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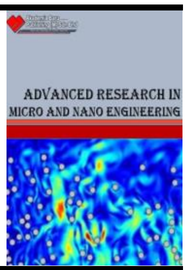

Formulation of Graphene Nanoplatelets Water-Based Nanofluids using Polyvinylpyrrolidone (PVP) as Surfactant

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ABSTRACT

Graphene nanoplatelets (GNP) have garnered significant interest owing to their remarkable mechanical properties, making them ideal materials for enhancing the heat transfer properties of nanofluids. However, the agglomeration of GNP in water-based nanofluids remains a significant challenge, limiting their practical applications. In this study, a comprehensive investigation into the formulation of water-based nanofluids using GNP is presented, with a focus on the incorporation of polyvinylpyrrolidone (PVP) as a surfactant to enhance their stability and dispersion. The nanofluid formulations were systematically prepared with varying concentrations of GNP and PVP to optimize their thermal properties. Additionally, the thermal conductivity and viscosity of the nanofluids were examined over a range of temperatures and concentrations. The results show that the formulation of GNP with 0.6 wt.% shows great potential as a nanofluid with 0.6611 W/mK and 1.22 mPa.s for thermal conductivity and viscosity, respectively. The findings also demonstrate that the incorporation of PVP significantly improves the stability and dispersion of GNP in water, leading to enhanced thermal conductivity and manageable viscosity. These findings contribute to the application of this nanofluid in various industries, specifically solar energy.

1. Introduction

Over the last few decades, solar energy has arisen as one of the best promising renewable energy sources, meeting the demands of current generations without jeopardizing future generations' ability to satisfy their own [1]. There has been a great deal of study on new types of heat transfer fluids [2], notably nanofluids. The utilization of nanofluids as working fluid in photovoltaic thermal (PV/T) also has obtained a lot of interest and is recognized as a promising class of heat transfer medium [3]. It is due to their superior thermal characteristics as compared to traditional heat transfer fluids. The

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thermal conductivity, heat transfer coefficient, and stability of nanofluids have significantly improved [4].

Nanofluid is a fluid that consists of solid particles that are nanometer in size, typically less than 100 nm. The nanofluid developed by Choi *et al.,* [5] has been shown to have higher heat transfer efficiency than traditional fluids. Nanofluids are being used in the design of PV/T collectors to improve their performance and it also have been explored as coolants for cooling photovoltaic (PV) panels to mitigate the decrease in efficiency caused by high operating temperatures. Increased thermal and electrical outputs are possible with the presence of nanoparticles to the base fluid. Compared to conventional devices, the PV/T system generates more energy per unit area and offers better aesthetics in terms of uniformity, making it suitable for limited effective space [6]. The working principle of the PV/T system is simple, where it simultaneously generates electrical and thermal energy. To increase the efficiency of heat transfer in PV/T systems, several techniques such as employing different cooling fluids and modifying the absorber arrangement are used [7]. The improved heat dissipation of nanofluids can help maintain optimal PV panel performance [8].

The utilization of nanofluids as working fluids in solar systems has been investigated in several studies. A numerical study by Rosli *et al*., [9] compared the experimental and simulation results by investigating the PV surface temperature and nanofluid outlet temperature while utilising aluminium oxide (Al_2O_3) water nanofluid as the coolant. Another study by Hu *et al.,* [10] mentions that carbon-based hybrid nanofluids improve energy efficiency in solar energy harvesting systems. For example, when compared to systems using water as the working fluid, the overall efficiency of a PV/T system using MWCNT/water nanofluids was shown to be more significant [11].

The use of nanofluids has led to significant improvements in electrical power output, exergy efficiency, and reduction in exergy losses and entropy generation. Additionally, the economic analysis has shown better payback periods when cooling with nanofluids compared to uncooled PV modules. Vaka *et al.,* [12] stated that the current research and development of nanofluids in PV/T systems have shown significant output, but there is a need for further advancements and investments to scale down designs and maximize electrical production.

