** also includes some more details about the type of treatment, nanoparticles dimensions and concentration range, temperature, and main results. Zhang *et al.* [96] studied the controlled reduced graphene oxide (crGO)/deionized water nanofluids. They obtained that the viscosity decreases with the increase of temperature from 283 to 333 K, increases with the increase of rGO concentration from 0.2 to 1 mg/mL. The samples exhibit shear-thinning viscosity at shear rates lower than ~60-80 s^{-1}, while behaving in a Newtonian manner at high shear rates. Shear-thinning degree and dynamic viscosity increase with nanoparticle concentration. Likewise, viscosity depletion with temperature is more noticeable with rising crGO loading. On the other hand, Mehrali *et al.* [97] prepared free-surfactant aqueous nanofluids containing 1-4 vol.% concentrations of red wine reduced graphene oxide (W-rGO), which were synthesized by a modified version of Hummers' method. Samples were Newtonian and although dynamic viscosity increased with nanoparticle loading, pressure drops (at different flow velocity conditions) did not overcome 15%. Similar viscosity reductions with increasing temperature (between 86.2% and 87.9% in the entire 293-343 K range) were observed for low and high concentrations. Reduced graphene oxide (rGO), distilled water and PVA as a surfactant were used by Akhavan-Zanjani *et al.* [98] to prepare nanofluids in the rGO concentration range 0.005-0.02 vol%. Measurements in the temperature interval between 298 and 308 K showed a non-linear behavior of viscosity ratio with respect to concentration and maximum augmentations in this property of 4.9% at 298 K and for 0.02 vol% content. Sadeghinezhad *et al.* [99] interested in the study of rGO-Fe_3O_4/water nanofluids that contain Tannic acid as a surfactant. In their study, authors focused on one concentration 0.5 wt% of nanofluid and measured rheological properties

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in the temperature range 293-333 K. The results showed that the viscosity of the nanofluid is higher than that of the base fluid, and showed a Newtonian behavior above the shear rate of 100 s^{-1}. Chen *et al.* [100] investigated the PEG-functionalized reduced graphene oxide (rGO-PEG)/water nanofluids with 0.025 mg/mL concentration and at a temperature range 295-379 K. The authors found that the dynamic viscosity of water and rGO-PEG nanofluid decreases with increasing temperature, and that the percentage enhancement of dynamic viscosity increases with the addition of rGO-PEG nanoparticles with increasing temperature. Reduced graphene oxide (rGO) and reduced graphene oxides decorated with silver nanoparticles (Ag-rGO) were dispersed in deionized water without using any surfactant by Mehrali *et al.* [101], who did the viscosity measurements between 298 and 333 K and for 100 ppm as concentration. The results showed a non-linear increase of relative viscosity with temperature for all fluids, a highest viscosity enhancement of 22% for rGO nanofluid within the considered temperature range, and a lower relative viscosity enhancement for Ag-rGO nanofluids. Mishra *et al.* [90] investigated the case of reduced graphene oxide (rGO)/SAE10W40 nanofluid with concentration of 0.1 mg/mL, and showed a shear thinning flow behavior for this nanofluid at low shear rates (less than 1 s^{-1}). Observed reduction in dynamic viscosity with increasing shear rate (which contrasts with the Newtonian behavior obtained in the same article for neat SAE10W40 or a 0.03 mg/mL dispersion of GO/SAE10W40) were attributed to the effective inter-sheet shearing of rGO. Graphene oxide (GO)/water nanofluids in the volume concentration range 0.0005-0.1 % were prepared by Cabaleiro *et al.* [92] who studied their rheological properties at 293 and 303 K. The measurements showed that the nanofluids with concentrations lower than 0.01 vol% were Newtonian, and those higher than 0.01 vol% exhibited shear-thinning non-Newtonian behavior. Additionally, maximum increases by 70-80 % were obtained for the non-Newtonian nanofluids, and the largest increase was observed for the 0.1 vol% concentration at 303 K. Relative viscosity results showed an increase by about 70% in the considered

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loading range, and that temperature had no significant influence on the relative viscosity. On the other hand, by comparison to the GO nanofluids, Cabaleiro *et al.* [92] showed that 0.1 vol%, rGO nanofluids exhibit lower apparent viscosities and weaker shear-thinning behaviors.

Insert here Table 3

3.3 Graphene-based nanofluids

An important proportion of the studies on the topic of this review was on pure graphene-based nanofluids. For example, Wang *et al.* [102] worked on 0.03 wt% mass loading of graphene G/[HMIM]BF4 ionanofluid. They obtained that the viscosity of the nanofluid is lower than that of the base fluid and decreases from 217.4 to 40.6 cP with increasing temperature from 298 to 348 K. Graphene (G) was dispersed also in ionic liquid [HMIM]BF4 without using surfactant by Liu *et al.* [103], who prepared 0.03-0.06 wt% mass concentrations of these suspensions and did the viscosity measurements between 303 and 488 K. The results showed that the addition of G can slightly decrease the viscosity of the base fluid, and reductions in viscosity by 4.6%-13.1% for 0.06 wt% loading from room temperature to around 473 K were obtained. On the other hand, Sadeghinezhad *et al.* [104], Mehrali *et al.* [105], and Iranmanesh *et al.* [106] prepared different weight concentrations (between 0.025 and 0.1 wt%) of graphene nanoplatelets (GnP)/distilled water nanofluids. Nanoplatelets had 2 μm as larger dimension, 2 nm as thickness and specific surface areas (SSA) of either 300, 500 or 750 m^2/g. When the viscosity of nanofluids was measured between 293 and 333 K, the authors found that the viscosity increases with rises in concentration and decreases with increasing temperature, with a Newtonian and non-Newtonian behavior (for high concentrations and SSA of 300 and 500 m^2/g) of nanofluids found by Mehrali *et al.* [105]. Agarwal *et al.* [71] studied graphene nanoplatelets (GnP)-Kerosene nanofluids with using oleylamine as a surfactant. They used nanoparticles with 300, 500, 750

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m^2/g specific surface areas (SSA), to prepare dispersions at 0.005-0.2 wt% concentrations. Their results showed an increase in viscosity by 8% for 750 SSA, 0.2 wt% GnP-kerosene nanofluid at room temperature. Moreover, Askari *et al.* [72] prepared 0.1-0.5 wt% concentrations of nanofluids by nanoporous graphene and water with the addition of Tween 80 as a surfactant. They performed measurements at temperatures between 293 and 313 K and showed an increase in viscosity at 293 K from 0.99 cP (water) to 1.31 cP (nanofluid at nanoporous graphene concentration of 0.5 wt%). Additionally, in the case of 0.1 wt% concentration, viscosity decreases from 1.15 cP (at 293 K) to 0.76 cP (313 K). Chai *et al.* [107] used a two-step method to prepare nanofluids based on a hydrogenated oil and containing graphene nanosheets concentrations ranging from 25 to 100 ppm. In addition to the shear thinning behavior obtained for nanofluids at very low shear rates, and the slight shear thickening behavior at higher shear rate between 303 and 323 K, Chai *et al.* found an increase in viscosity and shear stress up to 33% at 303 K and at the highest nanoparticle concentration. Ghozatloo *et al.* [108] worked with graphene nanosheets that were prepared by a catalytic decomposition CVD method and used the two-step method to prepare aqueous nanofluids containing 0.05-0.1 % weight concentrations of nanosheets. They obtained from the viscosity measurements an increase by 11.97% at 0.1% weight fraction of graphene in water. Graphene nanoplatelets (GnP) nanofluids prepared with distilled water (DW) as base fluid without any use of surfactant were investigated by many researchers who studied rheology properties at temperatures between 293 and 333 K and at weight concentrations in the range of 0.025-0.1 wt%. For example, Iranmanesh *et al.* [109] found a Newtonian behavior of the nanofluids containing 0.025-0.1 wt.% graphene nanoplatelets with a specific surface area (SSA) of 750 m^2/g. The authors also observed an increase and decrease of viscosity with rising concentration and temperature, respectively. On the other hand, Mehrali *et al.* [110] mentioned in their paper that the viscosity of nanofluids with different SSA of GnP of 300, 500 and 750 m^2/g, reduced at higher temperatures by 4-

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44% compared with DW at a high shear rate of 500 s^{-1}. Additionally, Sadeghinezhad *et al.* [111] reported a decrease in nanofluid viscosity of up to 38% with rising the temperature between 293 and 333 K at shear rate of 500 s^{-1}. Moghaddam *et al.* investigated experimentally [112] and theoretically [113] the glycerol-based nanofluids containing graphene nanosheets with few layers and size of about 15-50 nm. The prepared nanofluids were in the concentration range of 0.0025-0.02 wt% and their viscosity was measured at temperatures between 293 and 333 K. In addition to a shear thinning behavior of suspensions (more appreciable with rising nanoparticle concentration), which authors attributed to the complex interactions between the glycerol and graphene nanosheets, a maximum increase in dynamic viscosity of 401.49% was observed for 2% graphene nanosheets load at 293 K and the shear rate of 6.32 s^{-1}. In their theoretical work, the highest viscosity value calculated using molecular dynamic simulation was 1739.95 MPa.s for number of layers (N_{layer}) equal to 6, and they found a high level of agreement with experiment for N_{layer}= 6. The results also showed that the viscosity increases as the number of layers of graphene increases. In addition, a theoretical work was also conducted by Sandeep and Malvandi [114] who investigated graphene/water nanofluids. The authors used Deborah number with relaxation (δ_1) and retardation times (δ_3), which plays a major role in the shear thinning and thickening behavior of the non-Newtonian nanofluid. Sarsam *et al.* [62] was focused on the preparation of 0.1 wt% of graphene nanoplatelets (GnP)/water nanofluids using different surfactants like sodium dodecylbenzene sulfonate (SDBS), sodium dodecyl sulfate (SDS), cetyl trimethylammonium bromide (CTAB), and gum arabic (GA). At temperature conditions between 298 and 328 K and shear rates from 20 to 200 s^{-1}, the authors remarked a Newtonian behavior for nanofluids with all surfactants except GA, a non-Newtonian behavior for GA-GnPs nanofluid (surfactant-GnPs ratio: 0.5-1), and a non-Newtonian behavior for pristine GnPs nanofluids except at 35℃ the behavior was Newtonian. Additionally, the (1-1) SDBS-GnPs sample showed the highest stability and nearly the lowest viscosity (7.4% higher than

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distilled water). And based on the average values of viscosity, the water based nanofluids can be sequenced as (0.5-1) GA-GnPs >> pristine GnPs > (1-1) SDBS-GnPs > (1-1) SDS-GnPs > (1-1) CTAB-GnPs. The dispersion of graphene nanoplatelets (GnP), with thickness of about 5-10 nm and 15 μm of larger dimension, in deionized water + ethylene glycol (70:30 volume ratio) with using 0.75 vol% of sodium deoxycholate (SDC) as surfactant was investigated by Selvam *et al.*[65,66]. Authors performed the study between 303 and 323 K for nanofluids in the concentration range of 0.1-0.5 vol%. The obtained results showed that the viscosity ratio (μ_{nf}/μ_{bf}) of nanofluids increases from 1.06 to 1.16 and 1.13 to 1.39 at 0.1 vol% and 0.5 vol%, respectively [66]. They mentioned also that the viscosity values for water + ethylene glycol mixture were in good agreement with ASHRAE standard (values within ± 2% deviation), and a maximum enhancement of viscosity was found equal to about 40% for 0.5 vol% nanofluid [65]. In another paper from the same research group [67] the same nanofluids with the same conditions except that used graphene nanoplatelets (GnP) had average thicknesses in the range of 4-8 nm, the presented values of viscosity showed an increase by increasing concentration, and a decrease with increasing temperature. In another paper from the same research group [67] this same base fluid (an ethylene glycol+water mixture 70:30 volume ratio) was used to produce graphene nanofluids in the concentration range 0-0.5 vol.%, this time using nanoplatelets with thicknesses in the range of 4-8 nm. Reported viscosity values in the temperature range from 303 to 323 K showed an increase by increasing concentration (up to 43.8% for 0.5 vol.% at 323 K), and decreases with increasing temperature (in the range from 21-35%, depletion degree with temperature gets reduced as concentration increases). Wang *et al.* [115] investigated graphene/heat-transfer oil nanofluids with 0.02-0.2 mg/mL concentrations prepared by a two-step method. The viscosity of nanofluids was shown to increase with the temperature between 298 and 333 K. In addition, they showed that adding small amounts of graphene to heat-transfer oil could reduce kinetic viscosity of pure oil, but an increase of viscosity can be found by adding too

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much graphene to the oil. In another work, Wang *et al.* [116] investigated the single-layer graphene (GnP)/water nanofluids that contain a special dispersant. The authors found that the viscosity logarithmically decreases with increasing temperature, founding a correlation with nanoparticle concentration in the temperature range from 278 to 298 K. Also, an increase with rising concentration from 0.2 to 1 wt% with viscosity increment ratios between GnP nanofluids and water ranging from 1.24 to 2.35 was found. Additionally, observed shear thinning effect and non-Newtonian behavior were described with the Power Law viscosity model with a flow behavior index ~ 0.85-0.95. Graphene nanoplatelets (GnP)/deionized water nanofluids were prepared by Vakili *et al.* [117] who found a decreasing of viscosity with increasing temperature from 293 to 333 K, and an increase with rising concentration from 0.025 to 0.1 wt%. Pamies *et al.* [118] investigated nanofluids based on graphene (G) with 0.5-1 wt% concentration range, where two ionic liquids, [EMIM][DCA] and [EMIM][TFSI] were used as base fluids without any surfactant assistance, between 298 and 400 K. The results showed an increase in viscosity with increasing loading to 1 wt% by 151% and 269% with respect to [EMIM][DCA] and [EMIM][TFSI], respectively, in both cases at 500 s^{-1} and 298 K, constant viscosities for nanofluids with 0.5 wt% graphene concentration. Flow curves at 298 K and in the shear rate range from 10^{-3} to 500 s^{-1} show a Newtonian behavior for neat ionic liquids, while a pseudo-plastic non-Newtonian behavior (particularly pronounced under low shear) was observed for nanofluids. Temperature sweeps at constant shear rate (50 s^{-1}) measured for neat ionic liquids and low-concentrated dispersions show a decreasing trend with increasing temperature. In the case of $\mu(T)$ curves obtained for samples containing large amounts of graphene, a decreasing-increasing trend is possible to observe. Mehrali *et al.* [119] studied graphene nanoplatelets nanofluids based on distilled water. The concentrations were between 0.025 and 0.1% in mass, and their viscosity was tested in the temperature range 293-333 K. The results showed that the viscosity increases with the increase of nanoparticle

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content and decreases strongly with the rise of temperature. In the temperature range 293-333 K, viscosity decreases with increasing temperature are 4-44% higher that corresponding temperature depletion of water in such temperature range. Dhar *et al.* [120] prepared different volume fractions of graphene nanosheets (main dimensions: 5 nm-1.5 µm) nanofluids up to 0.5% using water as base fluid. Nanosheets were further sulfonated with sulfanilic acid to improve their dispersibility in water. Viscosity measurements of all nanofluids were done in the temperature range 298-343 K. The authors obtained a decreasing of the viscosity with the increase of temperature and with the decrease of nanoparticle loading. More recently, Hamze et al. [121] investigated a few-layer graphene (FLG)-based nanofluids in the weight concentration range 0.05-0.5% where a commercial fluid, Tyfocor® LS, was used as base fluid. The measurements were done at different temperatures from 283.15 K to 323.15 K. The authors tested three series of nanofluids that differ by the type of surfactant used (Triton X-100, Pluronic® P-123, and Gum arabic). Different kind of measurement showed that the stability under shear of nanofluids depends on the shearing condition, the duration of shearing, and the temperature as well as the type of surfactant and its content. In addition, the shear flow behavior of nanofluids varied with the same parameters and can be related to the visual aspect of the nanofluids after shear. Finally, the variation of dynamic viscosity of stable nanofluids with temperature was well correlated to the Vogel-Fulcher-Tammann (VFT) viscosity model.

