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Experimental Investigation of Thermal Properties of Ternary Nanofluids in Water-Ethylene Glycol (60:40) Mixture

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ABSTRACT

In recent years, research is directed towards enhancing the thermo-physical properties of single-component nanofluids. Hence, a hybrid or composite nanofluid is developed to improve heat transfer performance. The thermophysical properties of the Al_2O_3-TiO_2-SiO_2 nanoparticles suspended in the base of water (W) and ethylene glycol (EG) blends with vol 60:40 or Ternary Nanofluids for various volume concentrations are investigated. The experiments were undertaken for the concentration volume of 0.5, 1.0, 1.5, 2.0, 2.5 and 3.0% of Al_2O_3-TiO_2-SiO_2 nanofluids with 30, 40, 50, 60 and 70 °C. Thermal conductivity measurements and dynamic viscosity are carried out at temperatures ranging from 30-70 °C. The highest thermal conductivity of Ternary nanofluids was obtained at a concentration of 3.0%, and the maximum increase was up to 27.1% higher than the base fluid (EG/W). Ternary nanofluids at a concentration of 0.5% give the lowest effective thermal conductivity of 14.4% at 70°C. Meanwhile, the evidence from the dynamic viscosity of the Ternary nanofluids is influenced by concentration and temperature. Furthermore, Ternary nanofluids behaviour as Newtonian fluid in volume concentration from 0.5-3.0%. The development of a new correlation for thermal conductivity and dynamic viscosity of Ternary nanofluids are precise. In conclusion, the combination of enhancement in thermal conductivity and a dynamic viscosity at a concentration of 3.0% has optimum conditions, which have more advantages for heat transfer than at other concentrations.

INTRODUCTION

Nanofluid is a suspension of a liquid containing metal or non-metallic nanoparticles with a typical size of 1-100 nm dispersed in a base liquid first introduced by Choi, et al. [1]. The nanofluids have proven to enhance heat transfer by increasing their thermo-physical properties. Nanofluids are known for their utility in the heating and cooling process. The primary cooling process is an integral part of industrial applications such as power plants, chemical processes, microelectronics, transportation, and automotive cooling systems [2-6]. The existence of solid particles leads to exciting characteristics in the fundamental thermophysical properties of nanofluids. Many researchers have investigated thermal conductivity, viscosity, density, and stability in recent years [7-9].

The method of nanofluids preparation is essential, which minimizes the agglomeration of the nanoparticles and improve stability. The most common procedure used in nanofluids preparation is the one-step and two-step method. The one-step way is the process of synthesizing nanofluids with simultaneous dispersion of particles in a base liquid. However, this method is not practical in an industry, which only applies to low vapour pressure liquids. Another way of nanofluid preparation is the two-step method. There are two processes involved, namely (i) the synthesis of nanoparticles in powder form (ii) spreading the nanoparticles in a base liquid to form a stable and homogeneous solution [10-13]. Most nanofluids used oxide particles, and the

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preparation of carbon nanotubes is through a two-step method [14-17]. The two-step method is preferable for the production of nanofluids on a large scale required for industrial applications. The challenge of using the two-step method lies in preventing agglomeration, which results in sedimentation of the nanoparticles rapidly [18]. The two-step method is the most dominant compared to the one-step process for nanofluid preparation.

Recent studies discuss the topic of hybrid or composite nanofluids [19-21]. Hybrid or a composite nanofluid is considered an extension of research work for single nanofluids, which can be carried out through combining two or more different nanoparticles - either in mixed or dispersed composites in liquids [22]. A composite or hybrid materials are elements that combine chemical and physical properties. Synthesis of hybrids or nanofluid composites aims to improve the properties of single nanoparticles through which the thermal properties or rheological properties are achieved. A hybrid nanofluid is expected to achieve good thermal performance compared to a single nanofluid [23]. Recently, Hamzah, et al. [24], Sidik, et al. [25], specific journal papers [23], [24] present a detailed presentation on hybrid nanofluid preparation, performance and application. Hence, investigations on thermal conductivity and viscosity are essential in understanding hybrid nanofluids behaviour for further implementation in heat transfer applications. Thermal conductivity is a crucial factor affecting the increase in heat transfer [26]. Several factors influence thermal conductivity: concentration, temperature, particle size, surface ratio to nanoparticle volume, and nanofluid stability [27-31]. Turgut, et al. [32] showed that thermal conductivity increased by 7.4% above the alkaline liquid. Experimental investigation with Al2O3-Cu composite nanofluid with water as a base liquid was carried out by Suresh, et al. [33], who report an increase of up to 12% with increasing volume concentration. In another paper, Hamid, et al. [34] the thermo-physical properties of TiO2-SiO2 nanoparticles suspended in a mixture of water (W) and ethylene glycol (EG) mixture of 60:40 ratio by volume has been reported. They found that the highest thermal conductivity for TiO2-SiO2 nanofluids was obtained with a 20:80 ratio and the maximum enhancement exceeded up to 16% higher than the base fluids.