Carbon nanostructures, such as graphene, have higher heat transfer properties than metals or metal oxides. Graphene nanoplatelets (GNP), one of several forms of nanoparticles, have drawn a lot of interest because of their remarkable thermal, mechanical, and electrical capabilities [13]. A study by Ridha *et al.,* [14] shows that heat transfer can be improved by utilizing graphene nanofluids as a working fluid in a tube channel. While prior studies have explored the potential of nanofluids in enhancing the performance of PV/T systems, this study distinguishes itself through a comprehensive investigation into the use of GNP as heat transfer enhancers. This study seeks to characterize GNPs before formulating them as nanofluids and identify potential uses for them in various heat control systems.

1.1 Application of Surfactant in Nanofluids

Surfactants play a vital part in avoiding nanoparticle agglomeration and settling and ensuring uniform dispersion within the base fluid, which is commonly water in this case. A surfactant, also known as a dispersing agent, is a surface-active compound that is added to a solution, typically a colloid, to improve particle separation and prevent settling or clumping. A study by Morsy [15] shows that surfactants can also act as stabilizers, decreasing the tension on the surface between two liquids or interfacial tension between a liquid and a solid.

Notably, this study focuses on the formulation of GNP water-based nanofluids using PVP as a surfactant to overcome agglomeration issues. Previously, Ilyas *et al.,* [16] used sodium dodecyl sulphate (SDS) in their study and found that SDS is suitable for stabilizing GNP with the ratio of 1.5:1 to the GNP. However, SDS causes a lot of foaming so the concentration used must be low to avoid foaming. It is important to consider the compatibility between surfactants and nanoparticles to ensure optimal foam stability. Despite the growing interest in the formulation of GNP for enhanced heat transfer properties in water-based nanofluids, there remains a significant research gap concerning the optimization of PVP as a surfactant for stable and homogeneous GNP dispersion. While previous research has acknowledged the role of PVP in preventing GNP agglomeration, there has been no systematic exploration of the longstanding stability of PVP-stabilized GNP in water based nanofluids.

This study aims to bridge this knowledge gap and unlock the full potential of GNP as heat transfer enhancers. By formulating GNP nanofluids with the aid of PVP surfactant, this study is expected to overcome the agglomeration issues encountered by other researchers. This innovative approach not only contributes to the field of thermal management but also holds promise for applications in renewable energy systems.

2. Methodology

2.1 Characterization of Graphene Nanoplatelets Powder

Before formulating the GNP powder into nanofluids, the GNP powder undergoes some characterization analysis to determine the morphology of the GNP. The GNP is thoroughly characterized and examined to ensure the nanofluids to be produced are stable and evenly dispersed in a base fluid. In this study, the characterization analysis done on the GNP powder are field emission scanning electron microscopy (FESEM), particle size analyzer and X-ray diffraction (XRD).

The GNP powder as shown in Figure 1 was obtained from Shanghai Harza Electromechanical Technology Co., Ltd. with a thickness of 1-10 nm and grain size of ~160.4 µm. Table 1 below shows the properties given by the supplier.

Table 1
Properties of GNP powder

Product name	Industrial graphene nanoplate
Carbon content	>99 at% (EDS)
Ash content	<1 wt%
Lateral size	1-10 µm HRTEM
Thickness	1-10 nm HRTEM
Conductivity	800-1100 S/cm
Moisture content	<2 wt%
Grain size	~160.4 µm
Tap density	0.13-0.16 g/cm^3
Apparent density	0.09-0.13 g/cm^3
Appearance	Black, grey powder

The GNP powder is first depicted by field emission scanning electron microscopy (FESEM) in a model Hitachi SU5000. This analysis is done to investigate the morphology of the GNP. The magnification used to analyze the morphology of the GNP is 50.0 K [17].

A particle size analyzer is a device or method used to determine and characterize the size distribution of particles in a sample [18]. The particle size of the GNPs is measured by Malvern Mastersizer 2000.

To analyze the crystal structure in the nanomaterial, an approach called X-ray diffraction (XRD) is utilized in this study by diffracting an X-ray beam in all directions [19]. Both qualitative and quantitative analysis can be done with XRD.

To calculate the crystallite size of the particle, the Scherrer equation as referred to by Fatimah *et al.,* [19] is used as shown in Eq. (1)

$$D = \frac{K\lambda}{\beta\cos\theta} \tag{1}$$

where D represents the crystallite size, K is the Scherrer constant (0.9), λ is the wavelength of the X-rays used, β is the full width at half maximum (FWHM in radians), and θ is the peak position (radians).

