International Communications in Heat and Mass Transfer 126 (2021) 105402

Contents lists available at ScienceDirect

International Communications in Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ichmt

# Stability and thermo-physical properties of green bio-glycol based $TiO_2$-$SiO_2$ nanofluids

S.N.M. Zainon [a], W.H. Azmi [a,b,*]

[a] Department of Mechanical Engineering, College of Engineering, Universiti Malaysia Pahang, Lebuhraya Tun Razak, 26300 Gambang, Kuantan, Pahang, Malaysia
[b] Centre for Research in Advanced Fluid and Processes, Lebuhraya Tun Razak, 26300 Gambang, Kuantan, Pahang, Malaysia

ARTICLE INFO

Keywords:
Hybrid $TiO_2$-$SiO_2$ nanofluids
Green bio-glycol
Thermal conductivity
Dynamic viscosity
Density

ABSTRACT

The investigation on thermal properties of nanoparticles dispersed in the mixture of water and green Bio-glycol are limited in the literature and only available for single nanofluids. The purpose of this study is to investigate the stability and thermo-physical properties of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids. In the present study, the hybrid nanofluids were prepared at various volume concentrations of 0.5 to 3.0% by dispersing $TiO_2$ and $SiO_2$ nanoparticles (20:80) in water and Bio-glycol (40:60) mixture base fluids. The stability of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids showed physically to be in good range of stability for suspension nanoparticles with zeta potential of −53.46 mV. The thermal conductivity of the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids was enhanced up to 12.52% higher than the mixture of W/BG at 3.0% volume concentration and temperature of 70 °C. Meanwhile, there is insignificant dynamic viscosity increment of the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids with temperature. As a conclusion, a stable $TiO_2$-$SiO_2$ hybrid in green Bio-glycol/water mixture nanofluids was formulated in the present study with significant enhancement in thermal properties and recommended for heat transfer application. Further heat transfer investigation is required to confirm the feasibility of the green coolant.

## 1. Introduction

The conventional heat transfer fluid such as water, ethylene glycol, and oil has reached its limitation in increasing the performance of heat transfer due to its poor thermal conductivity properties. Since the fluids that improve the performance of heat transfer are required in many engineering applications, many researchers have been actively studying the thermo-physical properties of nanoparticles in the base fluids [1–9]. Until the emergence of nano-sized particles, metal and metal oxide particles were also dispersed in micro sizes. Pourhoseini and Naghizadeh [10] in their study used micro-sized silver particles dispersed in water. They concluded that micro-particles have the same properties as nanoparticles at a reduced cost compared to the nanoparticles that need to undergo a complex process before it can be used, resulting in high production cost. However, Paul et al. [11] reported that micro-sized particles exhibited unstable characteristics such as sedimentation that will cause clogging in the channel and result in added flow resistance in the system. In line with the developments of nanotechnology, the use of milli- and micro-sized particles has now been shifted to nano-sized particles due to its promising enhancement of thermo-physical properties with exceptional suspension stability [12–15].

Nanofluids are famous for their stability and improved thermal conductivity, with the addition of a small amount of nanoparticles in the base fluids. Since then, it has become a promising heat transfer fluid for the future [16,17]. Nanofluids are brand new fluids composed of nanoparticles with sizes less than 100 nm dispersed in the base fluids such as water, ethylene glycol, propylene glycol, and oil. Sidik et al. [18] defined nanofluids as an advanced type of fluid containing a small quantity of nanoparticles sized less than 100 nm, which are uniformly and stably suspended in a liquid. The concept of dispersing metallic and non-metallic nanoparticles began when [2] and Choi and Eastman [19] demonstrated a promising suspension of nanoparticles in conventional fluids to improve the heat transfer. Common nanoparticles that are used in the preparation of nanofluids are Silicon oxide ($SiO_2$), Titanium oxide ($TiO_2$), Aluminium oxide ($Al_2O_3$), Gold (Au), Carbon nanotubes (CNTs) and Zinc oxide (ZnO). Recently, many experiments have been conducted and reported that nanofluids show a significant increase in thermal conductivity compared to conventional heat transfer fluids [4,12,13]. Their notable properties have great potential to be used in many heat

* Corresponding author at: Department of Mechanical Engineering, College of Engineering, Universiti Malaysia Pahang, Lebuhraya Tun Razak, 26300 Gambang, Kuantan, Pahang, Malaysia.
E-mail address: wanazmi2010@gmail.com (W.H. Azmi).

https://doi.org/10.1016/j.icheatmasstransfer.2021.105402

Available online 19 June 2021

**Nomenclature**

| | |
|---|---|
| $\overline{A}$ | Final absorbance |
| $\overline{A_r}$ | Ideal ratio of absorbance ratio |
| $\overline{A_o}$ | Initial absorbance |
| BG | Bio-Glycol |
| $C_{bf}$ | Specific heat of base fluid |
| $C_{hnf}$ | Specific heat of hybrid nanofluids |
| $C_{nf}$ | Specific heat of nanofluids |
| $C_p$ | Specific heat of nanoparticle |
| $C_{SiO_2}$ | Specific heat of silicone oxide |
| $C_{TiO_2}$ | Specific heat of titanium oxide |
| $k$ | Thermal conductivity |
| $k_{bf}$ | Thermal conductivity of base fluid |
| $k_{nf}$ | Thermal conductivity of nanofluids |
| $k_r$ | Ratio of nanofluid to base fluid thermal conductivity ($k_{nf}$ / $k_{bf}$) |
| $m_{bf}$ | Mass of base fluid |
| $m_p$ | Mass of particle |
| $\rho_{bf}$ | Density of base fluid |
| $\rho_{hnf}$ | Density of hybrid nanofluids |
| $\rho_p$ | Density of nanoparticle |
| $\rho_{SiO_2}$ | Density of silicone oxide |
| $\rho_{TiO_2}$ | Density of titanium oxide |
| $\rho_w$ | Density of water |
| TEM | Transmission Electron Microscopy |
| $W$ | Water |
| $V$ | Volume |
| $V_1$ | Initial volume |
| $V_2$ | Final volume |
| $\Delta T$ | Temperature drops |
| $\Delta V$ | Additional volume |

*Greek symbols*

| | |
|---|---|
| $\phi$ | Volume concentration in percent |
| $\varphi$ | Volume concentration in fraction |
| $\omega$ | Weight concentration in percent |
| $\rho$ | Density |

*Subscript*

| | |
|---|---|
| $bf$ | Base fluid |
| $nf$ | Nanofluid |
| $hnf$ | Hybrid nanofluids |
| $p$ | Nanoparticle |
| $r$ | Ratio |

transfer applications such as microelectronics, engine cooling, and heat exchanger [20–25].

The act of dispersing nanoparticles started as an alternative to substitute conventional heat transfer fluids with poor thermal conductivity. Nowadays, hybrid nanofluids and its applications have also been explored in nanoparticle-related research. The innovations of hybrid nanofluids began when researchers saw potential demonstrated by single nanofluids in improving the thermo-physical properties. Single nanofluids refer to the suspension of one type of nanoparticle in the base fluids. In contrast, the hybrid nanofluid was prepared by dispersing two dissimilar nanoparticles into the conventional fluid like oil, water, propylene glycol, and ethylene glycol. Previous studies have shown significant enhancement in thermo-physical properties by adding a small amount of nanoparticles in the base fluid. Thus, it is believed that combining two or more nanoparticles can modify or increase the thermal conductivity of the nanofluid and indirectly increase the heat transfer performance. Koçak Soylu et al. [26] studied the heat transfer performance of an automobile radiator using single $TiO_2$ nanofluids, $TiO_2$-Cu, and $TiO_2$-Ag hybrid nanofluids at different volume concentrations. They found that the combination of $TiO_2$ and Ag nanoparticles produced a higher heat transfer coefficient than single nanofluids while the hybridization of $TiO_2$ nanoparticles with low concentrations of Cu nanoparticles produced a lower heat transfer coefficient than single nanofluids.

According to Sundar et al. [27], the enhancement of the heat transfer using nanofluids is initially affected by the thermal conductivity of the nanoparticles, particle volume concentration, and flow rate. However, if the volume concentration and mass flow rate were to be kept constant, the only property that will influence the heat transfer enhancement is the thermal conductivity of the nanoparticles [28–31]. However, the performance of heat transfer and fluid flow also depends on other properties such as dynamic viscosity [32–35], density [36–38], and specific heat [39–42]. With the production of nanofluids that grows on a daily basis, the researchers are also developing a novel approach to improve the thermo-physical properties by dispersing the nanoparticles synthesized from organic and waste materials into conventional heat transfer fluids. The effort is made to produce nanofluids from renewable materials and reduce the production costs. Bio-glycol is a green and renewable thermal fluids. The fluids is produced by the plant such as grown corn. It is non-toxic and environmental friendly coolant. The green Bio-glycol provides 30% lower viscosity at low temperatures compared to the traditional ethylene glycol [43]. Lower viscosity means less energy required to pump the thermal fluid. Bio-glycol offers greater thermal stability while possessing similar or better physical properties compared to ethylene glycol, it protects the surface against corrosion as a coolant and provide cost-effective rather than using oil [43]. It also stabilizes the pH of the fluids, keeping it in the range that is suitable for the metals in the system.

Poor thermal properties of conventional heat transfer fluids become a cause for the heat transfer enhancement to be stunted [44,45]. However, there is potential shown by the dispersion of single nano-sized particles in conventional coolants towards heat transfer augmentation and being triggered by numerous experimental investigations for heat transfer performance and pressure drop of heat transfer fluids. Their notable thermal properties have indicated feasibility for heat transfer applications. The performance of single nanofluids was proven to be better than the existing heat transfer fluids in transferring heat. However, many studies reported short-term stability for the single nanofluids [28]. Hence, this concern led to the development of hybrid nanofluids using the suspension of two different nanoparticles into the base fluids to improve the thermal properties and stability of single nanofluids. However, the previous hybrid nanofluids in the literature mostly used based fluids of water and ethylene glycol [46–49]. The thermal properties evaluation of nanoparticle dispersion in the green Bio-glycol based fluids is limited for single nanofluids in the literature [50–53]. Therefore, the present study is intended to formulate stabilized $TiO_2$-$SiO_2$ hybrid nanoparticles dispersed in a mixture of water/green Bio-glycol at various volume concentrations. Further, comprehensive investigation is performed on the stability and thermo-physical properties of green Bio-glycol based of $TiO_2$-$SiO_2$ hybrid nanofluids.

## 2. Methodology

### 2.1. Preparation of green bio-glycol based $TiO_2$-$SiO_2$ nanofluids

In the present study, the two-step method was employed in the formulation of the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids. The present nanofluids was formulated by utilising the mixture of the single $TiO_2$ and $SiO_2$ nanofluids at composition ratio of 20:80 by volume to produce $TiO_2$-$SiO_2$ nanofluids. Hamid et al. [47] and Hamid et al. [32] conducted comprehensive studies on the thermo-physical properties and experimental forced convection heat transfer of $TiO_2$-$SiO_2$ nanofluids in water/EG base mixture at various composition ratios. They observed significant enhancement of thermal conductivity and heat transfer performance with minimum increments at 50:50 composition ratio while maximum enhancement was recorded at 20:80 composition ratio. Similar results were obtained by Kumar and Sarkar [48] for the $Al_2O_3$-$TiO_2$ hybrid nanofluids. Therefore, the present study investigated the green Bio-glycol based nanofluids at 20:80 ($TiO_2$:$SiO_2$) composition ratio. Two types of nanoparticles namely Titanium oxide ($TiO_2$) and Silicone dioxide ($SiO_2$) were used in the present investigation. The properties of the nanoparticles are summarized in Table 1. The nanoparticles were acquired from US Research Nanomaterials, Inc. (USA). On the other hand, the mixture of water and Bio-glycol (W/BG) at 60:40 ratio was considered in the present study as the based fluids. Distilled water was produced in the laboratory while green Bio-glycol was purchased from Dynalene, Inc.