Ho, et al. [35] have determined the nanofluid dynamic viscosity of Al_2O_3-MEPCM nanocomposites. In another study, Esfe, et al. [19] using Ag-MgO/water composite nanofluid. They found that the composite nanofluids dynamic viscosity (VST) increased with an increase in volume fraction and build correlation for viscosity. In comparison, their correlation predicts a higher value than the existing correlation in the literature. Soltani, et al. [36], conducting a VST 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 significant for nanofluids at high concentrations. Their findings show that nanofluid behaves as a Newtonian fluid.

The thermo-physical properties of various hybrid nanofluids are fundamental requirements. It aims to understand the behaviours and factors that affect properties that can improve heat transfer performance. The authors concluded from their study that mixed ratios for the three nanoparticles in the shape of hybrid nanofluids are limited in the literature. Furthermore, the use of hybrid nanofluids with two different nanoparticles will result in increased viscosity relative to a single nanofluids component [37]. Based on this problem, the study was conducted by emphasizing the influence of the ratio of three nanoparticles on thermal-physical properties. In addition, a new correlation is proposed to determine the thermal conductivity and dynamic viscosity of Al_2O_3-TiO_2-SiO_2 nanofluids or Ternary nanofluids for heat transfer applications.

EXPERIMENTAL METHOD

Preparation of Al_2O_3-TiO_2-SiO_2 nanofluids

The preparation of ternary nanofluid involved three different types of single component nanofluids, namely Al_2O_3, TiO_2, and SiO_2 mixed and dispersed in the base liquid mixture of water and EG. All the nanofluids were procured from US Research Nanomaterials, Inc. The respective nanoparticle size of Al_2O_3, TiO_2, and SiO_2 are 11, 50 and 22 nm, are of exceptionally high purity greater than 99%. The properties of nanoparticles are given in **Table 1**. The base fluid mixture of water and EG is used in the present study in a 60:40 ratio by volume. The ethylene glycol properties are presented in **Table 2**. The characterization of nanoparticles is undertaken with the field scanning electron microscope (FESEM) with the image of nanoparticles shown in **Figure 1**.

Table 1. Properties of Al_2O_3, TiO_2 and SiO_2 nanoparticles [41]

Properties	**Al_2O_3**	**TiO_2**	**SiO_2**
Molecular mass, g mol^{-1}	101.96	79.86	60.08
Average particle diameter, nm	13	50	22
Density, kg m^{-3}	4000	4230	2220
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) [41]

Properties	**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

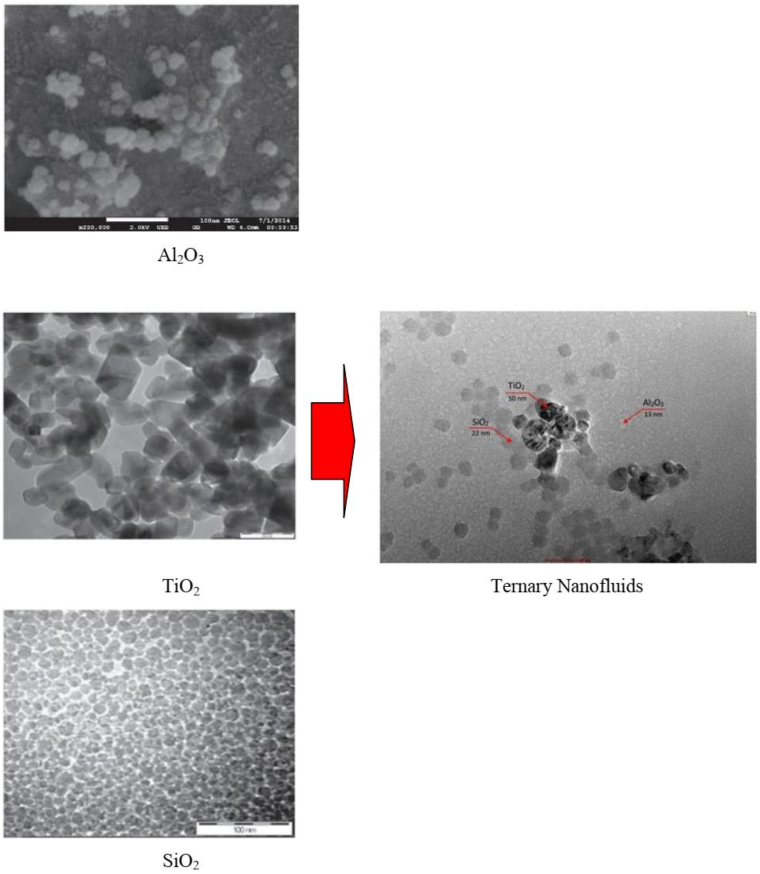


Figure 1. TEM Images for Al_2O_3, TiO_2, SiO_2 nanoparticles and ternary nanofluids [9, 41]

