

Water:Ethylene Glycol Properties Alteration Upon Dispersion Of Al_2O_3 and SiO_2 Nanoparticles

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ABSTRACT

Proton exchange membrane fuel cell (PEMFC) seems to be a popular option as a green energy carrier due to its high efficiency and pollutant-free operation. However, the slight temperature difference between the working temperature and surroundings requires innovation in cooling strategy. Active thermal management strategy is limited due to the larger space requirement. Alternatively, utilizing nanofluids as coolant as the passive cooling strategy tends to be a viable quick fix. In this research, thermophysical properties of $Al_2O_3:SiO_2$ hybrid nanofluids in the base fluid of water: Ethylene Glycol (EG) were discussed comprehensively concerning alterations made in thermal conductivity, dynamic viscosity, and electrical conductivity properties. There were four mixture ratios of 0.5% volume concentration of hybrid nanofluids considered ranging from 10:90, 30:70, 50:50, and 70:30 $Al_2O_3:SiO_2$. Upon completion of the study, there is an improvement of 9.8% shown in 10:90 $Al_2O_3:SiO_2$ hybrid nanofluids for thermal conductivity measured at 60 °C in comparison to the base fluid. Meanwhile, 10:90 $Al_2O_3:SiO_2$ hybrid nanofluids are also favorable with the lowest values of viscosity as compared to other mixture ratios resulting in lower parasitic loss. Electrical conductivity on the other hand also showed an increment in 10:90 $Al_2O_3:SiO_2$ hybrid nanofluids as compared to base fluid and other mixture ratios.

Keywords: *Dynamic Viscosity; Electrical Conductivity; Thermal Conductivity; Aluminium Oxide; Silicon Dioxide*

Introduction

Researchers have recently become highly concerned with the replacement of fossil fuels with renewable energy. Fuel cell applications are among the popular candidates for renewable energy which has been massively used in many sectors such as communication, stationary power, and transportation [1]. A Proton Exchange Membrane Fuel Cell (PEMFC) is an electrochemical instrument that produces electricity by converting chemical energy. The chemical energy from the reaction of hydrogen that acts as a fuel with oxygen has generated electricity as the product. The molecules of hydrogen split into protons and electrons at the anode side while the electrons flow through the outer circuit to produce electricity. Excessive heat is also produced during the reaction. The PEMFC is favorable as a potential energy generation device due to its excellent energy conversion efficiency of 60% in comparison to 20%-30% in Internal Combustion Engines (ICE) [2]. The PEMFC attracts researchers especially in the area of transportation due to its numerous advantages including high energy efficiency, and a quick start-up time which allows for almost instantaneous power generation. The PEMFC is also lightweight and compact in design. Its zero-emission operation is one of the most significant advantages as the only byproducts of PEMFC operation are water vapor and heat, making them environmentally friendly [3]. As concerns over climate change and air pollution intensify, the zero-emission nature of PEMFCs positions them as a better substitute for conventional internal combustion engines, helping to lessen reliance on fossil fuels and carbon dioxide emissions. These advantages make PEMFC a promising technology for achieving sustainable and clean transportation solutions in the future [4].

In PEMFC, thermal management focuses on achieving the ideal temperature for its membrane's electrochemical process [5]. In order to maximize the PEMFC performance, it is crucial to ensure that the working conditions are balanced in the aspect of humidity, temperature, and reactant characteristics [6]. A typical Sankey diagram of PEMFC shows that there are 50% of the hydrogen reaction output is converted to power, while 45% is released as waste heat and 5% is from the excessive hydrogen [7]. Therefore, thermal management in PEMFC is quite challenging. The PEMFC also has to have the capability to remove excessive heat at a smaller temperature difference between the operating and the ambient temperature. Several mechanisms of heat removal were discovered by researchers namely liquid cooling, phase change cooling, air cooling, and edge cooling method [8]. Among these mechanisms, the liquid cooling method is believed as the most effective method in the PEMFC heat transfer mechanism [9].