The single $TiO_2$ and $SiO_2$ nanofluids were prepared at volume concentrations of 0.5 to 3.0%. Then, both single nanofluids were mixed at a composition ratio of 20:80 ($TiO_2$: $SiO_2$). The basic expressions for the concentration of nanofluids by weight percent, $\omega$ and volume percent, $\phi$ are represented by Eqs. (1) and (2), respectively. The new expression of Eq. (3) is used to convert the weight concentration, $\omega$ of suspended nanoparticles to volume concentration, $\phi$. Subsequently, the dilution process is applied with the aid of Eq. (4), where $V_1$ is a predetermined volume for preparation of the single nanofluids from higher concentration to lower concentration. The volume of the base fluid, $\Delta V$ to be added to the nanofluids at initial volume concentration, $\phi_1$ is calculated to obtain the desired volume concentration, $\phi_2$.

$$\omega = \frac{m_p}{m_p + m_{bf}} \times 100 \quad (1)$$

$$\phi = \frac{\frac{m_p}{\rho_p}}{\frac{m_p}{\rho_p} + \frac{m_{bf}}{\rho_{bf}}} \times 100 \quad (2)$$

$$\phi = \frac{\omega \rho_w}{\frac{\omega}{100}\rho_w + \left(1 - \frac{\omega}{100}\right)\rho_p} \quad (3)$$

$$\Delta V = (V_2 - V_1) = V_1\left(\frac{\phi_1}{\phi_2} - 1\right) \quad (4)$$

The green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids was prepared with a minimum of 100 ml for each sample by applying the dilution process. The dilution process is used frequently in the literature for the preparation of nanofluids [9,45]. Then, the nanofluids were agitated using a stirrer for 30 min to ensure uniform mixture of single $TiO_2$ and $SiO_2$ nanofluids. The magnetic stirrer was used for sample preparation of small volumes of nanofluids whereas the bulk volume of nanofluids were mixed using a digital overhead stirrer. After that, the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids were subjected to the sonication process at different sonication times of 0.5, 1.0, 1.5, 2.0, 2.5 and 3.0 h using ultrasonic bath. The ultrasonic bath generated an ultrasonic pulse of 80 W at a frequency of 36 Hz to break particle agglomeration and reduce particle size. This method can improve the stability of nanofluids.

### 2.2. Stability of nanofluids

Stability plays a crucial role in prolonging the lifespan and maintaining the properties of the nanofluids [28]. In the present study, the stability of the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids was evaluated qualitatively using visual observation, transmission electron microscopy (TEM), and quantitatively determined by ultraviolet-visible (UV–Vis) spectroscopy, zeta potential, and density.

#### 2.2.1. Visual observation

Numerous investigations considered the visual method to observe the stability of the solution with suspended nanoparticles. This technique requires daily observation on the visibility of the sedimentation or separation layer in the sample. The sample with 10 ml volume for each sample were placed in a test tube for visual observation of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids. The images for each sample was observed right after preparation, daily and then weekly until the sedimentation was visible in the test tube. In this study, the stability of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids was evaluated using visual observation for more than 30 days.

#### 2.2.2. Ultraviolet-visible spectroscopy measurement

The absorbency analysis for the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids was performed using Drawell (DU-8200) UV–Vis spectrophotometer. The sample of nanofluids for 0.1% volume concentration at different sonication times was stored in the quartz cuvettes with 3 ml of each sample. The absorbance of sample is strongly dependent on the input wavelength. The high absorbance is recommended for stability evaluation. The peak absorbance was obtained at 600 nm wavelength. The absorbance ratio $\overline{A}_r$, can be determined from the formula of $\overline{A}_r = \left(\frac{\overline{A}}{\overline{A}_0}\right)$. The absorbance, $\overline{A}_0$ indicates the absorbance of the reference base fluids while $\overline{A}$ refer to the absorbance of the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids. The ideal $\overline{A}_r$ is equal to 1.0 (100%). This condition will reflect the constant absorbance value with time and is perfectly stable for the suspended particle without any sedimentation occurring during the sedimentation period [6]. Hence, the value of $\overline{A}_r$ to the nearest ideal of 1.0 will determine the stability condition of the sample and naturally decrease with the sedimentation time.

#### 2.2.3. Transmission electron microscopy observation

The stability of the nanofluids also can be evaluated by measuring the particle size distribution using the Transmission electron microscope (TEM). Instead of light, TEM uses a beam of electrons to create an image of the specimen. It generates a high-resolution digital image and is known as electron micrograph with size approximately 0.1 nm [54]. The TEM magnification is higher than other methods and possesses greater resolving power compared to a light microscope, thus generating better image quality and details for nanoparticle characterization.

#### 2.2.4. Density measurement

Another approach to measure the stability of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids is by measuring the changes of the density over time. The method was conducted by evaluating the density of the nanofluids on the first day and up to 30 days after preparation using DA-130 N portable density meter. The density of the nanofluids was measured at least three times for each concentration to obtain consistent

**Table 1**
Properties of nanofluid by US Research Nanomaterial, Inc.

| Type of nanofluid | Average Diameter, (nm) | Weight Concentration, $\omega$ (%) | Volume Concentration, $\phi$ (%) | Specific Heat, $c$ (J/kg°C) | Density, $\rho$ (kg/m$^3$) |
|---|---|---|---|---|---|
| $TiO_2$ | 50 | 40 | 13.62 | 692 | 4230 |
| $SiO_2$ | 22 | 25 | 13.06 | 745 | 2220 |

results.

### 2.2.5. Zeta potential measurement

Further evaluation for the stability of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids was done by measuring the zeta potential. In the present study, the stability evaluation of nanoparticle dispersion in the base fluids was performed using Litesizer™ 500 supplied by Anton Paar. The zeta potential of the solution can be directly obtained from the instrument with several input parameters of the based green Bio-glycol. The input data for viscosity, refractive index, and dielectric constant of the green Bio-glycol at temperature of 25 °C are given by 3.611 cP, 61.8 and 1.3751. The adoption of zeta potential in determining nanofluid stability is widely used by previous researchers. Prior investigations conducted by Zawawi et al. [25], Hamid et al. [32] and Zakaria et al. [29] also presented the stability of the nanofluids through zeta potential measurement.

## 2.3. Thermal conductivity measurement

The measurement of thermal conductivity in the present study was performed using the C-Therm TCi Thermal Conductivity Analyzer. The instrument employed (MTPS) techniques. The analyzer provides rapid measurements of thermal conductivity with accuracy and precision of 5% and 1%, respectively. This equipment consists of a TCi control unit, TCi software (v4.0), a weight, a sensor, and a stainless steel sensor plate. The equipment used the modified transient plane source (MTPS) techniques with capability to measure the thermal conductivity from 0 to 500 W/m.K in a broad range of temperature from −50 to 200 °C. The sensor was calibrated by the manufacturer, hence no further calibration required since the calibration data was already stored in the calibration chip. However, a validation process on the sensor is necessary to confirm an accurate measurement. Before starting the measurement, the sensor was validated using the reference material, namely Pyrex that is provided by the manufacturer. The sensor with base plate was placed inside a Memmert Universal Oven (UN30) in order to control and maintain the operating temperature during the thermal conductivity measurement. The sample with 1.5 ml volume was positioned on the heating element. Furthermore, a set of thermal conductivity measurement consisting of 20 readings was recorded using the TCi software in the temperature range from 30 to 80 °C. A similar flow of measurement was repeated for other samples at different volume concentrations of 0.5 to 3.0%.

## 2.4. Dynamic viscosity measurement

The measurement of dynamic viscosity in the present study was done using the Rheolab QC Rotational Rheometer. The device is equipped with a Peltier temperature device and capable of controlling the sample temperature from 0 to 180 °C using air. This device is an engineered ball bearing rotational rheometer and operated using the Searle principle. In the present study, a double gap measuring system (DG42) consists of a cup and measuring bob. The system was used for low viscosity fluid measurements. The 12 ml sample was transferred to the measurement cup. Then, the measuring bob was carefully placed into the cup to create a double gap measuring system. The measuring bob was ensured to be immersed in the sample before starting the measurement. Consequently, the measuring bob was mounted to the rheometer motor, and the measuring system was operated using the Rheocompass software. In the software, the operating speed was set in the range of 0 to 1000 rpm. In addition, the dynamic viscosity of the base fluids and green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids was measured for temperatures of 20 to 80 °C.

## 2.5. Density estimation and measurement

The estimation density of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids was determined by using mixture relation. The mixture relation presented in Eq. (5) was used to calculate the density of single nanofluids. The equation requires the volume fraction, $\varphi$ the density of the base fluid, $\rho_{bf}$ and density of nanoparticles, $\rho_p$ for the density estimation of single nanofluids. Various researchers used the equation to calculate the density of the single nanofluid [34,55]. Meanwhile, the density of hybrid nanofluids was estimated using another mixture equation as shown in Eq. (6) [32,47].

$$\rho_{nf} = \phi\rho_p + (1-\phi)\rho_{bf} \tag{5}$$

$$\rho_{hynf} = (1-\phi)\rho_{bf} + (0.2\phi)\rho_{TiO_2} + (0.8\phi)\rho_{SiO_2} \tag{6}$$

The density of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids was measured by using a Portable Density meter (Model: DA-130 N). The device functions to measure the density of liquid sample by adapting the resonant frequency oscillation method with ±0.0001 g/cm$^3$ accuracy in the temperature range from 0 to 70 °C. The reading was validated using distilled water to obtain density of 1.000 g/cm$^3$. The density of nanofluids was measured at various concentrations ranging from 0.5 to 3.0% and temperature of 30 to 70 °C. The data is manually recorded for each sample and repeated for a minimum of three times at different concentrations.

## 2.6. Estimation of specific heat

The specific heat of green Bio-Glycol based $TiO_2$-$SiO_2$ nanofluids was estimated using mixture relation equation presented in Eqs. (7) and (8). Eq. 3.7 was proposed by Zhou et al. [41] and was used by several investigators including Azmi et al. [34] and Khdher et al. [52] to estimate the specific heat of the single nanofluids. Meanwhile, Eq. (8) was used to estimate the specific heat of the hybrid nanofluids using volume fraction, density and specific heat of base fluid and nanoparticles. The estimation of specific heat of the hybrid nanofluids using this equation can be seen in various research articles [27,46,47].

$$C_{nf} = \frac{(1-\phi)(\rho C)_{bf} + \phi(\rho C)_p}{\rho_{nf}} \tag{7}$$

$$C_{hyb} = \frac{(1-\phi)\rho_{bf}C_{bf} + (0.2\phi)\rho_{TiO_2}C_{TiO_2} + (0.8\phi)\rho_{SiO_2}C_{SiO_2}}{\rho\ hnf} \tag{8}$$

## 2.7. Uncertainty of properties instruments

The uncertainties evaluation is performed to evaluate the error of instruments for the related thermo-physical quantities. Table 2 presents the uncertainties of the instrumentation of these quantities. From the table, the maximum percentage of the uncertainty was observed to be up to 1.27% for dynamic viscosity measurement. On the other hand, the lowest uncertainty was recorded for the thermal conductivity measurement with 0.02%.