The preparation of ternary nanofluids is based on **Figure 2**. The procedure of the two-step method is used for the preparation of ternary nanofluids, which is prepared by mixing three single nanofluids (Al_2O_3, TiO_2, and SiO_2), undergo a mixing and sonication process. The nanofluid preparation is initiated with the calculation of the required volume for a specific concentration. In the present study, ternary nanofluids were prepared for volume concentrations of 0.5 - 3.0%. The ternary nanofluids were prepared at the highest concentration of 3.0% and diluted to lower concentrations later. The supply of Al_2O_3, TiO_2, and SiO_2 nanofluids were in weight concentrations of 20, 40, 25% for Al_2O_3, TiO_2 and SiO_2, respectively, in base liquid water. Eq. (1) [38] is made to convert from weight concentration to volume concentration. The quantity of base liquid (ΔV) to be added for dilution from higher volume concentration to lower volume concentration is determined with Eq. (2).

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

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

All single nanofluids were mixed at a volume ratio of 1/3:1/3:1/3 to form a ternary nanofluid. A 100 mL volume sample prepared for each concentration of the ternary nanofluid. The combined solution of the three single Al_2O_3, TiO_2, and SiO_2 nanofluids were mixed using a magnetic stirrer for 240 minutes. Then, the mixture solution has undergone a sonication process using the ultrasonic bath to enhance the stability.

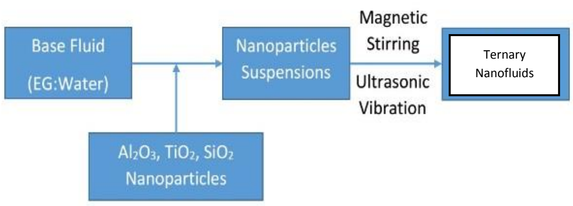


Figure 2. The preparation of ternary nanofluids [41]

Stability of Al_2O_3-TiO_2-SiO_2 nanofluids

The stability investigation of the ternary nanofluids in the present study is conducted through visual observation, measurement of a UV-Vis Spectrophotometer. The sedimentation through visual observation was performed for up to 30 days. Nanofluids will be considered stable when the concentration is constant. Previously, the observed visible sedimentation of the prepared nanofluids Azmi, et al. [39]. The UV-Vis was conducted for 30 days (720 h) by varying the sonication time. The wavelength of the UV-Vis spectrophotometer is set at 900 nm following the study by Hamid, et al. [40]. The UV-Vis measures scattering nanofluid's absorption and light intensity by comparing intensity level with 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 was also used by previous studies [38, 41].

Thermal conductivity measurement of Al_2O_3-TiO_2-SiO_2 nanofluids

The thermal conductivity measurement method followed the ASTM D5334 and IEEE 442-1981 standards, using KD2 Pro Property Analyzer (Decagon Devices) shown in **Figure 3**. Part of the thermal test sample conductivity, KS-1 sensor to read of k [W/m K], measuring bottle to put samples to be tested, KD2 Pro controller is an integral part of the 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. To maintain a constant sample temperature is used a water bath. Previously, validation of thermal conductivity values from thermal conductivity sensors using standardized liquid glycerine supplied by Decagon Devices. The measured k is 0.286 W/m K with accuracy ± 0.35%. Thermal conductivity measurements are performed several times and taken the average measurement time of 15 minutes for each data set at different temperatures. It is crucial to minimize errors in measurements with free convection due to temperature variation and the sensor directly touching the liquid sample.

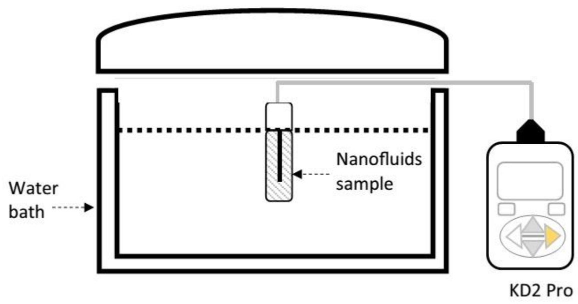


Figure 3. KD2 Pro thermal properties to the measurement of thermal conductivity

Dynamic viscosity measurement of Al_2O_3-TiO_2-SiO_2 nanofluids

Viscosity measurement used water bath circulation at Brookfield LVDV III Ultra Rheometer. Operating conditions of Rheometer for viscosity measurement from 1 to 6 × 106 mPa.s. The sample of 16 mL was added to the cylinder jacket and pasted into a Rheometer. The RheoCal program is used for determining the VST at various spindle velocities. Dynamic viscosity measurements were performed in the temperature range of 30~70°C. The use of a circulating water bath is made to control the sample temperature. The VST measurements are repeated five times, and the average value reported. Dynamic viscosity measurements used the apparatus shown in **Figure**

4. Water: EG in 60:40 mixture ratio at different temperatures is validated by the values reported in the literature. Furthermore, dynamic viscosity measurements are performed for Al_2O_3-TiO_2-SiO_2 or ternary nanofluids.