Fig. 1. Graphene nanoplatelets powder

2.2 Preparation of Graphene Nanoplatelets Nanofluids

The primary goal of this research is to describe the process of creating a graphene nanoplatelets nanofluid with the best formulation for improved thermal conductivity. It is critical to obtain the optimal nanofluid concentration since the mass flow rate of the working fluid in PV/T systems varies based on the kind of fluid employed, which might affect the performance of the PV/T systems [20]. There are two techniques for preparing nanofluids: the two-step method and the one-step method [21]. In this study, the two-step method is utilized as shown in Figure 2 as this method is the common method used in another research studies [22]. The nanoparticles powder employed in this technology are made via chemical or physical procedures. The nanoparticles were first weighed using digital analytical balance EJ610-E. Due to its solubility in water and other polar solvents, polyvinylpyrrolidone (PVP) was utilised as a surfactant to allow for effortless dispersion of nanoparticles in nano coolant. The details of the PVP surfactant as presented in Table 2. The GNP with concentrations of 0.2 wt.%, 0.4 wt.%, 0.6 wt.%, 0.8 wt.% and 1.0 wt.% were formulated with the surfactant PVP. The weight percentage of surfactant used is 40% of the GNP weight percentage. The nanofluids will then be homogenized using HG-15D homogenizer for 15 minutes with the speed of 1500 rpm and ultrasonically agitated for 30 minutes under room temperature using Elmasonic E 30 H. This method is the best cost-effective method to producing nanofluids on a wide scale because techniques for the synthesis of nanopowder have already been scaled up to levels of production that are commercially viable [23]. The details of all materials used in this study as in Table 3.

Table 2
Properties of PVP surfactant

Product name	Polyvinylpyrrolidone (PVP)
Chemical name	$(C_6H_9NO)_n$
Appearance (colour)	White to off white
Appearance (form)	Powder
Solubility in water	Soluble
pH	3-7

Table 3
Details of materials used

Name	Category	Function
Graphene nanoplatelets	Dispersion medium	Nanoparticle
Polyvinylpyrrolidone (PVP)	Dispersion medium	Surfactant
Distilled water	Dispersion phase	Base fluid

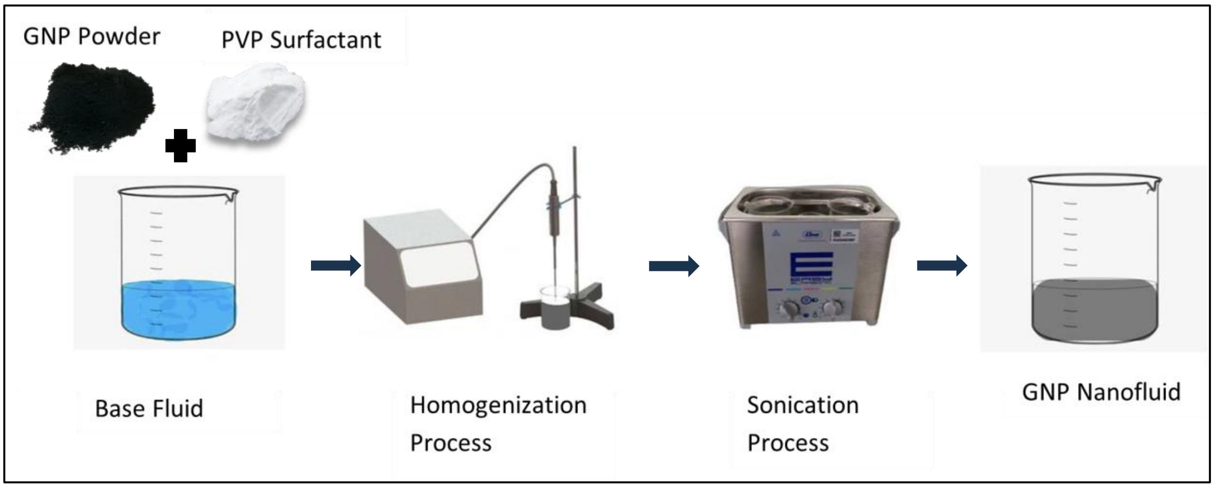

Fig. 2. Overview of two-step method

The mass/volume fraction or concentrations are calculated using the equation as stated in Eq. (2) [24]

$$\varphi = \left[\frac{\frac{w}{\rho_p}}{\frac{w}{\rho_p}+\frac{w_{bf}}{\rho_{bf}}}\right] \times 100 \tag{2}$$