**Table 2**
Uncertainty of properties measurements.

| Properties | Range | Least division | Data experiment | | % uncertainty | |
|---|---|---|---|---|---|---|
| | | | Min. | Max. | Min. | Max. |
| Thermal conductivity (W/m·K) | 0–500 | 0.0001 | 0.3212 | 0.5097 | 0.020 | 0.031 |
| Dynamic viscosity (cP) | 0–$10^9$ | 0.00001 | 0.00079 | 0.00693 | 0.144 | 1.270 |
| Density (kg/m$^3$) | 0–2000 | 1 | 1014 | 1100 | 0.091 | 0.099 |

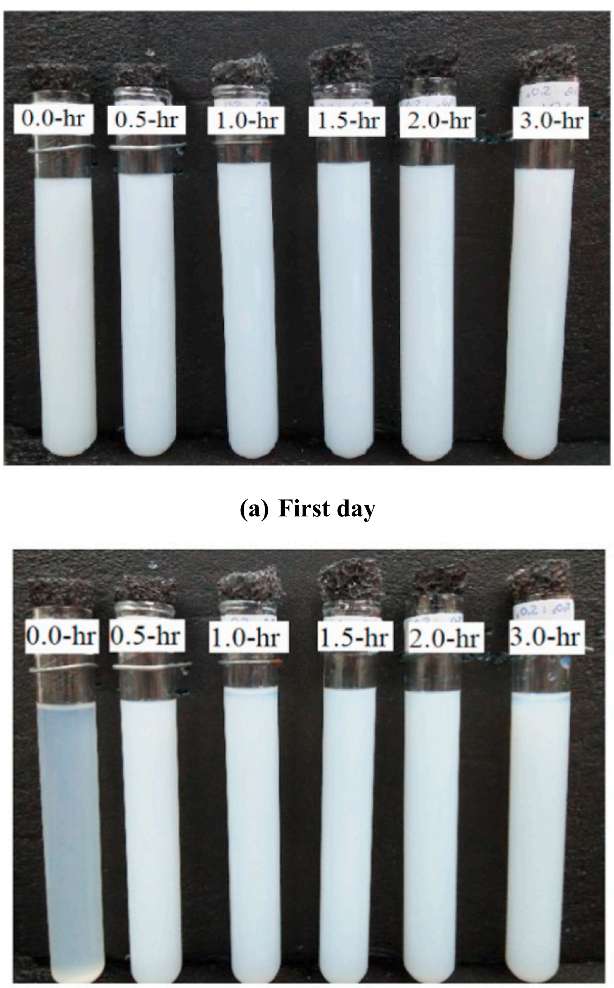
## 3. Results and discussion

### 3.1. Stability analysis of green bio-glycol based nanofluids

#### 3.1.1. Visual evaluation

The stability analysis of the green Bio-glycol based nanofluids was performed qualitatively using visual observation. The nanofluid samples in the test tube were observed visually within a designated duration to observe for any sedimentation formation at the bottom of the test tube. The visual image of observation was recorded during the first day of preparation, after a week, and after a month. This method was commonly used by other researchers [25,29,52] in their stability evaluation. The sample of green Bio-glycol based nanofluids at 0.1% volume concentration was prepared at different sonication times between 0.0 and 3.0 h. These samples were used in the visual evaluation (qualitative) and Ultraviolet-visible (UV–Vis) spectrophotometer evaluation (quantitative) to investigate the effect of sonication time on stability condition. The finding of stability condition will be supported by the both methods.

The visual technique was undertaken for six samples of the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids. The samples were prepared at different sonication times of 0.0 to 3.0 h and shown in Fig. 1. A small separation layer was displayed in the test tube for sample of 0.0 h during the first day of observation. Meanwhile, the samples of 0.5 to 3.0 h exhibited no visible separation layer on the same day. Instantaneously, the level of sedimentation increased dramatically in the sample of 0.0 h for up to 30 days observation. Furthermore, the samples of 0.5 and 1.0 h were beginning to form sedimentation after a week of preparation. The formation trend of sedimentation was followed by samples of 1.5 and 2.0 h after a week of preparation and almost invisible for separation layer. Nevertheless, the height of supernatant layer in the samples of 1.0 to 2.0 h were unchanged for several weeks of observation. On the other hand, the sample of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids with 3.0 h sonication time exhibited limited sedimentation in the test tube for up to four weeks after preparation. The most prolonged stability with minimum sedimentation appeared in the sample of 3.0 h during the fourth week after preparation and persisted in a stable condition after a month of preparation. The UV–Vis spectrophotometer analysis supported the outcomes from the present visual observation on the effect of sonication time towards stability behaviour of green Bio-glycol based nanofluids. The longer sonication time produced more stable nanofluids than shorter time with the optimum sonication time found at 3.0 h.




**(a) First day**

**(b) 30 days**

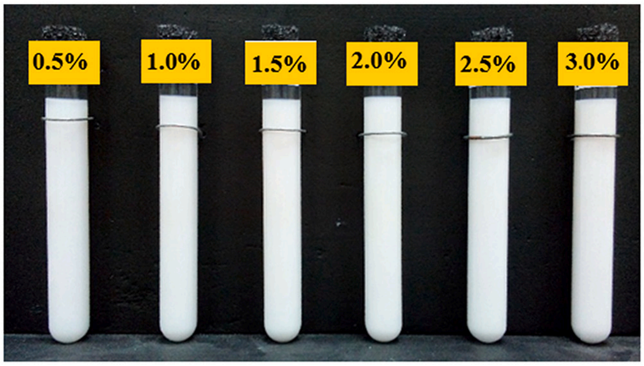
**Fig. 1.** Green Bio-glycol based nanofluids at 0.1% volume concentration and different sonication times. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The fact that the sonication process helps to break down and reduce the size of particle agglomeration explains the reason for the different stability conditions in each sample with variation of sonication time. Therefore, the current findings confirmed the influence of sonication time on the stability of the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids. Another researcher also presented similar stability condition in their works including Zawawi et al. [25], Sharif et al. [56], and Hamid et al. [32]. The stability evaluation by visual observation for any formation of sedimentation over time continued for green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids at various concentrations. Fig. 2(a) - (c) showed samples of green Bio-glycol based nanofluids in the test tube at different concentrations. All samples were prepared and sonicated up to 3 h sonication time. The samples were observed immediately just after preparation (first day), after a week (7 days), and after a month (30 days). The samples were found to have dispersed well with no settlement of nanoparticles at the bottom of the test tube for the observation on the first day of preparation as shown in Fig. 2(a). After several days of preparation, the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids displayed excellent stability condition without any visible sedimentation as shown in Fig. 2(b). Lastly after 30 days observation, a small separation layer was found at the top of the sample for each concentration and shown in Fig. 2(c). Nevertheless, the liquids at the top of the layer was observed to be not completely clear. These circumstances confirmed that the nanoparticles were not totally separated from the mixture.

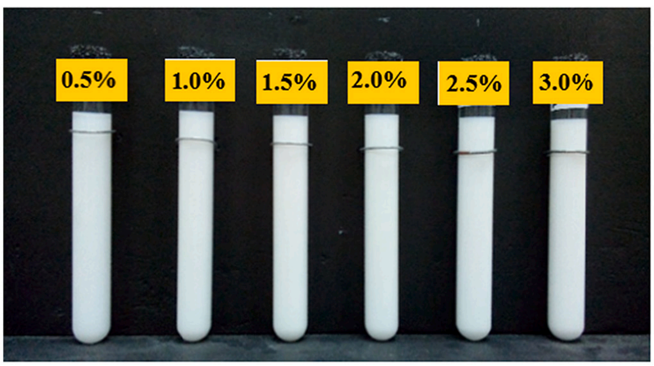
The small visible separation layer at the upper part of the samples exhibited different thicknesses of the visible layers. Fig. 2(c) displayed the sequence of thickness layer by volume concentration and given by 0.5% > 1.0% > 1.5% > 2.0% > 2.5% > 3.0%. The thickness of the visible layers at low volume concentration is higher than the sample at higher concentrations. The thickness layer decreases with increasing volume concentration. The small amount of particles at lower concentrations is inadequate to produce the repulsive force to counteract the attraction between the particles caused by Van der Waals forces; hence contributing to unstable suspension over time [57,58]. Although no surfactant was used in the preparation, all samples were considered to be in a stable condition with minimal thickness layer in the test tubes. The present green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids displayed little sedimentation for all concentrations, which is probably due to the attraction between particles to form agglomeration caused by the Van der Waals forces. After 30 days, a small portion of the agglomerate particles settled down at the bottom of the test tube due to the gravitational forces [28,59].

#### 3.1.2. Ultraviolet-visible spectrophotometer evaluation

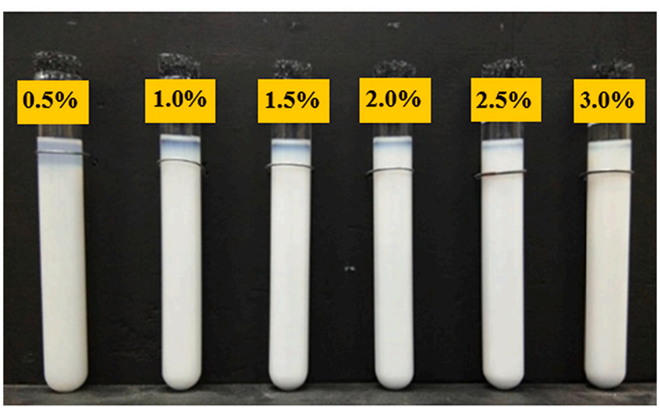
In the stability analysis, the UV–Vis spectrophotometer was used in determining the change in absorbance ratio over a specified period of time. The similar samples of green Bio-glycol based nanofluids at 0.1% volume concentration and different sonication times between 0.0 and




(a) First day




(b) 7 days




(c) 30 days

**Fig. 2.** Nanofluid samples at different volume concentrations.

3.0 h were used in the UV–Vis spectrophotometer evaluation. Fig. 3 indicates the absorbance ratio of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids relative to the base fluids of water/Bio-glycol (W/BG) mixture. The optimal value for the absorbance ratio is equal to one or 100%. At this condition, the suspended nanoparticles was known to be stable without any sedimentation taking place throughout the sedimentation period [6]. Hence, the actual absorbance ratio of the green Bio-glycol based nanofluids relative to the ideal absorbance ratio condition of 1.0 will determine the stability condition of the sample, which varies with the sonication and sedimentation time.