On another note, engineered colloids are termed as nanofluids which consist of a base fluid and nanoparticles that range from 1 to 100 nm in size [10]. The existence of the metallic nano-sized particles that are dispersed homogeneously in the base fluid, has shown a significant improvement in

thermal conductivity property due to greater total surface area and improvement in the Brownian motion of the base fluid [11]. The higher the surface area, the higher the heat transfer contact surfaces, which leads to a higher rate of heat transfer [12]. Apart from thermal conductivity, other thermo-physical properties that interest the researchers are alterations in density, dynamic viscosity, specific heat, and electrical conductivity.

Single nanofluids thermo-physical properties of thermal conductivity and electrical conductivity of Al_2O_3 nanofluids were reported by Zakaria et al. [13] who highlighted that the enhancement of thermal conductivity and electrical conductivity was up to 12.8% and 14.3%, respectively. Azmi et al. [14] studied the thermos-physical properties of TiO_2 nanofluids in water-based and discovered an enhancement of thermal conductivity up to 24.2%. The performance of thermo-physical properties of EG-based ZnO nanofluids was also reported by Li et al. [15] who reported a 9.1% enhancement in its thermal conductivity.

The nanofluids study then emerged from single nanofluids to hybrids and also ternary nanofluids [16]. Saifuddin et al. [17] researched the relationship of thermal conductivity, viscosity, and electrical conductivity for the hybrid ratio of Al_2O_3 : SiO_2 in water. He established a ratio known as thermal-hydraulic-ratio and proposed that the best mixture ratio was 10:90 (Al_2O_3 : SiO_2). In comparison to another hybrid nanofluids study by Hamid et al. [18] which was TiO_2 : SiO_2 hybrid nanofluids, thermal conductivity values of Al_2O_3 : SiO_2 has exceeded the thermal conductivity of TiO_2 : SiO_2 significantly at all ratios studied up to 43.5% enhancement.

The advantage of nanofluids seems promising to be incorporated as an alternative cooling medium to a liquid cooled PEMFC. Unlike the active cooling heat transfer method, this passive cooling heat transfer strategy will reduce the possibility of a bulky cooling system of a PEMFC with a more compact heat exchanger size. The general guidelines for the PEMFC coolant have been outlined by McMullen et al. [19] and Zakaria et al. [20]. Among the highlighted general criteria of coolant for PEMFC are a boiling point of less than 90 °C, thermal conductivity to be higher than 0.4 W/m.K, viscosity of less than 1cP at 80 °C, and also electrical conductivity value of less than 2 $\mu\text{S}/\text{cm}$.

The use of nanofluids in PEMFC has been among the favorite subjects of many research studies. Zakaria et al. [21] tested Al_2O_3 in a 2.4 kW_e PEMFC and reported that the 0.1 vol% of Al_2O_3 has the highest Thermal-Electrical-Ratio (TER) which concluded to be the most feasible for adoption in PEMFC. A similar study was performed with the adoption of ZnO in water: ethylene glycol base fluid [22]. The finding reported that there was a possible radiator's frontal area reduction by 27% with the adoption of 0.5 vol% of ZnO to the base fluid studied. An extensive review of the usage of nanofluids in PEMFC has also been conducted by Islam et al. [23]-[24].

Unlike the existing work of Saifudin et al. [17] who studied hybrid nanofluids properties in the base fluid of water, and base fluid of water:

BioGlycol by Johari et al. [5], this paper reports comprehensive thermo-physical properties of Al₂O₃:SiO₂ hybrid nanofluids in the base fluid of water: EG. The hybrid nanofluids that have been investigated were four different mixture ratios namely 10:90, 30:70, 50:50, and 70:30 (Al₂O₃:SiO₂). The thermo-physical properties measured were thermal conductivity, dynamic viscosity, and electrical conductivity. These properties were then summarized and represented in thermal-hydraulic and thermal-electrical ratios for better understanding in terms of the effect on the respective thermo-physical properties to each other in terms of PEMFC application. Hybrid nanofluids with a low concentration of 0.5% volume were used because of the PEMFC's low electrical conductivity limit [25]. In comparison to its base fluid and its single nanofluids, hybrid nanofluids showed a higher capability of heat transfer enhancement due to their higher thermal conductivity value. A recommendation on the potential mixture ratio of Al₂O₃:SiO₂ hybrid nanofluids as an advanced coolant replacement of PEMFC is presented at the end of this study.