In this equation, ‘φ’ is the concentration, ‘w’ is the weight of nanoparticles, ‘ρ_p’ is the density of nanoparticles, ‘w_{bf}’ and ‘ρ_{bf}’ are the weight and density of the base fluid, respectively.

2.3 Determination of the Thermophysical Properties

The determination of the thermophysical properties of nanofluids involves measuring various physical properties that influence the behavior of these colloidal suspensions of nanomaterials in a base fluid. In this study, the thermal conductivity of the nanofluid is measured by using a TEMPOS thermal analyzer under room temperature using a KS3 sensor. This physical property must be measured since it determines the ability of nanofluids to store and transmit heat. Enhanced thermal conductivity in nanofluids is beneficial for heat exchanger or cooling system applications since it can lead to enhanced heat transfer efficiency.

The viscosity of the nanofluids were also measured in this study using the IKA ROTAVISC lo-vi viscometer model with the VOL-SP-6.7 spindle. This rotor can measure the viscosity above 1 mPa.s. The viscosity test was carried out at a temperature of about 22°C and a rotational speed of 150 rpm with a stop condition of 2 minutes. Measuring the viscosity of the nanofluids is important as it helps to understand and optimize their flow behaviour. Viscosity affects how easily the nanofluid flows through pipes and channels, influencing heat transfer efficiency and overall performance in various applications.

3. Results

3.1 Characteristics of the Nanoparticles

3.1.1 Field Emission Scanning Electron Microscopy (FESEM)

The structure of the GNPs particles was observed through Field Emission Scanning Electron Microscopy (FESEM) and the results as shown in Figure 3. The FESEM analysis of the GNPs powder reveals valuable insights into its morphology. The GNPs appear to have irregular shapes with varying sizes and thicknesses. The average particle size of the GNPs found was around 98 to 400 µm. The results also show that the nanoparticles are stacking on each other. The stacking of nanoparticles can affect the properties such as surface area, reactivity, and stability [25].

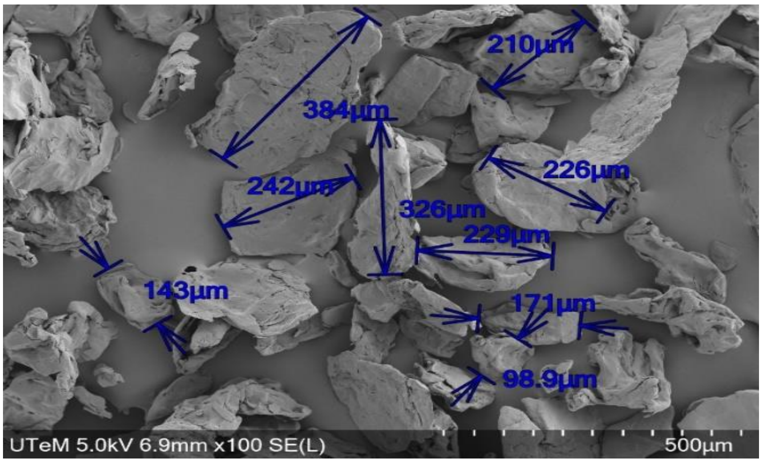

Fig. 3. Results of FESEM

3.1.2 Particle size analyser

Based on Figure 4, it is found that the particle size of GNPs varies substantially, with the presence of the smallest particles being around 100 µm and the largest particles around 1000 µm. The distribution of the powder shows a high percentage of volume particles in a range of 700 µm where this shows that a substantial volume of the sample consists of particles around that size.