According to Fig. 3, the sample without sonication (represented by 0.0 h) was recorded with the lowest absorbance ratio and found to be unstable. After a few hours of preparation, the absorbance ratio declined rapidly to 90% (0.9 ratio) and after 50 h continues to fall up to 20% (0.2 ratio). The nanofluid samples with sonication method showed significantly improved absorbance ratio for longer sedimentation time. These

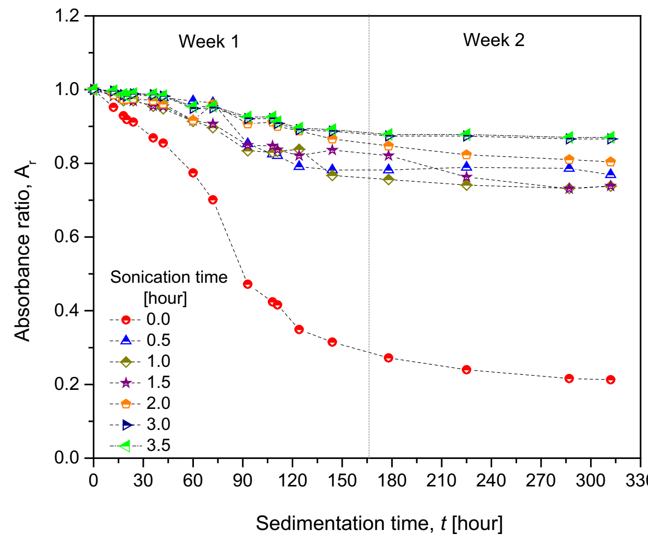



**Fig. 3.** Variation of absorbance ratio for different sonication time.

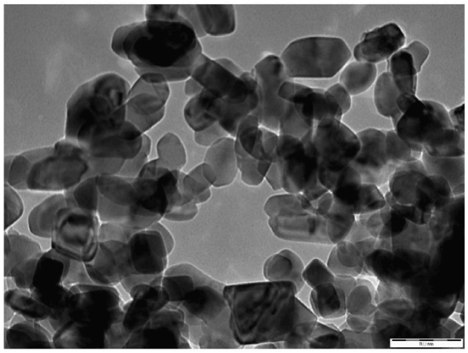
findings imply that the stability of the nanofluids is strongly influenced by the sonication time. The sample with 3.0 h sonication was observed to have the most stable condition. The absorbance ratio of the sample was maintained at more than 80% for up to 300 h of sedimentation time. The sonication time of 3 h or 180 min was recommended in the next stage of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluid preparation, thereby leading to the improvement of the stability condition. Therefore, the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids at different volume concentrations were prepared with sonication time up to 3.0 h to formulate the coolants with long-lasting stability. However, it was noticed that additional sonication time of more than 3.0 h did not give any substantial improvement in the stability as shown in Fig. 3 for 3.5 h. Hence, the evaluation results of absorbance ratio were presented for up to 3.0 h only.

### 3.1.3. *Transmission electron microscopy evaluation*

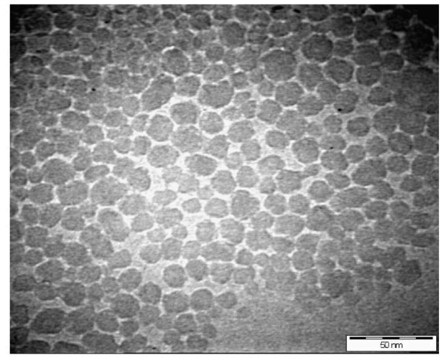
The physical characterization of $TiO_2$-$SiO_2$ nanoparticles was acquired using the transmission electron microscopy (TEM) technique. The arrangement of the single nanoparticles, $TiO_2$ and $SiO_2$ in the base fluids are shown in Fig. 4(a) and (b), respectively. The shape of $TiO_2$ nanoparticles is non-spherical as shown in Fig. 4(a) whereas $SiO_2$ nanoparticles is almost spherical as observed in Fig. 4(b). Fig. 4(c) displayed the dispersion of hybrid $TiO_2$-$SiO_2$ nanoparticles in the base fluids. In the figure, the large particles represent $TiO_2$, while the smaller particles represent $SiO_2$. Minimum agglomeration can be seen in the figure when the nanoparticles stick to one another for the same type of nanoparticles. It was happened as a result of collision from their random Brownian motion meanwhile different size of the two nanoparticles lead to the gravitational agglomeration [60]. However, no significant agglomeration and clustering was observed for the suspended nanoparticle suspension in the base fluids. The present nanofluids were sonicated for up to 3.0 h, which eventually causes the separation of agglomerated particles. Based on Fig. 4(c), the $TiO_2$ and $SiO_2$ nanoparticles showed good dispersion in the base solution.

### 3.1.4. *Zeta potential analysis*

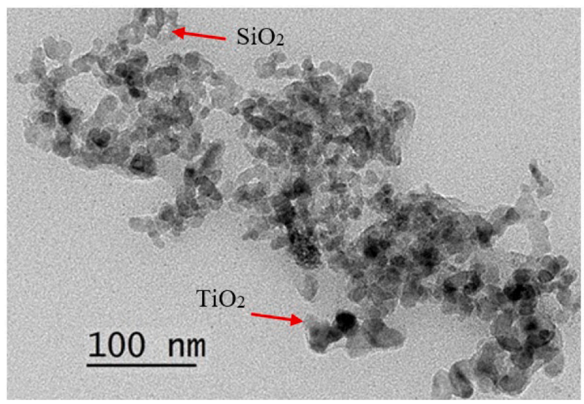
Further confirmation on the stability of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids was done by measuring the zeta potential. Zeta potential is defined as the potential difference between the electrical double layer attached to the particles [61]. It is a parameter that determines the stability condition of the colloidal suspension by measuring the magnitude of electrostatic repulsion and attraction between the particles. The value of zeta potential serves as an indicator to determine and



(a) $TiO_2$ nanoparticles (X 140,000)




(b) $SiO_2$ nanoparticles (X 35,000)




(c) $TiO_2$-$SiO_2$ nanoparticles (X 39,000)

**Fig. 4.** Characterization of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids using TEM images. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

predict the potential stability of the nanoparticle suspension in the base fluids. By knowing the magnitude of zeta potential of a suspension, it will reduce the time required to produce a trial sample of the nanofluids. Based on the zeta potential value, the colloidal suspension can have different stability behaviors, which are pronounced aggregation ($\zeta < |5|$ mV), limited stability ($\zeta < |20|$ mV) and physically stable ($\zeta > |30|$ mV). When $\zeta > |60|$ mV, the colloidal suspension is considered to be in excellent stability [62]. Nanofluids with small negative or positive zeta potential are said to be less stable compared to the nanofluids with large negative or positive zeta potential due to the repulsive force between particles. This force prevents the particles from adhering to one another and form agglomeration, thus resulting in a stable colloidal suspension.

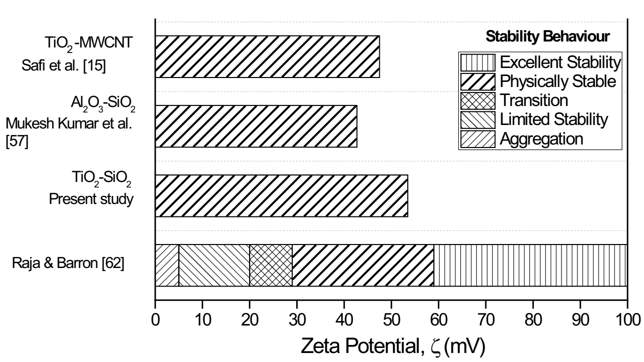



**Fig. 5.** Zeta potential of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids in comparison to literature. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 5 displays the zeta potential of the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids for the present study. The zeta potential of the nanofluids in the present study is −53.46 mV, which falls under the category of physically stable. This result supports the outcomes from previous stability evaluations and confirms the long-lasting formulation of green Bio-glycol based nanofluids in the present study. The stability analysis by zeta potential was used by various researchers such as Mukesh Kumar et al. [57] and Safi et al. [15] to measure the degree of stability. The value of the zeta potential from their investigations is presented in Fig. 5 and compared with the present study. All the hybrid nanofluids in the figure exhibited good stability conditions. However, among the hybrid particles in the figure, the combination of $TiO_2$-$SiO_2$ particles from the present study provides the highest zeta potential.

### 3.1.5. *Density observation*

At stationary fluid conditions, the settling rate was influenced by density of the final solution [63]. The difference in density between nanoparticles and base fluids will cause sedimentation of particles in the solution. By minimizing the density difference between nanoparticles and base fluids, the sedimentation velocity can be reduced [30]. Hence, the present study also attempted to study the stability condition of the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids by observing the density over time. The density of green Bio-glycol based nanofluids was measured at various concentrations from 0.5 to 3.0%. The variation in the density of green Bio-glycol based nanofluids is illustrated in Fig. 6. From the figure, the density of the green Bio-glycol based nanofluids was observed to increase with volume concentrations. Besides that, there was no significant drop of the density measurement during the 30 days

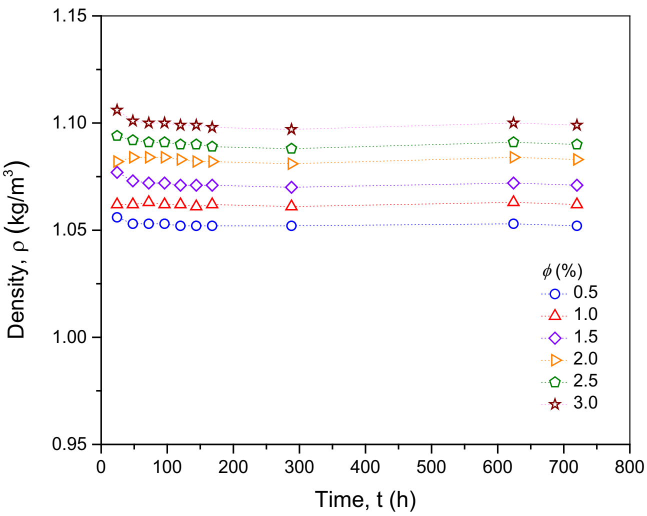



**Fig. 6.** Density of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids at room temperature. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

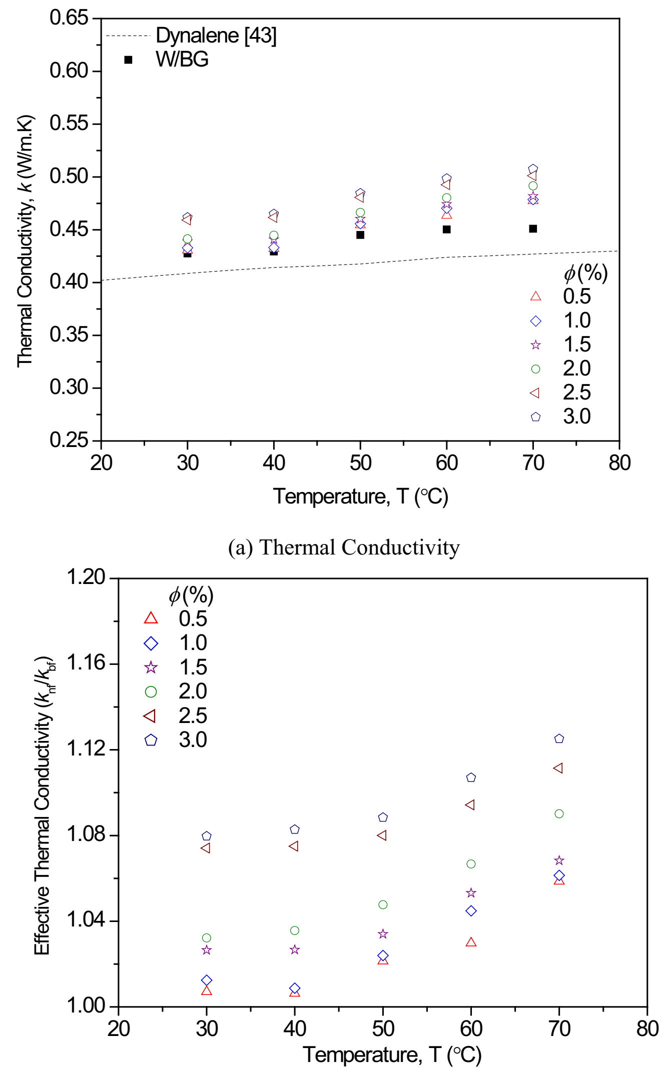
of the evaluation. The measurements of the minimum and maximum drop in the density from the first day until the last day were 0.0% and 0.81%, respectively. The results implied that the nanoparticles remained suspended in the base fluids without significant settling throughout the observation. Therefore, the present green Bio-glycol based nanofluids samples at different volume concentrations were concluded to be stable over time.