As a summary, the reader is referred to **Table 4** where key information about graphene-based nanofluids preparation and rheological trends are also compiled.

Insert here Table 4

3.4 Graphene quantum dots (GQDs)-based nanofluids

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GQDs-based nanofluids were less studied than previous graphene derivatives. The following description of the works in this field is coupled with **Table 5**. For example, Amiri *et al.* [122] worked on 0.001 wt% of the amine-treated graphene quantum dots (A-GQD) dispersed in transformer oil. In the studied temperature range (between 293 and 353 K), no significant penalization in dynamic viscosity of the transformer oil (increases in the property were less than 1.3%) was observed after the addition of 0.001 wt% of A-GQD. Amine-treated graphene quantum dots (A-GQD) were used also by Amiri *et al.* [123], but here, distilled water was utilized as base fluid without the need of any surfactant. The investigation was done for 0.001-0.002 weight concentrations and at temperature conditions from 293 to 323 K. Viscosity versus shear rate curves (in the range from 20 to 300 s^{-1}) showed the Newtonian behavior of studied nanofluids with insignificant differences between the average viscosities of water and measured values for water-based A-GQD nanofluids. Goharshadi *et al.* [124] studied the graphene quantum dots GQD/glycerol nanofluids where the nanoparticles were prepared by one-step green pyrolysis of citric acid. The authors worked at temperatures between 293.15 and 333.15 K, and for concentrations between 0.25 and 2 wt%. The dynamic viscosity reduced with the addition of GQDs, with diminutions ranging from -3.6% for a 0.25% at 2.64 s^{-1} and 313.15 K to -49.97% for the 2% suspension at 0.66 s^{-1}. In addition, the authors found a shear-thinning behavior for GQDs-glycerol suspensions at low shear rates and temperatures, which was more obvious at greater mass fractions.

Insert here Table 5

3.4 Functionalized graphene and doped graphene-based nanofluids

Functionalized and doped graphene-based nanofluids were also studied from a rheological point of view as explained below, few results with graphene hybrid nanofluids are also introduced. Thus, nitrogen-

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doped graphene (NDG) was used by Mehrali *et al.* [68–70] when dispersed 0.01-0.06 wt% of these nanoparticles in distilled water with the addition of 0.025 wt% of Triton X-100 as a surfactant. Like distilled water, nanofluids act as Newtonians in the studied shear rate range (0.1 and 500 s^{-1}) [70]. The addition of NDG exhibit little impact on increases of fluid viscosity and pressure drop (with penalties in the range from 0.08 to 14.4%). In the temperature range 293-333 K, viscosity declines with rising temperature by between 51.2% and 51.5% following the general trend of the distilled water viscosity as a function of temperature. Askari *et al.* [125] studied Fe_3O_4/Graphene-deionized water hybrid nanofluids. For 0.1-1% mass loading of nanoparticles, and at temperatures between 293 and 313 K, authors found that the viscosity increases with increasing concentration (from 0.99 mPa.s of pure water to 1.03, 1.06 and 1.15 mPa.s at concentrations of 0.1, 0.3 and 0.5 wt%. Fe_3O_4/Graphene nanofluids, respectively) and with decreasing temperature. Similar trends were obtained also by Amiri *et al.* [126] who focused on the ethylene glycol-functionalized graphene nanoplatelets (EGGnP) which were synthesized covalently, dispersed in water+ethylene glycol (40:60 volumetric ratio) without using any surfactant. Amiri *et al.* [126] confirmed the Newtonian behavior of EGGnP dispersions loaded with nanoparticle concentrations in the range of 0.01-0.2 wt.% at 298 K and shear rates between 20-160 s^{-1}. Additionally, viscosity measurements in the temperature range from 298 to 338 K showed that, even though the viscosity increased with the concentration of nanoparticles, those rises were almost insignificant when compared with the water–EG mixture. Other authors interested in the study of functionalized graphene, as Arzani *et al.* [127] who prepared aqueous nanofluids loaded with 0.025-0.1 wt% of either covalent (GnP-COOH) and noncovalent (GnP-SDBS) functionalized graphene nanoplatelets. Reported viscosity measurements at 140 s^{-1} under variation of temperature condition from 293 to 353 K, showed a decreasing in viscosity with increasing temperature (with reductions in the entire temperature range of ~53-58% and ~62-67% for GnP-SDBS/water and GnP-COOH/water series,

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respectively) and an increase with increasing concentration which was higher for GnP-SDBS set (maximum μ_r= 2.36 at 0.1 wt.% and 353 K) than for GnP-COOH set (maximum μ_r= 1.36 at 0.1 wt.% and 338 K). Ma *et al.* [128] prepared nanofluids based on a silicon oil and containing 0.01 and 0.05 wt.% loads of functionalized graphene nanosheets (f-GnS) by following a two-step method. In addition to the Newtonian behavior observed in the shear rate range from 1 to 1000 s^{-1} for the base fluid and the two suspensions, the authors reported close viscosity values at the 0.01 and 0.05 wt.% loadings (with μ increases that almost reached 100% in the highest concentration). Decreases with rising temperature in 293-333 K range reached 49.95% and 48.11% for 0.01% and 0.05% nanofluids, respectively. On the other hand, gallic acid-functionalized graphene nanoplatelets (GAGnP) [129,130] and clove-treated graphene nanoplatelets (cGnP) dispersed in distilled water were investigated by Sadri *et al.* [131]. A GAGnP nanofluid at a 0.05 vol.% graphene concentration was studied between 293 and 313 K, finding that the viscosity of this low volume concentration was close to that of distilled water [129]. The investigation was extended to the 0.02-0.1 wt% nanoparticle range and the 293-323 K temperature interval in a later work [130]. In that last investigation [130], a comparison of the dynamic viscosity measured at 150 s^{-1} for the base fluid and nanofluids shows slight μ increases (when compared to suspensions stabilized with surfactants such as SDBS, triton X-100 or gum arabic). This relatively low rises in viscosity were confirmed by the increases in the friction factor (1.46-3.9% higher than water at studied conditions). Recently, similar rises in friction factor (up to 3.79%) were also reported by the same authors [131] when studied aqueous dispersions of clove-treated graphene nanoplatelets (cGnP) at 0.025-0.1 wt.% mass contents. Sarsam *et al.* [132] focused on the study of triethanolamine-treated graphene nanoplatelets (TEA-GnP)/water nanofluids. The used GnPs had a diameter of 2 μm, and different specific surface areas (SSA) 300, 500 and 750 m^2/g. The rheology measurements were done between 293 and 313 K and for different mass fractions 0.025-0.1 wt%. Sarsam *et al.* [132] found that

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the nanofluids act like Newtonian fluid in studied shear rate range (from 40 to 200 s^{-1}) with increases in measured viscosity values both with increasing weight concentration and with decreasing temperature. Very close μ values were observed when comparing nanofluids containing 0.1 wt.% of different specific surface areas, with only slightly higher values for higher SSAs (especially at temperatures below 303 K). Yarmand *et al.* [133] worked on functionalized graphene nanoplatelets (f-GnP) and functionalized graphene nanoplatelets-platinum (GnP-Pt) [134] dispersed in distilled water nanofluids. The mass concentrations of nanoparticles used in both cases were 0.02-0.1 wt% and they were studied between 293 and 313 K. The authors obtained that viscosity increases by about 24% and 33% at 0.1 wt% of f-GnP and GnP-Pt, respectively, compared to the viscosity of distilled water at 313 K. Functionalized graphene (f-HEG) which was prepared from high purity graphite powder by Hummers method followed by exfoliation and reduction by hydrogen gas and then functionalized using acid, was dispersed in distilled water + ethylene glycol (70:30 in volume) by Kole and Dey [88] without using surfactant. The concentration and temperature ranges worked on were 0.041-0.395 vol% and 283-343 K, respectively. Nanofluids exhibit a non-Newtonian behavior that becomes more prominent with increasing graphene nanosheets (GnS) loading. Increases of relative viscosity with increasing f-HEG loading at 303 K reach nearly 100% (compared to the base fluid) for 0.395 vol% loading of f-HEG. Hussien *et al.* [64] studied the case of graphene nanoplatelets/multi-walled carbon nanotubes (GnP/MWCNT) nanoparticles dispersed in distilled water with the use of polyvinylpyrrolidone (PVP) as a surfactant. The used concentrations were 0.075, 0.125 and 0.25 wt% for MWCNTs, and 0.035 wt% for GnPs. The measurements were done at different temperatures between 300 and 330 K and results showed reductions in viscosity with rising temperature as well as average viscosity increases (compared to distilled water) of 2.8%, 6.4%, and 10.3% for 0.075, 0.125, and 0.25 wt% MWCNTs/GnPs hybrid nanofluids, respectively. Sani *et al.* [63] prepared two concentrations of nanofluids, 0.005 and 0.05 wt%,

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with polycarboxylate chemically modified graphene nanoplatelets (f-GnP), Havoline® XLC Premixed 50/50 base fluid, and using 0.125 wt% of SDBS as surfactant following a two-step method. The rheological measurements were conducted in the temperature range of 293.15-323.15 K, and results showed the Newtonian behavior of the samples (in the shear rate range between 10 and 1000 s^{-1}) and a maximum viscosity increase of 6.6% for the highest nanoparticle concentration (0.05 wt.%) and temperature (323.15 K). Polycarboxylate chemically modified graphene nanoplatelets (f-GnP) were also used by Vallejo *et al.* [135] who investigated the f-GnP dispersions based on water, propylene glycol:water mixture at 30:70 wt%, and propylene glycol:water mixture at 50:50 wt%. Authors analyzed the rheological behavior of different samples at different concentrations in the range of 0.25-1.0 wt% and at temperatures between 283.15 and 353.15 K. A pesudoplastic behavior was obtained in the shear rate range of 10-100 s^{-1}, while samples become Newtonian in the range from 100 to 1000 s^{-1}. In addition, they remarked that the higher the viscosity of the base fluid, the higher the viscosity depletion with temperature of nanofluids, and that the lower the viscosity of the base fluid, the higher the viscosity increase due to the weight loading. Sulfonic acid-functionalized graphene nanoplatelets were dispersed in an ethylene glycol:water mixture 50:50 vol% this time by Vallejo *et al*. [136] who focused on the study of their rheological properties. Nanofluids with different concentrations between 0.25 and 2.0 wt% were prepared and measured in the temperature range 283.15-353.15 K and at shear rates from 1 to 1000 s^{-1}. The results showed a decreasing in the viscosity by around 80% for nanofluids and 82% for the base fluid with increasing temperature 70 K step. It was found also that viscosity increases with concentration. For example, nanofluid with 0.50 wt% had a viscosity higher than the base fluid by 16% without any temperature dependence. In addition, a non-Newtonian shear thinning behavior, more evident with increasing nanoadditive concentration, was observed at low shear rates, and after that sample became quasi Newtonian. Agromayor *et al.* [137] studied the case of aqueous nanofluids

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containing concentrations between 0.25 and 1.0% of sulfonic acid-functionalized graphene nanoplatelets.Viscosity measurements at the temperature range 293-313 K showed a decreasing with the temperature increase (~48-55% in entire temperature range), and the addition of nanoparticles led to significant viscosity increases (~74-80% higher than distilled water). In addition, rheological tests showed Newtonian behavior at all concentrations. Vallejo *et al.* [138] investigated propylene glycol:water (30:70) wt% based nanofluids prepared with functionalized graphene nanoplatelets (f-GnP). Viscosity was tested for different f-GnP mass concentrations in the range of 0.25-1.0%, at temperatures between 293.15 and 323.15 K and constant shear rates from 1000 to 4000 s^{-1}. Measurements showed that the viscosity is independent on the testing time (100 s) and also on the imposed shear rate, which means that all nanofluids are Newtonian. Besides, the viscosity decreased between 31% and 57% with the temperature rise. As for nanoparticle dependence, viscosity was observed to increase with the increase of nanoparticle loading, reaching 44% for the lowest concentration (0.25 wt.%) and 214% for the highest one (1 wt.%). Details of the previous investigations on functionalized and doped graphene- nanofluids are summarized in **Table 6**.

Insert here Table 6

4. Viscosity enhancement: impact of base fluids, temperature and graphene derivatives concentration

After the description in the previous sections of the impact of the nature of graphene derivatives on the rheological properties and viscosity of graphene-based nanofluids, it is proposed in this part to compare the viscosity enhancement and the relative viscosity (*i.e.* the ratio of nanofluid viscosity to base fluid viscosity, $\mu_r=\mu_{nf}/\mu_{bf}$) variation of these nanofluids with graphene content. Particular

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attention is given to analyze the influence of base fluids, volume or mass fraction, temperature and presence of surfactant.