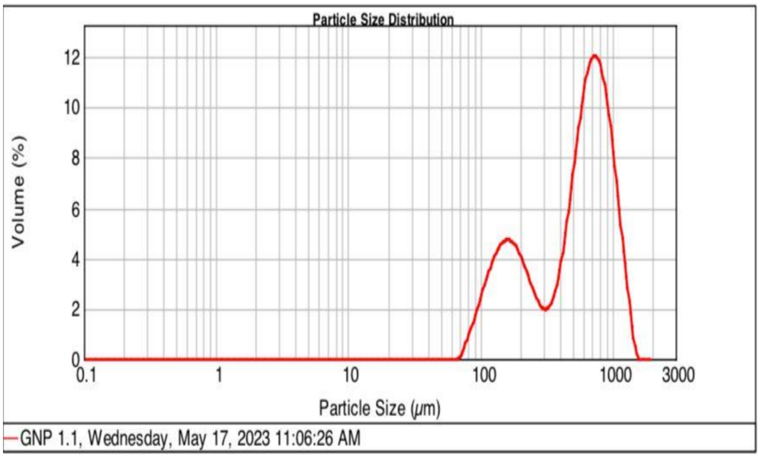

Fig. 4. Results of particle size analyzer

3.1.3 X-ray diffraction

The XRD pattern that was obtained is recorded at 2θ = 20° until 90°. In Figure 5, the XRD pattern of GNPs clearly reveals that graphene has a crystalline structure because it exhibits the highest peak of intensity at 2θ = 26.44°, which is consistent with observations by Vakili *et al.,* [26]. The smallest peak of the GNP was recorded at 2θ = 86.96°. With the obtained results, the evaluated material has a crystallite form due to the sharp peak obtained [27]. This result also shows that the powder contains high graphite that denotes as (002) as referred to by Siburian *et al.,* [28].

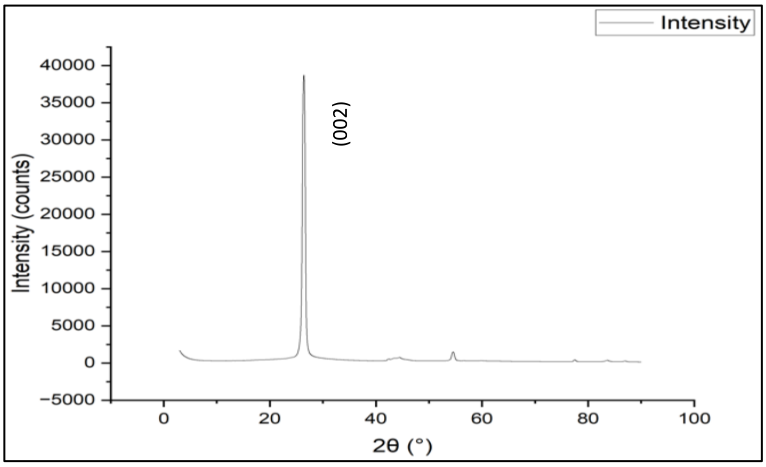

Fig. 5. XRD Results on graphene nanoplatelets

Based on the characterization analysis done, the GNPs powder shows a particle size ranging from 100 to 1000 μm which is in line with the characterizations provided by the supplier. This characterization analysis also shows the suitability of these nanoparticles to be formulated into nanofluids to achieve optimal stability.

3.2 Stability of the Nanofluids

Nanofluids' sustained stability is crucial in their application. The size of the nanoparticles is another factor that impacts the stability of nanofluids, with nanoparticles of lesser size being more stable [29]. Nanofluids have a tendency to agglomerate, which can reduce their efficient heat transfer power [30,31]. The agglomeration of nanoparticles in nanofluids impacts thermophysical properties such as thermal conductivity and viscosity, which in turn influences the efficiency of heat transfer [32]. In this study, the stability of the nanofluids were observed by visual observation method. Through this method, the sedimentation and agglomeration of the nanofluids were observed. The sample of nanofluids were left static for two weeks and the sedimentation of the nanofluids were observed as shown in Table 4. From observations, PVP surfactant portrays a good property as an additive as it prevents the agglomeration in GNP nanofluids from occurring due to its ability to disperse and stabilize GNP in aqueous solutions [33]. Additionally, PVP also can prevent rapid agglomeration and sedimentation of GNP, leading to highly stable colloidal mixtures.

