

Experimental Investigation of Thermo-Physical Properties of Tri-Hybrid Nanoparticles in Water-Ethylene Glycol Mixture

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Abstract

In recent years, research has focused on enhancing the thermo-physical properties of a single component nanofluid. Therefore, hybrid or composite nanofluids have been developed to improve heat transfer performance. The thermo-physical properties of the Al₂O₃-TiO₂-SiO₂ nanoparticles suspended in a base of water (W) and ethylene glycol (EG) at constant volume ratio of 60:40 and different volume concentrations were investigated. The experiment was conducted for the volume concentrations of 0.05, 0.1, 0.2, and 0.3% of Al₂O₃-TiO₂-SiO₂ nanofluids at different temperatures of 30, 40, 50, 60, and 70 °C. Thermal conductivity and dynamic viscosity measurements were carried out at temperatures ranging from 30 to 70 °C by using KD2 Pro Thermal Properties Analyzer and Brookfield LVDV III Ultra Rheometer, respectively. The highest thermal conductivity for tri-hybrid nanofluids was obtained at 0.3% volume concentration, and the maximum enhancement was increased up to 9% higher than the base fluid (EG/W). Tri-hybrid nanofluids with a volume concentration of 0.05% gave the lowest effective thermal conductivity of 4.8 % at 70 °C temperature. Meanwhile, the dynamic viscosity of the tri-hybrid nanofluids was influenced by volume concentration and temperature. Furthermore, tri-hybrid nanofluids behaved as a Newtonian fluid for volume concentrations from 0.05 to 3.0%. The properties enhancement ratio (PER) estimated that the tri-hybrid nanofluids will aid in heat transfer for all samples in the present. The new correlations for thermal conductivity and dynamic viscosity of tri-hybrid nanofluids were developed with minimum deviation. As a conclusion, the combination of the enhancement in thermal conductivity and dynamic viscosity for tri-hybrid at 0.3% volume concentration was found the optimum condition with more advantage for heat transfer than other concentrations.

Keywords: Ethylene glycol-water, Dynamic viscosity, Thermal conductivity, Tri-hybrid nanofluids

Introduction

Nanofluid is a suspension of liquid containing metal or non-metallic nanoparticles of typical size (1 - 100 nm) dispersed into the base liquid. In 1995, the concept of nanofluids was first introduced by Choi *et al.* [1]. This new method was proven to increase heat transfer by improving the thermo-physical properties of the nanofluids. Nanofluids are known for their application in the heating and cooling process. The main cooling process is an important part of industrial applications such as power plants, chemical processes, microelectronics, transportation, and automotive cooling systems [2,3]. The existence of solid particles leads to interesting characteristics in the fundamental thermo-physical properties of nanofluids. Thermal conductivity, viscosity, density, and stability have been investigated in recent years by many researchers [4,5].

The method of nanofluid preparation is important to minimize the agglomeration of the nanoparticles, and hence improve the stability. The most common processes used in nanofluid preparation

are 1-step and 2-step methods. The 1-step method is the process of synthesizing nanoparticles and simultaneously dispersing them in a base liquid. However, this method is not practical for industry, which only applies to low vapor pressure liquids. Another method of nanofluid preparation is known as the 2-step method. There are 2 processes in this method, namely (i) the synthesis of nanoparticles in powder form and (ii) spreading the nanoparticles into the base liquid to form a stable and homogeneous solution [6,7]. Most nanofluids that use oxide particles and carbon nanotubes are produced through a two-step method [8-10]. The 2-step method is preferable for the production of nanofluids on a large scale and is thus applicable for industry. However, the challenge of using the 2-step method is that agglomeration and nanoparticles tend to settle quickly [11]. The 2-step method is the most dominant method compared to the 1-step method for nanofluid preparation.

Several recent studies have discussed the topic of hybrid or composite nanofluids [12,13]. Hybrid or composite nanofluids are considered an extension of research work for single nanofluids, which can be carried out through a combination of 2 or more different nanoparticles- either in mixed or dispersed composites in liquids [14]. Composite or hybrid materials are elements that combine chemical and physical properties. The aim of synthesizing hybrids or nanofluid composites is to improve the properties of single nanoparticles, in which a better increase in thermal properties or rheological properties can be achieved. Hybrid nanofluid is expected to achieve good thermal performance when compared to a single nanofluid [15]. Recently, some papers regarding nanofluid hybrids have been reviewed by Hamzah *et al.* [16] and Sidik *et al.* [17]. Both papers were presented on hybrid nanofluid preparation, performance, and application methods. Therefore, investigations on thermal conductivity and viscosity are important in understanding hybrid nanofluid behavior for further implementation in heat transfer applications. Thermal conductivity is an important factor affecting the increase in heat transfer [18,44]. There are several factors that influence thermal conductivity, such as: concentration, temperature, particle size, surface ratio to nanoparticle volume, and nanofluid stability [19-22]. Turgut *et al.* [23], proved that thermal conductivity increased by 7.4 % with a volume fraction of particles above alkaline liquids. Investigation of Al_2O_3 -Cu composite nanofluid with water as a basic liquid was carried out by Suresh *et al.* [24]; they reported an increase of up to 12 % with increasing volume concentration. In another paper, Hamid *et al.* [25], the thermo-physical properties of TiO_2 - SiO_2 nanoparticles suspended in a base fluid of water (W) and ethylene glycol (EG) mixture with 60:40 volume ratio were investigated. They found the highest thermal conductivity for TiO_2 - SiO_2 nanofluids was obtained with a ratio of 20:80, and the maximum enhancement exceeded up to 16 % higher than the base fluids.