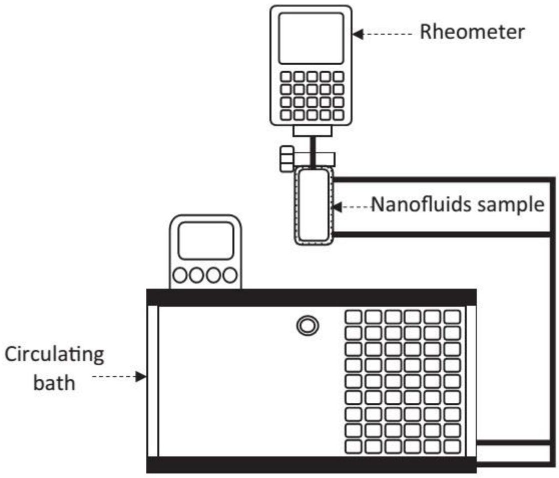


Figure 4. Brookfield LVDV III Ultra Rheometer to the measurement of dynamic viscosity

RESULTS AND DISCUSSION

Stability of Al_2O_3-TiO_2-SiO_2 nanofluids

The absorbance ratio determines the stability of Al_2O_3-TiO_2-SiO_2 nanofluids. *A*s shown in **Figure 5**, A_o is the initial absorbance, referring to the reference liquid absorbance value (base liquid). *A* is the final absorbance, referring to the absorbance value of Al_2O_3-TiO_2-SiO_2 nanofluids. The ideal A_r value is one (100%). This condition will reflect the constant absorbance value with time and perfectly stable for the suspended particle without any sedimentation occurring during the sedimentation period. Therefore, the variation of *Ar* value with the sedimentation time determines the state of the stability of the sample [42]. **Figure 5** shows that the duration of sonication time in preparation also contributes to the different effects on stability. From **Figure 5**, one can observe that the 10 hour sonication time shows the best absorbance ratio compared to others. Thus, the preparation of the ternary nanofluids in the present study used 10 hours for the sonication process. Preparation of nanofluids that experienced sonication time up to 10 hours was found to be the most stable sample with A_r more than 88% for sedimentation 120 h - 720 h time.

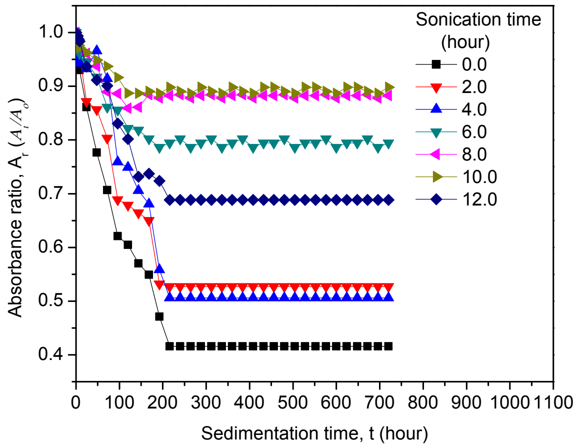


Figure 5. The absorbance ratio of ternary nanofluids for different sonication times within a 30 day

Meanwhile, other samples made with 6 and 12 hours of different sonication periods are considered less stable, with A_r lower than 80% starting at 120 h until 720 h. Then, samples began to decline with sedimentation time. Samples without sonication process were observed unstable with less than 50% absorption ratio after 240 h condition. The UV-Vis spectrophotometer determines the dispersion condition of suspended nanoparticles. Therefore, ternary nanofluids stability is influenced by sonication and sedimentation times. For thermal conductivity and dynamic viscosity rating, the remaining samples at different concentrations have been provided with a time of 10 hours of sonication. According to Yu, et al. [43], nanofluids are considered stable when the concentration or size of the suspended nanoparticles part remains fixed. The observation of the Al_2O_3-TiO_2-SiO_2 nanofluids sedimentation is shown in **Figure 6**. After 30 days of preparation, the Al_2O_3-TiO_2-SiO_2 nanofluids observed after 30 days is stable, as shown in **Figure 6 (d)**. The ternary nanofluids stability assessment is also determined by Ultra Violet-Visible (UV-Vis) spectrophotometer. The absorption and diffusion of light were measured by comparing the intensity of the ternary nanofluids light in a primary fluid of 60:40 (water: EG) [44].

