Assessment of thermophysical properties of hybrid nanoparticles (Graphene nanoplatelets (GNPs) and Cellulose nanocrystal (CNC)) in a base fluid for heat transfer applications

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Abstract

This article comprehensively investigates single (GNP) and hybrid nanofluids (GNP/CNC nanoparticles), including nanofluid preparation and thermophysical properties. Nanoparticles were characterized using FESEM and XRD analyses. A two-step approach for the preparation of nanofluids was employed, and the prepared nanofluids were determined by various analytical techniques. The thermal conductivity of nanofluids was measured in the range of 20–50 °C of the temperature using the ASTM D2717–95 norm, and the volume concentration range of the Nano-fluid in this research ranged from 0.01-0.2%. For the single GNP nanofluid, temperatures at room level indicated the thermal conductivity value in the range of 0.366-0.441 W/m-K, and for hybrid nanofluid, the thermal conductivity values are in the range of 0.501-0.551 W/m-K. In addition, the viscosity, density, and specific heat of the nanofluids are also measured and discussed. The theoretical and experimental density values come in pact with a minor error percentage increasing with the concentration of nanoparticles with a value of 1050 kg/m^3 & 1060 kg/m^3 for 0.01 % concentration of mono/hybrid nanofluids, respectively. Finally, based on the findings, it can be determined that the thermal conductivity properties of the selected nanoparticles are beneficial, and hybrid Nano-fluid is an acceptable alternative to conventional/water-based fluids in terms of thermal properties in operational systems.

Keywords: Thermal conductivity, viscosity, Graphene nanoplatelets, crystal nanocellulose, Hybrids

1. Introduction

The utilization of solid nanoscale particles distributed in the base fluid is a groundbreaking approach for enhancing the thermal functioning of heat transfer solutions. Nanofluids in this research were produced as a modern heat transfer solution by combining solid nanometer sized particles of graphene nanoplatelets/cellulose nano crystals at minimal concentrations with the base fluid (ethylene glycol: water; 60:40). Heat transfer is a concern of practical significance and prominence in the industries [1]. The potential of fluids to heat flow performs a significant responsibility in the quantity of heat loss and, in general, thermal conduction. Many industries rely on water, ethylene glycol, and oil [2, 3] kind of fluids. Considering current innovations and new technologies in certain sectors, it is important to enhance the efficiency of this type of fluids' thermal properties and ability to use them. Researchers are currently experimenting with Nano-fluids and ensuring appropriate stabilization of Nanoparticles in base fluids to enhance their heat properties [4].

Nanoparticles are distributed in a “traditional” operating fluid such as water or the anti-freeze ethylene glycol to create a efficient substitute working fluid for enhanced heat transfer called “nanofluid” [5]. Choi and Eastman [6] first proposed the term "nano-fluid" in 1995, referring to the presence of nanoparticles with diameters of 1–100 nm in base fluids. Investigators have discovered that application to a working fluid by introducing nanoparticles change its thermophysical properties dramatically in the new decade [7]. The certainly changed thermal properties of the dispersed nanoparticles in the base fluid in evaluation to the traditional fluid have resulted in some noteworthy improvements in the nanofluids thermal properties [8], such as thermal conductivity and convective efficiency of the heat transfer(CHT). Metal oxide nanoparticles, such as Al_2O_3, CuO, ZnO, and TiO_2, or carbon-based particles, such as carbon nanotubes (CNTs), graphene oxide (GO), and graphene nanoplatelets (GNPs), are examples of nanoparticles [9]. Since single/multi-wall carbon nanotubes, graphite, graphene/graphene oxide are carbon-based nanoparticles, are sometimes referred to as miraculous nanoparticles, many scientists are currently focusing on them to develop nanofluids with large-aspect-ratio nanoparticles with improved thermal, mechanical, and catalytic characteristics [10]. As all nanoparticles with carbon-base have a superior thermal conductivity, and these nanofluids have significantly enhanced thermal properties like thermal conductivity including coefficients of heat transfer. For improving heat transfer coefficient and thermal conductivity of heat exchanging fluid, the majority of preliminary research has been conducted on single/mono nanoparticles for the reason that of its unusual physical properties or thermal properties and mechanical or electrical properties [11, 12]. Graphene has fascinated a lot of consideration as a two-dimensional of carbon atoms with single layer [13]. Graphene nanoplatelets, on the other hand, (which are made up of numerous layers of graphene) bring the advantages together of monolayer property, such as the area of surface a high and great thermal conductivity, alongside of tightly packed graphitic carbon advantages, also such as strong stable nature and low budget. Due of strong Van der Waals interactions, GNPs, on the other hand, be likely to accumulate between the cause of large specific surface area [14, 15]. Below figure 1 show the important properties related to nanofluids for the thermal application obtained from the Scopus data.

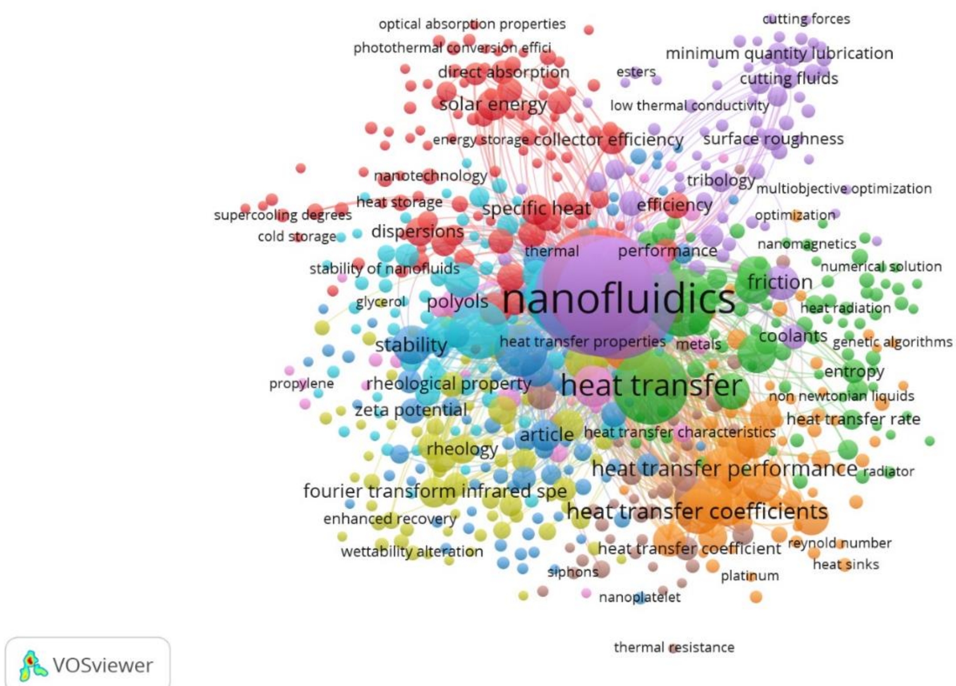

Figure 1 Bibliographic representation of accomplished properties related to nanofluids