Ho *et al.* [26], determined the nanofluid dynamic viscosity of Al_2O_3 -MEPCM composites. They observed an increase in the mass fraction of the nanoparticles. In another study, Esfe *et al.* [12] used Ag-MgO/water composite nanofluid. They found that the dynamic viscosity of the composite nanofluid increased with an increase in volume fraction and build correlation for viscosity. In comparison, their correlation predicted a higher value than the existing correlation in the literature. Soltani *et al.* [27] conducted a viscosity experiment with MgO-MWCNT composite nanofluid within the range of 0.1 - 1.0 % concentration and temperature range of 30 - 60 °C. The observed temperature effect is important for nanofluids at high concentrations. Their findings showed that nanofluid acts as a Newtonian fluid.

The thermo-physical properties of various types of hybrid nanofluids are very important to study. It aims to understand the behaviors and factors that affect the properties that can improve heat transfer performance. Based on the information obtained by the authors, the study of the effect of mixed ratios for the 3 nanoparticles in the shape of hybrid nanofluids is limited in the literature. Furthermore, the use of hybrid nanofluids with 2 different nanoparticles will result in increased viscosity relative to single nanofluid components [28]. Based on this problem, this study was conducted by emphasizing the influence of the ratio of three nanoparticles on thermal-physical properties. The thermal conductivity and dynamic viscosity of Al_2O_3 - TiO_2 - SiO_2 nanofluids or tri-hybrid nanofluids for heat transfer applications was investigated.

Materials and methods

Preparation of tri-hybrid nanofluids

The preparation of tri-hybrid nanofluids involved 3 different types of single nanofluids, namely, Al_2O_3 , TiO_2 , and SiO_2 mixed together and dispersed in the base fluid of water/EG mixture. All the single nanofluids were procured from US Research Nanomaterials, Inc. The respective nanoparticle sizes for Al_2O_3 , TiO_2 , and SiO_2 were 13, 50, and 30 nm, with purity of 99.8, 99, and 99.99 %, respectively. The properties of each nanoparticle are given in **Table 1**. The base fluid used in the present study was a mixture of water and EG at a ratio of 60:40 (vol%). The properties of ethylene glycol are presented in **Table 2**. The nanoparticle size characterization of the tri-hybrid nanofluids was performed using the field scanning electron microscope (FESEM) technique for Al_2O_3 , TiO_2 , and SiO_2 nanoparticles. The FESEM images for nanoparticles are shown in **Figure 1**.