### *3.2. Thermo-physical properties of green bio-glycol based nanofluids*

#### *3.2.1. Thermal conductivity evaluation*

The thermal conductivity was validated by comparing the present data of the W/BG (60:40) mixture with the data provided by Dynalene [43]. Fig. 7(a) indicates the thermal conductivity of the base fluids and green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids at different temperatures and volume concentrations. The thermal conductivity of the W/BG mixture increased with increasing temperature. The thermal conductivity measurement of the base fluids in the present study showed a good agreement with the data provided by Dynalene [43], with a maximum deviation of 6.6%. Therefore, the present thermal conductivity measurement was considered reliable due to the small deviation in comparison to the standard data from Dynalene [43].

The variation of thermal conductivity for green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids with volume concentrations of 0.5 to 3.0% and temperatures of 30 to 70 °C are demonstrated in Fig. 7(a). The thermal conductivity of the green Bio-glycol based nanofluids was observed to increase with volume concentration and temperature. The maximum thermal conductivity was measured at a temperature of 70 °C and 3.0% volume concentration, while the lowest thermal conductivity was recorded at volume concentration of 0.5% and 30 °C temperature. The mechanism behind the enhancement of thermal conductivity with temperature in the present study was due to a frequent collision between the particles caused by the fast-moving molecules in the fluids; hence magnifying the elevated temperatures [31]. The phenomenon of the random movement of particles in the fluids that collide with each other is called Brownian motion. The particles can transfer heat from one to another directly, and at high temperatures, the rate of heat transport increases with increasing kinetic energy [64]. Overall, significant thermal conductivity increment of the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids that is higher than the base fluids of W/BG mixture was observed for all volume concentrations. A similar trend has been reported by other scholars, including Kumar and Sonawane [65] and Nabil et al. [6].




(a) Thermal Conductivity

(b) Effective Thermal Conductivity

**Fig. 7.** Thermal conductivity of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

It is crucial to understand the variation of thermal conductivity with the addition of nanoparticles in the base fluids. Hence, the thermal conductivity of green Bio-glycol based nanofluids in the present study was compared to the thermal conductivity of the base fluids of W/BG mixture. The relative thermal conductivity of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids or effective enhancement of the nanofluids is shown in Fig. 7(b). The relative thermal conductivity of the green Bio-glycol based nanofluids increased with the increase of volume concentration and temperature. Fig. 7(b) also revealed that temperature plays a significant role in the improvement of effective thermal conductivity of green Bio-glycol based nanofluids. The thermal conductivity of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids was found to increase with temperature for all volume concentrations; however, with exception at 0.5% and 1.0% volume concentrations at 40 °C temperature. The lowest relative thermal conductivity was observed at 0.5% volume concentration, with an improvement of 0.71% at 30 °C. Meanwhile, the maximum enhancement in thermal conductivity was recorded at 3.0% volume concentration and temperature of 70 °C. The increment reached up to 12.52% higher than the base fluids. Generally, the relative thermal conductivity of the present green Bio-glycol based nanofluids at all volume concentrations and temperatures recorded were always higher than the base fluids of W/BG mixture. The result indicates the applicability of the present green Bio-glycol based nanofluids for heat transfer applications. Further investigation on heat transfer performance is required to confirm the pertinence of the present green nanofluids.

### 3.2.2. Dynamic viscosity evaluation

Initially, a validation process was carried out on the equipment by measuring the dynamic viscosity of the base fluids of W/BG mixture. This process was undertaken in order to confirm the reliability of the present measurement data by comparing the experimental result of the base fluids with the standard data provided by Dynalene [43]. The present experimental data was found to be in good agreement with the data from the manufacturer [43] with a maximum of 0.75% deviation as shown in Fig. 8(a). The measured data for the base fluids of W/BG mixture exhibited a similar trend to the standard data given by Dynalene [43], thereby confirming the reliability of the viscometer instrument in the present study.

The dynamic viscosity variation for green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids in the temperature range of 20 to 80 °C and volume concentrations of 0.5 to 3.0% are demonstrated in Fig. 8(a). Based on the figure, the dynamic viscosity of the green Bio-glycol based nanofluids was observed to be slightly higher than the base fluids. The reason behind the increment is possibly due to the increase in the fluid internal shear stress with the dispersion of nanoparticles in the base fluids [35]. The figure also implied that the dynamic viscosity of the green Bio-glycol based nanofluids increased with volume concentration due to the increase in particle loading at high volume concentrations. The highest dynamic viscosity was observed at 3.0% volume concentrations, while minimum dynamic viscosity increment was recorded at the lowest volume concentration of 0.5%. In contrast to the volume concentration, the dynamic viscosity of the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids decreased exponentially with the operating temperature. The minimum dynamic viscosity was noticed at the maximum operating temperature of 80 °C. At the elevated temperature, the kinetic energy increases, and the intermolecular interactions between the molecules become weak; hence lowering the dynamic viscosity [33,66]. Therefore, the present findings confirm the dynamic viscosity dependency of green Bio-glycol based nanofluids on the operating temperatures and volume concentrations.

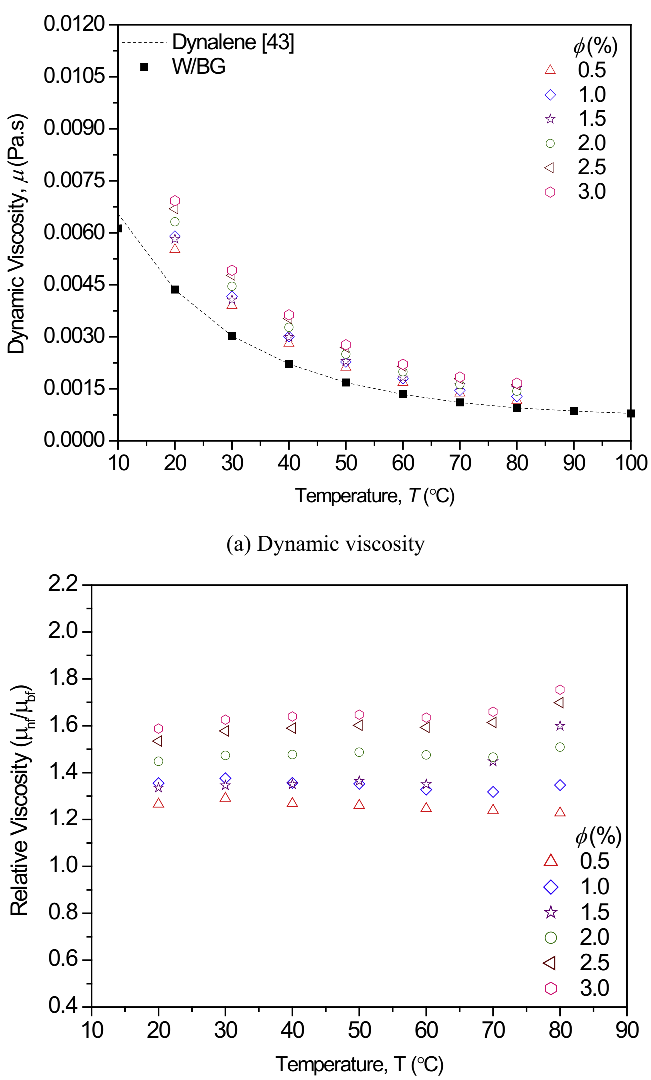


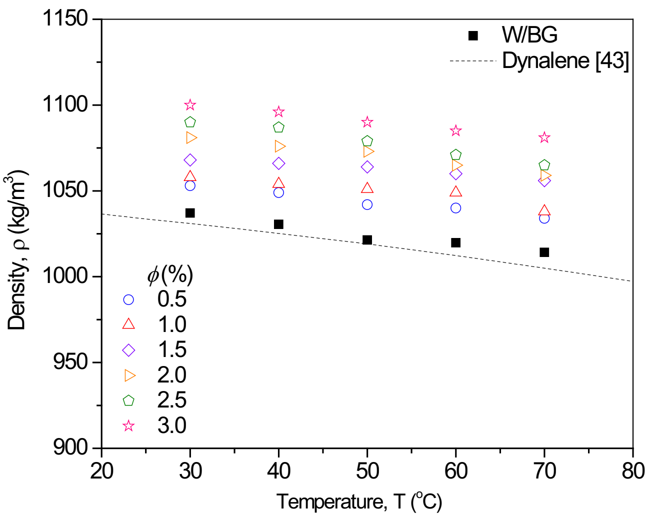

**Fig. 8.** Dynamic viscosity of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 8(b) illustrates the variation in the relative viscosity of green Bio-glycol based nanofluids with volume concentration and temperature. From the figure, it was confirmed that the addition of nanoparticles in the base fluids caused an increase in the dyamic viscosity of the base fluids. At the temperature of 20 to 70 °C, the relative viscosity of the nanofluids are almost constant for all volume concentrations. In contrast, a slight increase of relative viscosity was observed at 80 °C temperature. The dynamic viscosity of green Bio-glycol based nanofluids increased up to 1.75 times relative to the base fluids at temperature of 80 °C and volume concentration of 3.0%. From the analysis, it can be inferred that the relative viscosity increased with concentration while the increment rate of relative viscosity is significant at high temperatures when compared to the low temperatures. The reason for the fluctuation of the relative viscosity within the temperature and volume concentration ranges was undetermined. Regardless, this behaviour which affects the effective volume concentration and the viscosity of the suspension may be related to the difference in structure and thickness of the diffused fluid layers around the nanoparticles in the base fluids [67].

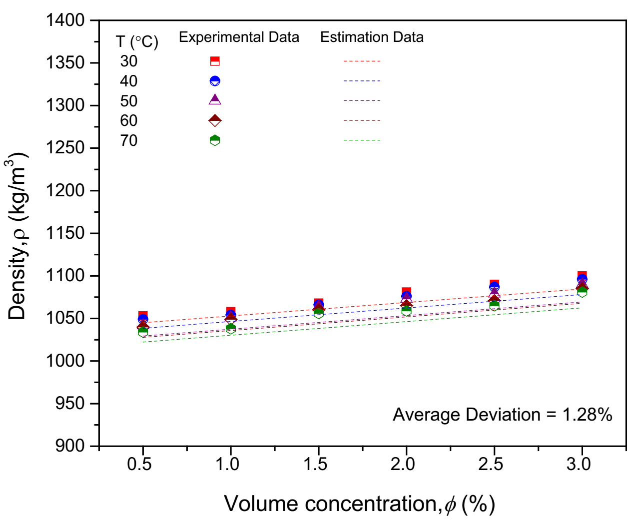
### 3.2.3. Density evaluation

The density of the mixture was validated by comparing the present experimental data for the W/BG mixture with the standard data provided by Dynalene [43]. Fig. 9(a) indicates the density of the base fluids and green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids at different volume concentrations and temperatures. In general, the density decreased with increasing temperature. The maximum deviation for the density measurement of W/BG mixture was found to be less than 1.0% at the temperature of 40 °C. Therefore, the density measurement of the present study using density meter is considered reliable with minimum error. Fig. 9(a) also illustrated the density measurement for the present green Bio-glycol based nanofluids at different volume concentrations and temperatures. The results show that the density of the nanofluids increased with increasing volume concentrations and decreased with increasing temperatures. At the elevated temperatures, the molecules in the fluid tends to excite and move randomly at higher speed; hence allowing the collision rate to increase. This behaviour alters the volume of the liquids and leads to a decrease in density. This trend is consistent with the published data available in literatures such as that presented by Nabati Shoghl et al. [37] and Kishore et al. [3].