The most common base fluid used for the preparation of graphene nanofluids is water, as for many of other types of nanoparticles. The relative viscosity of aqueous nanofluids made of graphene is shown in **Figures 4 and 5**, distinguishing pristine graphene derivatives (**Fig. 4**) to GO, rGO and functionalized graphene (**Fig. 5**). Similar analyses are done in **Figures 6** and **7** for graphene dispersions in glycols and glycol+water mixtures (**Fig. 6a & 6b**) and for oil-based nanofluids and ionanofluids (**Fig. 7**). First at all, as generally expected with nanofluids [33], it is observed in **Figures 4-7** that the inclusion of graphene nanostructures usually raises the viscosity of the host base fluid. Thus, increases with nanoparticle concentration can reach 20% for very low contents in graphene dispersed in water, typically with 0.075% with pristine graphene and for similar or lower content with GO, rGO or functionalized graphene, see **Figures 4 and 5**. The Figures also evidence that main studies were performed for graphene contents lower than 0.1% in wt. It is also shown that, for pristine graphene, the results are well comparable between the studies and the enhancements are in the same order of magnitude in the concentration range 0-0.1 wt.%. This can be explained by both the similar treatment applied to the graphene and the preparation of nanofluids as these works mainly involved the same group of authors. With other derivatives, the results are much more scattered evidencing the impact of chemical treatments applied to these graphene derivatives and the preparation methods of nanofluids. Viscosity studies on nanofluids prepared using larger graphene concentrations (>0.2 wt%) are more scarce. Although most of the samples loaded with 1 wt.% of graphene or graphene derivatives exhibit viscosity enhancements larger than 100% [116,118,138], some researchers [99,116,135,138] obtained rises in dynamic viscosity of less or around 40% for

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that same concentration of functionalized graphene nanoplatelets [99,116] or graphene decorated with Fe_3O_4 [135,138].

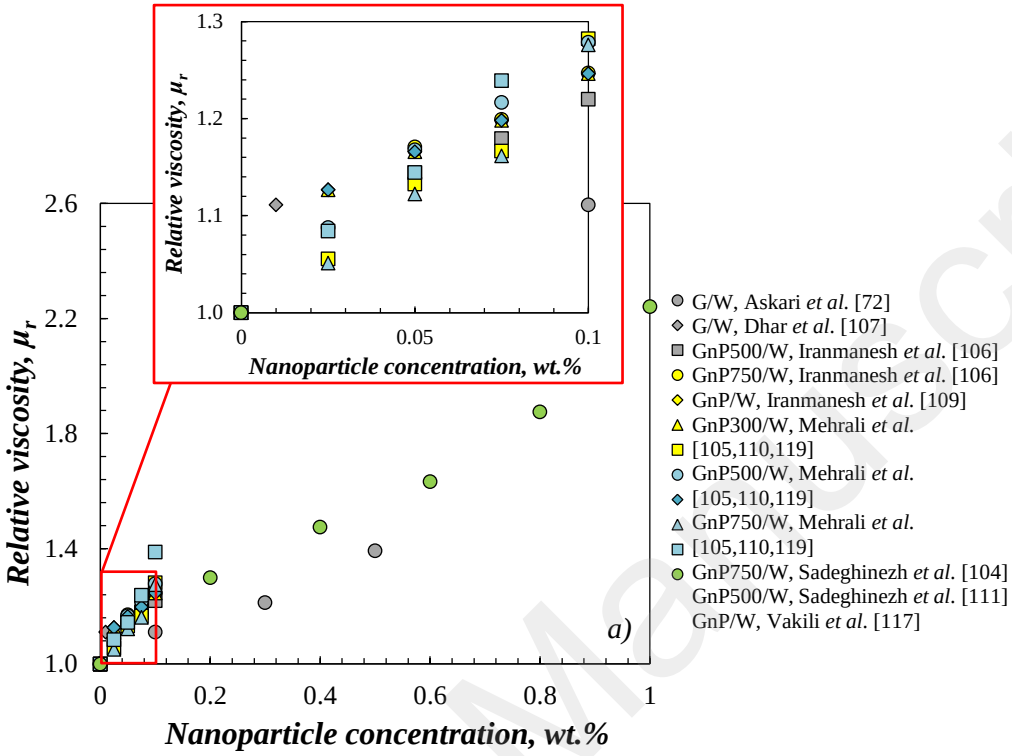

Figure 4. Relative viscosity, $\mu_r=\mu_{nf}/\mu_{bf}$, of aqueous nanofluids containing pristine graphene as a function of mass fraction at 293~303 K.

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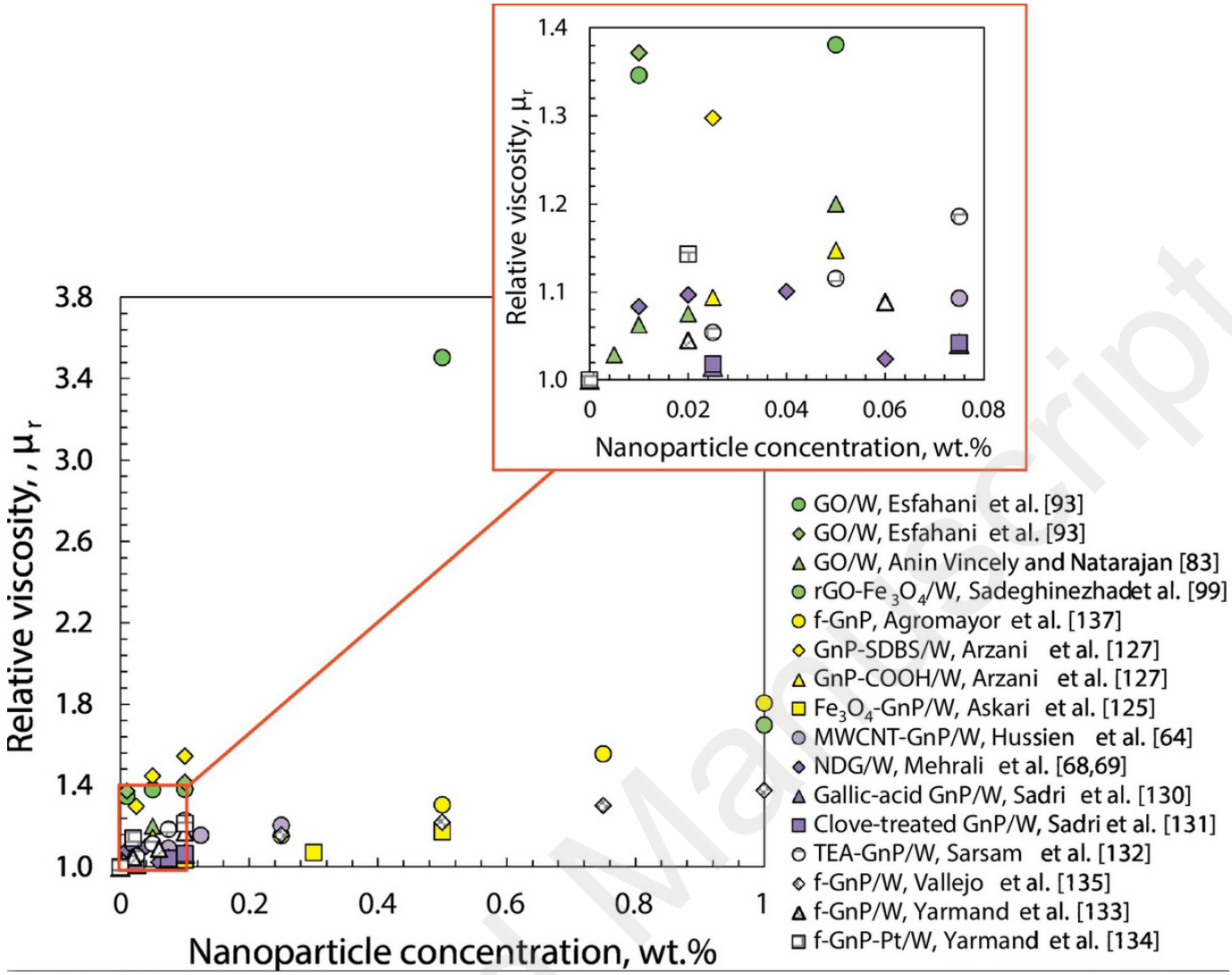
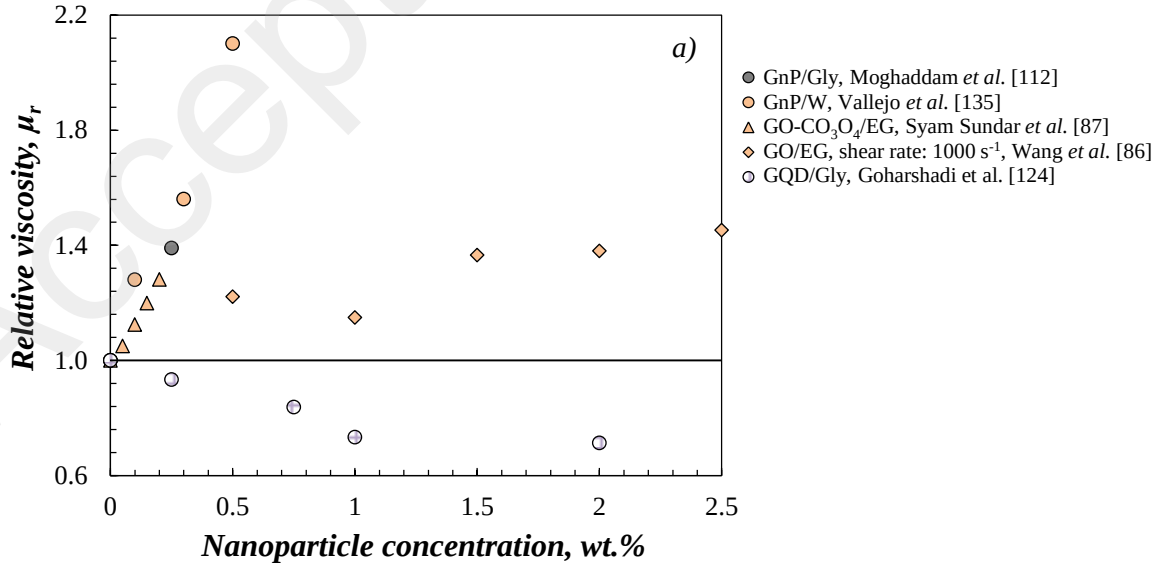


Figure 5. Relative viscosity, $\mu_r=\mu_{nf}/\mu_{bf}$, of aqueous nanofluids containing graphene oxide, reduced graphene oxide or functionalized graphene as a function of mass fraction at 293~303 K.



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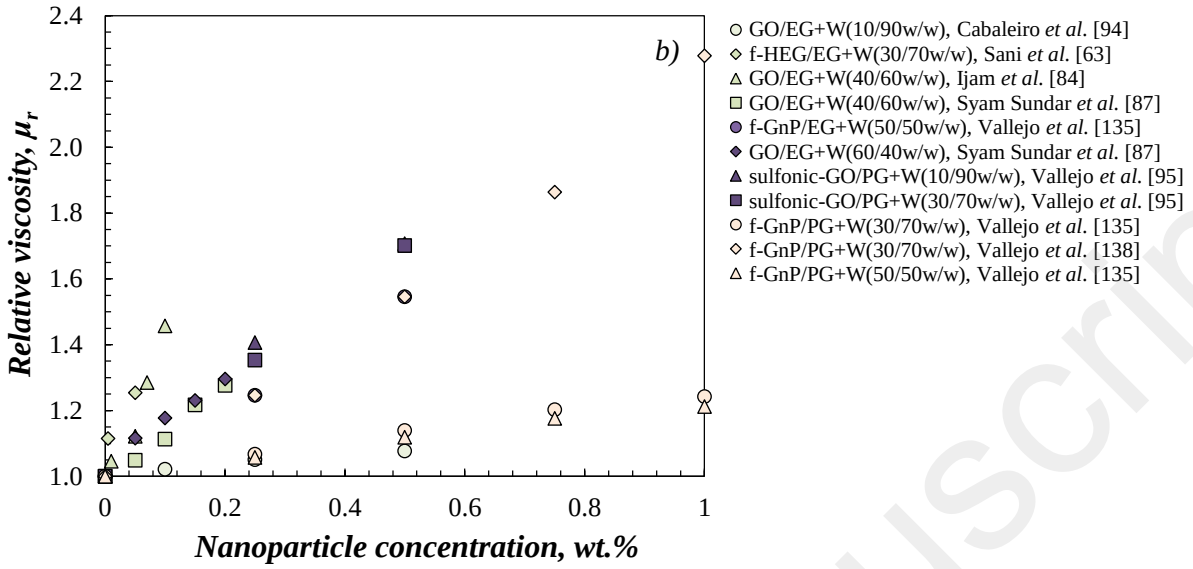

Figure 6. Relative viscosity, $\mu_r=\mu_{nf}/\mu_{bf}$, of *(a)* glycol-based at 293~303 K and *(b)* glycol+water-based nanofluids as a function of nanoparticle mass concentration at 303 K.

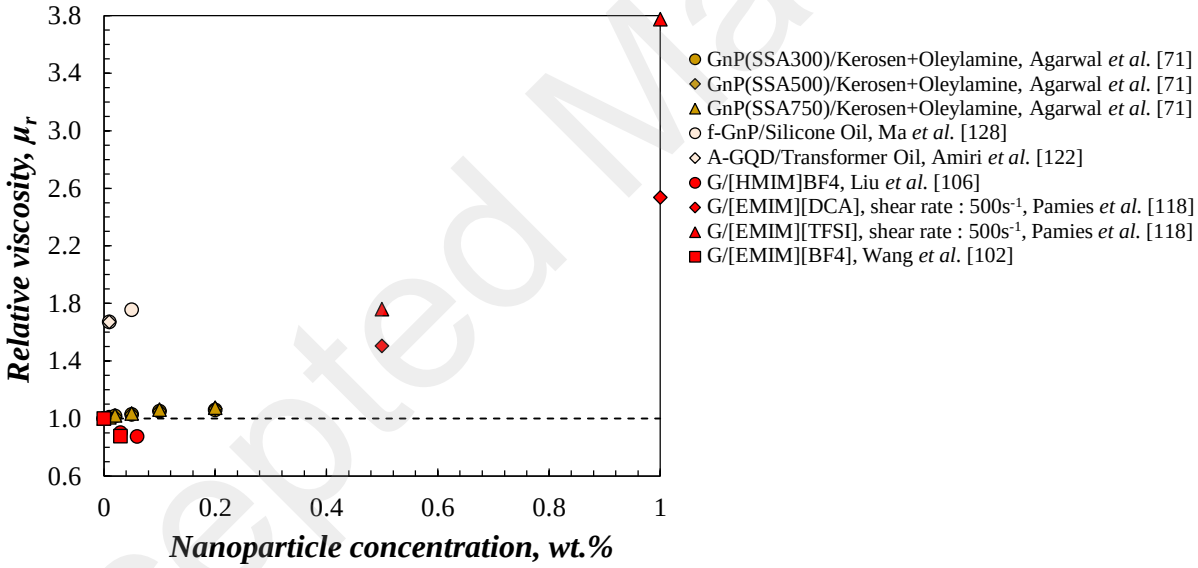

Figure 7. Relative viscosity, $\mu_r=\mu_{nf}/\mu_{bf}$, of oil-based nanofluids and ionanofluids as a function of nanoparticle mass concentration at 303 K.