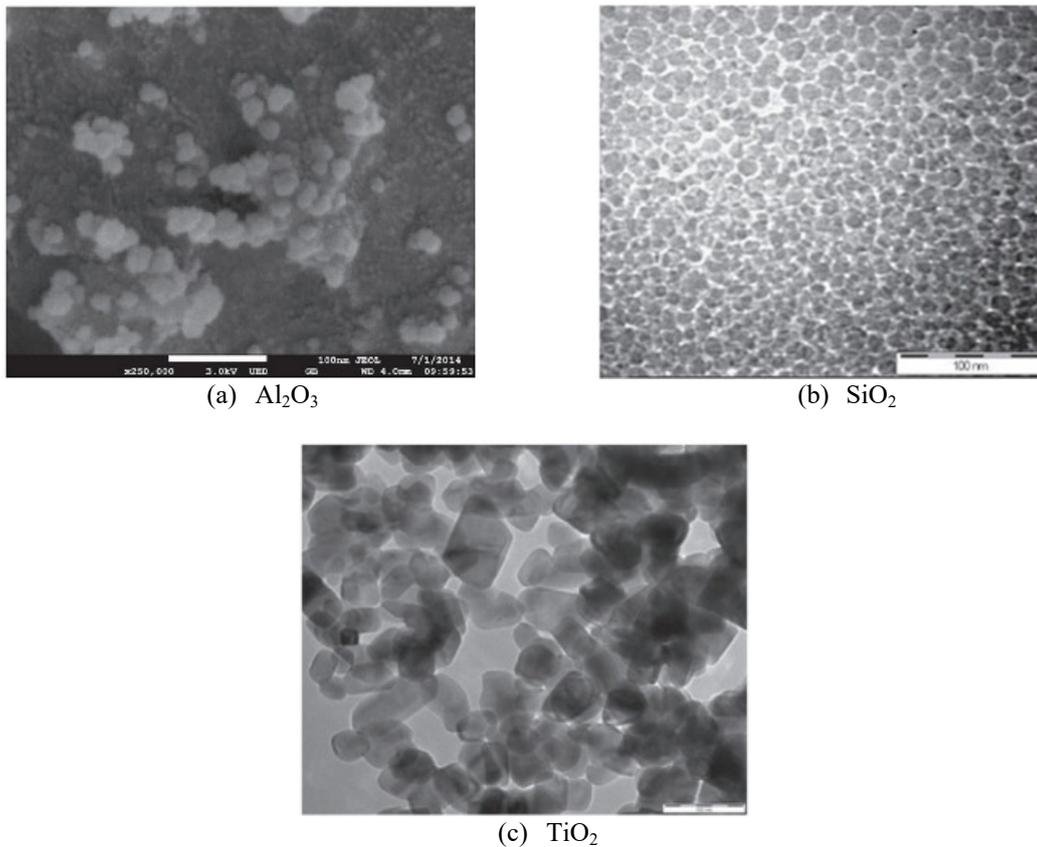


Figure 1 Field Scanning Electron Microscope (FESEM) images for Al_2O_3 , TiO_2 , and SiO_2 nanoparticles [9].

Table 1 Properties of Al₂O₃, TiO₂, and SiO₂ nanoparticles [9].

Properties	Al ₂ O ₃	TiO ₂	SiO ₂
Molecular mass, g mol ⁻¹	101.96	79.86	60.08
Average particle diameter, nm	13	50	30
Density, kg m ⁻³	4,000	4,230	2,220
Thermal conductivity, W m ⁻¹ K ⁻¹	40	8.4	1.4
Specific heat, J kg ⁻¹ K ⁻¹	773	692	745

Table 2 Properties of ethylene glycol (EG) [9].

Properties	Ethylene Glycol (EG)
Vapour pressure, mmHg at 20 °C	0.08
Boiling point, °C	195 - 198
Melting point, °C	-13
Density, g ml ⁻¹ at 25 °C	1.113

The 2-step method was used for the preparation of tri-hybrid nanofluids. Tri-hybrid nanofluids are prepared by mixing the 3 nanofluids (Al₂O₃, TiO₂ and SiO₂) together, undergoing mixing- and sonication-based processes as per Ramadhan *et al.* [43]. The preparation of the nanofluid initially begins with the calculation of the required volume according to the concentration performed for the experiments. Further, in this study, the tri-hybrid nanofluids were prepared at volume concentrations of 0.05, 0.1, 0.2, and 0.3 %. The nanofluids were first prepared at the highest concentration, 0.3 %, and then diluted to lower concentrations.

The single nanofluids Al₂O₃, TiO₂, and SiO₂ were supplied in a water suspension with weight concentrations of 20, 40, and 25 % for Al₂O₃, TiO₂, and SiO₂, respectively. Eq. (1) [29] is used to convert from weight concentration to volume concentration. The dilution from higher volume concentration to lower volume concentration utilized Eq. (2) [30, 46, 47, 48] by adding the base fluid (ΔV).

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

$$\Delta V = (V_2 - V_1) = V_1 \left(\frac{\phi_1}{\phi_2} - 1 \right) \tag{2}$$

All single nanofluids were mixed together at a volume ratio of 1/3:1/3:1/3 to form a tri-hybrid nanofluid. Total volumes of 100 mL were prepared for each concentration of the tri-hybrid nanofluids. The combined solution from the three single Al₂O₃, TiO₂, and SiO₂ nanofluids were mixed together using a magnetic stirrer for 120 min. Then, the solution underwent a sonication process using an ultrasonic bath to enhance stability.

Stability of tri-hybrid nanofluids

The investigation on the stability of the tri-hybrid nanofluids in the present study was conducted through visual observation, via measurement by UV-Vis Spectrophotometer. The sedimentation through visual observation was conducted up to 14 days. Nanofluids were considered stable when the

concentration was constant. Previously, the same method to observe visual sedimentation of prepared nanofluids was used Azmi *et al.* [31]. The UV-Vis spectrophotometry measurement was conducted for 10 days (250 h) by varying the sonication time. The wavelength of the UV-Vis spectrophotometer was set at 900 nm, following the study by Hamid *et al.* [32]. The UV-Vis spectrophotometer measures the absorption and light intensity of the scattering light after passing the nanofluid by comparing the intensity level to what it was when it passed the base fluid. The absorbance ratio of sonication times is different during sedimentation time at a constant wavelength (λ) of 900 nm. The stability evaluation by UV-Vis spectrophotometer was also used by previous studies [29, 49].

Thermal conductivity measurement of tri-hybrid nanofluids

The method of thermal conductivity measurement followed the ASTM D5334 and IEEE 442-1981 standards, using KD2 Pro Property Analyzer (Decagon Devices), is shown in **Figure 2**. Part of the thermal test sample conductivity consists of a KS-1 sensor to read of k [W/m K], a measuring bottle to put samples to be tested, and a KD2 Pro controller, which is an important part of thermal conductivity measuring device. The KD2 Pro instrument uses a transient line heat source to measure thermal properties. Thermal conductivity measurements are performed for temperatures varying from 30 to 70 °C. A water bath was used to maintain a constant sample temperature during the experiment. Previous to the sample measurement, the validation of thermal conductivity values from thermal conductivity sensors was evaluated using standardized liquid glycerine supplied by Decagon Devices. The measured k was 0.286 W/m K with an accuracy ± 0.35 %. Thermal conductivity measurements were performed several times for each sample and temperature, and an average value was taken; the measurement time for each set data was about 15 min for each set data at different temperatures. It is important to minimize the occurrence of errors in measurements with free convection due to temperature variation along the sensor that directly touches the liquid sample.