“Nanocomposite” refers to the synthesis of at least two distinct nanoparticles into one. Sundar, Singh [16] produced MWCNT-Fe_3O_4 nanocomposite and developed a hybrid nanofluid, achieving a 29 percent increase in thermal conductivity at 0.3 percent concentration by volume in water at 60 °C. Theres Baby and Sundara [17] developed a hybrid nanofluid and observed an increase of 8% in thermal conductivity for Ag/MWCNT-HEG at a volume fraction of 0.04 percent and at 25 °C. Amiri, Shanbedi [18] studied the properties in rheological terms of MWCNT–Ag nanocomposite using both covalent and noncovalent polymerization methods and discovered that the covalent method is better for sustained thermophysical properties of nanofluid. As the Graphene nanoplatelets are hydrophobic in nature [19], the functionalization process which used to generate suspension of stable nanofluids with graphene is appropriate for nanofluids applications. With a yearly output of approximately $7.6x10^{10}$ ton, cellulose is the most abundant renewable organic substance [20]. Nanosized cellulose have recently attracted attention due to their extraordinary high specific strength and modulus, low density, chemical adaptability, renewable "green" nature, and affordable cost [21]. Few of the research has been made on finding the mechanical properties of the prepared hybrid nanocellulose fluid but there is no or limited research on hybrid cellulose for thermophysical properties for thermal application [22, 23]. Cellulose nanocrystals (CNC) are fibrillar form, with a diameter of about 5 nm and a length that varies depending on their source and fabrication process [24]. cellulose, the most common organic substance as from ecosystem, is renewable, biodegradable, biocompatibility, non-toxic, and environmentally friendly attributable to its recyclability, biodegradability, cytocompatibility, and environmental friendliness [25], has drawn increasing attention in several disciplines and could serve as a notable alternative to thermal applications. The benefits of cellulose can also be advanced by investigating its nonmetric size, which results in nanocellulose, which is regarded as a capable class of forthcoming materials expected to its remarkable physicochemical capabilities. Nano cellulose has a low density, dilatation morphology, inertness, wide surface area and aspect ratio, and is abundant and easy to bio-conjugate. Due of their unique physicochemical, mechanical, thermal, rheological, and optical properties, CNC-based nanomaterials have been widely studied. CNC could provide acceptable features to hybridization or nanocomposites (metallic, ceramics, and polymeric) however at low concentrations for a wide range of applications. Fullerenes, carbon nanotube (single-walled, double-walled, few-walled, or multi-walled), nano diamonds, as well as graphene-based materials like graphene, oxide form of graphene, reduced form of graphene oxide, and graphene quantum dots have evolved into a new category of hybrid materials with a synergetic effect or synergetic effect in a variety of applications. Despite the fact that several potentially possible techniques to produce effective Graphene nanoplatelets are now being developed, there are still several practical difficulties to overcome. GNPs, for example, are further normally generated from aqueous dispersals, although they can effortlessly agglomerate. This type of agglomeration can limit surface area and have a detrimental impact on properties. As a result, the addition of CNC not only overcomes this disadvantage due to its exceptional disseminative properties, but similarly converses additional assistances to the resulting GNP/CNC hybrids, such as quick dispersion and thermal stability, as well as improved adsorption capability, photothermal interaction, sustainability, intrinsic luminosity and diffraction, optical transparency, and thermal conductivity. Considering these facts, it is clear that using CNC as a companion material in GNP nanoparticles could be more effective and beneficial in improving the nanocomposite's thermal conductivity as well as thermal properties. We detail the preparation, and thermal properties of GNPs/CNC hybrid fluids in this study. This paper presents a forward-considering perspective on GNPs/CNC hybrids for a variety of applications. Nonetheless, the progression of

GNPCNC hybrid-based nanomaterials is a comparatively innovative belief that is largely restricted to scholarly disciplines. However, it is expected that several hybrid nanofluids (graphene based) research will become more attractive in the potential, attracting more study consideration not only in several functions but additionally in achieving multifunctional systems and opening new perceptions. Furthermore, the sensible implementation of such hybrids as next-group materials necessitates significant functional and performance enhancements. The present study focuses on the comparison of nanofluids thermophysical properties with single and hybrid Graphene based nanofluid. As there is no data available in the literature for the novel work as this kind on the thermophysical properties assessment of hybrid nanoparticles including the Graphene nanoplatelets and cellulose nano crystals in a base fluid of ethylene glycol and water at a ratio of 60:40.

2. Methodology

This research process offers comprehensive details about the analysis, the materials as well as equipment utilized for the characterization of nanofluids (Water & Ethylene glycol -based GNPs / CNC), nanoparticles of Single/ Hybrid and accompanied by an examination on stability.

2.1 Materials

In this investigation, graphene nanoplatelets (GNPs) with 800 m^2/g specific surface area (S.A) were employed, which were purchased from Nanografi nanotechnology (USA) with 99.9% purity, 3nm Size, and1.5 m in diameter, and crystalline nanocellulose from the country Malaysia by MY Biomass Sdn. Bhd. CNC remained challenging to separate in powder type from the produced pulp because of its hydrophilic character. A spray-drying approach with a tiny fan was employed for CNC handling in the form of powder. When the pulp or suspensions reached into connection with heated air from the nozzle spray dryer's entering space, the moisture quickly evaporated, resulting in steady CNCs flake. The flakes of CNC are collected and ground into powder. The specific parameters of the obtained CNC nanoparticles crystallinity index with 80%, 100-150nm crystal length, 9-14nm crystals diameter, and the hydrodynamic diameter is 150nm.

2.2 Preparation of nanofluid

At a concentration by volume as 0.01%, 0.05%, 0.1%, & 0.2%, the graphene nanoplatelets are weighed by means of the Internal Sartorius Analytical Balance (Model : BSA24S-CW) and were scattered in the Ethylene glycol-distilled water which is at a ratio of 60:40 by using a magnetic stirrer with rotating magnetic probe (Thermo-fisher, USA) and is was allowed to stir for about 2 hours and later the probe of the ultrasonication (CE ISO Ultrasonic Homogenizer Sonicator Processor Cell Disruptor Mixer 20-1000mL) having an productivity control over power of 950 W and a frequency choice as 20kHZ supply of power with a φ13mm diameter probe. By using Eq (1), the density of hybrid nanoparticles is calculated.

$$\rho_{GNP/CNC} = \frac{\phi_{GNP}\rho_{GNP} + \phi_{CNC}\rho_{CNC}}{\phi_{total}} \tag{1}$$

Where, GNP denoted Graphene Nano Platelets, CNC denote Cellulose Nano Crystal, ‘ϕ’ denotes the volume concentration of nanoparticles in nanofluids and ‘ρ’ denotes density respectively.

In the absence of a surfactant, since nanoparticles of carbon-based having a hydrophobic nature, they cannot be sustainably distributed in base fluid. Graphene nanoplatelets, as a result of their electrical and thermal conductions, are graphite form. The GNPs are recommended that they can be scattered in medium with stirrer & sonication through a probe without utilizing surfactants. Therefore, 5 hours of ultrasonication time was used to make the particles properly disperse and stable with a power utilization of 50%. Likewise, preparation of hybrid nanofluid contains the particles GNPs / CNCs at 1:1 ratio is disseminated in the base fluid Ethylene Glycol-distilled water (60:40) with a magnetic stirrer (Thermo-fisher, USA). This high-speed stirrer operated at a range of 400-500 rpm until proper blending/mixing for about 120-180 minutes and altered for every 15 mins followed by ultrasonication procedure with a probe for 5 hours with a power output of 50% with interval gap of 5 mins after every 15 minutes of sonication process to maintain the temperature of the fluid. This break avoids the nanofluid to heat up and losing the properties of particles, this process followed for single nanoparticle dispersion as well. For hybrid nanoparticles, the weight of nanoparticles was validated using Eq (2).

$$W_{G\text{-}CNC} = \left(\frac{\varphi}{100-\varphi}\right) \times \left(\frac{\rho_{(GNP/CNC)}}{\rho_{(bf)}}\right) W_{bf} \qquad (2)$$

Where 'W' is weight of hybrid nanoparticles 'φ' implies the concentration of single/hybrid nanofluids by volume, 'w' stands for weight and 'ρ' defines the density. The subscripts 'GNP' denotes Graphene nanoplatelets, 'CNC' is cellulose nanocrystal, 'bf' represents base fluid, respectively. Figure 2 below provides a schematic illustration of the development of nanofluid.