Figures 6a and 7 also show unusual reductions in viscosity with nanoadditive loading reported by Goharshadi *et al.* [124] when studied dispersions of graphene quantum dots in glycerol or by Wang *et al.* [102] and Liu *et al.* [103] when investigated graphene ionanofluids based on [HMIM]BF_4. As

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comprehensively reviewed by Minea and Murshed [139], unusual reductions in viscosity with rising nanoparticle loading were also reported for other ionanofluids containing different nanoadditives such as multi-walled carbon nanotubes [140,141] or Ru nanoparticles [142]. In the case of base ionanofluids, such unexpected reductions were previously attributed in the literature to: *i)* a certain self-lubrication effect of nanoparticles [102,143,144] or *ii)* the possible formation of hydrogen bonds between carbon-based nanoparticles and the cation of the IL which, in turn, may reduce Coulombic attractions between cations and anions present in the ionic liquid used as base fluid [141]. As discussed throughout the article, some investigators used a dispersant to improve the stability of their samples. **Figure 8** graphically presents the relative viscosities of graphene nanofluids stabilized with surfactants. The addition of a surfactant usually modifies the viscosity of a colloidal dispersion. To strictly analyze the effect that the addition of graphene has on the viscosity of graphene nanofluids, μ_r values were calculated considering the surfactant as a part of the base fluid. A sharp increase in viscosity ratio within the nanoparticle loading range of 0.025-0.1 wt.% was observed by Arzani *et al.* [127] for aqueous dispersions of graphene stabilized with SDBS at a graphene:SDBS ratio of 0.5:1. This result contrasts with the maximum μ_r ratio of 1.194 obtained for 1.0 wt.% of f-GnP nanofluids based on a commercial mixture of Havoline® XLC Premixed 50/50 and a concentration of 0.125 wt.% of that same surfactant (SDBS) by Vallejo *et al.* [145], for example. According to most models reported in the literature to describe the influence of particle concentration on dynamic viscosity of colloidal dispersions [4,34], a proper comparison of the viscosity of different nanofluids sets should be carried out based on the nanoparticle volume fraction, in which density of nanoparticle is included, and not only mass fraction. However, only few authors (see **Figure 9**) reported their viscosity results indicating nanoparticle content in volume that needs to know the nanoparticle density. Strong differences are also observed when relative viscosities are presented as a function of volume fraction. Hence, maximum increases in

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viscosity range from 9.3% for an (ethylene glycol+water)-based nanofluid loaded with 0.5 vol.% of GnP [65–67] to 180% in the case of aqueous nanofluids containing only 0.01 vol.% of GO:SiO_2 (50:50 vol.%) [91].

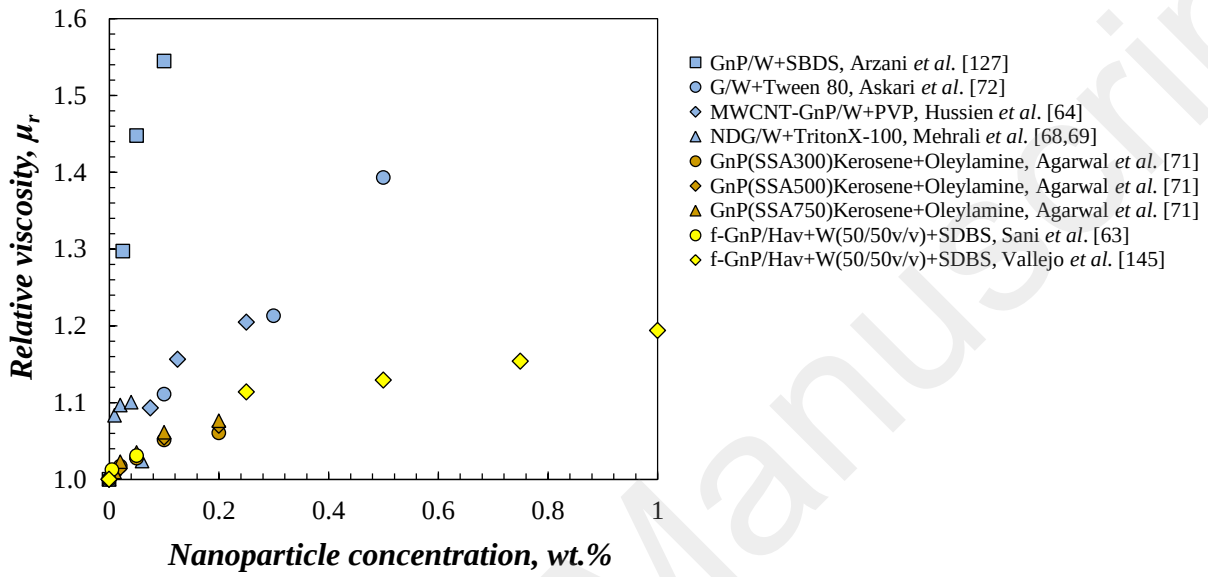

Figure 8. Literature relative viscosities, $\mu_r=\mu_{nf}/\mu_{bf}$, of graphene-derivative nanofluids stabilized by means of several surfactants. Values were collected at a temperature in the range from 293-303 K.

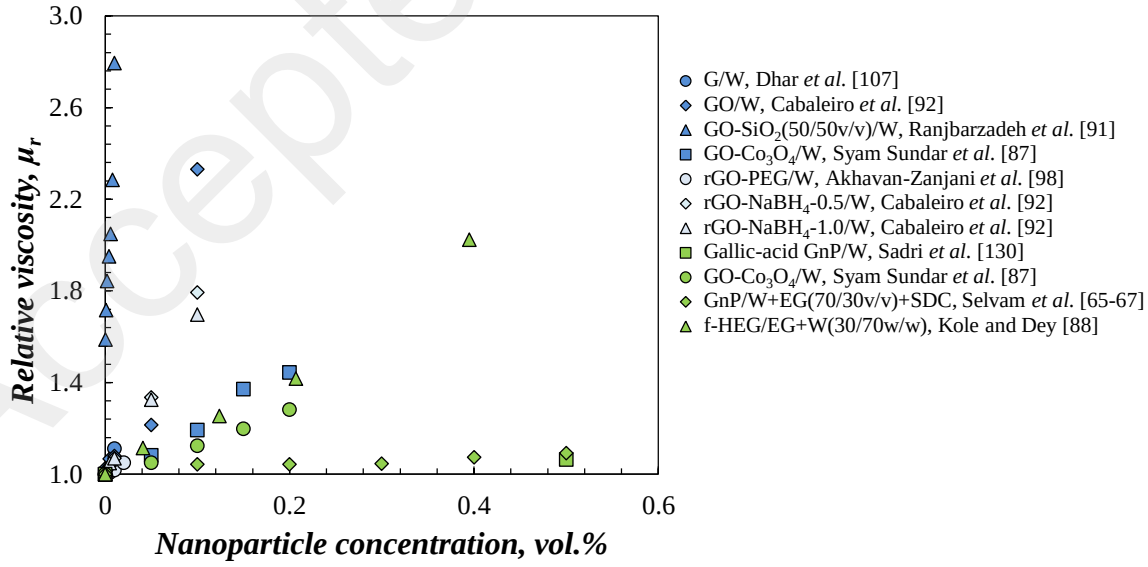

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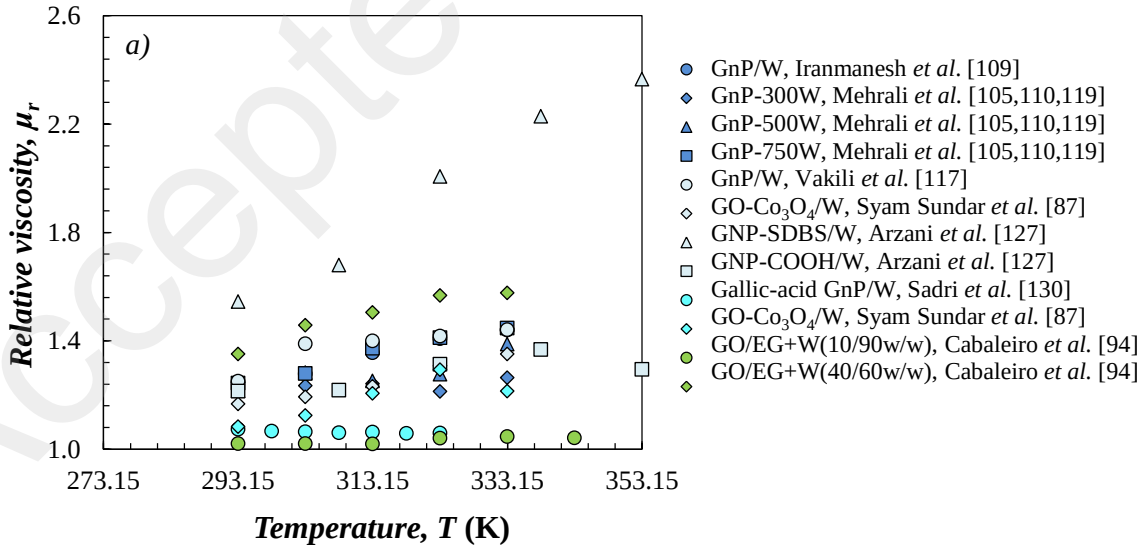
Figure 9. Relative viscosity, $\mu_r=\mu_{nf}/\mu_{bf}$, of graphene-derivative nanofluids as a function of nanoparticle volume fraction.

Figure 10 graphically presents the temperature dependence of relative viscosity (calculated as the ratio between the viscosity of the nanofluid and the value of the corresponding base fluid at the same temperature) for several nanofluids prepared at nanoparticles concentrations of 0.1 and 0.5 wt.%. It can be observed that temperature has a mixed effect on relative viscosity and can lead to increases, decreases or no change on μ_r results. Authors such as Sadri *et al.* [130], Mehrali *et al.* [105,110,119], Vallejo *et al.* [135] or Cabaleiro *et al.* [94] did not observe any special temperature-based mechanism that alters the viscosity of their nanofluids. However, other researchers reported reductions [95] or increases [84,87,105,109,110,117,119,127] in relative viscosity with increasing temperature.



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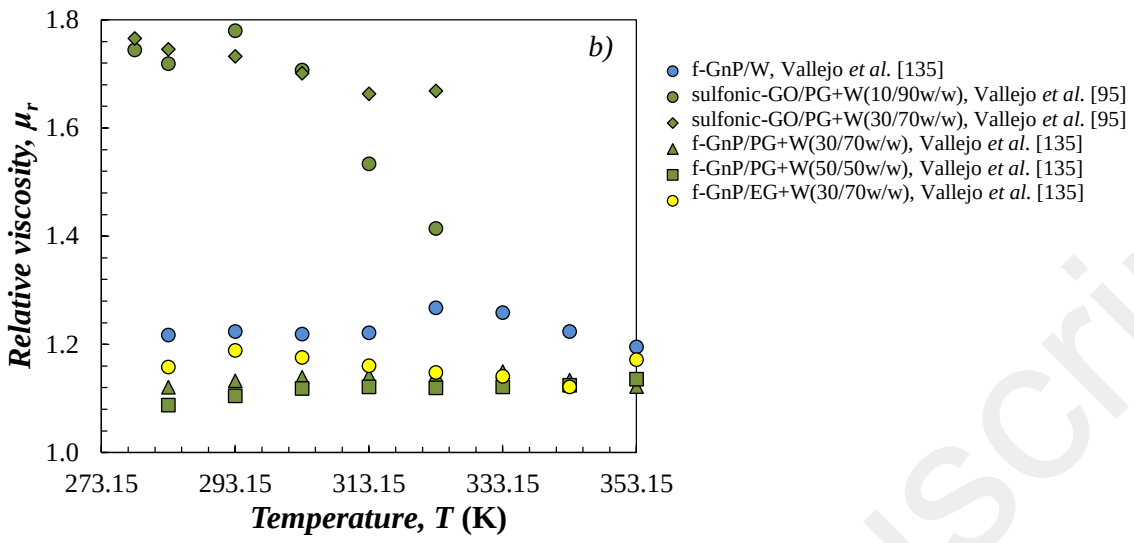

Figure 10. Temperature dependence of relative viscosities, $\mu_r=\mu_{nf}/\mu_{bf}$, for nanofluids based on water or glycoled water and containing: *(a)* 0.1 wt.% and *(b)* 0.5 wt.% nanoparticle concentrations of graphene-derivatives.