Table 4
GNP-PVP nanofluids stability

Duration	Nanofluid state
1 day	
14 days	
30 days	

3.3 Thermophysical Properties of Nanofluids
3.3.1 Thermal conductivity

A nanofluid with enhanced thermal conductivity is generally favoured since it can greatly improve the fluid's heat transfer capabilities. Furthermore, nanoparticles with increased thermal conductivity can transmit heat through the fluid more effectively, resulting in improved thermal performance. The nanofluids with higher thermal conductivity also can effectively dissipate heat and maintain lower operating temperatures [34].

Figure 6 shows the results of the thermal conductivity of each concentration of GNP. These results indicate that the concentration of 0.2 wt.% has the highest thermal conductivity, 1.3673 W/mK while 0.8 wt.% have the lowest thermal conductivity which is 0.4780 W/mK. The thermal conductivity decreases as the concentration increases. However, as depicted in Figure 6, the thermal conductivity shows an increment at 1.0 wt.%. Depending on the unique properties of the nanofluid, the thermal conductivity of the fluid can either rise or decrease with increasing concentration. According to research by Borode *et al.,* [35] and Darabian *et al.,* [36], as concentration increases, a nanofluid's thermal conductivity decreases. On the other hand, additional research has demonstrated that as concentration increases, the nanofluids' thermal conductivity also can increase [37,38]. The type and size of the nanoparticles, the base fluid, and the existence of surfactants are some of the variables that affect how thermal conductivity behaves with concentration.

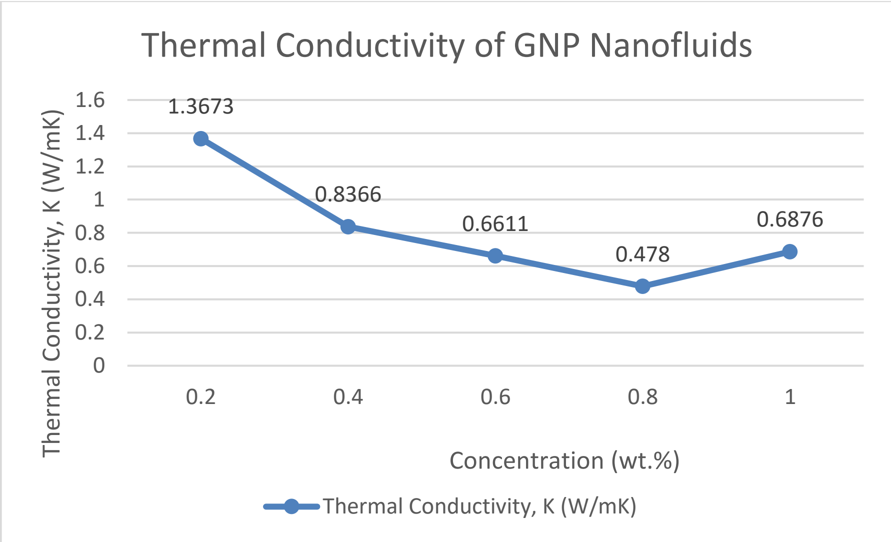

Fig. 6. Thermal conductivity of GNP nanofluids

3.3.2 Viscosity

In this case, a nanofluid with low viscosity is preferred as lower viscosity can lead to better flow and heat transfer rates, which can be beneficial for efficient cooling in PV/T systems. High viscosity nanofluids can increase the loss of pressure and pumping power, posing a challenge to the system. [39]. In a study by Bindu *et al.,* comparing different nanofluids [40], it was found that using carbon nanotubes (CNT) nanofluid with low viscosity resulted in higher system efficiency compared to using base fluid or Ag-MgO nanofluid with higher viscosity. Therefore, low viscosity nanofluids are preferable for photovoltaic thermal systems as they enhance system efficiency and reduce pressure drop. The entire performance of the system is negatively impacted when the pressure drops increase. It can be difficult for nanofluid-based systems to deal with the higher pressure drop and pumping power caused by the higher viscosity of nanofluids [41]. Reduced system efficiency and greater energy usage may arise from this increased pressure drop. It may also result in a higher friction factor and pressure drop, both of which could worsen the system's performance [39].