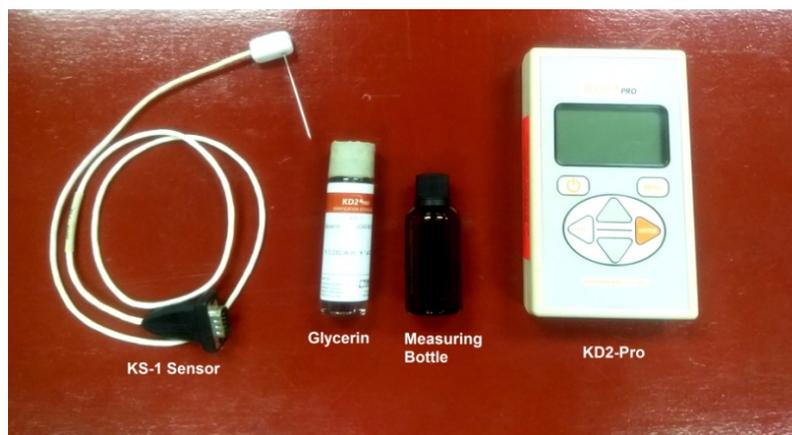


Figure 2 KD2 Pro thermal property measurement apparatus for thermal conductivity measurement.

Dynamic viscosity measurement of tri-hybrid nanofluids

Viscosity measurement was done using a Brookfield LVDV III Ultra Rheometer with a water bath circulation. Operating conditions of Rheometer for viscosity measurement range from 1 to 6×10^6 mPa.s. The sample of 16 mL was added to the cylinder jacket and pasted into the Rheometer. The RheoCal program was used for measuring the viscosity connected to the controller. Sample viscosity was evaluated by varying the velocity of the spindle. Dynamic viscosity measurements were performed with a temperature variation of 30 ~ 70 °C. The circulating water bath was used to control the sample

temperature. The measurements were repeated 5 times and the average value was reported. Dynamic viscosity measurement apparatus is shown in **Figure 3**. Basic liquid 60:40 (water: EG) at different temperatures were validated by the data contained in the literature. The dynamic viscosity measurements were performed for $\text{Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$ or tri-hybrid nanofluids.



Figure 3 Brookfield LVDV III Ultra Rheometer for the measurement of dynamic viscosity (from left: temperature sensor, controller device and water bath).

Results and discussion

Stability of tri-hybrid nanofluids

The stability of the $\text{Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$ nanofluids was further confirmed by the concentration ratio, ϕ_r , presented in **Figure 4**. The concentration ratio of 0.1 % vol concentration of the tri-hybrid nanofluids was used for 6 different sonication hours. The ideal absorbance ratio of one (100 %) presented the ideal stability of the fluid. The concentration ratio of mixture sonication at 0.5, 1, and 2 h started to decrease after being stored for 24 h and kept decreasing until 10 days (240 h). With sonication time of 5 and 10 h, it remained at good concentration ratio value at about (70 ~ 80 %) until 10 days. From this figure, it can be seen that the 10 h sonication time shows the best absorbance ratio compared to others. Thus, the preparation of the tri-hybrid nanofluids in the present study used 10 h for the sonication process. The findings of concentration ratio obtained 80 % from 150 to 240 min. This condition was stable. **Figure 5** shows images of tri-hybrid nanofluids ($\text{Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$) -W/EG at volume concentrations of 0.05, 0.1, 0.2, and 0.3 %. Tri-hybrid nanofluids ($\text{Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$) -W/EG images were taken only after preparation and after 14 days. From **Figure 5(a)**, no sedimentation of particles was observed after the nanofluids were prepared. The sedimentation of particles started to be noticeable at 7 days. After 14 days, the sedimentation could be clearly seen, as shown in **Figure 5(d)**. The sedimentation from this observation was affected by the gravity falling motion of the particles in the tube.

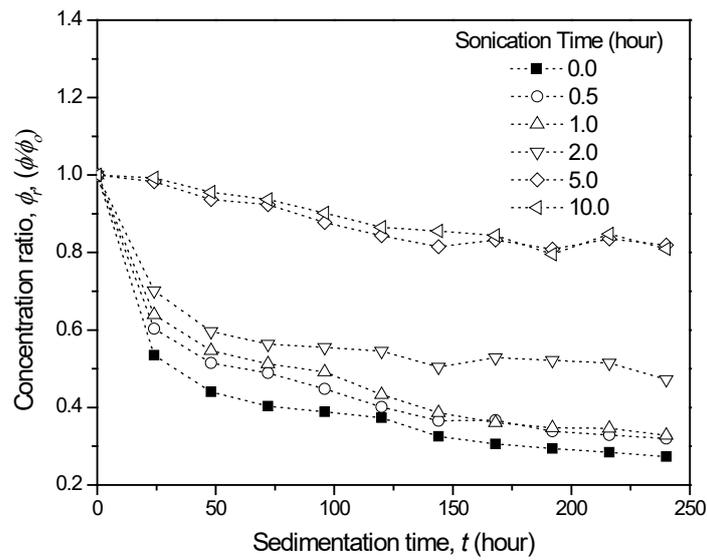


Figure 4 The concentration ratio of tri-hybrid nanofluids for different sonication times within 10 days.