The mixture relation equation in the literature was used to estimate the density of the green Bio-glycol based nanofluids. The data of density for green Bio-glycol based nanofluids obtained from the density meter can be predicted accurately using the mixture relation and comparing with the present experimental data. Fig. 9(b) presents the comparison between the present experimental data and the mixture relation equation for the density of green Bio-glycol based nanofluids at temperatures of 30 to 70 °C. The experimental data showed a marginal increment value of density compared to the mixture relation model. As seen in the




(a) Density of nanofluids




(b) Comparison with mixture relation model

**Fig. 9.** Density of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

figure, the mixture relation slightly underestimates the density in comparison to the experimental data. However, the deviation of mixture relation is still acceptable in the prediction of the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluid density for the present study. The average deviation between the experimental and mixture relation equation was recorded at 1.28%. Therefore, the present data from the measurement is considered reliable.

### 3.2.4. Specific heat evaluation

Fig. 10 showed the specific heat variation of the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids with temperature, which was estimated using mixture relation equation. From the figure, the specific heat of green Bio-glycol based nanofluids was observed to be lower than the base fluid at all concentrations. The increase in particle concentration reduced the specific heat of the nanofluids. He et al. [68] stated that the increase or decrease in specific heat was influenced by the specific heat of nanoparticles. If the specific heat of nanoparticles is higher than the base fluid, the specific heat of nanofluids will increase. The results observed in the present study are consistent with other studies conducted by Akilu et al. [40] and Wole-Osho et al. [39]. They reported a

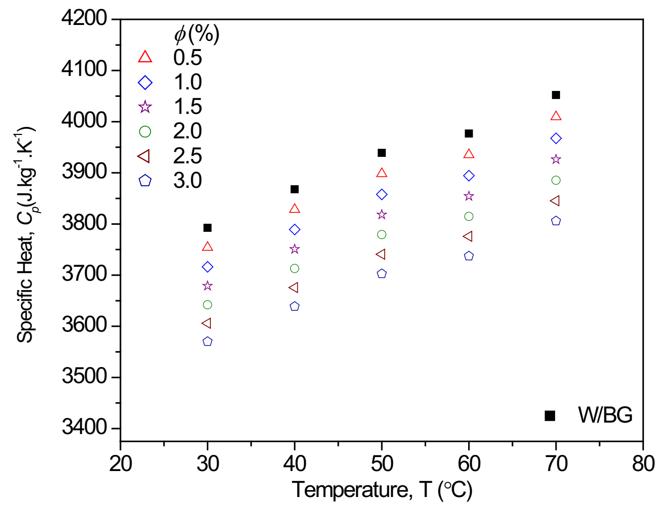
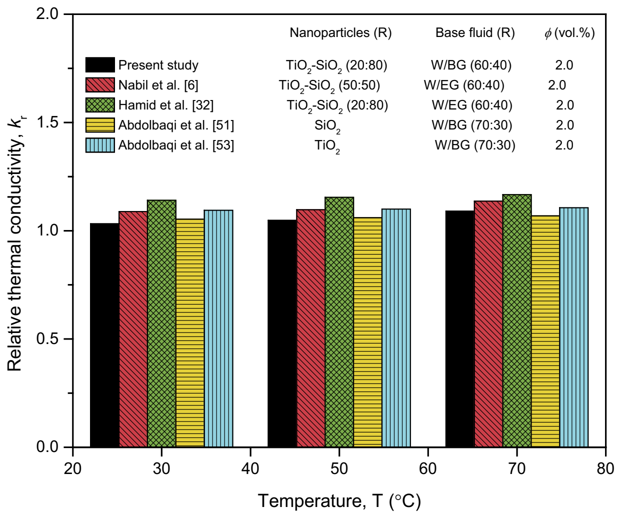



**Fig. 10.** Specific heat of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

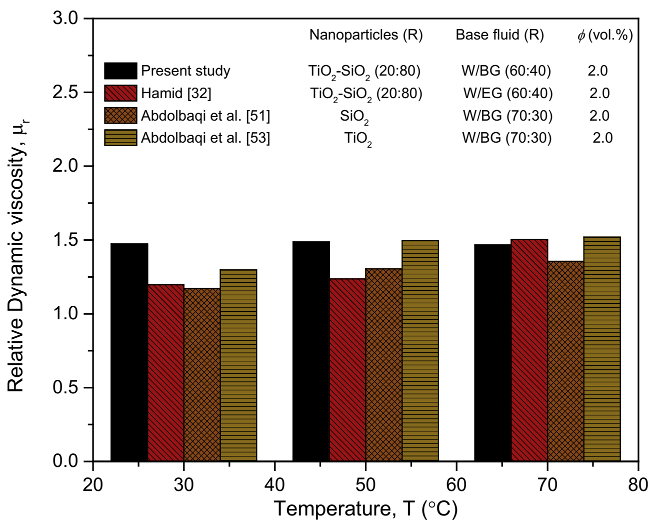
decrease in specific heat of $SiO_2$ nanofluids and $Al_2O_3$-ZnO water hybrid nanofluids, respectively when the volume concentration increases. According to Wole-Osho et al. [39], the specific heat of $Al_2O_3$-ZnO hybrid nanoparticles are lower than the base fluid, and therefore increasing the concentration of nanoparticles will decrease the specific heat of the nanofluids. In contrast, it was observed that the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids was positively influenced by the temperature. Vajjha and Das [42] stated that nanoparticles possess a higher thermal diffusivity compared to the base fluid; thus, allowing them to absorb more heat and obtain a higher temperature than the base fluid. From the energy balance, the increase in temperature of particles in the base fluid resulted in a higher specific heat of nanofluids.

### 3.2.5. Comparison with literature

Fig. 11(a) and (b) show the comparison of effective thermal conductivity and relative viscosity of the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids, respectively with the previous investigations in literature. In Fig. 11(a), the comparison of the effective thermal conductivity for nanofluids was made at 2.0% volume concentration. The present study used a combination of $TiO_2$-$SiO_2$ nanoparticles at a composition ratio of 20:80 and dispersed in 60:40 W/BG mixture. The thermal conductivity of the green Bio-glycol based nanofluids in the present study increased from 3.22 to 9.01% higher than the W/BG base mixture for the temperature range between 30 and 70 °C. At a similar nanoparticle composition ratio of $TiO_2$-$SiO_2$ in the mixture of W/EG, Hamid et al. [32] found the relative thermal conductivity to be 1.85 times higher than the present study. In another study conducted by Nabil et al. [6] using W/EG as the base fluid, the relative thermal conductivity of $TiO_2$-$SiO_2$ nanoparticles at composition ratio of 50:50 was observed to be 1.52 times higher than the present study at a temperature of 70 °C. Meanwhile, Abdolbaqi et al. [53] and Abdolbaqi et al. [45] measured the thermal conductivity of $TiO_2$ and $SiO_2$ nanofluids in 70:30 W/BG and found that in comparison to the present study, the enhancement of $SiO_2$ nanofluids was slightly lower and $TiO_2$ nanofluids was observed to be slightly higher than the present study at a temperature of 70 °C. Overall, the present study shows a slightly lower enhancement in thermal conductivity when compared to the nanofluids in the literature. However, the present study used green Bio-glycol as the base fluid, which is completely renewable in comparison to other studies that mostly used ethylene glycol as the main base solution. In addition, the discrepancy in the enhancement of thermal conductivity is possibly due to various factors such as the nanoparticle composition ratio and the base fluids that were employed in the present study.




(a) Comparison of thermal conductivity



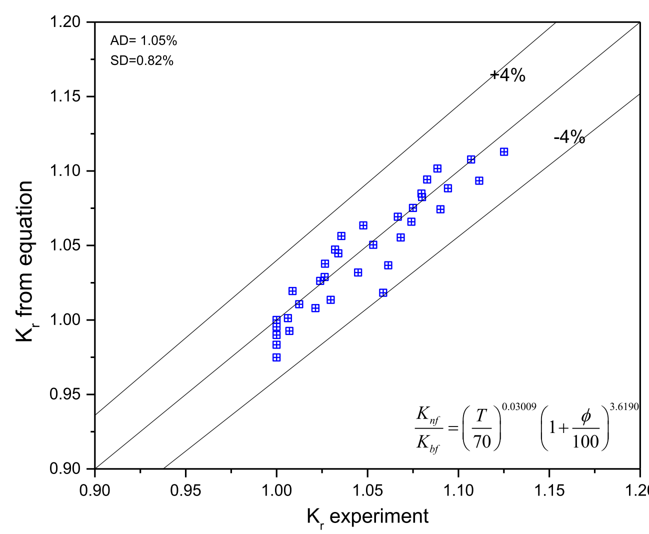

(b) Comparison of dynamic viscosity

**Fig. 11.** Comparison of $TiO_2$-$SiO_2$ nanofluids properties with data from literatures.

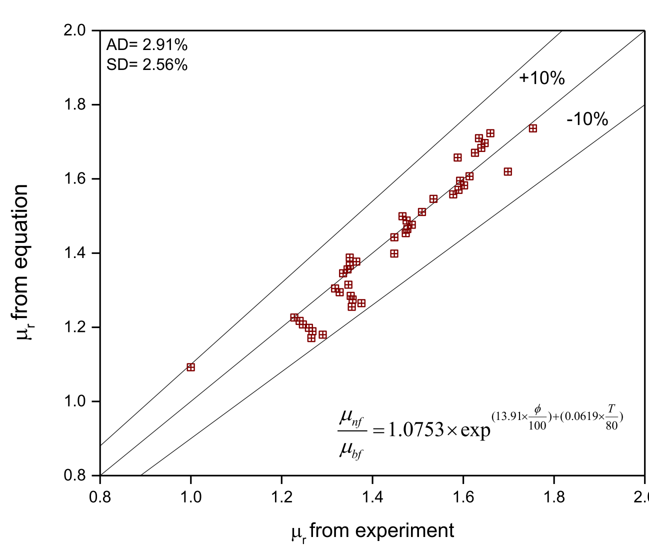
Fig. 11(b) displays a comparison of relative viscosity between the present study and previously published data in literature. The relative viscosity of the nanofluids is almost constant as the temperature increases. Based on Fig. 11(b), the present study demonstrates a higher relative viscosity than Hamid et al. [32] for $TiO_2$-$SiO_2$ nanoparticles in 60:40 W/EG mixture. However, the viscosity difference becomes smaller with the increase of temperature and at 70 °C, the $TiO_2$-$SiO_2$ nanoparticles in W/EG mixture was 1.08 times higher than the present study. The relative viscosity of present green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids was also compared with the single $TiO_2$ and $SiO_2$ nanofluids in 70:30 W/BG base mixture [45,53]. The relative dynamic viscosity of the present study is 1.30 times higher than the $SiO_2$ nanofluids; however, 1.11 times lower than the $TiO_2$ nanofluids at a temperature of 70 °C. While the studies of nanofluids with the mixture of water and Bio-glycol as base fluids is limited in literature, the present findings indicate that the type of base fluid plays an important role in the increment of the dynamic viscosity. The present results suggest that the magnitude of enhancement in thermal conductivity and relative viscosity is strongly dependent on the type of nanoparticles, type of base fluids [27], and nanoparticle composition ratio, as demonstrated in Fig. 11(a) and (b).