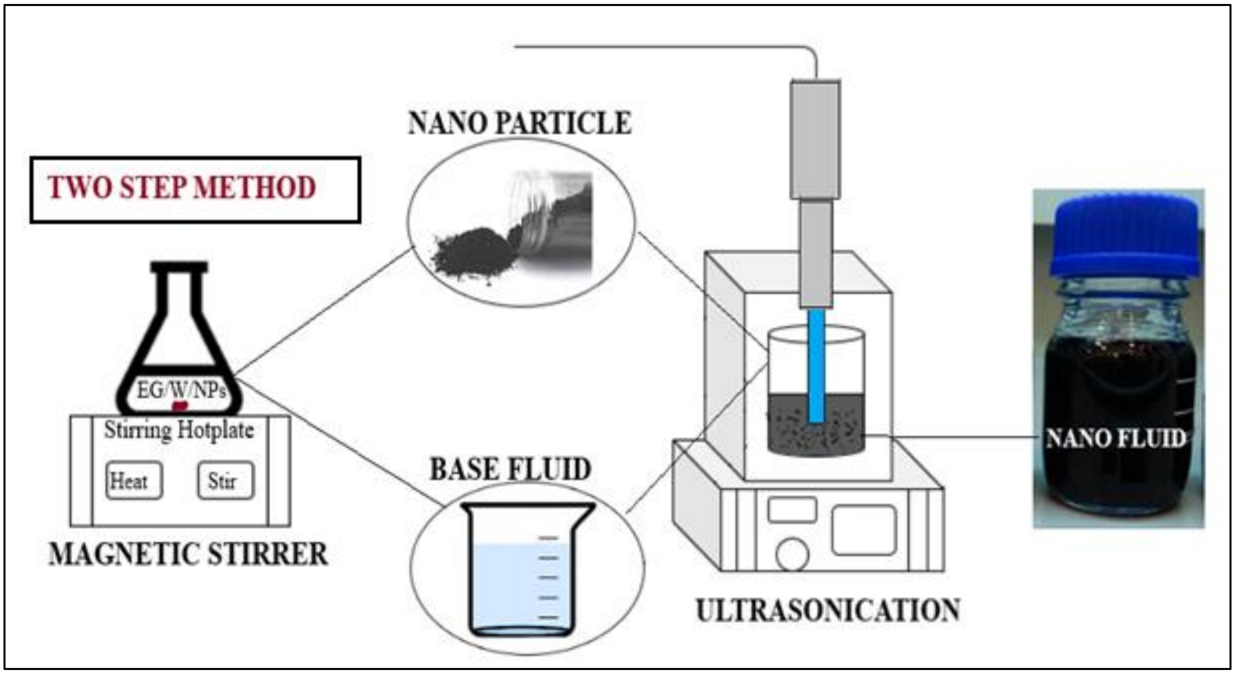

Figure 2 Two-step method preparation method representation [26].

2.3 Measurement devices

2.3.1 Evaluation of stability

The clustered nanoparticles get agglomerated and interrupt the hybrid nanofluids stability due to their large surface area, which is a critical condition for their utilization. The GNP/CNC nanoparticles stability and dispersibility in the nanofluids were evaluated applying the method of sedimentation with photographs captured at different periods, and by using UV–Vis spectroscopy, and Zeta potential analysis. The spectrum was obtained using PerkinElmer's LAMBDATM UV/Vis with operational array of UV-spectrometer wavelengths of 200 nm–800 nm and specific cuvettes (quartz) appropriate for measuring light absorption for all the samples. For proper light transmission, all the samples with base fluid are diluted. The single / hybrid nanofluids Zeta potential is determined using the Anton Paar light sizer 500. In nanofluid dispersion, the measurement of zeta potential displays the repulsion degree between nearby particles with the same charge.

2.3.2 Characterization

The characterization of nanoparticles microstructure in the nanofluids is done using a transmission electron microscope (TEM). The size of the particle and dispersion of W/EG developed GNPs and hybrid nanofluids of GNPs were measured using a digital TEM. Before TEM examination, the samples of the hybrid nanofluids are sonicated for 15 minutes. The TEM apparatus (Tecnai G2 20 S-TWIN, USA) with 210KV of accelerating voltage evaluated the solution of nanofluid constituted of GNPs and CNC of the nano-base fluid. GNPs and CNC nanofluids were analyzed using an X-ray diffractometer (Rigaku D/MAX-2500PC, Japan) with Cu K α radiation (λ= 1.54056 Å) at 40 KV and 30 mA, with 0.02/s rate of scan. The nanoparticle's phase was assessed using X-ray Diffraction (XRD) analysis. The produced nanofluid trials are coated to assess the superficial morphology for microstructure characterization. SEM scanning electron microscopy (HITACHI/TM 3030 PLUS, Czech Republic) was used to examine the dispersion of nanoparticles in the fluid. Field emission scanning electron microscopy (FESEM, Zeiss Sigma HD VP, Germany) was used to examine the structure of developed filaments at 0.5 kV acceleration voltage. Prior to observation, each sample has platinum sputtered. The samples were morphologically inspected before being seen using a FESEM scope for capturing the topographical representations of the powder as received [27, 28].

2.3.2.1 Thermal conductivity measurement

Various strategies for evaluating the thermal conductivity of nanofluids have been proposed in recent years. Transients hot-wire is the highly accurate and quick of all these approaches (THW). In this research, for the measurement of thermal conductivity a hot wire-type KD2-Pro (Decagon devices Inc., USA) is used for GNPs/ base fluid(W/EG), GNP-CNC/based hybrid nanofluid is established. The below Table 1 gives the list of studies that indicates the thermal conductivity estimates obtained by authors at distinct volume concentrations. These values of thermal conductivity are used to validate with the thermal conductivity estimates attained in the present study at different temperatures and volume concentrations.

Table 1: Nanofluids Thermal Conductivity Enhancement Summary.

NP's	**Conc-Wt (%)**	**Surfactant**	**Thermal Conductivity(W/m-K)**				**References**
			30 ˚C	**40 ˚C**	**50 ˚C**	**60 ˚C**	
Graphene nanoplatelets	0.1	SDS	0.559	0.618	N/A		[29]
		CTAB	0.635	0.648			
		SDBS	0.64	0.66			
		Gum Arabic	0.645	0.676			
Graphene nanoplatelets	0.01	-	0.31	0.34	0.36	0.37	[30]
	0.05		0.38	0.4	0.41	0.42	
	0.1		0.43	0.44	0.45	0.46	
	0.2		0.46	0.48	0.5	0.52	
Graphene	0.05	-	1.02	1.019	1.03	N/A	[31]
	0.08		1.052	1.066	1.078		

Graphene oxide	0.1	SDS	0.63	0.65	N/A		[32]
		TX-100	0.62	0.64			
Graphene	0.124	Not used	0.315	0.318	0.319	0.325	[33]
	0.207		0.324	0.327	0.33	0.339	
	0.395		0.335	0.339	0.342	0.345	
Graphene nanoplatelets	0.02	Gum acacia	0.63	0.66	N/A		[34]
	0.1		0.72	0.77			
Carboxyl graphene	0.04	SDS	-	0.383	0.385	-	[35]
Graphene nanoparticles (750m2/g)	0.024	Not used	0.68	0.71	N/A		[36]
	0.05		0.71	0.75			
	0.1		0.75	0.8			
Graphene NP-Ag	0.2	Not used/Acid treatment	0.63	0.651	N/A		[37]
	1.0		0.72	0.77			
Graphene nano-platelets	0.1	NPE 400 (ionic)	0.5	0.51	0.525	N/A	[38]
	0.2		0.54	0.55	0.565		
	0.3		0.62	0.64	0.66		
Graphene nanoplatelets	0.01	Gum Arabic	0.63	0.64	0.657	0.663	[39]
	0.05		0.64	0.642	0.67	0.682	
	0.1		0.641	0.68	0.7	0.712	
Graphene nanoplatelets	0.1	Not used	0.187	0.18	0.179	0.17	[40]
	0.25		0.20	0.20	0.199	0.19	

	0.5		0.215	0.213	0.21	0.209	
Graphene nanoparticles	0.25	SDBS	0.40	0.405	0.419	0.42	[41]
	0.5		0.41	0.415	0.421	0.43	
	1.0		0.42	0.425	0.435	0.44	

Table 2: Physical form of Properties of Nanoparticles. (Nanografi nanotechnology (USA), MY Biomass Sdn. Bhd (MALAYSIA))

Properties	GNP	CNC
Color	Black	White (dry powder)
Purity	99.9%	-
Density (kg/m^3)	2267	1050
Structure	Platelet shaped sheets	Crystalline form
Specific surface area (m^2/g)	800	-

Table 3: Thermophysical Properties at 20 °C temperature of base fluid [42, 43].

Properties	Water	Ethylene glycol
Chemical formula	H_2O	$C_2H_6O_2$
Vapor pressure (kPa)	3.169	0.007
Molar mass (g/mol)	18.0153	62.07
Density (kg/m^3)	1000	1100

A temperature bath (WNB7-MEMMERT, Germany) is used to sustain and monitor the thermal conductivity measurement by the temperature control. Probe vibration must be regulated to minimize experimental errors. To position vertically the KS-1 probe in the middle point of the sample vial, a horizontal support was mounted adjacent to the temperature bath. To examine the reproducibility of the data, the measurements were repeated twenty times in all planned volume concentrations and temperatures with a 5-minute intervening period. The Table 2 shows the physical properties of selected Graphene nanoplatelets and CNC nanoparticles, and Table 3 gives the information about the thermophysical Properties of base fluid water and ethylene glycol at 20 °C temperature. Table 4 presented few specifications of thermal conductivity measuring device KD2 Pro information.

Table 4: Specifications of thermal conductivity measurement device (KD2 Pro).