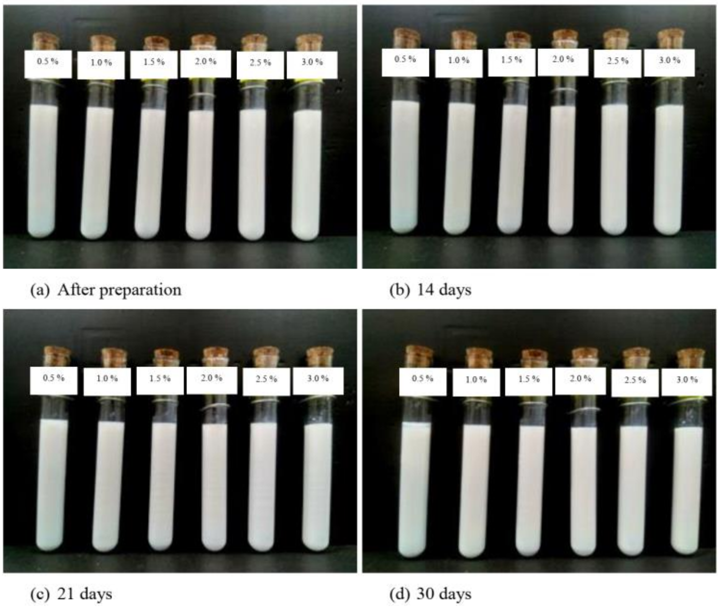
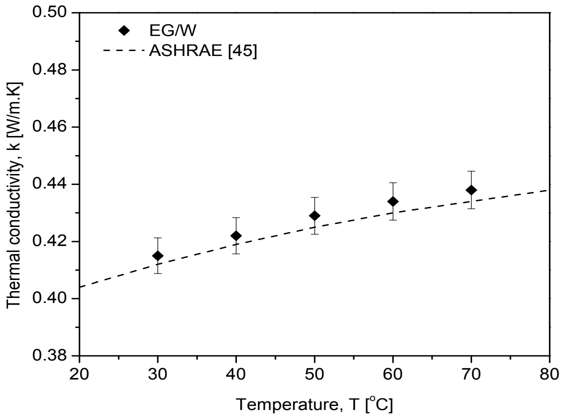


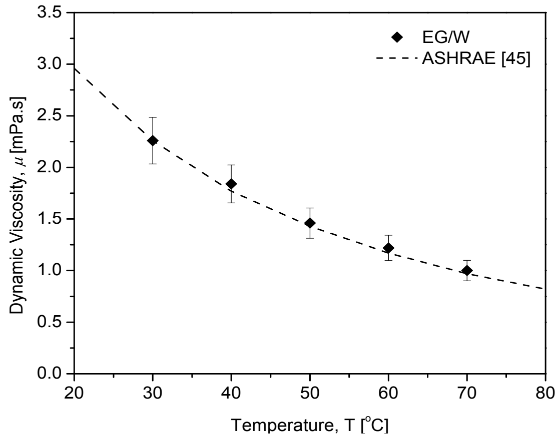
Figure 6. Sedimentation observation of ternary nanofluids: (a) after preparation, (b) 14 days, (c) 21 days, and (d) 30 days

Properties measurement validations with ASHRAE

Thermal conductivity measurement data and viscosity need to be validated using ASHRAE [45] for EG/Water (60:40). Measurement of thermal conductivity using KD2 Pro, data validation results with 1.5% error bar is presented in **Figure 7 (a).** Deviations for measurable data are less than 1.0% compared to ASHRAE [45]. Reddy, et al. [46] performed validation tests for deviations from their base fluid up to 2.5% compared to ASHRAE [45]. Furthermore, for the similar temperature range and water/EG ratio on validation of current viscosity measurements as stated by other papers [47-49]. **Figure 7 (b)** indicates that the viscosity data as per ASHRAE [45]. In addition, data for a 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 ternary nanofluids are performed.



(a) Thermal Conductivity



(b) Dynamic Viscosity

Figure 7. Validation of EG/Water (40:60) with ASHRAE

The thermal conductivity of Al_2O_3-TiO_2-SiO_2 nanofluids

Figure 8 show the relationships between ternary nanofluids thermal conductivity with temperature for volume concentration of 0.5~3.0%. The thermal conductivity of the ternary nanofluids for volume concentration variations may increase with temperature and higher than base fluid. Furthermore, the highest thermal conductivity was obtained for volume concentrations of 3% at a temperature of 70 °C with 27.1% higher than the base fluid. Meanwhile, the volume concentration of 0.5% provided the lowest thermal conductivity among temperatures investigated. In this study, the relationship between the composition ratio of nanoparticles (1/3: 1/3: 1/3) in the ternary nanofluids to increased thermal conductivity is influenced by three 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 in size than TiO_2 nanoparticles of 50 nm. Al_2O_3 and SiO_2 nanoparticles play a role in conduction by fulfilling larger TiO_2 nanoparticle spaces. Increasing the contact area for conduction between molecules, resulting in a higher heat transfer rate during a collision by the Brownian motion [50], requires a particular arrangement of the three nanoparticles. The effective thermal conductivity of ternary nanofluids is presented in **Figure 9**. The results revealed that the effective thermal conductivity increases with 3.0% volume concentration in ternary nanofluids, except for 0.5% volume with effective thermal conductivity of nanofluids are lowest. Effective thermal conductivity in ternary nanofluids can significantly affect the relationship between volume concentration and each set of temperatures.