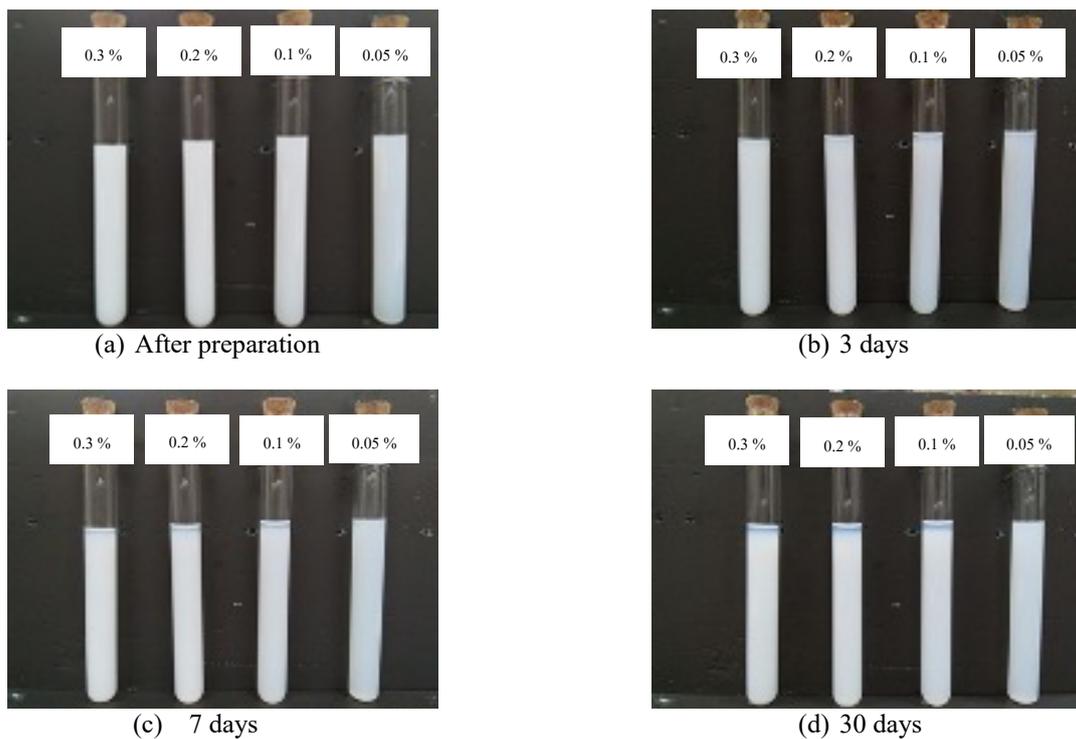


Figure 5 Sedimentation observation of tri-hybrid nanofluids: (a) after preparation, (b) 3 days, (c) 7 days, and (d) 30 days.

Validation of thermal conductivity and dynamic viscosity

Thermal conductivity data and viscosity need to be validated by comparing with ASHRAE [33] for EG/Water (60:40) medium. For the measurement of thermal conductivity using KD2 Pro, data validation resulted with 1.5 % deviation, presented in **Figure 6**. Deviations for measurable data were less than 1.0 % compared to ASHRAE [33]. Reddy *et al.* [34] performed validation tests for deviations from their base fluid up to 2.5 % compared to ASHRAE [33]. Furthermore, the same temperature range and water/EG ratio on validation of current viscosity measurements was investigated by other papers [30,35,36]. **Figure 6(b)** indicates that the viscosity data is in a good pattern and in accordance with ASHRAE [33]. In addition, data for base fluid composed of water and EG mixture is very accurate with ASHRAE data trends that lower viscosity on temperature. Therefore, further measurements and investigations for thermal conductivity and dynamic viscosity of tri-hybrid nanofluids were performed.

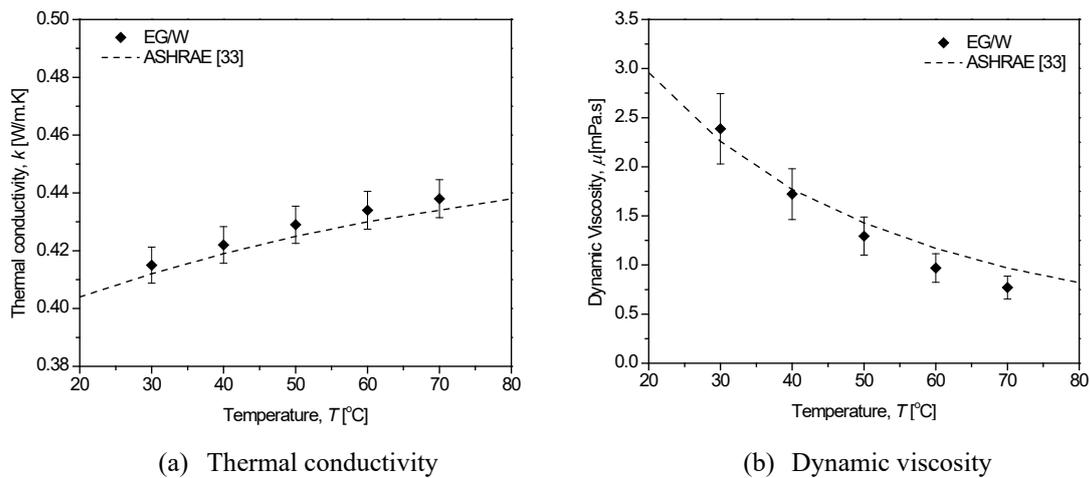


Figure 6 Validation (a) thermal conductivity and (b) dynamic viscosity data of EG/Water (40:60) with ASHRAE.