Some investigations also analyzed experimentally the shear viscosity dependence on shear stress of graphene nanofluids in order to provide an insight into the Newtonian or non-Newtonian behavior of the samples. Such information is useful to identify the flow nature of the nanofluids as well as to investigate the pressure drop at different flowing rates [34]. **Figures 11-12** show some representative examples of Newtonian and non-Newtonian nanofluids containing graphene derivatives and based on water, ethylene glycol or ionic liquids. Newtonian behaviors (within the studied ranges of shear rates and nanoparticle loadings) were reported by Vallejo *et al.* [95] for 0.25-0.50 wt% dispersions of f-GnP in two different propylene glycol-water mixtures, by Amiri *et al.* [123] for aqueous nanofluids containing 0.001 and 0.002 wt% of amine-treated graphene quantum dots (A-GQD), by Sarsam *et al.* [132] for 0.025-0.10 wt.% dispersions of triethanolamine-treated graphene nanoplatelets in water or by Ma *et al.* [128] for 0.01% and 0.05% loading of f-GnS in silicone oil, for example. Conversely, nanofluids that yield a non-linear relation between shear stress and shear rate were also obtained in the literature [62,82,84,85,91,93,107,118,124]. The Newtonian or non-Newtonian behavior strongly depends on the

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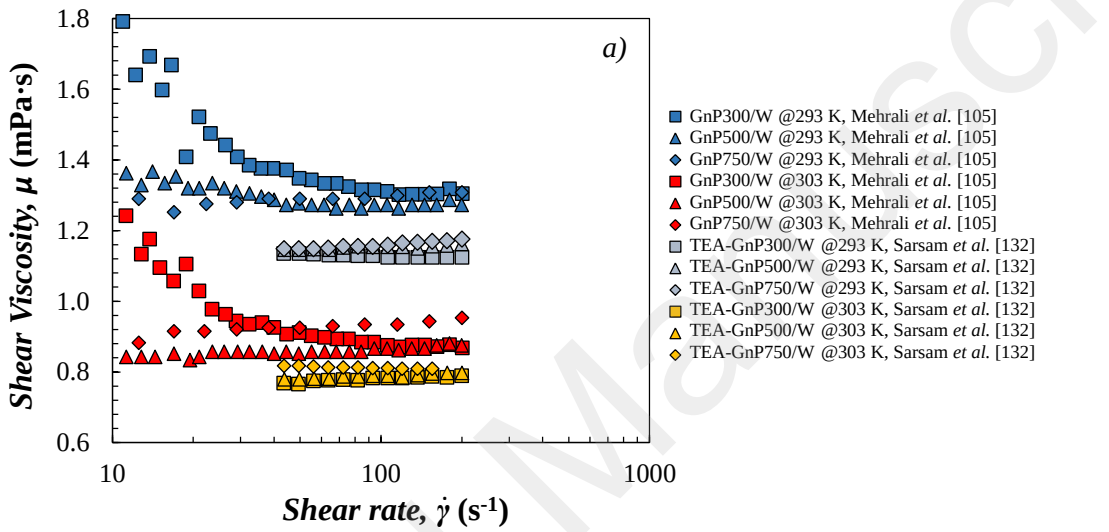
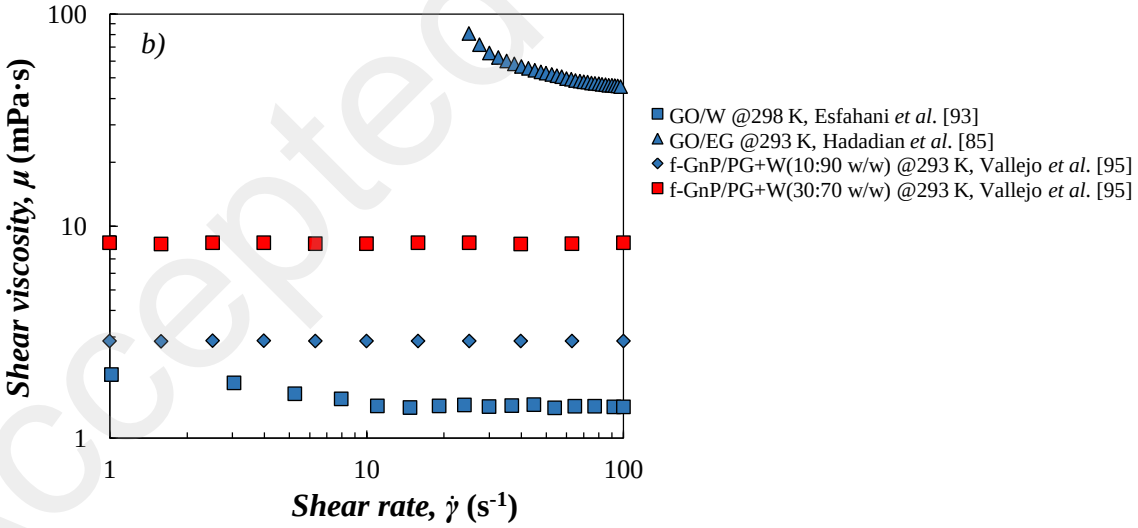
nanoparticle concentration (pseudoplastic or shear-thinning behavior being more evident at high contents of nanoparticles) as well as the functionalization/treatment of the nanoparticles or the use of surfactants. Thus, as it can be observed in **Figure 11c**, Sarsam *et al.* [62] found as the shear thinning degree was considerably reduced when an appropriate amount of surfactant (either SDBS, SDS, CTAB or GA) was incorporated to the sample.





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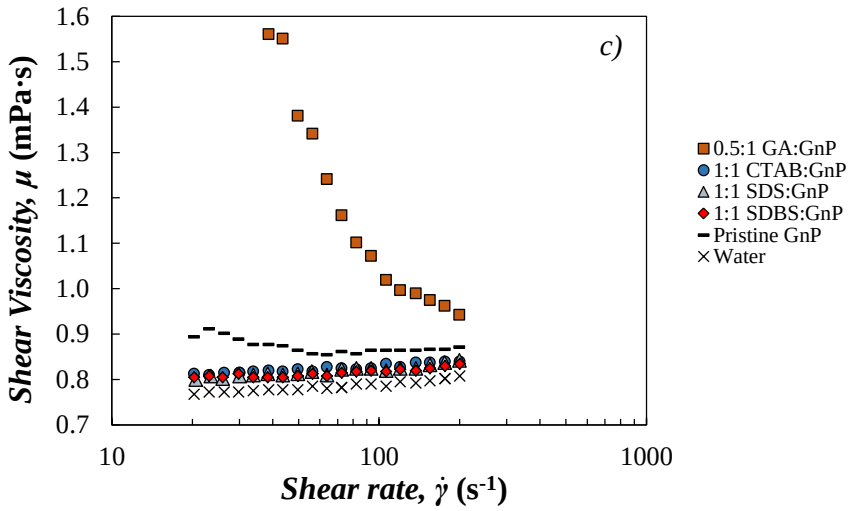
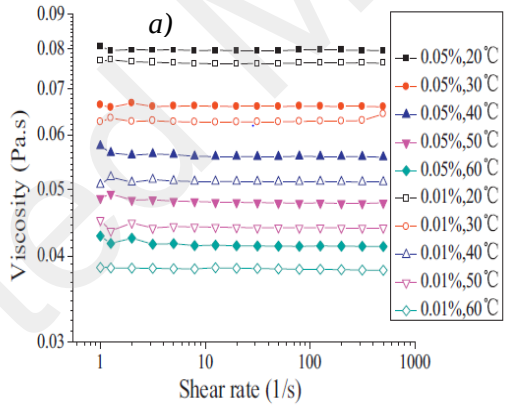


Figure 11. Representative examples of Newtonian and non-Newtonian nanofluids containing graphene derivatives and based on water or ethylene glycol. *(a)* Aqueous dispersions containing 0.1 wt.% loadings of pristine graphene or TEA-functionalized graphene with different SSAs at 293 and 313 K. *(b)* Graphene oxide suspensions at 0.5 wt.% in different base fluids at 293-298 K. (c) Water-based nanofluids containing 0.1 wt% concentrations of GNPs and stabilized with surfactant:GNPs ratios of 0.5:1 and 1:1.



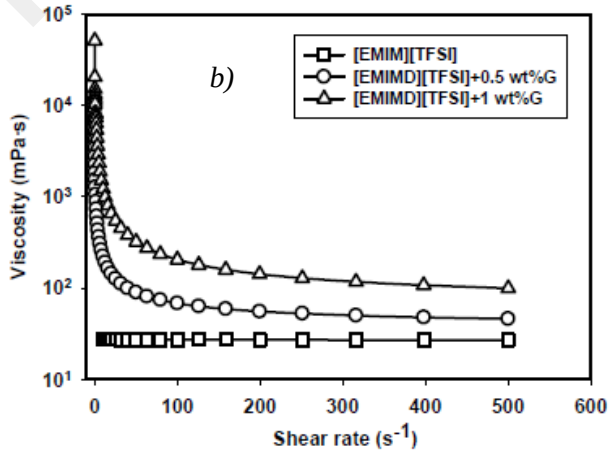


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Figure 12. Representative examples of Newtonian and non-Newtonian nanofluids containing graphene derivatives and based on ionic liquids. Reprinted with permission from a) Ma *et al.* [128] and Pamies *et al.* [118].

5. Viscosity models for graphene-based nanofluids

Several theoretical or empirical models have been used or developed to correlate or predict the viscosity variation of nanofluids under certain conditions, as reported in [34]. The majority of existing viscosity models are for nanofluids based on spherical nanoparticles, and few of them for rod-like nanoparticles. In this section, we only report the models associated to graphene-based nanofluids. As a summary, these models are gathered in Table 7.

Researchers have used the Andrade equation (Eq.1) to correlate the relationship between the viscosity of nanofluids and the temperature. This equation is defined as follows:

$$\mu = A.\, e^{\frac{B}{T}} \tag{1}$$

where A and B are adjustable parameters, and T is absolute temperature (K) [146,147].

A correlation was proposed by Wang *et al.* [116] to estimate the apparent viscosity of graphene (GnP) nanofluids as a function of nanoparticle mass fraction (φ) and fluid temperature T (K), using a multiple steps regression analysis. This equation is valid for $0.2 < \varphi < 1$ wt.% and $278.15 < T < 298.15$ K and writes as follows.

$$\mu = 0.004.\,(1-\varphi)^{-77.5}\,.\,e^{\frac{1652}{T}} \tag{2}$$

On the other hand, the Vogel-Fulcher-Tammann (VFT) equation [148,149] also known as the Vogel-Fulcher-Tammann-Hesse equation, is one of the most widely used models to describe the temperature dependence of viscosity:

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$$\mu = \mu_0 . e^{\frac{D \cdot T_0}{T - T_0}} \tag{3}$$

where μ_0, D, and T_0 are the fitting parameters.

Recently, the dynamic viscosity values of graphene nanofluids have been well modelled using Vallejo *et al.*'s equation [95] which includes, in the same expression, the dependence of the viscosity on the graphene volume fraction ϕ and the temperature T:

$$\mu = \mu_0 . e^{\frac{D.T_0}{T - T_0}} + \mathrm{E}.e^{\frac{F}{T}} . \phi - \mathrm{G}. \phi^2 \tag{4}$$

where E, F, and G are fitting parameters; ϕ is the volume fraction; and μ_0, D, and T_0 are the fitted parameters for the corresponding base fluid obtained previously from the VFT equation, see equation (3).

The Maron and Pierce model [150] defined by equation (5) has also been shown to be effective in the description of nanofluid viscosity, in particular with carbon nanotubes [17,94,151]. As an advantage, this model does not require the knowledge of intrinsic viscosity that includes nanoparticle shape.

$$\frac{\mu_{nf}}{\mu_{bf}} = \left(1 - \frac{\phi}{\phi_m}\right)^{-2} \tag{5}$$

where ϕ is the nanoparticle volume fraction, while ϕ_m is the maximum packing volume fraction.

This model was used by Cabaleiro *et al.* [92] to predict the viscosity enhancement of graphene oxide and reduced graphene oxide water-based nanofluids, ϕ_m being considered in this work as a fitting parameter.

Insert here Table 7

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6. Future remarks

Research groups around the world are stepping towards the development of advanced thermal fluids. The suspension of graphene derivatives in different base fluids has proven to be effective to improve the thermal performance of several thermal media. However, the investigation must not be limited to thermal conductivity and other thermo-physical properties must also be considered. Thus, more comprehensive analyses are still necessary to understand the complex mechanisms behind the modifications in dynamic viscosity and rheological behavior of graphene nanofluids before such dispersions can be used in real-life applications. Some remarks for future works are presented as follows:

- Decreased viscosity phenomenon with nanoparticle loading observed for graphene quantum dots or suspensions based on ionic liquids should be further investigated. Such behavior, which contradicts conventional theories such as the Einstein-Batchelor relationship (according to which effective viscosity shall increase with rising particle concentration), may be potentially interesting to achieve enhancements in thermal conductivity with no penalization in dynamic viscosity or even to improve the lubricity effect of some thermal fluids.

- Some interesting publications are analyzing the effect that nanoparticle specific surface area has on graphene-derivative nanofluids. However, such information is still limited. More investigations unfolding how size and aspect ratio of nanoparticles affect dynamic viscosity and rheological behavior of nanofluids containing graphene derivatives could help to better optimization of such suspensions.

- Functionalization of graphene derivatives is sometimes required to produce stable suspensions based on several polar solvents such as water. Information regarding the connections among thermal conductivity, dynamic viscosity and sample stability must be investigated for different functionalization

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processes. An insight on how functionalization affects stability and transport properties could help to optimize nanofluid preparation.

- Surfactant addition can also play an important role in sample stabilization and nanofluid viscosity. However, most analyses focused at around room temperature. More comprehensive studies are necessary to evaluate the effectiveness (or not) of using surfactants at high and low temperatures.

- A theoretical semi-empirical equation to predict the ratio between the effective dynamic viscosity of graphene-derivative suspensions and the dynamic viscosity of the base fluid as a function of nanoparticle formulation parameters (nanoparticle size, concentration, functionalization, etc.) is still one of the main challenges. The development of such a general model relies on a wide database of accurate dynamic viscosity measurements of nanofluids containing several graphene derivatives.

7. Conclusions

Literature research proves the potential of graphene and its derivatives as nano-enhancers to develop novel heat transfer media with increased thermal performance. However, the impact of nanoparticle addition on viscosity and pressure drop must be carefully considered before the practical implementation of nanofluids. In this paper, the dynamic viscosity and rheological behavior of suspensions containing graphene-derivatives have been reviewed, considering the effect of temperature as well as several nanofluid preparation parameters (base fluid type, surfactant type and concentration; or nanoparticle type, volume concentration, size and morphology). Available results and conclusions are critically discussed throughout the manuscript and presented in different graphs and tables. Graphene functionalization and base fluid were observed to play a main role. Thus, although most investigations

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reported dynamic viscosity increases with nanoparticle loading, some authors also observed reductions in this transport property when adding graphene quantum dots or when the suspensions were based on certain ionic liquids. Like their base fluids, nanofluids also showed a downward trend in dynamic viscosity with increasing temperature, which is characteristic of liquids. However, dissimilar effects were observed in the temperature dependence of relative viscosity. Thus, while some researchers observed non-temperature influence of rises in dynamic viscosity, others reported remarkable rises/depletions in relative viscosity with increasing temperature. The use or not of a dispersant also has significant effects on dynamic viscosity. Usually, nanofluid viscosity increases after the incorporation of a surfactant to the dispersion. However, comparative concentrations of different surfactants can lead to large differences in dynamic viscosity enhances or changes in the Newtonian or non-Newtonian behavior of the suspensions. Hence, special attention must also be paid to the selection of the optimum surfactant type and concentration to avoid undesirable penalizations in pumping power.