Accuracy ±5%	**Thermal conductivity**
Range of operation	0–50 °C
Range of measurement	0.02–2 W/m K
KS-1 Sensor	Needle length: 60 mm
	Needle diameter: 1.3 mm

2.3 Viscosity

Rheometer was used to assess the viscosity of all nanofluids in the range of 20 to 50 °C temperature at a constant shear rate (Brookfield DV-I prime viscometer) with varying volume concentrations. A circulating water jacket is connected to an RST coaxial cylinder rheometer to assess the temperature range and other uses. The rheometer can measure viscosities from 0.0001 to 5.4x106 Pa. s and temperatures from -200 to +180 °C. Experiment was carried out in a steady-state environment. Rotational measurement with a controlled shear rate was used as the method of measurement. To authenticate the rheometer, the base fluids viscosity was quantified, and the results were assessed to ASHRAE standard data. The viscosity is measured with 15.7 mL of fluid, and the results are compiled in a computer connected to an RST rheometer. To reduce the experimental error, five precision readings were acquired and averaged. Previously, several researchers used the Brookfield rheometer to determine viscosity [44-46]

2.4 Density & Specific heat measurement

The pumping power, friction factor, Reynolds number, and other properties of nanofluids are all affected by density. In this work, a digital density meter was employed to test the density of GNPs &GNP/CNC nanofluids with varying volume concentrations, similar to prior investigations by various researchers. The density meter used here is a KEM (model DA-640) from Kem Kyoto Electronics Co. Ltd. The density (gm/cm3) measuring range on this meter is 0.0000-3.000, with a ±0.0001gm/cm^3 precision along with repeatability of 0.00005 density (gm/cm^3). The temperature range for utilizing this meter is upto 35 degrees Celsius, with a humidity level of 85 percent RH or less. The density is measured using an ASTM D4052-18 digital density meter, which is recognized as a standard test method for density, relative density, and API gravity of liquids[47-49]. Differential Scanning Calorimetry (DSC) is a sensitive method for determining the specific heat capacity of viscoelastic fluids. PerkinElmer, Inc.'s DSC (model DSC 8000) was utilized to measure the specific heat in this study. The specific heat capacity of base fluid and GNP’s/CNC nano fluids was examined at room temperatures. The measurement solution was placed in an aluminum pan and weighed on an electrical balance with precision: 0.0001 before being covered with an aluminum lid and sealed with a universal crimper press. An empty pan filled with sapphire reference were placed in DSC before the actual sample measurement to get baseline and reference data. Following that, the sample pan was put in DSC beside an empty pan as a control. Following the standard DSC test procedure ASTM-E1269. the temperature range was set with a 100C/min temperature difference. For each sample, a minimum of 6 minutes was required. This test was carried out for all nanofluid and base fluid volume concentrations. The generated values are saved on a computer that is linked to DSC. Many previous studies employed DSC to conduct precise heat measurement tests on nanofluids[50-53].

3 Results and Discussions

Nanofluid preparation, characterization, and stability

The preparation method used is two step method for single graphene nanoplatelets, and hybrid nanoparticles dispersal. In the Faculty of Mechanical Engineering's Advanced Automotive Liquid Lab (A2LL) at University Malaysia Pahang, the needed graphene nanoplatelets & nanocellulose hybrid nanofluid was prepared successfully. Over a 5-hour ultrasonication duration followed by magnetic stirring, ultrasonication is the most influential way for generating very balanced dispersion of GNPs and hybrid nanoparticles. Figure 4 displays diffraction peaks for the CNC and graphene refraction planes, respectively, at 2θ =15.7°, 22.8°, 34.6° and 26.3°, 43.9°, 54.1°. The peak in graphene at 2θ=26.35° reflected a typical graphitic carbon diffraction pattern [37, 54, 55]. Furthermore, the connected carbon in cellulosic form was demonstrated by a negatively diffracted signal at 22.8375°. Further shows that the CNC peak intensity is higher to that of the peak of the graphene. The FESEM images for GNP and CNC are shown in Figure 3(a & b). A consistent dendrite forms uneven structure noticed for GNPs with platelet structure and CNC with porous microstructure with homogeneity and uniformity. TEM examination of CNC and GNP nanoparticle morphology and dispersion depicts in Figure 3 (c & d). It shows clearly distributed GNPs together with a CNC base due to the transparency. The images show that as the concentration of nanoparticles increased, resulting in a reduction in clarity, suggesting agglomeration. The structure of cellulose nanocrystals and the dispersion of graphene nanoplatelets in the base fluid (EG/W) is investigated using microstructure TEM analysis. Graphene platelet structure and CNC with a clear and gentle exterior in the base fluid, displaying the fragile structure behaviour. Finally, the morphology of the dispersed GNPs and CNC reveals that the nanoparticles were well prepared and dispersed in the ethylene glycol and water base fluid. The information more related to the preparation of the nanofluid in detail can be found in the previous article by authors related to preparation, characterization and permanence (stability) of the single and hybrid nano fluid that is prepared [56].

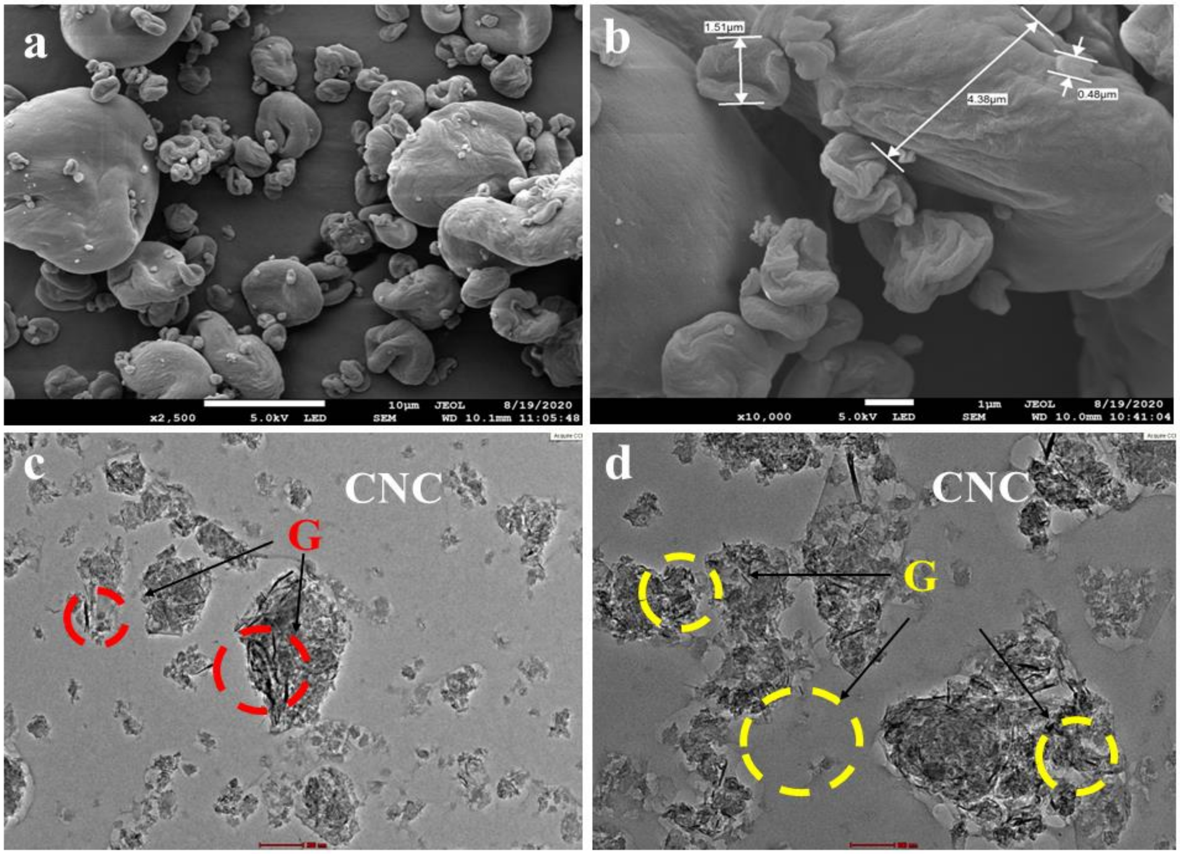

Figure 3 Images of FESEM of (a) GNP's-Graphene nanoplatelets with CNC hybrid nanofluid at 2500x (b) at 10000x magnification. (c) TEM images of hybrid nanofluids 0.2% GNP/CNC at lower enlargement, (d) at higher magnifications.