Thermal conductivity of tri-hybrid nanofluids

The relationships between tri-hybrid nanofluids thermal conductivity and temperature at volume concentrations of 0.05 ~ 0.3 % are presented in **Figure 7**. Thermal conductivity of the tri-hybrid nanofluids for the various volume concentrations increased as temperature increased and was higher than base fluid. Furthermore, the highest thermal conductivity was obtained for the volume concentration of 0.3 %. Meanwhile, the volume concentration of 0.05 % provided the lowest thermal conductivity among the investigated temperatures. In this study, the relationship between the composition ratio of nanoparticles (1/3:1/3:1/3) in the tri-hybrid nanofluids to increased thermal conductivity was influenced by 3 nanoparticles that have different sizes. The diameters of Al_2O_3 and SiO_2 nanoparticles are 13 nm and 22 nm, wherein both nanoparticles are smaller than TiO_2 nanoparticles that have a size of 50 nm. Al_2O_3 and SiO_2 nanoparticles play a role in conduction by fulfilling larger TiO_2 nanoparticle spaces. To increase the contact area for conduction between molecules, resulting in a higher rate of heat transfer during a collision by the Brownian motion [37], requires a special arrangement of the three type nanoparticles. The effective thermal conductivity of tri-hybrid nanofluids are presented in **Figure 8**. The results revealed that the effective thermal conductivity increased with 0.3 % volume concentration in the tri-hybrid nanofluids; except for 0.05 % volume, where the effective thermal conductivity of nanofluids were lowest. Effective thermal conductivity in tri-hybrid nanofluids can significantly affect the relationship between volume concentration and each set of temperatures.

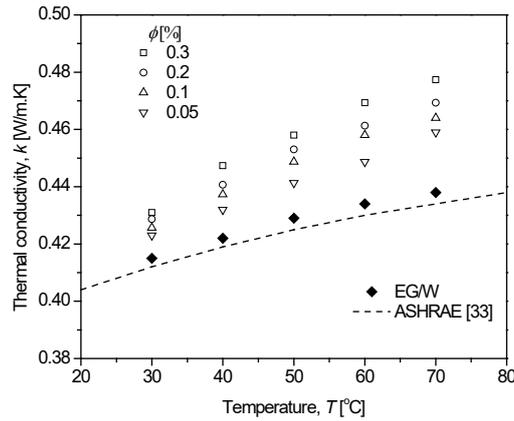


Figure 7 The experimental thermal conductivity of tri-hybrid nanofluids.

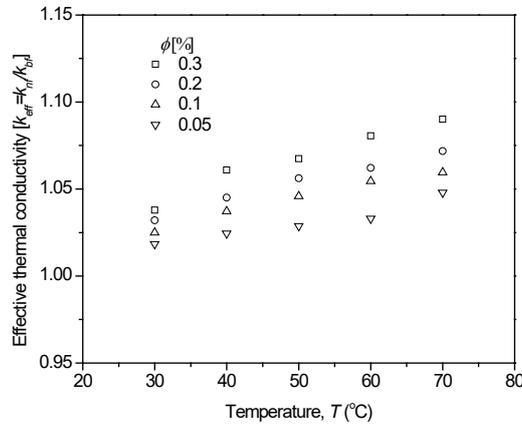


Figure 8 The effective thermal conductivity of tri-hybrid nanofluids.

Dynamic viscosity of tri-hybrid nanofluids

Figure 9 illustrates the dynamic viscosity against shear rate in the range of $920 \leq \gamma \leq 4320 \text{ s}^{-1}$ for 0.05 % vol concentration of tri-hybrid nanofluids. The results indicate that the dynamic viscosity remained constant with the increase of shear rate for 0.05 % vol nanofluid. The shear-independent viscosity demonstrates that the tri-hybrid nanofluids behaved as Newtonian fluids within the temperatures studied. The dynamic viscosity of the $\text{Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$ nanofluids for different temperatures had a significant effect on the concentration volume of 0.05 %. A temperature of 30 °C provides a higher dynamic viscosity than 40, 50, 60, and 70 °C, as shown in Figure 9. This is probably due to the different intensities of Al_2O_3 , TiO_2 and SiO_2 nanoparticles in the composition ratio (1/3:1/3:1/3) in tri-hybrid nanofluids.