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Table 1. Most common approaches to synthesize graphene derivatives[†].

Synthesis method	Number; transversal size	Precursor(s) used	Graphene derivative produced	Mechanism
Mechanical/micromechanical exfoliation	SLF, FLG; 5-10 µm	Graphite	Pristine	Peeling off graphene layers using an adhesive tape.
Chemical exfoliation	SLF, FLG; > 100 nm	Graphite with graphene-intercalated compounds	Chemically modified graphene	Increase interlayer spacing between graphite layers and applying sonication.
Chemical synthesis	SLF, FLG; < 20 µm	Graphite oxide	Graphene oxide (unless chemical reduction)	Exposing graphite oxide to solvents and applying sonication.
Chemical vapor deposition	SLF, FLG; > 100 µm	Polycrystalline Ni films, copper foils, transition metals	Pristine	Carbon segregation or precipitation over transition metals.
Epitaxial growth	SLF, FLG; > 50 µm	SiC or Ru	Pristine	Thermal decomposition of hydrocarbons on top of single-crystalline subtracts.
Solvothermal synthesis	SLF, FLG; < 20 µm	Solvothermal product (e.g. $Na+C_2H_5OH$)	Chemically modified graphene	Pyrolysis and filtering of

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solvothermal product.

[†]Partially adapted from Rasheed *et al.* [29].

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Table 2. Main information about rheological analysis of graphene oxide-based nanofluids

Type of nanoparticle†	Type of treatment	Base fluid⊥	Surfactant	Nanoparticle size⁑	Concentration range*	Temperature range‡	Key results	Rheological behavior	Reference
Graphene oxide (GO)	OH- and COOH-functional groups are used	Water	–	2 µm (diameter), 3.4-7 nm (thickness)	0.025-0.1 vol%	–	Dynamic viscosity increased with GO loading	–	Ranjbarzadeh *et al.* [80]
Graphene oxide (GO)	Modified Hummer method	Water	–	–	0.25-1.5 g/L	298 K	μ_r increased from 1.041 at 0.25 g/L to 1.248 at 1.5g/L.	–	Nazari *et al.* [81]
Graphene oxide (GO)	Oxidizing purified natural flake graphite via the modified Hummers method	Distilled water (DW)	–	650 ± 20 nm (Z average size of GO 0.01%), 1220 ± 15 nm (Z average size of GO 0.1%)	0.01-0.1 wt%	298-313 K	Viscosity of 0.01 wt% GO was in a similar range to DW; Viscosity of 0.1 wt% GO was 60% higher than DW at 298 K	Newtonian for 0.01 wt%, and non-Newtonian at low shear rates(under 20 s^{-1}) for 0.1 wt%	Esfahani and Languri [82]
Graphene oxide (GO)	From graphite by the modified Hummer's method	Deionized water	–	–	0.005-0.05 wt%	303-323 K	At 0.05wt%, μ_{nf} increased by 10.4% at 323 K to 20.2% at 303 K compared to the base fluid	Non-Newtonian at shear rates <2000 s^{-1} for 0.01 and 0.05 wt%	Anin Vincely and Natarajan [83]
Graphene oxide (GO)	Hierarchical method	Ethylene glycol +water (40:60 weight ratio)	–	0.5-2 µm	0.01-0.1 wt%	293-333 K	Viscosity of 0.1 wt% GO increased by 35% compared to the base fluid at 293 K and decreased by 48% from 293 to 333 K	Non-Newtonian at shear rates <100 s^{-1} for 0.05 wt% and <300 s^{-1} for 0.1 wt%.	Ijam *et al.* [84]

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Graphene oxide (GO)	GO NFs was prepared by exfoliation of GtO in the base fluids. GtO was fabricated using modified Hummers' method	Ethylene glycol	_	20 nm (average size)	0.001-0.005 wt%	293-323 K	μ_r = 3.4 for 0.005 wt% of GO at 293 K and shear rate of 27.5 s^{-1}. At 323 K and shear rate of 67.5 s^{-1}, μ_r increased from 1.52 at 0.001 wt% to 2.34 at 0.005 wt%.	Non-Newtonian at low shear rates and Newtonian at high shear rates	Hadadian *et al.* [85]
Graphene oxide (GO)	_	Ethylene glycol	_	1-5 µm (diameter), 0.8–1.2 nm (thickness)	0.5-2.5 wt%	293 K	Low viscosity increase (from 21.4 to 29.5 mPa.s for EG containing no and 2 wt% GO).	Newtonian behavior for 0.5-1 wt% loading and shear thinning behavior at low shear rates when the loading increased to 1.5 wt%	Wang *et al.* [86]
Graphene oxide-copper oxide hybrid nanoparticles (GO/Co_3O_4)	Graphene oxide (GO) nanosheets were obtained from exfoliation of high purity graphite in an acidic medium (modified Hummers method). In-situ growth and chemical co-precipitation method was used for the synthesis of uniform dispersion of Co_3O_4 nanoparticles on the graphene oxide (GO) nanosheet	Water, Ethylene glycol, Ethylene glycol+Water (20:80, 40:60, 60:40 weight ratios)	_	100 nm (GO)	0.05-0.2 vol%	293-333 K	μ increases with ϕ and decreases with T. μ_r=1.70 (water-based nanofluids) and μ_r=1.42 (ethylene glycol-based nanofluid) at 0.2 vol.% of GO/Co_3O_4 and T=333 K.	_	Syam Sundar *et al.* [87]

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Graphene oxide (GO)/carbon nanotube (MWCNTs)/magnetite Fe_3O_4 -(GMF) and shell of polyether (M2070)	Graphite oxide (GO) was synthesized with graphene as original material by a modified Hummers method. GO/MWCNTs/Fe_3O_4 (GMF) nanofluid was prepared through three reaction steps	Epoxy resin	_	Acidified MWCNTs: 100-500 nm (length), 20 nm (diameter) Fe_3O_4: 8 nm (diameter)	0.05 g/mL	Room Temperature-353 K	Viscosity was 3.26 Pa.s at 303 K and decreases with *T* to 0.81 Pa.s at 333 K	_	Yao *et al.* [89]
Graphene oxide (GO)	GO was prepared by a chemical method starting from graphite powder	SAE10W40 oil	_	_	0.03 mg/mL	_	_	Newtonian behavior for neat SAE10W40 and GO/SAE10W40 blend	Mishra *et al.* [90]
Graphene oxide silicon oxide (GO-SiO_2 at 50:50 volume ratio)	GO was synthesized using the modified Hummer's method, and the nanofluid was prepared using the two step method	Water	_	<100 nm (diameter)	0.5-1 vol%	293-333 K	Maximum increase in viscosity of water from 1.059 mPa·s to 2.421 mPa·s for 1 wt% nanofluid concentration at 293 K. μ decreases with increasing *T*. Addition of SiO_2 decreases the viscosity (regarding GO/water NFs). μ_r increases with *T* and ϕ.	Newtonian behavior in the shear rate range of 110-245 s^{-1}	Ranjbarzadeh *et al.* [91]

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Graphene oxide (GO)	GO was produced from graphite through a derived Hummers' method	Deionized water	_	_	0.0005-0.1 vol%	293-303 K	Maximum increases by 100-130 % for the non-Newtonian nanofluids. Larger increases are observed for the 0.1 vol% concentration at 303 K. Increase in relative viscosity by 130% in the considered concentration range. Relative viscosity is not significantly influenced by temperature	Newtonian behavior for nanofluids concentrations lower than 0.01 vol%, and shear-thinning non-Newtonian behavior for higher concentrations at shear rates between 10-1000 s^{-1}	Cabaleiro *et al.* [92]
Graphene oxide nanosheets	From graphite powder using the modified Hummers method	Deionized water	_	Three particle size groups: <200 nm, 200-1000 nm, and >1 μm.	0.01-0.5 wt%	298-333 K	μ increases linearly with the increase of nanofluid concentration with a severe increment for the highest one, and decreases with the rise of temperature. Viscosity increases at 298 K and shear rate of 100 s^{-1} are: 38% and 130%, respectively.	Non-Newtonian behavior at lower shear rates with higher concentrations.	Esfahani *et al.* [93]

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Sulfonic acid-functionalized graphene oxide nanoplatelets	Nanofluids were prepared following a two-step method	Ethylene glycol:water mixture 10:90 wt%	_	Apparent sizes from some nanometers to 4 µm	0.1-0.5 wt%	283.15-343.15 K	μ decreases with rising T and increases with nanoparticle concentration. Maximum viscosity increase of 12.6% at 343.15 K and 0.5 wt.%.	Newtonian behavior	Cabaleiro *et al.* [94]
Sulfonic acid-functionalized graphene oxide nanoplatelets	Nanofluids were prepared following a two-step method	Propylene glycol/water mixture at 10:90 wt% and 30:70 wt%	_	5-10 layers, 3-12 nm (thickness), up to 380 nm (long and width dimensions)	0.25 and 0.50 wt% with PG:W 10:90 wt%. 0.25, 0.50, 0.75 and 1.0 wt% with PG:W 30:70 wt%	278.15-323.15 K	PG:W 30:70 wt% based nanofluids are more viscous than PG:W 10:90 wt% based nanofluids by ~123% and ~106% for the same mass concentrations at 283.15 and 293.15 K, respectively. Viscosity depletions with increasing temperature are: 68% (for PG:W 10:90 wt%) and 80% (PG:W 30:70 wt% base fluids). Maximum viscosity increase by 58 and 99% for the PG:W 10:90 wt% base fluid and 39 and 76% for the PG:W 30:70	Newtonian behavior over all the analyzed concentration and temperature at shear rates from 1 to 100 s^{-1}.	Vallejo *et al.* [95]

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							wt% base fluid for the 0.25 wt% and 0.50 wt% nanofluids, respectively.		

†*Investigated nanoparticle types:* graphene oxide and graphene oxide nanosheets (GO), graphene oxide decorated with copper oxide (GO/Co_3O_4), graphene oxide (GO)/carbon nanotube (MWCNTs)/magnetite Fe_3O_4 (GMF) and shell of polyether (M2070) and graphene oxide silicon oxide hybrid nanoparticles ($GO\text{-}SiO_2$).

‡*Overall nanoparticle size*: thickness from 3-12 nm and largest dimensions up to 4 μm.

Overall nanoparticle concentration range: 0.0005-2.5% in mass and 0.025-1% in volume.

⸸*Investigated base fluids:* water (W), ethylene glycol (EG), ethylene glycol:water mixtures (EG:W at 20:80, 40:60 and 60:40 weight ratios), propylene glycol:water mixtures (PG:W at 10:90% and 30:70% in weight), epoxy resin and SAE10W40 oil.

‡*Overall temperature range:* 278-353 K.

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Table 3. Main information about rheological analysis of reduced-graphene oxide nanofluids

Type of nanoparticle†	Type of treatment	Base fluid‡	Surfactant	Nanoparticle size‡	Concentration range*	Temperature range‡	Key results/remarks	Rheology	Reference
Controlled reduced graphene oxide (crGO)	Modified Hummers' and chemical reduction methods	Deionized water	_	_	0.2-1 mg/mL	283-333 K	μ decreases linearly with the increase of T and increases with the increase of ϕ.	Newtonian behavior at higher rates	Zhang *et al.* [96]
Red wine reduced graphene oxide (W-rGO)	Modified version of Hummers' method	Distilled water	_	1.6 nm (thickness)	1-4 vol%	293-343 K	Viscosity of nanofluids with low concentrations were almost similar to its viscosity at higher concentrations	Newtonian behavior	Mehrali *et al.* [97]
Reduced graphene oxide decorated with magnetite (rGO-Fe_3O_4)	Simplified Hummers' method	Distilled water	Tannic acid	10-15 nm (average size of magnetic nanoparticles on graphene sheets)	0.5 wt%		Viscosity decreases with rise of temperature and it is higher than that of the base fluid	Newtonian behavior above the shear rate of 100 s^{-1}	Sadeghinezhad *et al.* [99]
Reduced graphene oxide rGO	Hummer's method	Distilled water	PVA	270 nm-1.5 μm (lateral size), 1.4-2.3 nm (thickness of composite sheets), 0.4-1.3nm (thickness of graphene sheets)	0.005-0.02 vol%	298-308 K	Increase in μ with the increase in concentration and the decrease in temperature. Maximum augmentation in viscosity of 4.9% at 298 K and 0.02 vol%. Viscosity ratio show nonlinear behavior with respect to concentration	-	Akhavan-Zanjani *et al.* [98]
PEG-functionalized reduced graphene oxide (rGO-PEG)	rGO-PEG was synthesized in a two-step process from GO, and GO was prepared from native graphitic flakes according to the modified Hummer's method	Water	_	_	0.025 mg/mL	295-379 K	293-333 K	_	Chen *et al.* [100]
Reduced graphene oxide (rGO),	Graphene oxide (GO) nanosheets	Deionized water	_	25-45 nm (particle size of	100 ppm	298-333 K	Non-linear increase of relative	_	Mehrali *et al.* [101]

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reduced graphene oxides decorated with silver nanoparticles (Ag-rGO)	were synthesized from graphite flakes by the modified Hummers method, and the silver decorated reduced graphene oxides (Ag-rGO) were prepared by means of the straightforward wet-chemical method			Ag)			viscosity of all fluids with temperature. Highest viscosity enhancement of 22% for rGO nanofluid within the considered temperature range. Ag-rGO nanofluids show a lower relative viscosity enhancement		
Reduced graphene oxide (rGO)	GO was prepared by a chemical method using graphite powder, and rGO was prepared by chemical reduction of GO	SAE10W40 oil	_	_	0.1 mg/mL	_	_	Shear thinning flow behavior at low shear rates <1s^{-1}	Mishra *et al.* [90]
Reduced graphene oxide (rGO)	rGO was chemically reduced from GO by using various concentrations of sodium borohydride, and GO was produced from graphite through a derived Hummers' method	Deionized water	_	_	0.0005-0.1 vol%	293-303 K	Maximum increases by 70-80 % for the non-Newtonian nanofluids. Larger increases are observed for the 0.1 vol% concentration at 303 K. Increase in μ_r by 70% in the considered concentration range. Relative viscosity is not significantly influenced by temperature. At 0.1 vol%, rGO nanofluids exhibit lower μ_r than corresponding GO NFs	Newtonian behavior for nanofluids concentrations <0.01 vol%, and shear-thinning non-Newtonian behavior for higher concentrations at shear rates between 10-1000 s^{-1}. rGO nanofluids at 0.1 vol% exhibit weaker shear-thinning behaviors compared to corresponding GO NFs	Cabaleiro *et al.* [92]

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†*Investigated nanoparticle types:* reduced graphene oxide rGO; controlled reduced graphene oxide (crGO), red wine reduced graphene oxide (W-rGO), reduced graphene oxide decorated with magnetite (rGO-Fe_3O_4), PEG-functionalized reduced graphene oxide (rGO-PEG) and reduced graphene oxides decorated with silver nanoparticles (Ag-rGO).