Figure 7 shows the results of viscosity that have been tested on every concentration of GNP. However, the viscosity pattern seems to show some fluctuation with increasing concentration. In nanofluids, viscosity can be influenced by various factors, including nanoparticle concentration, size, and surface chemistry. As the particle size increases, the viscosity of the nanofluid may either decrease or increase depending on the specific conditions and concentration levels [42,43]. Typically, at low concentrations, the viscosity is expected to be relatively low due to the dominance of the base fluid's properties. As the nanoparticle concentration increases, the viscosity can increase due to the influence of the nanoparticle suspension. However, once a certain concentration has been reached, further increases might lead to particle agglomeration, which can reduce viscosity. Another factor is the presence of aggregates or clusters of nanoparticles within the nanofluid. These aggregates can affect the flow behaviour and lead to fluctuations in viscosity [44]. Based on the results, it portrays that the formulation with 0.6 wt.% concentration appears to have the lowest viscosity making it a preferable concentration to be applied in PV/T systems. The summary of the thermophysical properties of GNP nanofluids are presented in Table 5.

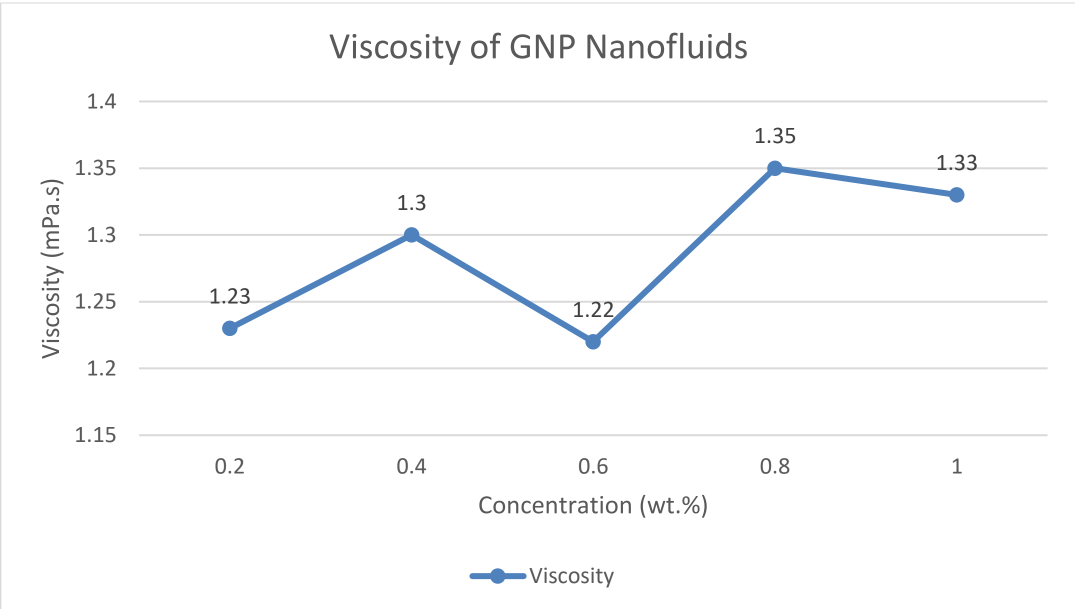

Fig. 7. Viscosity of GNP nanofluids

Table 5
Thermophysical properties of GNP nanofluids

Concentration (wt.%)	K (W/mK)	Viscosity (mPa.s)
0.2	1.3673	1.23
0.4	0.8366	1.30
0.6	0.6611	1.22
0.8	0.4780	1.35
1.0	0.6876	1.33

4. Conclusions

This study analyses the formulation of graphene nanoplatelets water-based nanofluids by varying the concentration and using PVP as the surfactant. From this study, several conclusions are discussed. To determine the best formulation of nanofluids, a formulation with relatively high thermal conductivity and lower viscosity is considered as the heat transfer efficiency is better as lower viscosity can lead to better flow and heat transfer rates, which can be beneficial for efficient cooling in PVT systems. A formulation with 0.6 wt.% is chosen as the best formulation after considering both thermal conductivity and viscosity. This chosen formulation appears to offer a slightly lower viscosity (1.22 mPa.s) while still maintaining a reasonably good thermal conductivity (0.6611 W/mK). More thermophysical properties can be considered in future studies to further ensure the properties of GNP nanofluids.

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