Methodology

Nanofluids preparation

The Al₂O₃ nanoparticles in 13 nm size with 99.8% purity were used in this experiment. The nanoparticles were procured from Sigma Aldrich and were dispersed via a 2-step method in a 60:40 (water: EG) base fluid to form single Al₂O₃ nanofluids. Meanwhile, SiO₂ nanofluids were purchased in dispersion form and the size of 30 nm with 99.8% purity. The SiO₂ nanofluids were also prepared using the 2-step method. No surfactant was added during the preparation of both single nanofluids. Table 1 shows the properties of nanoparticles and base fluid used in this experiment.

Table 1: Properties of Al₂O₃ and SiO₂ nanoparticles and water:EG

Fluid name	Thermal conductivity, k (W/mK)	Specific heat, C_p (J/kg.K)	Viscosity, μ (Pa.s)	Density, ρ (kg/m ³)	Ref.
Al ₂ O ₃	36	765	-	4000	[26]
SiO ₂	1.4	745	-	2220	[27]
Water	0.615	4180	0.00085	996	[28]
W:EG 60:40	0.4096	3491.80	0.00245	1056.72	[29]

Each nanoparticle's quantity was initially measured and added to a predetermined volume of a base fluid. Equation (1) was used to calculate the required amount of Al₂O₃ nanoparticles to form a specific vol % concentration.

As for SiO₂, it was in 25% weight dispersion form, and Equations (2) and (3) were used to measure the dilution required to form a specific vol% concentration [30].

$$\phi = \frac{\left(\frac{m_p}{\rho_p}\right)}{\left(\frac{m_p}{\rho_p} + \frac{m_{bf}}{\rho_{bf}}\right)} \times 100 \quad (1)$$

$$\phi = \frac{\omega \rho_{bf}}{\left(1 - \frac{\omega}{100}\right) \rho_p + \frac{\omega}{100} \rho_{bf}} \quad (2)$$

$$\Delta V = (V_2 - V_1) = V_1 \left(\frac{\phi_1}{\phi_2} - 1\right) \quad (3)$$

where ω is the mass concentration of the nanoparticles, ρ is the density in kg/m³, and subscript p and bf refer to the nanoparticle and base fluid respectively. This ΔV is the volume of water required to be added to the current base fluid volume V_1 with volume concentration ϕ_1 to reach volume V_2 with volume concentration ϕ_2 of nanofluids.

To assess the existence of nanoparticle dispersion in the base fluid, Transmission Electron Microscopy (TEM) was employed. The TEM images for Al₂O₃ and SiO₂ are shown in Figure 1. The image demonstrated that there was a presence of particles with sizes of 13 nm for Al₂O₃ and 30 nm for SiO₂ in the hybrid Al₂O₃:SiO₂ nanofluids prepared.

In the initial mixing process, Al₂O₃ nanoparticles were dispersed in water: EG base fluid which was then stirred slowly using a glass stirrer until the white Al₂O₃ powder dissolved in the base fluid. In order to get good dispersion of nanofluids, the stirring process was further continued using a magnetic stirrer for 30 minutes, before being subjected to a sonication process for three hours. Both single Al₂O₃ and SiO₂ nanofluids at 0.5% volume concentration were prepared prior to mixing to form the required mixture ratio. Four mixture ratios of Al₂O₃:SiO₂ hybrid nanofluids in w:EG were prepared which were 10:90, 30:70, 50:50, and 70:30.

Upon completion of the preparation process, stability measurement was performed to ensure the level of stability of the samples. There were two methods employed which were zeta potential and physical observation.