### 3.3. Regression correlations for properties

Eqs. (9) and (10) were developed for the estimation of thermal conductivity and dynamic viscosity of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids, respectively. The equations were developed with consideration of the important input parameters of volume concentration and temperature. The average deviation (AD), standard deviation (SD), and maximum deviation of Eq. (9) were obtained at 1.05%, 0.82%, and 3.82%, respectively, for the estimation of thermal conductivity. Meanwhile, Eq. 10 was developed with 2.91%, 2.56%, and 9.21% of average deviation (AD), standard deviation (SD), and maximum deviation, respectively. These equations are applicable for green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids with nanoparticle composition ratio of 20:80 dispersed in the base fluids of W/BG mixture at 60:40 ratio with volume concentrations up to 3.0% and temperature range from 30 to 70 °C. The experimental data are observed to be in good agreement with the estimated values from Eqs. (9) and (10). Fig. 12(a) and (b) illustrate the comparison of the estimated value for thermal conductivity and dynamic viscosity plotted against the present experimental data, respectively. Fig. 12(a) presents the estimation for thermal conductivity of




(a) Comparison of thermal conductivity ratio with Eq. (6)




(b) Comparison of dynamic viscosity ratio with Eq. (7)

**Fig. 12.** Properties regression equations for thermal conductivity ratio and viscosity ratio.

green Bio-glycol based nanofluids with accuracy within 4% deviation interval whereas Fig. 12(b) shows the estimation for dynamic viscosity of green Bio-glycol based nanofluids with precision of less than 10% deviation.

$$k_r = \frac{k_{cnf}}{k_{bf}} = \left(\frac{T}{70}\right)^{0.03009}\left(1+\frac{\phi}{100}\right)^{3.619} \tag{9}$$

$$\mu_r = \frac{\mu_{nf}}{\mu_{bf}} = 1.0753exp^{\left(13.91\times\frac{\phi}{100}\right)+\left(0.0619\times\frac{T}{80}\right)} \tag{10}$$

## 4. Conclusions

This study reported the enhancement in heat transfer using $TiO_2$-$SiO_2$ nanofluids at 20:80 composition ratio for 0.5 to 2.5% volume concentrations. The green Bio-glycol based nanofluids were observed to be in the most stable condition with the highest absorbance ratio at 3.0 h sonication process time. The preparation of the green Bio-glycol based nanofluids with sonication time up to 3.0 h also improved and maintained the absorbance ratio for more than 80% after 300 h of preparation. Furthermore, the stability of the $TiO_2$-$SiO_2$ nanofluids at different volume concentrations were observed to be in good stability for more than 30 days with minimum sedimentation. In addition, the TEM imaging analysis also confirmed the existence of $TiO_2$ and $SiO_2$ nanoparticles in the suspension without significant agglomeration. On the other hand, the quantitative evaluation by density evaluation over time was recorded with no significant changes in density reading for 30 days of observation whereas the quantitative evaluation by zeta potential evaluation reported up to −53.46 mV measurement. The high value of zeta potential exhibited by the green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids indicated a physically stable condition. The results from these stability evaluations by qualitative and quantitative methods confirmed the good stability condition of the present green Bio-glycol based nanofluids, and therefore it is ready for further properties characterizations and heat transfer evaluations.

The thermal conductivity was evaluated at operating temperatures of 30 to 70 °C. The maximum thermal conductivity was observed at 70 °C temperature and volume concentration of 3.0%. The green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids enhanced the thermal conductivity up to 12.52% higher than the base fluids of W/BG mixture. In contrast, the dynamic viscosity of the green Bio-glycol based nanofluids exponentially decreased with temperature. In the range of temperatures between 20 and 80 °C, the minimum dynamic viscosity of green Bio-glycol based $TiO_2$-$SiO_2$ nanofluids appeared at 80 °C temperature and 0.5% volume concentration. Besides that, the dynamic viscosity increased up to 75.38% higher than the base fluids at 80 °C temperature and 3.0% volume concentration. In addition, the development of regression correlations for the estimation of thermal conductivity and dynamic viscosity showed good agreement with the present experimental data with average deviations of 1.05% and 2.91%, respectively. However, these equations are only applicable for the green Bio-glycol based nanofluids for the range of temperature between 20 and 80 °C and volume concentrations of 0.5 to 3.0%. The green Bio-glycol based nanofluids was observed to be temperature-dependent fluids at all volume concentrations.

## Declaration of Competing Interest

None.

## Acknowledgements

The authors are grateful to the Ministry of Higher Education Malaysia and Universiti Malaysia Pahang (www.ump.edu.my) for financial supports given under FRGS/1/2018/TK10/UMP/02/10, RDU190146 and PGRS1903110. The authors also thank the research team from Centre for Research in Advanced Fluid and Processes (Pusat Bendalir) and Advanced Automotive Liquids Laboratory (AALL), who provided insight and expertise that greatly assisted in the present research work.

## References

[1] S.N.M. Zainon, W.H. Azmi, Recent progress on stability and thermo-physical properties of mono and hybrid towards green nanofluids, Micromachines 12 (2) (2021) 1–35.

[2] H. Masuda, A. Ebata, K. Teramae, N. Hishinuma, Y. Ebata, Alteration of thermal conductivity and viscosity of liquid by dispersing ultra-fine particles (dispersion of γ-$Al_2O_3$, $SiO_2$ and $TiO_2$ ultra-fine particles), Netsu Bussei 7 (4) (1993) 227–233.

[3] P.S. Kishore, V. Sireesha, V. Sree Harsha, V. Dharma Rao, A. Brusly Solomon, Preparation, characterization and thermo-physical properties of Cu-graphene nanoplatelets hybrid nanofluids, Mater. Today 27 (2020) 610–614.

[4] A. Huminic, G. Huminic, C. Fleacă, F. Dumitrache, I. Morjan, Thermo-physical properties of water based lanthanum oxide nanofluid. An experimental study, J. Mol. Liq. 287 (2019) 1–9.

[5] N. Ali, J.A. Teixeira, A. Addali, A review on nanofluids: fabrication, stability, and thermophysical properties, J. Nanomater. 2018 (2018) 33.

[6] M.F. Nabil, W.H. Azmi, K.A. Hamid, N.N.M. Zawawi, G. Priyandoko, R. Mamat, Thermo-physical properties of hybrid nanofluids and hybrid nanolubricants: a comprehensive review on performance, Int. Commun. Heat Mass Transf. 83 (2017) 30–39.

[7] W.H. Azmi, K. Abdul Hamid, R. Mamat, K.V. Sharma, M.S. Mohamad, Effects of working temperature on thermo-physical properties and forced convection heat transfer of $TiO_2$ nanofluids in water – ethylene glycol mixture, Appl. Therm. Eng. 106 (2016) 1190–1199.

[8] M.M. Elias, I.M. Mahbubul, R. Saidur, M.R. Sohel, I.M. Shahrul, S. S. Khaleduzzaman, S. Sadeghipour, Experimental investigation on the thermo-physical properties of $Al_2O_3$ nanoparticles suspended in car radiator coolant, Int. Commun. Heat Mass Transf. 54 (2014) 48–53.

[9] H.W. Chiam, W.H. Azmi, N.A. Usri, R. Mamat, N.M. Adam, Thermal conductivity and viscosity of $Al_2O_3$ nanofluids for different based ratio of water and ethylene glycol mixture, Exp. Thermal Fluid Sci. 81 (2017) 420–429.

[10] S.H. Pourhoseini, N. Naghizadeh, An experimental study on optimum concentration of silver-water microfluid for enhancing heat transfer performance of a plate heat exchanger, J. Taiwan Inst. Chem. Eng. 75 (2017) 220–227.

[11] G. Paul, J. Philip, B. Raj, P.K. Das, I. Manna, Synthesis, characterization, and thermal property measurement of nano-Al95Zn05 dispersed nanofluid prepared by a two-step process, Int. J. Heat Mass Transf. 54 (15) (2011) 3783–3788.

[12] X. Li, W. Chen, C. Zou, The stability, viscosity and thermal conductivity of carbon nanotubes nanofluids with high particle concentration: a surface modification approach, Powder Technol. 361 (2020) 957–967.

[13] A. Hameed, A. Mukhtar, U. Shafiq, M. Qizilbash, M.S. Khan, T. Rashid, C.B. Bavoh, W.U. Rehman, A. Guardo, Experimental investigation on synthesis, characterization, stability, thermo-physical properties and rheological behavior of MWCNTs-kapok seed oil based nanofluid, J. Mol. Liq. 277 (2019) 812–824.

[14] W.U. Rehman, Z.M.A. Merican, A.H. Bhat, B.G. Hoe, A.A. Sulaimon, O. Akbarzadeh, M.S. Khan, A. Mukhtar, S. Saqib, A. Hameed, N. Mellon, H. Ullah, S. Ullah, M.A. Assiri, Synthesis, characterization, stability and thermal conductivity of multi-walled carbon nanotubes (MWCNTs) and eco-friendly jatropha seed oil based nanofluid: an experimental investigation and modeling approach, J. Mol. Liq. 293 (2019) 1–15.

[15] M.A. Safi, A. Ghozatloo, N.M. Shariaty, A.A. Hamidi, Preparation of MWNT/$TiO_2$ nanofluids and study of its thermal conductivity and stability, Iran. J. Chem. Eng. 11 (4) (2014) 3–9.

[16] S. Suresh, K.P. Venkitaraj, P. Selvakumar, M. Chandrasekar, Effect of $Al_2O_3$–Cu/water hybrid nanofluid in heat transfer, Exp. Thermal Fluid Sci. 38 (2012) 54–60.

[17] W.H. Azmi, N.A. Usri, R. Mamat, K.V. Sharma, M.M. Noor, Force convection heat transfer of $Al_2O_3$ nanofluids for different based ratio of water: ethylene glycol mixture, Appl. Therm. Eng. 112 (2017) 707–719.

[18] N.A.C. Sidik, I.M. Adamu, M.M. Jamil, G.H.R. Kefayati, R. Mamat, G. Najafi, Recent progress on hybrid nanofluids in heat transfer applications: a comprehensive review, Int. Commun. Heat Mass Transf. 78 (2016) 68–79.

[19] S.U.S. Choi, J. Eastman, Enhancing thermal conductivity of fluids with nanoparticles, in: D.A. Siniger, H.P. Wang (Eds.), Developments and Applications of Non-Newtonian Flows, American Society of Mechanical Engineers (ASME), New York, 1995, pp. 99–105.

[20] M. Kole, T.K. Dey, Thermal conductivity and viscosity of $Al_2O_3$ nanofluid based on car engine coolant, J. Phys. D. Appl. Phys. 43 (31) (2010) 315501.

[21] M. Ali, A.M. El-Leathy, Z. Al-Sofyany, The effect of nanofluid concentration on the cooling system of vehicles radiator, Adv. Mech. Eng. 6 (2014) 962510.

[22] M. Fares, M. Al-Mayyahi, M. Al-Saad, Heat transfer analysis of a shell and tube heat exchanger operated with graphene nanofluids, Case Stud. Thermal Eng. 18 (2020) 100584.

[23] P.S.A. Banu, A. Krishnan, S.M. Shafee, A.G.S. Raj, Numerical investigation of micro-pin-fin heat exchanger using nanofluids, Mater. Today 22 (2020) 1020–1025.

[24] P. Selvakumar, S. Suresh, Use of $Al_2O_3$-Cu/water hybrid nanofluid in an electronic heat sink, IEEE Trans. Compon. Packag. Manuf. Technol. 2 (10) (2012) 1600–1607.