‡*Overall nanoparticle size*: thickness from 3-12 nm and largest dimensions up to 4 µm.

**Overall nanoparticle concentration range:* 0.5% in mass and 0.0005-4% in volume.

⸸*Investigated base fluids:* water (W) and SAE10W40 oil.

‡*Overall temperature range:* 273-379 K.

Table 4. Main information about rheological analysis of graphene-based nanofluids

Type of nanoparticle†	**Type of treatment**	**Base fluid**⸸	**Surfactant**	**Nanoparticle size**‡	**Concentration range***	**Temperature range**‡	**Key results/remarks**	**Rheology**	**Reference**
Graphene (G)	Graphite oxide (GO) was synthesized using Hummers' method	Ionic liquid	_	_	0.03 wt%	298-348 K	Viscosity lower than that of the base fluid and decreases from 217.4 to 40.6 cP as the temperature increases from 298 to 348 K	_	Wang *et al.* [102]
Graphene nanoplatelets (GnP)	Nanofluid samples are prepared by a two-step preparation method	Distilled water	_	2 µm (diameter), 2 nm (thickness), 750 m^2/g (specific surface area)	0.025-0.1 wt%	293-333 K	μ decreases between 4 and 44% with rising T and increases with GnP concentration. μ_r increases with rising T.	_	Sadeghinezhad *et al.* [104]
Graphene nanoplatelets (GnP)	_	Distilled water	_	< 2 µm (diameter), 2 nm (thickness), 300, 500, 750 m^2/g (specific surface area	0.025-0.1 wt%	293-333 K	Viscosity decreased for higher temperatures. Viscosity increased for higher concentrations of GnPs and viscosity of nanofluid improved by 44% compare to the viscosity of the base fluid	Newtonian and non-Newtonian behavior (for high concentrations)	Mehrali *et al.* [105]

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							for 0.1 wt.% of GnPs.		
Graphene (G)	The procedure for preparing G from graphite was reported in [29]	Ionic liquid [HMIM]BF4	_	_	0.03-0.06 wt%	303-488 K	μ decreases to 6.3 cP with the temperature increasing to 483 K and addition of G can slightly decrease the viscosity of the base fluid. Reduction in viscosity by 4.6%-13.1% for 0.06 wt% loading from room temperature to around 473 K	_	Liu *et al.* [103]
Graphene nanoplatelets (GnP)	_	Distilled water	_	2 µm (diameter), 2 nm (thickness), 500, 750 m^2/g (specific surface area)	0.05-0.1 wt%	293-333 K	Viscosity increases with rises in concentration and decreases with increasing temperature	_	Iranmanesh *et al.* [106]
Graphene nanoplatelets (GnP)	Nanofluids are prepared using a two-step technique	Kerosene	Oleylamine	300, 500, 750 m^2/g (specific surface area)	0.005-0.2 wt%	293-343 K	Viscosity increase by 8% at room temperature for 750 SSA, 0.2 wt% kerosene–GnP nanofluid	_	Agarwal *et al.* [71]
Nanoporous graphene	Nanoporous graphene was prepared by CVD method in catalytic Basis, and stable nanofluids were prepared through two-step procedure	Water	Tween 80	< 2-5 sheets (graphene)	0.1-0.5 wt%	293-313 K	Viscosity increase from 0.99 cp for water to 1.31 cp for nanofluid at concentration of 0.5 wt% and at 293 K. Viscosity of nanofluid with concentration of 0.1 wt% decreases from 1.15 cp at 293 K to 0.76 cp 313 K	_	Askari *et al.* [72]

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Graphene nanosheets (Gns)	Graphene nanosheets powder were dispersed through two-steps method	Hydrogenated oil	_	0.06-0.1 µm (X-Y dimensions), 0.002-0.005 µm (Z dimension)	25-100 ppm	303-323 K	Viscosity and shear stress increase up to 33% at 303 K and at the highest nanoparticle concentration	Shear thinning behavior at very low shear rates, and slight shear thickening behavior at higher shear rate	Chai *et al.* [107]
Graphene nanosheets (Gns) (alkaline graphene oxide)	Graphene nanosheets were prepared by catalytic decomposition CVD method, and the two-step method was used in the preparation of nanofluids	Deionized water	_	_	0.05-0.1 wt%	_	Viscosity increase by 11.97% at 0.1% weight fraction of graphene in water	_	Ghozatloo *et al.* [108]
Graphene nanoplatelets (GnP)	_	Distilled water	_	750 m^2/g (specific surface area)	0.025-0.1 wt%	293-333 K	Viscosity increase with increasing concentration and with decreasing temperature	Newtonian behavior	Iranmanesh *et al.* [109]
Graphene nanoplatelets (GnP)	The GnP nanofluid was prepared using a two-step method	Distilled water	_	2 µm (diameter), 2 nm (thickness), 300, 500, 750 m^2/g (specific surface area)	0.025-0.1 wt%	293-333 K	Viscosity decrease at higher temperatures by 4-44% compared with DW at a high shear rate of 500 s^{-1}	_	Mehrali *et al.* [110]

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Graphene nanosheets (GnS)	Production of graphene nanosheets from burning magnesium metal in dry ice	Glycerol	_	15-50 nm (size of few layers)	0.0025-0.02 wt%	293-333 K	Enhancement in viscosity of glycerol by 401 % was achieved by loading of 2% graphene nanosheets at shear rate 6.32 s^{-1} at 293 K.	Non-Newtonian behavior	Moghaddam *et al.* [112]
Graphene nanosheets (GnS)	_	Glycerol	_	_	_	_	The highest μ calculated using molecular dynamic (MD) simulation was 1739.95 MPa.s for Nl_{ayer}=6 (N_{layer} is the number of layers). The high level of agreement with experiment (previous work) for N_{layer}=6 is an indication of our accuracy in MD simulations. As number of layers of graphene increases, the viscosity increases	_	Moghaddam *et al.* [113]
Graphene nanoplatelets (GnP)	The GnP nanofluid was prepared by using a two-step method	Distilled water	_	2 µm (diameter), 2 nm (thickness), 500 m^2/g (specific surface area)	0.025-0.1 wt%	293-333 K	Viscosity decrease between 9% and 38% with the raising of temperature at shear rate of 500 s^{-1}	_	Sadeghinezhad *et al.* [111]
Graphene (G)	_	Water	_	_	_	_	Deborah number with relaxation and retardation times (δ_1, $\delta 3$), plays a major role in deciding the shear thinning and thickening	_	Sandeep and Malvandi [114]

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							behavior of the non-Newtonian nanofluid		
Graphene nanoplatelets (GnP)	The nanofluids were prepared via the two-step method	Water	SDBS, SDS, CTAB, GA	2 nm (thickness), 2 µm (lateral size), 300 m^2/g (specific surface area)	0.1 wt%	298-328 K	The (1-1) SDBS-GnPs sample showed the highest stability and nearly the lowest viscosity (7.4% higher than distilled water). Based on the average values of viscosity, water based 0.1 wt% GnPs nanofluids can be sequenced as (0.5-1) GA-GnPs >> pristine GnPs > (1-1) SDBS-GnPs > (1-1) SDS-GnPs > (1-1) CTAB-GnPs	Newtonian behavior for nanofluids with surfactant (all except GA). Non-Newtonian behavior for pristine GnPs nanofluids except at 308 K (Newtonian behavior). Non Newtonian behavior for (0.5-1) GA-GnPs nanofluid (shear rates 20-200 s^{-1})	Sarsam *et al.* [62]
Graphene nanoplatelets (GnP)	We used non-covalent functionalization approach to prepare stable dispersions. The graphene nanoplatelets were added directly to the EG+W mixture	Ethylene glycol:Water (EG+W at 70:30 volume ratio)	SDC (0.75 vol%)	5-10 nm (thickness), 15 µm (diameter)	0.1-0.5 vol%	303-323 K	The viscosity ratio (μ_{nf}/μ_{bf}) of nanofluids at 0.1 vol% increases from 1.06 to 1.16 and at 0.5 vol% it increases from 1.13 to 1.39	–	Selvam *et al.* [66]
Graphene nanoplatelets (GnP)	–	Ethylene glycol + Water (EG+W at 70:30 volume ratio)	SDC (0.75 vol%)	5-10 nm (thickness), 15 µm (diameter)	0.1-0.5 vol%	303-323 K	Viscosity values were in good agreement with ASHRAE standard values within ± 2%	–	Selvam *et al.* [65]

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							deviation. Maximum enhancement of viscosity ≈ 40% for 0.5 vol% nanofluid		
Graphene nanoplatelets (GnP)	Stable nanofluids were synthesized by non-covalent functionalization method	Ethylene glycol + Water (EG+W at 70:30 volume ratio)	SDC (0.75 vol%)	4-8 nm (average thickness)	0.1-0.5 vol%	303-323 K	Viscosity increases by increasing concentration, and decreases with increasing temperature	_	Selvam *et al.* [67]
Graphene (G)	Nanofluids were prepared by two-step method	Heat-transfer oil	_	0.5-2 µm (average diameter), 0.8-1.2 nm (thickness)	0.02-0.2 mg/mL	298-333 K	Viscosity decrease with the increase of temperature. Adding small amount of graphene to heat-transfer oil could reduce kinetic viscosity of pure oil, but adding too much graphene to the oil can lead to the increase of viscosity	_	Wang *et al.* [115]
Single layer graphene (SLG)	_	Water	Special dispersant	0.55-1.2 nm (thickness), 1-12 µm (diameter), 500-1200 m^2/g (SSA)	0.2-1 wt%	278-298 K	Decrease of μ with rising T and increase with increasing nanoparticle mass fraction. μ_r between GnP nanofluids and water ranges from 1.24 to 2.35.	Shear thinning effect and non-Newtonian behavior	Wang *et al.* [116]
Graphene nanoplatelets (GnP)	The two-step method has been chosen for the preparation of nanofluids	Deionized water	_	< 2 µm (diameter), 2 nm (thickness), 750 m^2/g (SSA)	0.025-0.1 wt%	293-333 K	μ increases with increasing concentration and decreasing temperature	_	Vakili *et al.* [117]

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Graphene (G)	_	Ionic liquids ([EMIM][DCA], [EMIM][TFSI])	_	1-10 layers	0.5-1 wt%	298-400 K	μ increases with increasing nanofluids concentration to 1 wt% by 151% and 269% with respect to [EMIM][DCA] and [EMIM][TFSI] respectively at 500 s^{-1} and 298 K. Decrease of μ with increasing T for ionic liquids. Constant viscosity values for nanofluids with 0.5 wt% graphene concentration, and linear increasing of viscosity for higher graphene concentrations under increasing temperature	Newtonian behavior for ionic liquids between 10^{-3} and 500 s^{-1} shear rates, and non-Newtonian for nanofluids at low shear rates and at 298 K.	Pamies *et al.* [118]
Graphene nanoplatelets (GnP)	_	Distilled water	_	2 µm (diameter), 2 nm (thickness), 750 m^2/g (SSA)	0.025-0.1 wt%	293-333 K	μ increases with concentration and decreases by between 4 and 44% with the increase of temperature in the tested range	_	Mehrali *et al.* [119]
Graphene nanosheets (GnS)	Two-step process: oxidation of graphite using the modified Hummer's method, then reduction of the graphene oxide	Water	Sulfanilic acid	5 nm-1.5 µm (dimensions)	0.01-0.5 vol%	298-343 K	μ increases with the concentration and decreases with temperature	_	Dhar *et al.* [120]

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Few-layer graphene (FLG)-		Commercial fluid, Tyfocor® LS	Either Triton X-100, Pluronic® P-123, or Gum arabic		0.05-0.5 wt%	283-323 K	μ increases with the concentration and decreases with temperature	Under shearing conditions NF stability depends on: shearing strength and time; temperature; surfactant type and content	Hamze et al. [121]

†*Investigated nanoparticle types:* graphene (G), graphene nanoplatelets (GnP), nanoporous graphene, graphene nanosheets, single-layer graphene (SLG) and few-layer graphene (FLG).

‡*Overall nanoparticle size*: thickness from 0.55-1.2 nm, largest dimensions up to 1-12 μm and specific surface areas (SSA) of 300-1200 m^2/g.

Overall nanoparticle concentration range: 0.0025-1% in mass and 0.01-0.5 % in volume.

⸸*Investigated base fluids:* water (W), ionic liquids ([HMIM]BF4, [EMIM][DCA] and [EMIM][TFSI]), kerosene, hydrogenated oil, heat transfer oil, glycerol, ethylene glycol:water (EG:W at 70:30% volume ratio) and commercial Tyfocor® LS.

‡*Overall temperature range:* 278-488 K.