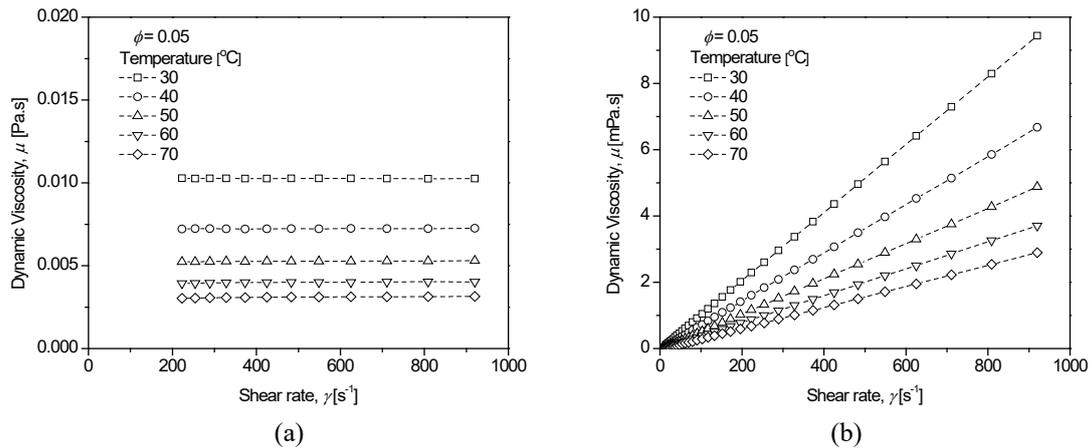


Figure 9 Variation of dynamic viscosity with shear rate.

Figure 10 shows the dynamic viscosity for various volume concentrations of tri-hybrid nanofluids in the temperature range of 30 ~ 70 °C. The viscosity for all volume concentration followed the base fluid trend, whereby it decreased exponentially with temperature. The viscosity of the volume concentration of 0.3 was higher than the values of those of 0.2, 0.1, and 0.05 %. The volume concentration of 0.3 % showed the highest value for viscosity at all temperatures. The dynamic viscosity of the tri-hybrid nanofluids decreased slightly and varied by the differences in the composition ratio of Al₂O₃, TiO₂, and SiO₂ nanoparticles of 1/3:1/3:1/3 in the tri-hybrid nanofluids contributing to the difference in the interactions of those particles with a base fluid. However, the effect of temperature on the Al₂O₃-TiO₂-SiO₂ nanofluid viscosity for all mixed ratios decreased with increasing temperature and was evidenced by Asadi *et al.* [38].

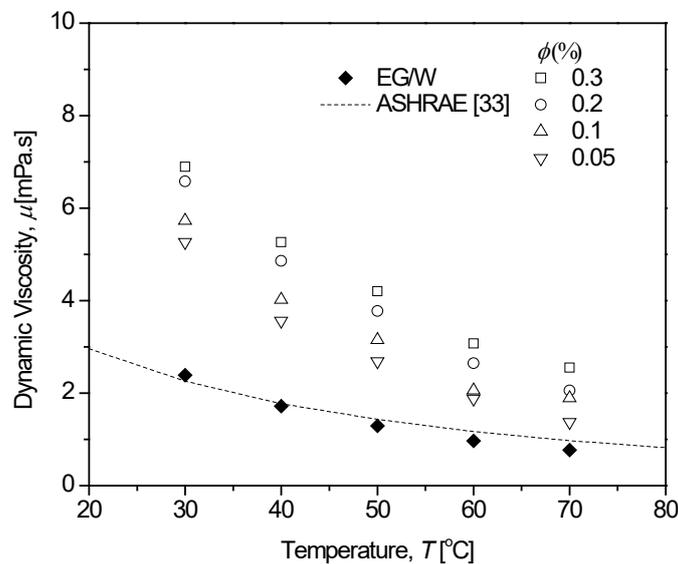


Figure 10 Variation of dynamic viscosity with temperature.

The variation of relative viscosity with temperature is shown in **Figure 11**. From the figure, the maximum relative viscosity for all volume concentrations occurred at the different temperatures of 30 ~ 70 °C. For the volume concentration of 0.3 %, the viscosity ratio was higher at 30 °C, and then increased at 40 ~ 70 °C. Similarly, this pattern also applied to the volume concentrations of 0.2, 0.1, and 0.05 %. At the volume concentration 0.2, relative viscosity value rose and fell at 30 °C at around 2.8, then increased from 40 to 70 °C. In addition, the values increased significantly for temperatures of 60 ~ 70 °C, with a maximum increase of 5.4 times compared to base fluid. It can be concluded that the relative viscosity distribution for all volume concentrations is always below 0.3 % vol in the investigated temperature range. The results also showed a specific tendency for viscosity at different volume concentrations. This may be due to different effects on shear flow resistance due to the presence of 3 different types of nanoparticles with different concentration volumes and particle sizes.