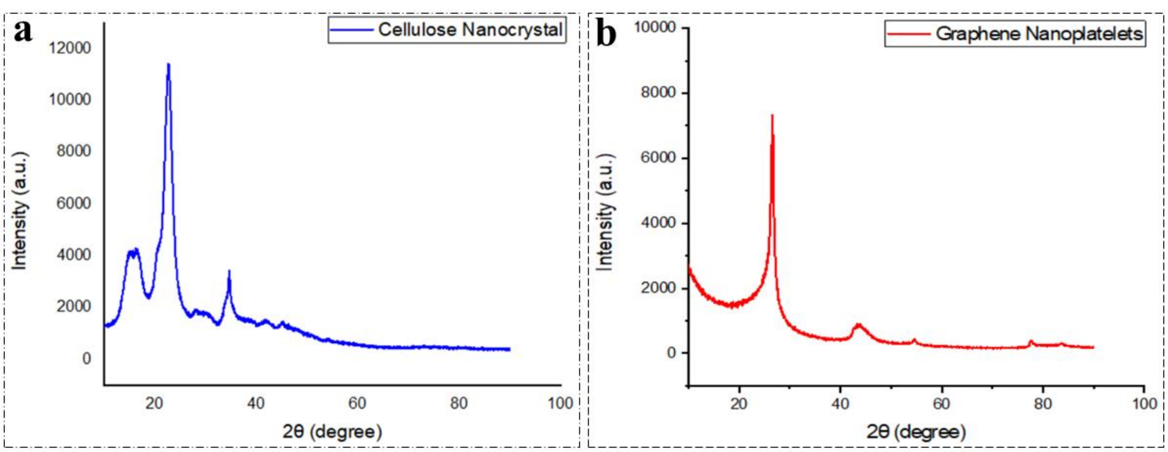

Figure 4 Analysis of XRD (a) CNC and (b) Nanoparticles of Graphene nanoplatelets.

3.3 Thermal conductivity

Thermal conductivity was measured by using the KD2 Pro thermal properties analyzer in the temperature range of 20–50 °C. Validation is a process of calculating the parameters in any laboratory work, for this the instrument must be adjusted. To calibrate the unit, the KD2 Pro Manufacturer recommends using a standard sample of glycerin. The accuracy of the measurement device must be tested as a condition before calculating the final thermal conductivity tests of nano-fluid. Besides the measured data for base fluid was compared with the data presented by different authors [29, 57]. As expected, the previous research suggests that the thermal conductivity value increases as the temperature increases, with a maximum inaccuracy below 10 percent, the KD2 Pro over/ underestimated the recorded values of thermal conductivity. The effect of temperature and the concentration based on volume for the thermal conductivity of graphene and hybrid GNPs/CNC nanofluids has been extensively investigated. The different volume percentages have variable thermal conductivity. GNP/CNC hybrid nanofluid samples are tested at temperatures ranging from 20 to 50 degrees Celsius as shown in Figure 5. The thermal conductivity of graphene nanofluids is shown in Fig. as a function of concentration in the range of 0.01–0.2 vol. percent at various temperatures. To avoid an increase in effective viscosity and sedimentation, low weight percentages are chosen. The thermal conductivity increases as the concentration of Graphene increases, which is to be expected. At a concentration of 0.01 % the thermal conductivity value is 0.3716 W/m-K for graphene nanoplatelets nanofluid at 20°c. At a concentration of 0.2 percent, the maximum enhancement was 27 percent with 0.4411 W/m-K at 50 °c. At the same temperature, from image contrasts the enhancement of thermal conductivity with concentrations of graphene and hybrid Graphene nanofluids. It is clear that the rate of enhancement increases with concentration of graphene and Cellulose nano crystals in comparable to metallic and ceramic nanofluids and is much superior to them. Temperature and volume concentration significantly increase the thermal conductivity of graphene nanofluids.

This is due to the fact that graphene nanofluids contain particles of varying sizes. In accordance with percolation theory, the larger particles contribute to the formation of a network-like chain structure. Brownian motion is contributed by the smaller particles, which travel spontaneously. As the temperature rises, Brownian motion creates micro convection, which provides thermal conductivity to increase. This has led to the strong suggestion of a hybrid character for thermal conduction in graphene nanofluids comprising micro convection and diffusion phenomena. With increases in both the weight proportion and the temperature, the rise in thermal conductivity is nonlinear. The nonlinearity/linearity of the variability of thermal conductivity with respect to weight fractions is influenced by the characteristics of the hybrid nanoparticle and even the base fluid. The increase in thermal conductivity is 14.91 percent at 20 °C and about 17.77 percent at 40 °C when using a 0.01 percent weight concentration of GNP–CNC nanofluid. The high thermal conductivity of GNP and CNC nanoparticles results in an increase in effective thermal conductivity. The spacing amongst nanoparticles (unrestricted passage) reduces as the volume fraction of nanoparticles increases. It occurs as a result of the percolation effect.

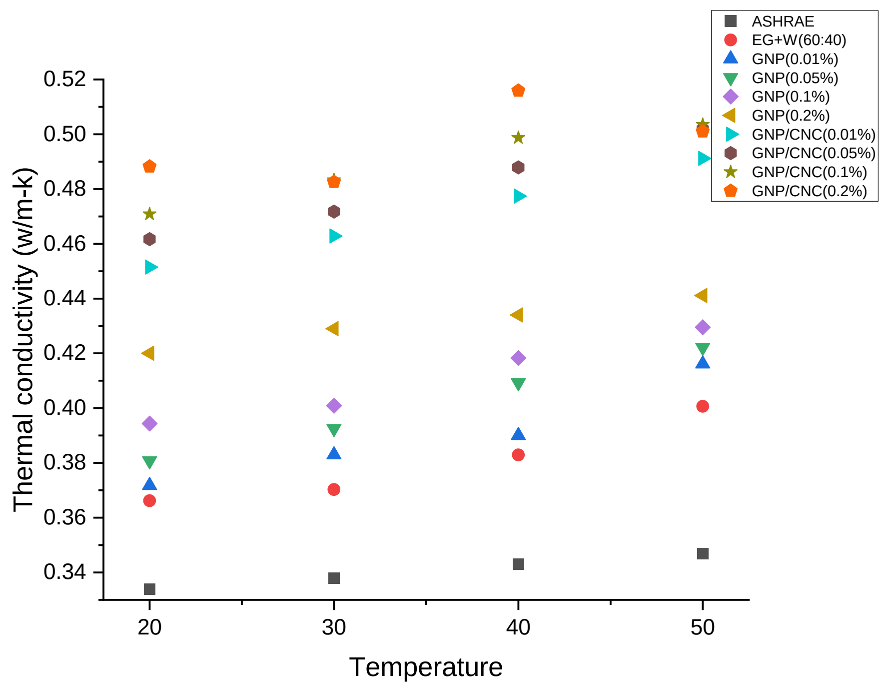

Figure 5: Thermal conductivity of GNP & GNP/CNC nanofluids at different concentrations and temperature

Other studies have also seen an increase in thermal conductivity of carbon-based nanofluids as the weight concentration increases.[58, 59]. To explain the reason for thermal conductivity of nanofluids has increased so dramatically, Nanoparticles move in a Brownian approach, and the liquid at the liquid/particle contact layers at the molecular level, the nature of heat transmission to the nanoparticles, and the impact of nanoparticle clustering are some of the hypothesized mechanisms [60]. They reached the conclusion that Brownian motion can be ignored because thermal diffusion has a greater influence than Brownian diffusion though it is the measure of immobile nanofluids. Although many contributing factors have been examined, such as the liquid–solid interfacial region, Brownian motion, charge carrier status, and ballistic dielectric transport, no overarching mechanism to govern the exceptional behaviour patterns of nanofluids, including that of the significantly improved effective thermal conductivity, has been discovered.

Similar to the graphene nanofluid thermal conductivity there is an increase in the hybrid GNP/CNC hybrid nanofluids with increase in volume concentration from 0.01 % to 0.2%. At 40 °C for 0.2% the thermal conductivity value is recorded as 0.465 W/m-K. At same volumetric concentration and temperature in comparison with single and hybrid nanofluid there is an increase of 5.2 % and 13.3 % with respect to base fluid. Below table 5 gives the validation of present study by comparing it with the previous studies based on graphene nanoparticles and hybrid nanoparticles. The present study base fluid experimental values at 60:40 EG:W ratio, agrees well with the author Sundar, Singh [61] at same base fluid ratio at the temperatures varying from 20 to 50°C. The thermal conductivity values are compared at around equal concentrations and temperature to give a clearer vision of the present study.