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Table 5. Main information about rheological analysis of graphene quantum dots (GQD)-based nanofluids

Type of nanoparticle†	Type of treatment	Base fluid‡	Surfactant	Nanoparticle size⁑	Concentration range*	Temperature range‡	Key results/remarks	Rheology	Reference
Amine-treated graphene quantum dots (A-GQD)	GQD were initially synthesized with a novel and cost-effective exfoliation approach. To eliminate the acidity, a covalently functionalization process was employed to change GQD to A-GQD	Transformer oil	_	5-17 nm (diameter), < 1 nm (average height)	0.001 wt%	293-353 K	Maximum of enhancement of viscosity due to the loading A-GQD is < 1.3%	_	Amiri *et al.* [122]
Amine-treated graphene quantum dots (A-GQD)	Modified Hummers method was used to produce the graphite oxide sheets from natural graphite powder [35]. The synthesis procedure for GQDs was done by adapting the procedure employed by Zhang et al. (2013) with slight modification [35]	Distilled water	_	5-20 nm (diameter), < 1 nm (average height)	0.001-0.002 wt%	293-323 K	Insignificant difference between the average viscosities of water and water-based A-GQD nanofluids	Newtonian behavior	Amiri *et al.* [123]
Graphene quantum dots (GQD)	The GQDs were prepared by one-step green pyrolysis of citric acid	Glycerol	_	20 nm	0.0025-0.02 wt%	293.15-333.15 K	Reduction in μ of glycerol by 50% was achieved by loading of 2% GQDs for shear rate of 0.66 s^{-1} at 293 K and it decreases with increasing the concentration at 303.15 K	Shear-thinning behavior for GQDs-glycerol suspensions at low shear rates and temperatures and it is more obvious at greater mass fractions	Goharshadi *et al.* [124]

†*Investigated nanoparticle types:* graphene quantum dots (GQD) and amine-treated graphene quantum dots (A-GQD).

⁑*Overall nanoparticle size*: diameters in the range of 5-20 nm.

Overall nanoparticle concentration range: 0.001-0.02% in mass.

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[⸸]*Investigated base fluids:* water (W), transformer oil and glycerol.

[‡]*Overall temperature range:* 293-353 K.

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Table 6. Main information about rheological analysis of functionalized or doped graphene-based nanofluids

Type of nanoparticle†	Type of treatment	Base fluid↓	Surfactant	Nanoparticle size‡	Concentration range*	Temperature range‡	Key results/remarks	Rheology	Reference
Nitrogen doped graphene (NDG)	Heat-treatment of graphene in ammonia solution. NDG was prepared by a hydrothermal process with GO as raw material. GO was synthesized from natural graphite powder by a simplified Hummers' method	Distilled water (DW)	Triton X-100 (0.025 wt%)	3-5 nm (uniform pore size)	0.01-0.06 wt%	293-333 K	μ decreased by 51.2~51.5 % with raising temperature and it is almost the same for the lower and higher concentrations	Newtonian behavior	Mehrali *et al.* [70]
Ethylene glycol-functionalized graphene nanoplatelets (EGGnP)	Synthesized covalently	Ethylene glycol: Water (EG:W at 40:60 volume ratio)	–	–	0.01-0.2 wt%	298-338 K	μ increased with concentration and decreased with rising T	Newtonian behavior at 298 K	Amiri *et al.* [126]
Covalent and noncovalent functionalized graphene nanoplatelets (GnP-SDBS and GnP-COOH)	Regarding covalent nanofluid (GNP-SDBS-based water nanofluid), based on the technique explained by Wang et al. [33] with slight modification, carboxylation of GnP was performed.	Water	–	–	0.025-0.1 wt%	293-353 K	μ decreased with increasing T and increased with increasing concentration and it is higher for GnP-SDBS	–	Arzani *et al.* [127]

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Graphene oxide decorated with magnetite (GO/Fe_3O_4)	Hummer's method for synthesis of graphene oxide	Deionized water	–	5 nm (average size of Fe_3O_4)	0.1-1 wt%	293-313 K	μ increased with increasing concentration and with decreasing *T*	–	Askari *et al.* [125]
Functionalized graphene nanosheets (f-GnS)	f-GnS were prepared by a two-step method. The first step was to prepare GO by the Hummers method. Functionalized graphene was prepared by oxidation–reduction and functionalized with KH-570	Silicone oil	–	1.1-2.3 nm (average thickness)	0.01-0.05 wt%	293-333 K	μ decreased with increasing *T*, and the lower percentage of viscosity reached 49.95% (for 0.01% nanofluid) and 48.11% (for 0.05% sample).	Newtonian behavior	Ma *et al.* [128]
Nitrogene-doped graphene (NDG)	A simplified Hummers' method was used to synthesize graphene oxide (GO) [33] and the NDG was prepared by a hydrothermal process with GO as raw material in an ammonia solution	Distilled water (DW)	Triton X-100 (0.025 wt%)	1.8 nm (thickness)	0.01-0.06 wt%	293-333 K	μ declines with rising *T* by 51.2~51.5%	–	Mehrali *et al.* [69]

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Nitrogene-doped graphene (NDG)	NDG nanosheet was synthesized by heat treatment of graphene oxide (GO) [36] in an ammonia solution	Distilled water (DW)	Triton X-100 (0.025 wt%)	–	0.01-0.06 wt%	293-333 K	μ reduction by almost 50% as T increased from 293 to 333 K. μ increasing as a function of NDG volume concentration	–	Mehrali *et al.* [68]
Gallic acid-functionalized graphene nanoplatelets (GAGnP)	Covalent functionalization	Distilled water (DW)	–	–	0.05 vol%	293-313 K	μ decreases with increasing T and it is only slight higher than that for DW	–	Sadri *et al.* [129]
Gallic acid-functionalized graphene nanoplatelets (GAGnP)	Covalent functionalization	Distilled water (DW)	–	GnPs: 2 µm (maximum particle diameter), 750 m^2/g (SSA)	0.025-0.1 wt%	293-323 K	μ decreases with an increase in T and increases with increasing concentration. There is a small increment in the measured μ of nanofluid with an increase in concentration of GAGnPs (compared to DW))	–	Sadri *et al.* [130]
Clove-treated graphene nanoplatelets (cGnP)	Covalent functionalization	Distilled water (DW)	–	GnPs: 2 µm (lateral size), 2 nm (thickness), 750 m^2/g (SSA)	0.025-0.1 wt%	293-323 K	μ decreases with the increase of T and there is only a slight increase in μ_r with the increase of concentration	–	Sadri *et al.* [131]
Triethanolamine-treated graphene nanoplatelets (TEA-GnP)	Covalent functionalization	Distilled water	–	GnPs: 2 µm (diameter), 300, 500, 750 m^2/g (SSA)	0.025-0.1 wt%	293-303 K	μ decreases as T increases, higher than water and increases as weight concentration increases with slightly higher viscosity for higher SSA for 0.1 wt% at temperatures above 303 K	Newtonian behavior	Sarsam *et al.* [132]

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Functionalized graphene nanoplatelets (f-GnP)	GnP nanopowder is functionalized by chemical reaction process. GnP was functionalized by acid treatment method	Distilled water (DW)	–	GnP: 2 µm (maximum particle diameter), 500 m^2/g (SSA)	0.02-0.1 wt%	293-313 K	μ decreases with rising T and an increase of about 24% is noticed at 0.1 wt% (compared to DW at 313 K).	–	Yarmand *et al.* [133]
Functionalized graphene nanoplatelets-platinum (GnP-Pt)	GnP-Pt uniform nanocomposite was produced from a simple chemical reaction procedure, which included acid treatment for functionalization of GnP	Distilled water	–	GnP: 2 µm (maximum particle diameter), 500 m^2/g (SSA)	0.02-0.1 wt%	293-313 K	μ decreases with rising T and an increase of about 33% is noticed at 0.1 wt% (compared to DW at 313 K).	–	Yarmand *et al.* [134]
Functionalized graphene (f-HEG)	Hummers method followed by exfoliation and reduction by hydrogen gas and then by functionalization using acid	Ethylene glycol: Water (EG:W at 70:30 volume ratio)	–	–	0.041-0.395 vol%	283-343 K	Increasing of μ_r with increasing f-HEG loading at 303 K. Decreasing of μ with increase in T. Nearly 100% viscosity enhancement compared to the base fluid (EG+W) with f-HEG loading of 0.395 vol%	Non-Newtonian behavior that becomes more prominent with increasing GnS loading	Kole and Dey [88]

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Graphene nanoplatelets/Multi-walled carbon nanotubes (GnP/MWCNT)	Two methods are performed in the preparation the hybrid nanofluids which are the one-step and two-step methods. The current experiment performed the two-step method to prepare the working fluids	Distilled water	PVP	GnPs: 6–8 nm (thickness), 5–25 µm (diameter) MWCNTs: 15± 2 nm (average outer diameter), 1-5 µm (average length)	0.075, 0.125, 0.25wt% (MWCNTs), 0.035wt% (GnPs)	303-330 K	μ decreases by increasing temperature and a slight increase in the measured μ was observed by increasing concentration. The average μ increases in the hybrid nanofluids compared to (DW) are found to be 2.8%, 6.4%, and 10.3% for 0.075, 0.125, and 0.25 wt% MWCNTs/GNPs, respectively	_	Hussien *et al.* [64]

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Polycarboxylate chemically modified graphene nanoplatelets (f-GnP)	Nanofluids have been prepared following a two-step method	Havoline® XLC Premixed 50/50	SDBS (0.125 wt%)	5-10 graphene stacks, 2-3 nm (height per layer), >200 nm (main dimensions)	0.005, 0.05 wt%	293.15-323.15 K	Maximum increase of $\mu$of 6.6% (at 323.15 K)	Newtonian behavior in the shear rate range between 10 and 1000 s^{-1}	Sani *et al.* [63]

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Polycarboxylate chemically modified graphene nanoplatelets (f-GnP)		Nanofluids were prepared following a two-step method	Water, propylene glycol:water mixture at 30:70 wt%, propylene glycol:water mixture at 50:50 wt%	_	2-18 nm (highs per layer), up to 530 nm (long and width dimensions)	0.25-1 wt%	283.15-353.15 K	μ decreases with the rise of T by 73%, 84% and 88% for water, PG:W 30:70 and PG:W 50:50 nanofluids, respectively, and increases with increasing concentration to reach around 38%, 23% and 21% for water, PG:W 30:70 and PG:W 50:50 nanofluid sets at 1 wt%, respectively	Newtonian behavior at shear rates between 100 and 1000 s^{-1}, and non-Newtonian behavior at low shear rates 10-100 s^{-1}	Vallejo *et al.* [135]

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Sulfonic acid-functionalized graphene nanoplatelets		Nanofluids were prepared following a two-step method	Ethylene glycol:water mixture 50:50 vol%	_	up to 500 nm (dimensions)	0.25-2 wt%	283.15-353.15 K	μ decreases by around 80% for nanofluids and 82% for the base fluid with increasing temperature 70 K step. μ increases with concentration. For example, nanofluid with 0.5 wt% has a viscosity higher than the base fluid by 16% without any temperature dependence.	Non-Newtonian shear thinning behavior at low shear rates withhigher pseudoplasticity for higher nanoadditive concentrations	Vallejo *et al.* [136]
Sulfonic acid-functionalized graphene nanoplatelets		Nanofluids were prepared following a two-step method	Water	-	Up to some micrometers	0.25-1wt%	293-313 K	Viscosity decrease with the increase of temperature and the decrease of nanofluid concentration	Newtonian behavior	Agromayor *et al.* [137]

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Functionalized graphene nanoplatelets		Nanofluids were prepared following a two-step method	Propylene glycol:water mixture at (PG+W at 30:70 by wt.%)	_	11–15 nm (thickness)	0.25-1wt%	293.15-323.15 K	μdecreases between 31% and 57% with the increase of *T*. Maximum viscosity increase by 214% for the highest concentration. Constant viscosity with testing time (100 s) at shear rate between 1000 and 4000 s^{-1} for all samples at all temperatures.	Newtonian behavior	Vallejo *et al.* [138]

†*Investigated nanoparticle types:* functionalized graphene nanosheets (f-GnS) and graphene nanoplatelets functionalized using ethylene glycol (EGGnP), functionalized with SDBS (GnP-SDBS), chemically modified with polycarboxylate or sulfonic acid (f-GnP), functionalized using gallic acid (GAGnP), clove-treated (cGnP), triethanolamine-treated (TEA-GnP), functionalized with platinum (GnP-Pt) decorated with magnetite (GO/Fe_3O_4), decorated with multi-walled carbon nanotubes (GnP/MWCNT) and nitrogen doped graphene (NDG).

⁑*Overall nanoparticle size*: thickness from 1.1-2.3 nm, largest dimensions up to 5-25 μm and specific surface areas (SSA) of 300-750 m^2/g.

⁎*Overall nanoparticle concentration range:* 0.005-2% in mass and 0.041-0.395% in volume.

⸸*Investigated base fluids:* water (W), ethylene glycol:water (EG+W at 40:60%, 50:50% and 70:30% volume ratio), propylene glycol:water (PG+W at 30:70% in weigh and 70:30% volume ratio), Havoline® XLC Premixed 50/50 and silicone oil.

‡*Overall temperature range:* 283-353 K.

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Table 7. Theoretical or empirical models used in the literature to correlate or predict the viscosity of graphene-based nanofluids.

Authors	Model
Andrade's equation [146,147]	$\mu = A.\ e^{\frac{B}{T}}$ where A and B are fitting parameters, and T is absolute temperature (K)
Vogel-Fulcher-Tammann (VFT) equation [148,149]	$\mu = \mu_0.\ e^{\frac{D \cdot T_0}{T - T_0}}$ where μ_0, D, and T_0 are the fitting parameters.
Wang *et al.* [116]	$\mu = 0.004.\ (1 - \varphi)^{-77.5}\ .\ e^{\frac{1652}{T}}$ where φ is the nanoparticle mass fraction and T is fluid temperature (K) equation valid for $0.2 < \varphi < 1$ wt.% and $278.15 < T < 298.15$ K and writes as follows.
Vallejo *et al.* [95]	$\mu = \mu_0.\ e^{\frac{D.T_0}{T - T_0}} + \mathrm{E}.e^{\frac{F}{T}}.\ \phi - \mathrm{G}.\ \phi^2$ where μ_0, D, and T_0 are VFT fitted parameters for the corresponding base fluid; ϕ is the volume fraction; while E, F, and G are the adjustable parameters.
Maron and Pierce model [150]	$\frac{\mu_{nf}}{\mu_{bf}} = \left(1 - \frac{\phi}{\phi_m}\right)^{-2}$ where ϕ is the nanoparticle volume fraction and ϕ_m is the maximum packing volume fraction

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Declaration of interests

☒ 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.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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Journal Pre-proof

- Research and knowledge on graphene-NFs rheology and viscosity are comprehensively reviewed
- Influence of type of graphene derivative on rheological properties is analyzed
- Base fluid, surfactant, temperature and graphene loading effects are discussed
- Viscosity enhancements are compared and literature viscosity models revised

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