Nanofluids stability observation

There were two methods used in this research to observe the stability of the nanofluids prepared. This is important to ensure the validity of the findings

from this study. The first method used was visual observation. Each sample of sonicated hybrid nanofluids was measured for 10 ml and poured into a test tube for observation. This visual inspection was conducted to observe the agglomeration effect after two hours of sonication [31]. Each sample was observed immediately upon completion of the preparation and also after 60 days of preparation. Anderson et al. [32] stated that the sedimentation at the bottom of the test tube is caused by the nanoparticles' agglomeration which will change the colloidal suspension's composition. The researcher also reported that the nanofluids' stability was also affected as the mixture color changed from white to transparent, especially at the top portion of the test tube. Nanofluids have higher stability if the sample maintains in white colour over time, but lower stability if the sample exhibits a transparent appearance as it shows evidence of agglomeration and sedimentation.

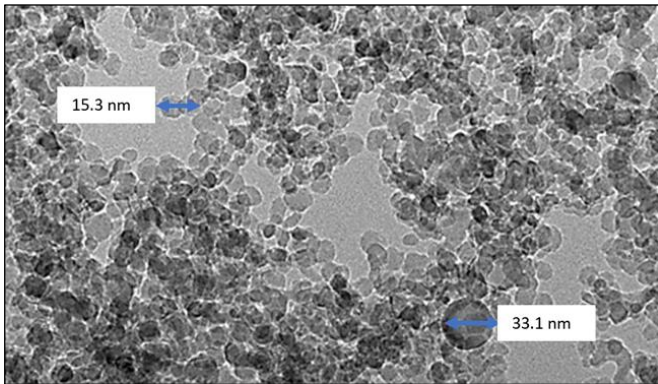


Figure 1: TEM images for dispersion of $\text{Al}_2\text{O}_3\text{-SiO}_2$ in water:EG

The second method employed was Zeta potential measurement. The Zeta potential is a measure of the electrical charge present at the surface of nanoparticles which arises because ions from the surrounding fluid have been adsorbed onto the particle surface, resulting in the formation of an electrical double layer [33]. The magnitude and sign of the zeta potential determine the stability of nanofluids. When the zeta potential is high, it creates electrostatic repulsion forces between the charged nanoparticles, preventing them from forming agglomerates, and thus hindering the aggregation and sedimentation of nanoparticles. Malvern Instruments Ltd.'s Zetasizer nano ZS was used to measure the zeta potential.

Nanofluids thermo-physical properties measurement

There were three properties of hybrid nanofluids measured in this study which were thermal conductivity, dynamic viscosity, and electrical conductivity. The

measurement methods for the thermos-physical properties are explained below.

Thermal Conductivity Measurement

Each sample of nanofluids was prepared in a small glass bottle of 25 ml before the measurements were taken. Then, each sample was placed in a water bath of WNB7L1 in order to maintain a constant required temperature range of 30 °C to 70 °C, with accuracy of 0.1 °C. The thermal conductivity of the nanofluids was measured using the KD2 Pro thermal property analyser of Decagon Devices Inc., USA as shown in Figure 2. The same device was also adopted in various studies of thermal conductivity [34]-[35]. The analyser complies with the standard of ASTM D5334 and IEEE 442-1981. The device operates on a transient hot-wire method to measure the thermal properties of solids and liquids in the range of 0.02 to 2 W/m.K with 5% accuracy. Ten readings were taken for each sample at the required temperature to ensure the measurement was within 5% accuracy.