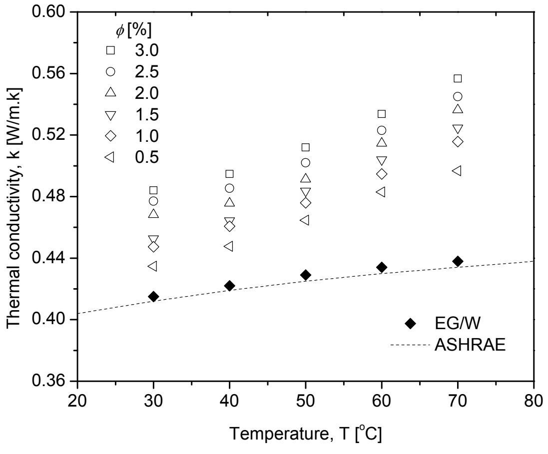


Figure 8. The experimental thermal conductivity of ternary nanofluids

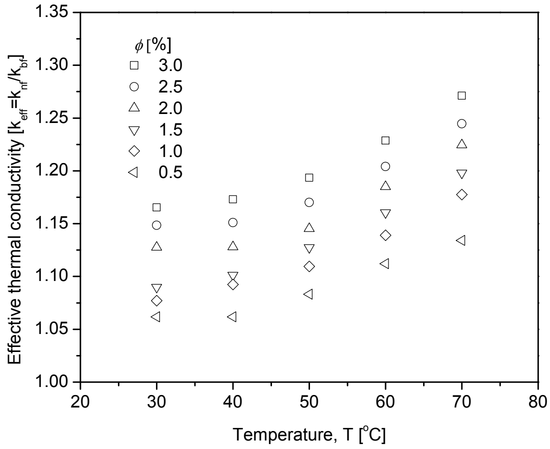


Figure 9. The effective thermal conductivity of ternary nanofluids

Dynamic viscosity of Al_2O_3-TiO_2-SiO_2 nanofluids

Figure 10 illustrated the dynamic viscosity against the shear rate in the range of $920 \leq \gamma \leq 4320$ s^{-1} for 0.5 % volume concentration of ternary nanofluids. The results indicate that the dynamic viscosity remains constant with the increase of shear rate for 0.5 % vol. The shear-independent VST demonstrates that the ternary nanofluids behaved as Newtonian fluids within the temperatures studied. The dynamic viscosity of the Al_2O_3-TiO_2-SiO_2 nanofluids for different temperatures has a significant effect on the concentration volume of 0.5%. A temperature of 30°C provides a higher dynamic VST than 40, 50, 60 and 70°C, as shown in **Figure 10.** It is probably due to the different intensities of Al_2O_3, TiO_2 and SiO_2 nanoparticles in both the composition ratio (1/3:1/3:1/3) in ternary nanofluids. **Figure 11** shows the dynamic viscosity for various volume concentrations of ternary nanofluids in the temperature range of 30~70°C. The VST at all volume concentrations follows the base fluid trend whereby it decreases exponentially with temperature. The viscosity of volume concentration of 3.0 is higher than the values of 0.5-2.5%. The volume concentration of 3% shows the highest value for VST at all temperatures. The dynamic VST of the ternary nanofluids decreased slightly. The VST of ternary nanofluids varied with the composition ratio of Al_2O_3, TiO_2 and SiO_2 nanoparticles mixture ratio, which might be due to the difference in the interactions of those particles with the base fluid. However, the effect of temperature on the Al_2O_3-TiO_2-SiO_2 nanofluid viscosity for all mixed ratios decreases with increasing temperature, evidenced by Asadi, et al. [51].