Table 5: Thermal conductivity of single and hybrid nanofluids attained by various researchers.

Nanoparticle	**Concentration/ Temperature**	**k_{NF}/k_{BF}**	**References**
Graphene nanoplatelets/EG-W	φ = 0.01% vol./50 °C.	1.038	Present study
	φ = 0.05% vol./50 °C.	1.053	Present study
	φ = 0.1% vol/50 °C.	1.071	Present study
	φ = 0.2% vol./50 °C.	1.100	Present study
Graphene nanoplatelets-CNC/EG-W	φ = 0.01% vol./50 °C.	1.225	Present study
	φ = 0.05% vol./50 °C.	1.252	Present study
	φ = 0.1% vol/50 °C.	1.256	Present study
	φ = 0.2% vol./50 °C.	1.250	Present study
3D-Graphene/EG	φ = 0.1% wt/25 °C.	1.149	Bing, Yang [62]
Graphene/DIW	φ = 0.1% wt. /25 oC	1.416	Ghozatloo, Rashidi [63]

Nanoparticle	Concentration/ Temperature	k_{NF}/k_{BF}	References
Graphene nanoplatelets/EG	φ = 0.5% vol./ 35 oC	1.208	Selvam, Lal [64]
		1.160	
	φ = 0.1% wt./60 °C (500 m2/g GNPs)	1.287	Iranmanesh, Mehrali [65]
	φ = 0.1% wt./60 °C (750 m2/g GNPs).	1.307	
Graphene/EG/DIW	φ = 0.2% wt./25 °C.	1.092	Contreras, Oliveira [66]
Graphene nanoplatelets	φ = 1% wt./25 °C (750 m2/g)	1.211	Wang, Wu [67]
Hybrid-Graphene wrapped MWNT			
TiO_2/Graphene/W	φ = 0.25% vol./25 °C.	1.098	Bakhtiari, Kamkari [68]
	φ = 0.25% vol./55 °C.	1.138	
Al_2O_3/Graphene oxide/W	φ = 0.25% vol./50 °C.	1.125	Taherialekouhi, Rasouli [69]
Fe-Si/DW	φ = 0.25wt %/50 °C.	1.109	Huminic, Huminic [70]
Graphene oxide/Co_3O_4/W	φ = 0.2wt %/50 °C.	1.156	Sundar, Singh [61]
Graphene oxide/Co_3O_4/EG	φ = 0.2vol %/50 °C.	1.113	Sundar, Singh [61]
Graphene oxide/Co_3O_4/EG/W	φ = 0.2vol %/50 °C.	1.120	Sundar, Singh [61]
Graphene oxide-CuO/EG-W	φ = 0.2vol %/50 °C	1.094	Rostami, Nadooshan [71]
Graphene nanoplatelets-platinum/DW	φ = 0.1vol %/40 °C	1.174	Yarmand, Gharehkhani [27]

3.4 Viscosity

The viscosity of EG/ distilled water (base fluid) at a ratio of 60:40 and GNP/CNC hybrid nanofluids at varying volume concentrations and temperatures ranging from 20 to 50 °C is shown in Figure 6. The viscosity has adverse effects on two factors for pressure drop and pumping power constraints, similar to density. Because of NPs/surface collisions and other inter-layer resistance and interfacial forces, the presence of Nano Particles in the Base fluid, i.e., constituting to the Nanofluid it increases friction at the fluid/surface contact. At 20 °C, the measured viscosity of the base fluid (EG/Water) is 5.485 (mPa-s), which is consistent with literature values. Since the increasing concentration has a direct effect on the fluid internal shear rate, the viscosity of nanofluids rises as the volume fraction of nanofluids rise [72]. The viscosity reduces as the temperature rises, as the intermolecular and interparticle adhesion forces weaken. When 0.2 percent volume concentration of GNP nanofluid is compared to the viscosity of EG/Water at 20 °C, the viscosity increases by around 21%. Similarly, there is an increase in viscosity by 24.5 % at 0.2 volume concentration of hybrid nanofluid (GNP/CNC) at 20°C. The viscosity values diminish as the temperature rises. The increased viscosity value at 0.2 percent volume concentration of GNP nanofluid at 50°C is only 14.7% as compared with base fluid and hybrid nanofluid of GNP/CNC at 0.2% volume concentration at 50°C is 18.3%. The GNP/CNC sample had the highest stability and caused the greatest increase in average viscosity of the base fluid. High colloidal stability and the lowest rise in base fluid viscosity are two of the most important factors to consider when using nanofluids as operating fluids in the applications of heat transfer. Accordingly, by the viscosity values the highest concentration of nanoparticles (single/hybrid) can be considered to be effective.

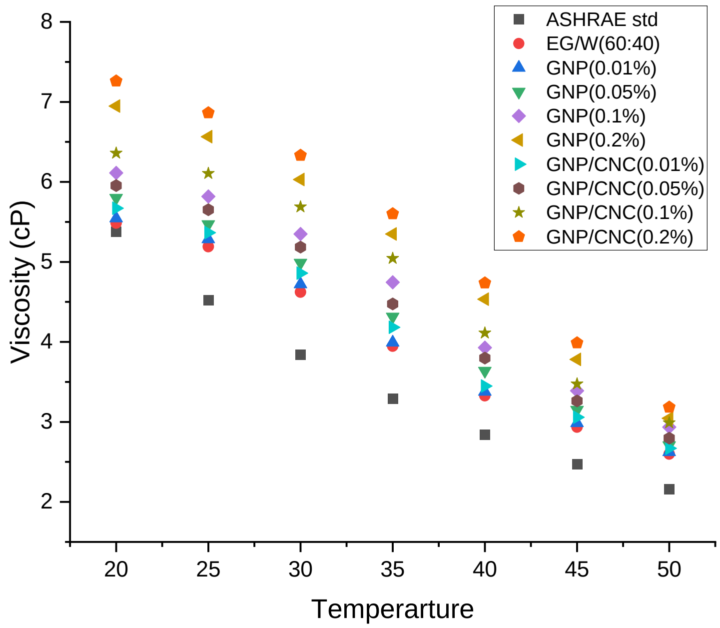

Figure 6: Viscosity of prepared nanofluids at different concentrations and temperature

Because a huge amount of nanomaterial has been disseminated, the friction factor appears to be high at high volume concentrations. The friction factor, literally, improves the value of dynamic viscosity. However, as the temperature of the nanofluid rises, the intermolecular adhesion force weakens, resulting in a lower dynamic viscosity value [73]. Figure 7 depicts the viscosity ratio of 60:40 (EG:W)-based fluids, as well as from author Sundar, Singh [61] data for 60:40 (EG: W) based fluids for the comparison of the study. The viscosity of the 60:40 (EG: W) fluid is found to be nearly identical throughout a wide range of temperatures.

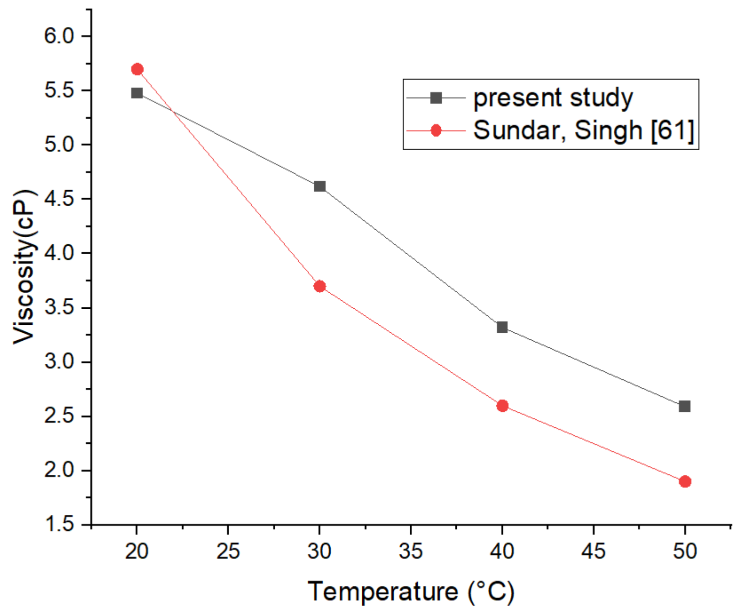

Figure 7: Viscosity comparison of prepared base fluid at different temperatures

3.5 Density

The volume concentration of nano particles and the distilled water with ethylene glycol base fluid equals the density of nanofluids. The base fluid has an impact on the nanofluids density. The density of nanofluids is also affected by temperature. The density of nanofluids drops as the temperature rises. Figure 8 shows the density of nanofluids determined at 20°C for the base fluid and varying volumetric concentrations of GNPs & GNP/CNC nanofluids. The result of density of base fluid is in good quality agreement with ASHRAE data and the deviation is below 1%. Table 2 shows theoretical density values for a range of fluids. The difference between experimental and theoretical density data is less than 1.0 percent, indicating that the two types of density values of base fluid and nanofluids are in good agreement. The density measurements of the nanofluids are compared to the projected values using Eq. (3), with the density of GNP, CNC, and base fluid being 1065 kg/m^3 ,1072 kg/m^3 and 1060 kg/m^3, respectively. The density of the nanofluid changed in direct proportion to the nanoparticle concentration in comparison to the base fluid.