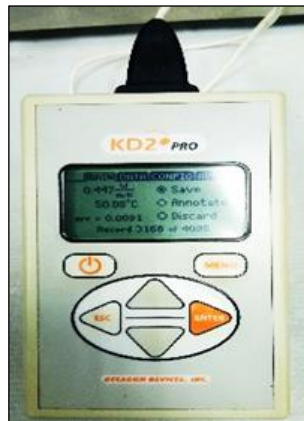


Figure 2: KD2 pro thermal property analyser

Dynamic viscosity measurement

This work employed the Brookfield LVDV-III Ultra Rheometer to test the dynamic viscosity of nanofluids. Renowned researchers who studied the dynamic viscosity of nanofluids also employed the same device [35]-[36]. The device's spindle was submerged in the nanofluid sample to measure the fluid's viscosity up to 100 centiPoise (cP). A rotary transducer then measured the spring deflection to measure the viscous drag of the fluid against the spindle. In this study, only a small amount of each sample was used to attain temperature equilibrium rapidly. The viscometer was equipped with a water bath device to allow the sample temperature to be adjusted. To reach the

temperature required, the water bath unit was connected to the inlet and outlet ports of the sample, and sufficient time was given. The spindle speed can vary up to 250 rpm. Results are good when spindle type and speed are combined as long as the torque applied is within 10% and 100% of the maximum allowable range. The data are discarded if the torque does not fall within the designated range. The Brookfield Engineering Laboratories was responsible for the Brookfield LVDV-III Ultra Rheometer annual calibration to confirm the reliability of its data.

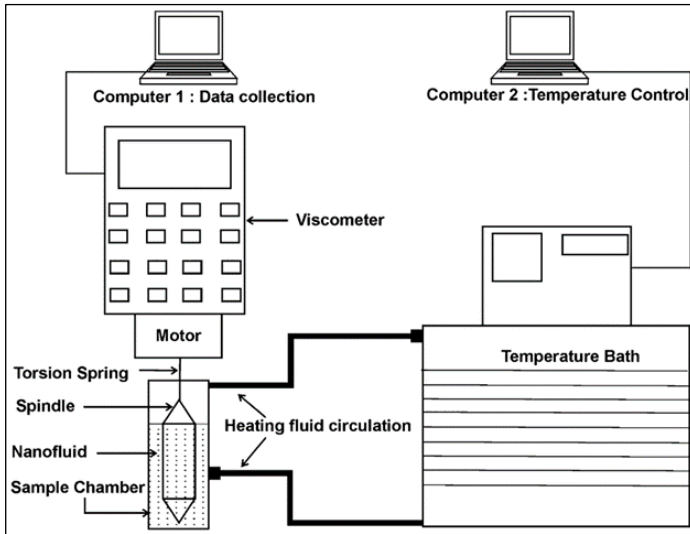


Figure 3: The Brookfield LVDV-III ultra rheometer setup

Electrical conductivity measurement

Each sample of nanofluids was prepared in 50 ml and was placed on a heater in order for the sample to reach and maintain a constant required temperature range of 30 °C to 70 °C. The electrical conductivity measurement setup can be seen in Figure 4. Three readings for each sample and temperature were recorded for the accuracy of the measurements. The electrical conductivity measurement of nanofluids was performed using the EUTEC Handheld Meter Kit PC450. It has a built-in thermistor for automatic temperature compensation. The measuring device was calibrated using standard solutions of distilled water and ethylene glycol as recommended in ASHRAE standards [29].

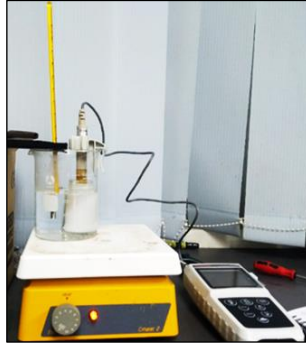


Figure 4: Electrical conductivity measurement

Mathematical model: Property-enhancement-thermo-hydraulic-ratio (PER_{t/v})