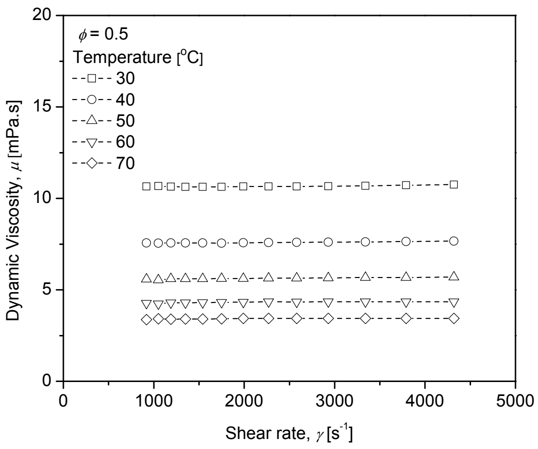


Figure 10. Variation of dynamic viscosity with shear rate

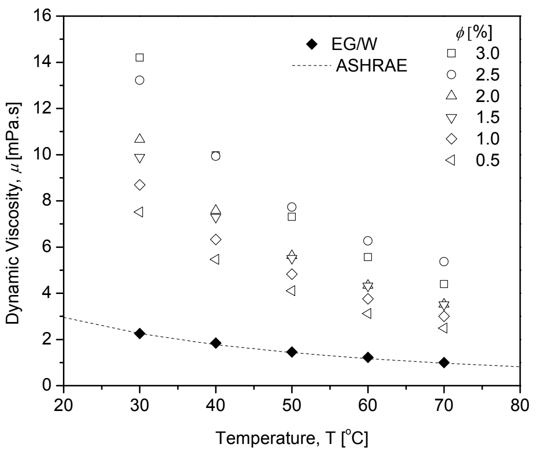


Figure 11. Variation of dynamic viscosity with temperature

The variation of relative viscosity with temperature is shown in **Figure 12**. From the figure, the maximum relative VST for all volume concentrations occurs at the different temperatures of 30~70 °C. For a volume concentration of 3%, the viscosity ratio is higher at 30 °C than 40 °C and then increases at 50~70 °C. Similarly, this pattern also applies to the volume concentration of 0.5, 1.0, 1.5, 2.0 and 2.5 %. From the results, the relative VST distribution for all volume concentrations is always lower than the value for the 3.0% vol sample in the range of temperature investigated. The results also show a specific tendency for VST at different volume concentrations. It may be attributed to varying effects on shear flow resistance due to the three nanoparticles of different concentrations and particle sizes.

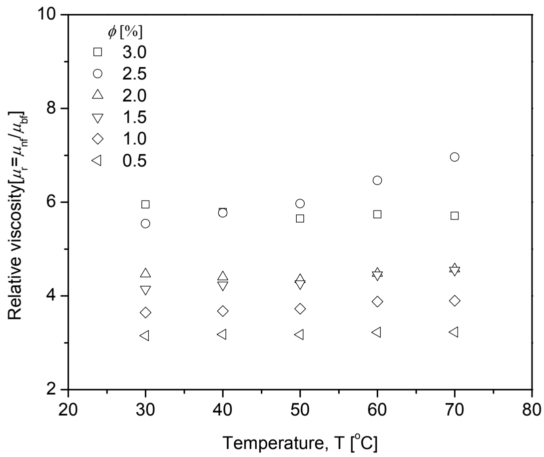
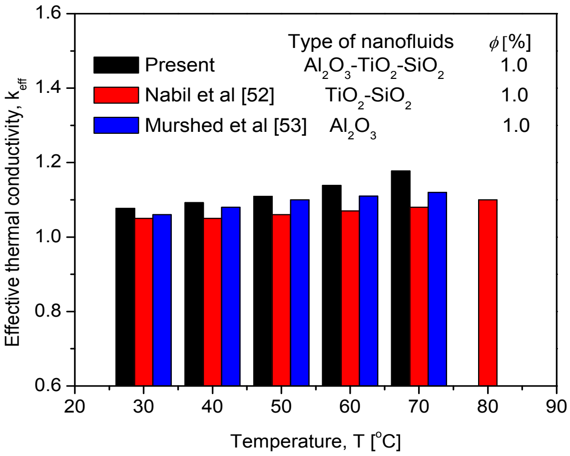


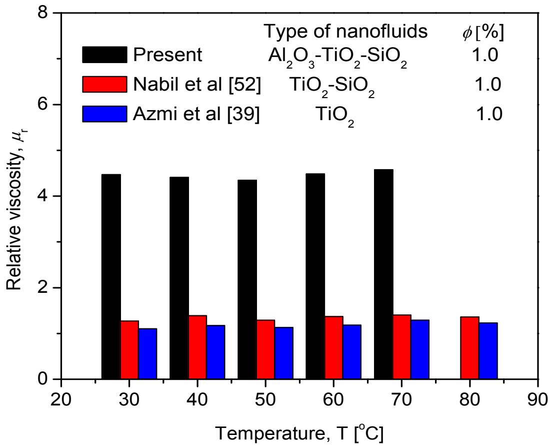
Figure 12. Variation of relative viscosity with temperature