$$\rho_{nf} = \phi\rho_s + (1 - \phi)\rho_f \quad (3)$$

Where ‘ρ’ denotes the density, is ‘φ’ volume concentration, the subscripts ‘nf’ is nanofluid, ‘s’ is solid nanoparticles, ‘f’ is the base fluid (W/EG)

According to the molecular dynamic simulation principle, the nanoparticles are filled with the molecules of the base fluid in various ways. In the case of nanofluids, increased Vander wall interaction causes non-uniform density to change in the interfacial region being the disparity in reported data. The density value is decreased for hybrid nanofluids (GNP/CNC) when compared with single nanofluid (GNP). The density value of Graphene nanoplatelets at 0.2 % volume fraction is 1304.2 Kg/m^3 & at same volume fraction for hybrid nanofluid of graphene nanoplatelets/cellulose nanocrystals (GNP/CNC) is 1182.32 Kg/m^3 respectively. It clearly shows density value increases with volume concentration. The density of base fluid (water/ethylene glycol) in comparison with 0.2% concentration of Graphene nanoplatelets is 18.6% and at same concentration and temperature of hybrid nanofluid it is 10.23%. This confirms that density decreased for hybrid nanofluid when compared with single nanofluid composition. Table 6 representing theoretical density values of fluids. The density of hybrid nanofluid increases as the volume concentration of nanoparticles increases and the temperature decreases. The nanofluid with a 0.02 % volume concentration and 70:30 Cu-GNPs hybrid nanoparticles had the maximum density in a research conducted by Kishore, Sireesha [74]. similar equation as in this study is used to compute the density of hybrid nanoparticles by the author. Because copper has a higher density than graphene, the densities of 70:30 Cu-GNPs is higher with respect to the author. A hybrid nanofluid's density is influenced by both the volume percentage and the densities of the nanoparticles. Following the similar trend in this research study, the density of the Graphene nanoplatelets in single nanoparticle fluid is higher compared to hybrid nanofluid as shown in the below figures of experimental density.

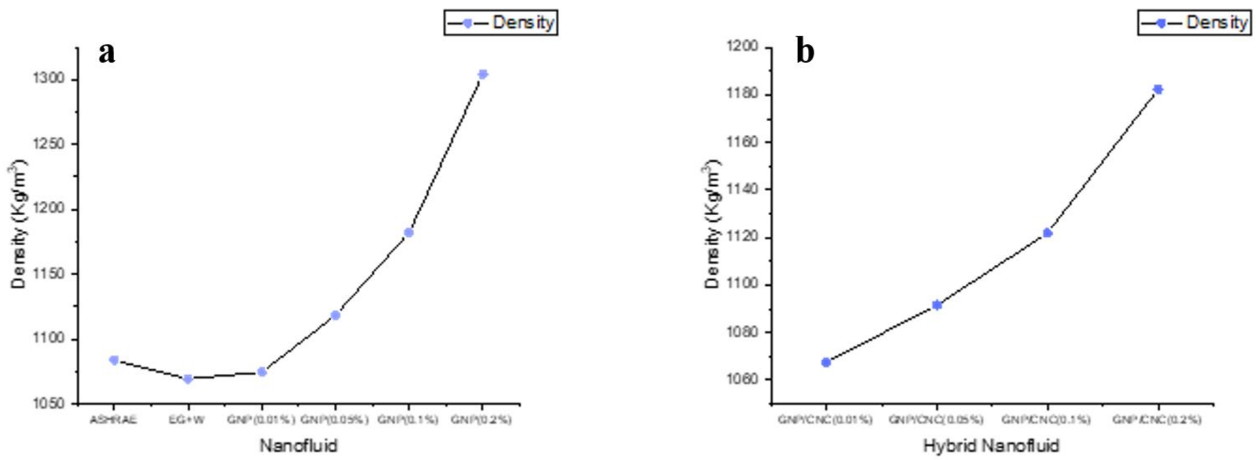

Figure 8: Density of nanofluids at different concentrations a) Graphene nanoplatelets b) Hybrid GNP/CNC

Table 6: Theoretical density values of fluids

Fluids		**Density (kg/m3)**	
EG+ Water (60:40)			
0.01% GNP	0.01% GNP/CNC	1052.2	1105.5
0.05% GNP	0.05% GNP/CNC	1101.3	1127.9
0.1% GNP	0.1% GNP/CNC	1162.7	1155.2
0.2% GNP	0.2% GNP/CNC	1285.4	1211.7

3.4 Specific heat

Differential Scanning Calorimetry was used to investigate the specific heat capacity characteristics of CNC nanofluids. Figure 9 shows the specific capacity of the base fluid and GNP & GNP/CNC nanofluids. Figure (b) depicts the effect of temperature and mass fraction on specific heat capacity when the GNP/CNC mass ratio is 1:1. There have not been enough mathematical and investigational research to estimate the nanofluids specific heat capacity at various temperatures and volume concentrations. The specific heat capacity of nanofluid samples is lower than that of base fluid, as can be observed from Figure 9. The specific heat capacity of particles decrease as their volume concentration increases. At 30 °C, the measured specific heat capacities of nanofluids demonstrate that they are roughly 0.56 percent and 7.52 percent lesser than those of the base fluid for 0.01 and 0.2 volume percent of nanoparticles, respectively. However, most previous studies have shown that adding nanoparticles reduces the specific heat capacity, although some unexpected outcomes have also been recorded [75]. The heat capacity of nanofluids appears to be affected by the specific heat capacity of both nanoparticles and the base fluid, and the interfacial energy released of solid liquid is altered when suspended nanoparticles are adjusted. The specific heat of nanocomposite materials is influenced by the surface free energy of nanoparticles since they have a higher surface area and a greater overall heat capacity. On one hand, this is due to the fact that water has a higher specific heat than nanoparticles; on the other hand, it demonstrates that the hybrid nanoparticle has a significant impact on specific heat capacity; even a small amount of mass fraction nanoparticle can significantly reduce specific heat capacity, especially at lower temperatures. The specific heats of the hybrid and single nanoparticle nanofluids, GNP-EG/W nanofluid and GNP/CNC-EG/W nanofluid, with the same mass fraction nanoparticles of 0.1 percent, are contrasted in both images. It means that as the temperature rises, all specific heat capacities rise as well. Besides water, it is seen that hybrid nanofluid has the highest specific heat.

This is owing to the GNP's low specific heat capacity and the nanofluid's lower GNP/CNC concentration.