Property enhancement of Thermo-Hydraulic-Ratio (PER_{t/v}) is an evaluation through comparison of enhancement in heat dissipation over the penalty of viscosity of a fluid. This method was also adopted by Saifuddin et al. [17] to investigate the effect of thermal conductivity on the dynamic viscosity of Al₂O₃:SiO₂ hybrid nanofluids in water. In their investigation into carbon nanotube-based nanofluids (CNTs), Garg et al. [37] concluded that an advantage's PER_{t/v} value for laminar flow shouldn't be greater than 5.0. As for the PER_{t/v} effect on PEMFC application, it signifies the impact of increasing the thermal conductivity of nanofluids to the higher requirement in pumping power to circulate the cooling fluids in the system. The PER_{t/v} equation used is shown as Equation (4) [17],

$$PER_{t/v} = \frac{\mu_r - 1}{k_r - 1} = \frac{k_{bf}(\mu_{hnf} - \mu_{bf})}{\mu_{bf}(k_{hnf} - k_{bf})} \quad (4)$$

where hybrid nanofluids viscosity and base fluid represent as μ_{hnf} and μ_{bf} , respectively.

Mathematical model: property-enhancement-thermo-electrical-ratio (PER_{t/e})

On the other hand, a relationship between the characteristics of electrical and thermal conductivity of hybrid nanofluids is established from the correlation of increment in thermal conductivity and stringent limit of electrical conductivity requirement in PEMFC application as in Equation (5) [17].

$$PER_{t/e} = \frac{\sigma_r - 1}{k_r - 1} = \frac{k_{bf}(\sigma_{hnf} - \sigma_{bf})}{\sigma_{bf}(k_{hnf} - k_{bf})} \quad (5)$$

$PER_{t/e}$ established in this study represents the effect of enhanced thermal conductivity of nanofluids on the electrical conductivity property. A lower $PER_{t/e}$ value is favorable due to the strict limit of electrical conductivity permissible by PEMFC [37].

Uncertainty analysis

Uncertainty analysis was performed to evaluate the uncertainty measurement of all related independent varying factors. According to Beckwith et al. [38], the uncertainties for the primary analytical parameter are evaluated. As for the thermophysical properties of thermal conductivity, dynamic viscosity, and electrical conductivity, the error analysis considered was $\pm 0.1\%$.

Results and Discussions

Stability test

To analyse the stability of the hybrid nanofluids, two tests were conducted to ensure the stability of the hybrid $Al_2O_3:SiO_2$ nanofluids prepared. The methods were visual inspection as well as zeta potential measurement.

Visual observation

Samples of hybrid nanofluids are shown in Figure 5 both showing immediately after preparation and 60 days later. It was examined that there was a bit of sedimentation occurred in the bottom area of the samples. Sedimentation build-up was noticeable as the top region of the test tube became more transparent than before. The higher content of Al_2O_3 showed an obvious sedimentation effect as the 70:30 hybrid ratio has a prominent separation issue after 2 months as compared to other hybrid ratios with a lower ratio of Al_2O_3 . This is expected as the Al_2O_3 nanofluids are whitish while SiO_2 is clear in color. However, sedimentation should not be an issue as the applications are mostly in a forced cooling loop where the sedimentation will be immediately dissolved upon forced circulation of the fluid. The monitoring indicates that the prepared hybrid nanofluids were stable and appropriate for further evaluation of their thermo-physical properties.

Zeta potential measurement

The second method deployed for stability measurement was zeta potential and the result is summarized as in Figure 6. An outline classified by Lee et al. [39] stated that the zeta value below 5 mV is pronounced aggregation, 5 to 20 mV for limited stability, values from 20 to 30 mV are stable, and absolute stability of zeta potential from 30 to 60 mV. The zeta potential measurements obtained for the hybrid nanofluids prepared for the research were in the absolute stability region. This suggests that the particles have larger electrostatic

repulsion forces which prevent the attraction between neighboring particles thus reducing the possibility of sedimentation in the hybrid nanofluids impact.