[25] N.N.M. Zawawi, W.H. Azmi, A.A.M. Redhwan, M.Z. Sharif, K.V. Sharma, Thermo-physical properties of $Al_2O_3$ -$SiO_2$ /PAG composite nanolubricant for refrigeration system, Int. J. Refrig. 80 (2017) 1–10.
[26] S. Koçak Soylu, İ. Atmaca, M. Asiltürk, A. Doğan, Improving heat transfer performance of an automobile radiator using Cu and Ag doped $TiO_2$ based nanofluids, Appl. Therm. Eng. 157 (2019) 1–9.
[27] L.S. Sundar, K.V. Sharma, M.K. Singh, A.C.M. Sousa, Hybrid nanofluids preparation, thermal properties, heat transfer and friction factor – a review, Renew. Sust. Energ. Rev. 68 (2017) 185–198.
[28] G.M. Moldoveanu, G. Huminic, A.A. Minea, A. Huminic, Experimental study on thermal conductivity of stabilized $Al_2O_3$ and $SiO_2$ nanofluids and their hybrid, Int. J. Heat Mass Transf. 127 (2018) 450–457.
[29] I. Zakaria, W.H. Azmi, W.A.N.W. Mohamed, R. Mamat, G. Najafi, Experimental investigation of thermal conductivity and electrical conductivity of $Al_2O_3$ nanofluid in water - ethylene glycol mixture for proton exchange membrane fuel cell application, Int. Commun. Heat Mass Transf. 61 (2015) 61–68.
[30] D. Wu, H. Zhu, L. Wang, L. Liu, Critical issues in nanofluids preparation, characterization and thermal conductivity, Curr. Nanosci. 5 (1) (2009) 103–112.
[31] N.N. Esfahani, D. Toghraie, M. Afrand, A new correlation for predicting the thermal conductivity of ZnO–Ag (50%–50%)/water hybrid nanofluid: an experimental study, Powder Technol. 323 (2018) 367–373.
[32] K.A. Hamid, W.H. Azmi, M.F. Nabil, R. Mamat, K.V. Sharma, Experimental investigation of thermal conductivity and dynamic viscosity on nanoparticle mixture ratios of $TiO_2$-$SiO_2$ nanofluids, Int. J. Heat Mass Transf. 116 (2018) 1143–1152.
[33] S. Zeroual, H. Loulijat, E. Achehal, P. Estellé, A. Hasnaoui, S. Ouaskit, Viscosity of Ar-Cu nanofluids by molecular dynamics simulations: effects of nanoparticle content, temperature and potential interaction, J. Mol. Liq. 268 (2018) 490–496.
[34] W.H. Azmi, K.V. Sharma, R. Mamat, G. Najafi, M.S. Mohamad, The enhancement of effective thermal conductivity and effective dynamic viscosity of nanofluids – a review, Renew. Sust. Energ. Rev. 53 (2016) 1046–1058.
[35] C.T. Nguyen, F. Desgranges, N. Galanis, G. Roy, T. Maré, S. Boucher, H. Angue Mintsa, Viscosity data for $Al_2O_3$–water nanofluid—hysteresis: is heat transfer enhancement using nanofluids reliable? Int. J. Therm. Sci. 47 (2) (2008) 103–111.
[36] E. Montazer, E. Salami, H. Yarmand, Z.Z. Chowdhury, M. Dahari, S.N. Kazi, A. Badarudin, Development of a new density correlation for carbon-based nanofluids using response surface methodology, J. Therm. Anal. Calorim. 132 (2) (2018) 1399–1407.
[37] S. Nabati Shoghl, J. Jamali, M. Keshavarz Moraveji, Electrical conductivity, viscosity, and density of different nanofluids: an experimental study, Exp. Thermal Fluid Sci. 74 (2016) 339–346.
[38] R.M. Mostafizur, R. Saidur, A.R.A. Aziz, M.H.U. Bhuiyan, Investigation on stability and density of methanol based $TiO_2$ nanofluids, IOP Conf. Ser. 88 (2015), 012057.
[39] I. Wole-Osho, E.C. Okonkwo, D. Kavaz, S. Abbasoglu, An experimental investigation into the effect of particle mixture ratio on specific heat capacity and dynamic viscosity of $Al_2O_3$-ZnO hybrid nanofluids, Powder Technol. 363 (2020) 699–716.
[40] S. Akilu, A.T. Baheta, K.V. Sharma, M.A. Said, Experimental determination of nanofluid specific heat with $SiO_2$ nanoparticles in different base fluids, in: AIP Conference Proceedings, AIP Publishing LLC, 2017, p. 090001.
[41] L.P. Zhou, B.X. Wang, X.F. Peng, X.Z. Du, Y.P. Yang, On the specific heat capacity of CuO nanofluid, Adv. Mech. Eng. 2010 (2010) 1–4.
[42] R.S. Vajjha, D.K. Das, Specific heat measurement of three nanofluids and development of new correlations, J. Heat Transf. 131 (7) (2009), 071601.
[43] Dynalene, Dynalene BioGlycol Technical Data Sheet, Dynalene, Inc, USA, 2020, pp. 1–5.
[44] S. Baskar, M. Chandrasekaran, T. Vinod Kumar, P. Vivek, L. Karikalan, Experimental studies on convective heat transfer coefficient of water/ethylene glycol-carbon nanotube nanofluids, Int. J. Ambient Energy 41 (3) (2020) 296–299.
[45] M.K. Abdolbaqi, W.H. Azmi, R. Mamat, N.M.Z.N. Mohamed, G. Najafi, Experimental investigation of turbulent heat transfer by counter and co-swirling flow in a flat tube fitted with twin twisted tapes, Int. Commun. Heat Mass Transf. 75 (2016) 295–302.
[46] K. Abdul Hamid, W.H. Azmi, R. Mamat, K.V. Sharma, Heat transfer performance of $TiO_2$–$SiO_2$ nanofluids in a tube with wire coil inserts, Appl. Therm. Eng. 152 (2019) 275–286.
[47] K.A. Hamid, W.H. Azmi, M.F. Nabil, R. Mamat, Experimental investigation of nanoparticle mixture ratios on $TiO_2$–$SiO_2$ nanofluids heat transfer performance under turbulent flow, Int. J. Heat Mass Transf. 118 (2018) 617–627.
[48] V. Kumar, J. Sarkar, Numerical and experimental investigations on heat transfer and pressure drop characteristics of $Al_2O_3$-$TiO_2$ hybrid nanofluid in minichannel heat sink with different mixture ratio, Powder Technol. 345 (2019) 717–727.
[49] W.H. Azmi, K.A. Hamid, N.A. Usri, R. Mamat, K.V. Sharma, Heat transfer augmentation of ethylene glycol: water nanofluids and applications - a review, Int. Commun. Heat Mass Transf. 75 (2016) 13–23.
[50] M.K. Abdolbaqi, W.H. Azmi, R. Mamat, K.V. Sharma, G. Najafi, Experimental investigation of thermal conductivity and electrical conductivity of BioGlycol–water mixture based $Al_2O_3$ nanofluid, Appl. Therm. Eng. 102 (2016) 932–941.
[51] M.K. Abdolbaqi, N.A.C. Sidik, M.F.A. Rahim, R. Mamat, W.H. Azmi, M.N.A.W. M. Yazid, G. Najafi, Experimental investigation and development of new correlation for thermal conductivity and viscosity of BioGlycol/water based $SiO_2$ nanofluids, Int. Commun. Heat Mass Transf. 77 (2016) 54–63.
[52] A.M. Khdher, N.A.C. Sidik, W.A.W. Hamzah, R. Mamat, An experimental determination of thermal conductivity and electrical conductivity of BioGlycol based $Al_2O_3$ nanofluids and development of new correlation, Int. Commun. Heat Mass Transf. 73 (2016) 75–83.
[53] M.K. Abdolbaqi, N.A.C. Sidik, A. Aziz, R. Mamat, W.H. Azmi, M.N.A.W.M. Yazid, G. Najafi, An experimental determination of thermal conductivity and viscosity of BioGlycol/water based $TiO_2$ nanofluids, Int. Commun. Heat Mass Transf. 77 (2016) 22–32.
[54] L. Kong, J. Sun, Y. Bao, Preparation, characterization and tribological mechanism of nanofluids, RSC Adv. 7 (21) (2017) 12599–12609.
[55] B.C. Pak, Y.I. Cho, Hydrodynamic and heat transfer study of dispersed fluids with submicron metallic oxide particles, Exp. Heat Transf. 11 (2) (1998) 151–170.
[56] M.Z. Sharif, W.H. Azmi, A.A.M. Redhwan, R. Mamat, T.M. Yusof, Performance analysis of $SiO_2$/PAG nanolubricant in automotive air conditioning system, Int. J. Refrig. 75 (2017) 204–216.
[57] P.C. Mukesh Kumar, K. Palanisamy, V. Vijayan, Stability analysis of heat transfer hybrid/water nanofluids, Mater. Today 21 (2020) 708–712.
[58] H. Huang, E. Ruckenstein, Repulsive force between two microparticles decorated with highly charged nanoparticles, Colloids Surf. A Physicochem. Eng. Asp. 436 (2013) 862–867.
[59] S.U. Ilyas, R. Pendyala, N. Marneni, Preparation, sedimentation, and agglomeration of nanofluids, Chem. Eng. Technol. 37 (12) (2014) 2011–2021.
[60] A. Singer, Z. Barakat, S. Mohapatra, S.S. Mohapatra, Chapter 13 - Nanoscale drug-delivery systems: in vitro and in vivo characterization, in: S.S. Mohapatra, S. Ranjan, N. Dasgupta, R.K. Mishra, S. Thomas (Eds.), Nanocarriers for Drug Delivery, Elsevier, 2019, pp. 395–419.
[61] S. Chakraborty, An investigation on the long-term stability of $TiO_2$ nanofluid, Mater. Today 11 (2019) 714–718.
[62] P.M.V. Raja, A.R. Barron, Rice University Physical Methods in Chemistry and Nano Science, LibreTexts, USA, 2020.
[63] S. Witharana, C. Hodges, D. Xu, X. Lai, Y. Ding, Aggregation and settling in aqueous polydisperse alumina nanoparticle suspensions, J. Nanopart. Res. 14 (5) (2012) 851.
[64] P. Keblinski, S.R. Phillpot, S.U.S. Choi, J.A. Eastman, Mechanisms of heat flow in suspensions of nano-sized particles (nanofluids), Int. J. Heat Mass Transf. 45 (4) (2002) 855–863.
[65] N. Kumar, S.S. Sonawane, Experimental study of thermal conductivity and convective heat transfer enhancement using CuO and $TiO_2$ nanoparticles, Int. Commun. Heat Mass Transf. 76 (2016) 98–107.
[66] M. Afrand, D. Toghraie, B. Ruhani, Effects of temperature and nanoparticles concentration on rheological behavior of $Fe_3O_4$ –Ag/EG hybrid nanofluid: an experimental study, Exp. Thermal Fluid Sci. 77 (2016) 38–44.
[67] E.V. Timofeeva, W. Yu, D.M. France, D. Singh, J.L. Routbort, Base fluid and temperature effects on the heat transfer characteristics of SiC in ethylene glycol/ $H_2O$ and $H_2O$ nanofluids, J. Appl. Phys. 109 (1) (2011), 014914.
[68] Q. He, S. Wang, M. Tong, Y. Liu, Experimental study on thermophysical properties of nanofluids as phase-change material (PCM) in low temperature cool storage, Energy Convers. Manag. 64 (2012) 199–205.