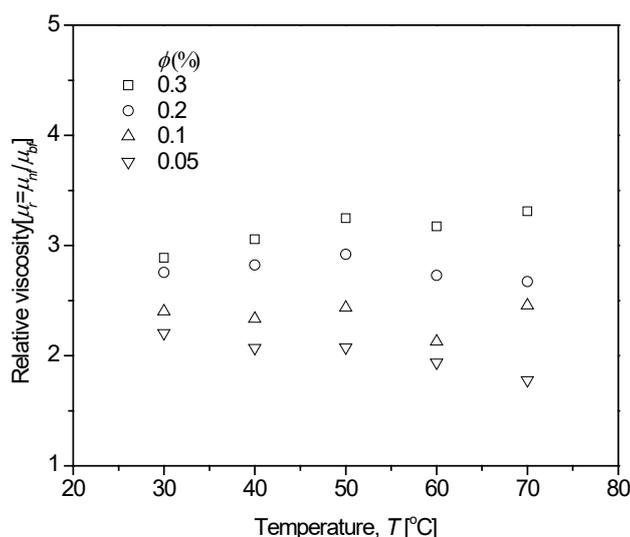


Figure 11 Variation of relative viscosity with temperature.

Comparison with literature

Figures 12(a) and **(b)** demonstrate comparisons of the effective thermal conductivity and relative viscosity of the present study with the data from Nabil *et al.* [39], Murshed *et al.* [40], and Azmi *et al.* [30]. In this study, the effective thermal conductivity of tri-hybrid nanofluids was increased by 1.02 - 1.06 times compared to a base fluid for concentration 0.1 %. Murshed *et al.* [40], used Al₂O₃ nanoparticle with 0.5 % vol concentration of a base fluid of EG/water mixture. They proved that the experimental values of the different thermal conductivities of nanofluid significantly increased with fluid temperature. This is due to the high fluid temperature, which increases the movement of Brownian nanoparticles and also decreases the base fluid viscosity. The influence of intense Brownian motion meant the contribution of micro connectivity to heat transport increased, resulting in increased thermal conductivity of nanofluid. A study by Nabil *et al.* [39] used TiO₂-SiO₂ with EG/water as a base fluid. They presented the results of the thermal conductivity of the TiO₂-SiO₂ nanofluids enhancement with increasing concentration and temperature. The relative viscosity in their research is almost identical compared to this study for temperatures between 30 and 50 °C.

In another paper, Azmi *et al.* [30] performed relative viscosity measurements for TiO₂ nanoparticles in EG/water based liquids at a concentration of 0.5%. The results showed that the relative viscosity in the range studied increased about 1.05 - 1.12 times compared to the water/EG mixture. The occurrence of

relative viscosity fluctuations in an unspecified temperature range. Their relative viscosity was the lowest value compared to the others, as shown in **Figure 12(b)**. According to Sundar *et al.* [28], the magnitude of the increase in thermal conductivity or relative viscosity depends on the type of nanoparticle and the base fluid, thus observed and illustrated in **Figures 12(a)** and **12(b)**.

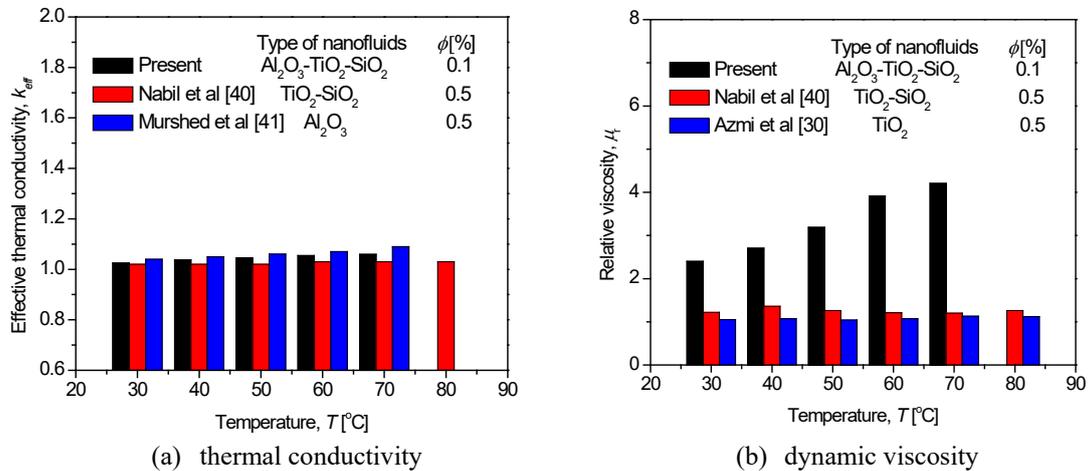


Figure 12 Comparison of tri-hybrid nanofluids properties with data from the literature.

Conclusions

In this study, thermal conductivity and dynamic viscosity of tri-hybrid nanofluids were investigated for four concentration volumes, and temperatures from 30 to 70 °C. The experimental results showed that the concentration volume of 0.3% obtained the best effective thermal conductivity and relative viscosity compared to 0.05 ~ 0.2%. Therefore, in this study, different concentration volumes became the control parameters and performance studied. In terms of increased thermal conductivity, 0.3% volume concentration provided a maximum increase of up to 9%, while it was observed that 0.05% had the least increase for dynamic viscosity, which was approximately 2.2 times the average compared to other ratios. The optimum of the concentration volume was 0.3%, where the increase in thermal conductivity and dynamic viscosity had more advantages than other concentration volumes. However, experimental investigations on heat transfer are required to determine the actual performance of this volume of concentration.

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