Comparison with literature

Figure 13 (a) and **(b)** demonstrate comparisons of the effective thermal conductivity and relative viscosity of the present study with the data from Nabil, et al. [52], Murshed, et al. [53], and Azmi, et al. [47]. In this study, the thermal conductivity of ternary nanofluids was increased by 1.07-1.17 times compared to a base fluid for a concentration of 1.0%. Murshed, et al. [53], using Al_2O_3 nanoparticle with 1.0% volume concentration of base fluid used by EG-Water mixture. They proved that the experimental values of the different nanofluid thermal conductivity increased significantly with fluid temperature, which enhances the nanoparticles' Brownian motion and decreases the base fluid viscosity. With the influence of intense Brownian action, the contribution of micro connectivity to heat transport increased, resulting in improved thermal conductivity of nanofluid. A study by Nabil, et al. [52] used TiO_2-SiO_2 with EG/Water as a base fluid. They present the results of the thermal conductivity of the TiO_2-SiO_2 nanofluids enhance with increasing concentration and temperature. However, 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. [47] conduct relative viscosity measurements for TiO_2 nanoparticles in EG/Water-based fluid with a concentration of 0.5%. The study results prove that the relative viscosity in the range studied increases around 1.35-1.75 times compared to the water/EG mixture. The reason for fluctuations in relative viscosity in an unspecified temperature range. Their relative viscosity is the lowest value compared to the others, as shown in **Figure 13 (b)**. According to Sundar, et al. [37], the magnitude of the increase in thermal conductivity or relative viscosity depends on the type of nanoparticle and the base fluid, thus being observed and illustrated in **Figure 13 (a)** and **(b)**.



(a) Comparison of thermal conductivity



(b) Comparison of dynamic viscosity

Figure 13. Comparison of ternary nanofluids properties with the data from the literature

Regression correlations

The properties regression equations (3) and (4) can aid in analyzing the experimental data for thermal conductivity and dynamic viscosity. The equation is combined with the volume concentration of ternary nanofluids for the range 0.5-3.0%. This equation applies to estimating ternary nanofluids at 60:40 (Water/EG) and temperatures from 30 to 70°C. The average deviation (AD) and standard deviation (SD) for thermal conductivity were 2.5% and 3.6%, respectively. For viscosity, the values are 11.7% and 17.1%, respectively. Measurement data is in good agreement with estimated values from Eq. (3) and (4) based on the statistical analysis shown in **Figure 14** and **15**.

$$\frac{k_{nf}}{k_{bf}} = 0.9191\left(1+\frac{\phi}{100}\right)^{5.4525}\left(1+\frac{T}{70}\right)^{0.2289} \quad (3)$$

$$\frac{\mu_{nf}}{\mu_{bf}} = 2.424\left(1+\frac{\phi}{100}\right)^{34.0253}\left(\frac{T}{70}\right)^{0.0827} \quad (4)$$

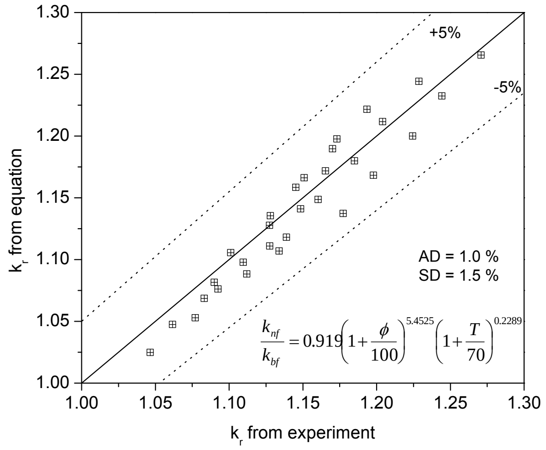


Figure 14. Comparison of effective thermal conductivity with Eq. (3).

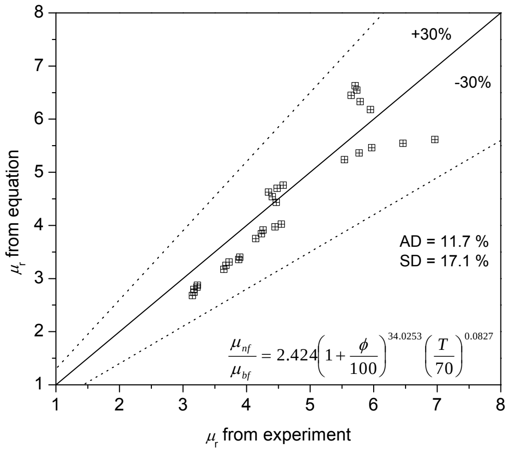


Figure 15. Comparison of relative viscosity with Eq. (4).

CONCLUSION

In this study, ternary nanofluids thermal conductivity and dynamic viscosity were investigated for six concentration volumes and temperatures from 30 to 70°C. The experimental results showed that the concentration volume of 3.0% obtained the best according to the effective thermal conductivity and relative viscosity compared to 0.5~2.5%. Therefore, in this study, different concentration volumes become the control parameters and performance studied. In terms of increased thermal conductivity, 3.0% vol provided a maximum increase of up to 27.1% while observed 0.5% had the most negligible increase for dynamic viscosity, which was approximately 1.19 times average compared to other ratios. The regression equations for the estimation of thermal conductivity ratio and relative VST were presented with reasonable accuracy. The equations are applicable for volume concentrations 0.5-3.0% and working temperatures of 30 to 70 °C.

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