The specific heat capacity of the 0.01and 0.2 volume % for mono nanofluid (GNP) reduces by 1.74% and 23.43 % as compared to the base liquid, respectively. The specific heat of the hybrid nanofluid (GNP/CNC) reduces by 0.38 % as compared at 0.01wt.% and it reduces by about 15.92% at 0.2wt%. The specific heat value when compared between Hybrid Nanofluid (GNP/CNC) and mono nanofluid GNP at 0.01wt% is increased by 1.35% and at 0.2wt% is increased by about 8.92%. It can be concluded that specific heat value is much higher for hybrid nanofluid than mono nanofluid at lower volume concentration. The Specific Heat Capacity of hybrid nanofluids has been demonstrated to be significantly affected by temperature. The reduced Specific Heat Capacity of hybrid nanofluids compared to water is universally agreed upon by all studies [76]. According to a study temperature has a mixed effect on specific heat that is rather inconsistent. Fazeli, Emami [77] found that as the temperature of the MWCNT-CuO increased from 20 to 35 °C, the Specific heat capacity of the MWCNT-CuO reduced. A similar finding was made by Mousavi, Esmaeilzadeh [78] who found that the CuO/MgO/TiO_2 triple hybrid nanofluid had a decreasing SHC as temperature increased across all volume concentrations studied. Few authors explained the effect of volume concentration on specific heat capacity of hybrid nanofluids that it exhibits a linear relationship with the volume concentration of hybrid nanofluids. The combined influence of the specific heat capacities of the nanoparticles and base fluids is responsible for this tendency. Furthermore, raising the volume concentration of nanoparticles appears to disrupt the solid–liquid phase's interfacial free energy. Because nanoparticles have a bigger surface area, their surface free energy has a stronger impact on overall heat power, which influences nanocomposite materials' specific heat [18, 75, 76]. When volume concentration was improved from 0.02 percent to 0.06 percent at the constant temperature of 20 °C, specific heat capacity decreased showing 7% drop [27]. Similar trend was recorded in different studies [79, 80]. Their research also found that when the volume concentration of the hybrid nanofluid increased, the specific heat capacity of the hybrid nanofluid decreased significantly. As liquids (base fluids) have a greater specific heat capacity than solids (nanoparticles), the base fluids have more hybrid nanocomposites added to them that affected the Specific heat capacity to drop, according to this analysis. When the volume concentration of the generated Graphene-Al_2O_3 hybrid nanofluid was increased 0.05 wt percent to 0.15 wt percent (as relative to the base fluid -20 °C), Gao, Xi [81] reported a Specific heat capacity reduction of 4 to 7 percent. At 30°C, Figure 10 depicts the fluctuation of specific heat capacity in relation to the volume fraction of GNP loadings. The specific heat of nanofluid is shown to decrease as GNP loadings increase. Because GNP has a lower specific heat capacity than the base fluid, the specific heat capacity of the nanofluid decreased when GNP is added. The most significant reduction in specific heat is determined to be 8% by Selvam, Mohan Lal [82]. The decreasing trend of specific heat value similar to the present study is plotted in image to validate the present study.

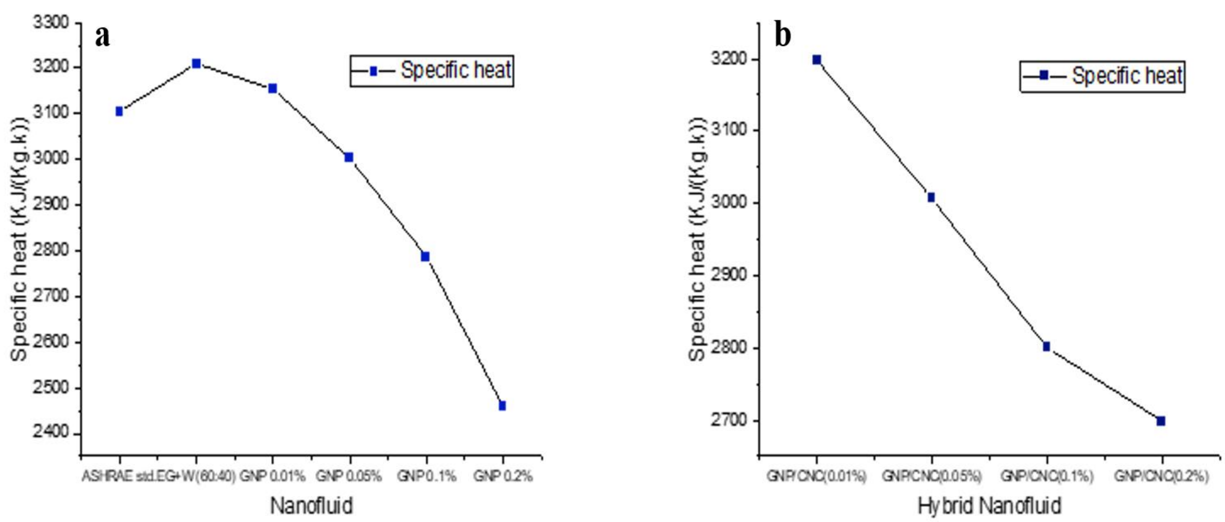

Figure 9: Specific heat of nanofluids at different concentrations a) Graphene nanoplatelets b) GNP/CNC

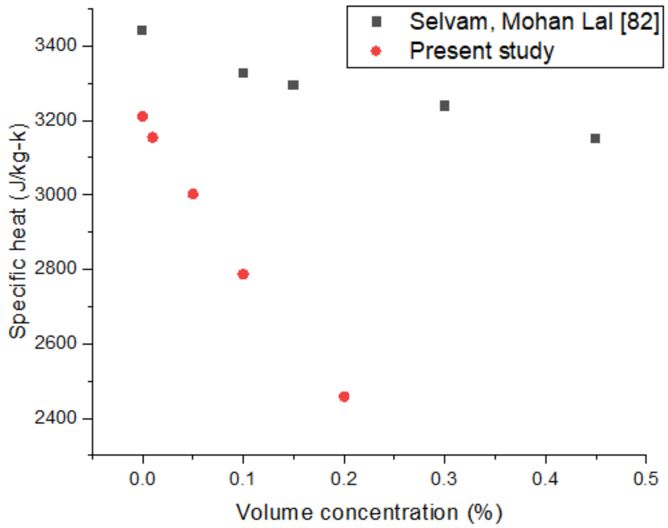

Figure 10: Comparison of graphene nanofluid Specific heat at different concentrations

4 Conclusion

A single & hybrid nanofluid of Graphene nanoplatelet (GNP) & GNP/CNC nanoparticles has been prepared by using two step method. Later the characteristic properties and thermophysical properties are studied at various volume concentrations in base fluid of EG/Water (60:40), with volume concentrations of 0.01 %, 0.05%, 0.1%, and 0.2% and it was concluded that,

- All GNP–CNC hybrid nanofluid samples give an increase in thermal conductivity with base fluid. At 0.2 vol % at 40 ˚C, experimental data reveals that thermal conductivity enhanced by 27%. At the room temperature for GNP nanofluid the values of thermal conductivity are in the range of 0.441W/m-K and for hybrid nanofluid in the range of 0.515

W/m-K. The viscosity of GNP/CNC nanofluids decreased with the increase in temperature. At 0.2 % of GNP nanofluid, the viscosity increased by 21%. Similarly, there is an increase in viscosity by 24.5% at 0.2% of hybrid nanofluid (GNP/CNC) at 20°C with comparison to base fluid.

- The experimental density of nanofluid obtained was consistent with theoretical values. The density value of GNP & GNP/CNC at 0.2 volume concentration is 1304.2 Kg/m^3 & 1182.32 Kg/m^3 respectively with an increase of 18.6% & 10.23% in comparison to base fluid. With an increased nanoparticle volume fraction, the nanofluid's specific heat capacity drops. At lower temperatures, the volume percentage of nanoparticles has a greater impact on the specific heat of hybrid nanofluid. The specific heat decreased with increase in nanoparticle concentration and when compared with Hybrid Nanofluid (GNP/CNC) and single nanofluid there is an increase by 1.35% and 8.92% at 0.01 and 0.2 vol % respectively.

The thermophysical characteristics of GNPs & GNP/CNC nanofluids obtained as a result suggest that this is an effective and useful approach for thermal engineering applications. Due to synergetic effects, GNP/CNC hybrid-based nanoparticles revealed properties that could not be achieved by using GNP or CNC nanoparticles independently. It is demonstrated that combining the diversity and uniqueness of both GNP and CNC not only enhances the number of applications available, but also provides undeniable benefits to their respective distinct characteristics. These hybrids have a several features that make them suitable for sensing, electronics, optical, biomedical, energy storage and heat transfer applications.

CRediT author statement

M. Sandhya: Conceptualization, Visualization, Investigation, Data curation, Writing- Original draft preparation, Reviewing and Editing. **D. Ramasamy:** Supervision, Writing-Reviewing and Editing. **K. Kadirgama:** Writing- Reviewing and Editing. **W.S.W. Harun:** Writing- Reviewing and Editing. **R. Saidur:** Writing- Reviewing and Editing.

Declarations

Competing interests

The authors claim they are conscious and say no financial or personal conflicts that might have affected the research provided in this study.

Authors' contributions

M. Sandhya: Conceptualization, Visualization, Investigation, Data curation, Writing- Original draft preparation, Reviewing and Editing. **D. Ramasamy:** Supervision, Writing-Reviewing and Editing. **K. Kadirgama:** Writing- Reviewing and Editing. **W.S.W. Harun:** Writing- Reviewing and Editing. **R. Saidur:** Writing- Reviewing and Editing.

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Availability of data and materials

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

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