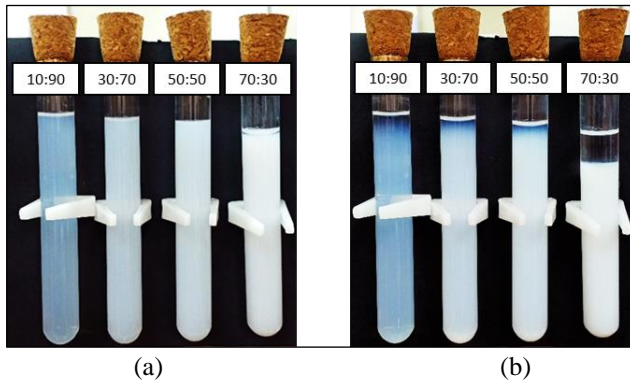


Figure 5: Samples of hybrid $\text{Al}_2\text{O}_3:\text{SiO}_2$ nanofluids; (a) right after preparation, and (b) after 2 months

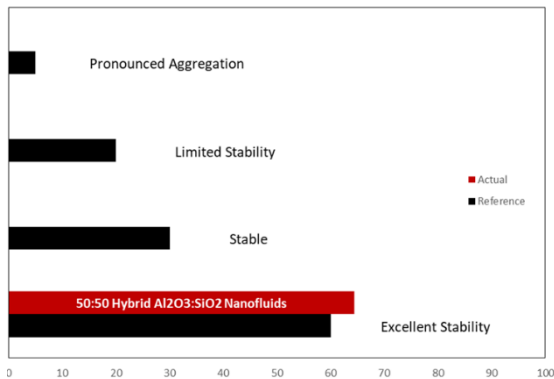


Figure 6: The zeta potential analysis for a mixture ratio of 50:50 ($\text{Al}_2\text{O}_3:\text{SiO}_2$) hybrid nanofluids

Thermophysical properties of hybrid $\text{Al}_2\text{O}_3:\text{SiO}_2$ nanofluids

The thermos-physical properties of hybrid $\text{Al}_2\text{O}_3:\text{SiO}_2$ nanofluids in water:EG based were measured in this study including thermal conductivity, dynamic viscosity, and electrical conductivity of the hybrid nanofluids.

Thermal conductivity measurement

Figure 7 shows the association of thermal conductivity with regards to the mixture ratio of $\text{Al}_2\text{O}_3:\text{SiO}_2$ and also temperature. It was noticed that the value

of thermal conductivity dropped as the Al_2O_3 content increased. This can be observed with every temperature measured. At 60 °C, the mixture ratio of 10:90 (Al_2O_3 : SiO_2) demonstrated the maximum thermal conductivity, with a 9.8% improvement over the base fluid, followed by 30:70, 50:50, and lastly 70:30 with 6.3, 5.1 and 1.6, consecutively. The higher portion of SiO_2 nanoparticles was seen to increase the thermal conductivity value of the hybrid nanofluids. This was a novel finding even though the thermal conductivity value of Al_2O_3 is higher than SiO_2 single nanofluids [40]. These results are consistent with the hybrid trends of Al_2O_3 : SiO_2 in water base fluid that was observed by Saifudin et al. [17].

Thermal conductivity was also noted to increase when temperature was increased. The internal energy of nanoparticles is impacted by temperature increase. This will cause the fluid's particles to eventually move and vibrate more quickly, causing the Brownian effect to occur and increasing the amount of particle contact [41].

However, not all hybrid ratios have an enhancement in thermal conductivity as compared to the base fluids. Only certain ratios of hybrid Al_2O_3 : SiO_2 exceed the thermal conductivity value of their single nanofluids as shown in Figure 8. The Al_2O_3 : SiO_2 hybrid nanofluids thermal conductivity was compared to the single Al_2O_3 and SiO_2 nanofluids [28] thermal conductivity at a temperature of 30 °C. The 10:90 and 30:70 (Al_2O_3 : SiO_2) ratios were among the ratios that showed enhancement as compared to single nanofluids of Al_2O_3 and SiO_2 while the other mixture ratios were lower than the single nanofluids' thermal conductivity. A higher ratio of SiO_2 shows advantages in terms of improved thermal conductivity of Al_2O_3 : SiO_2 hybrid nanofluids as compared to single nanofluids of Al_2O_3 and SiO_2 . The findings are similar to Khalid et al. [17] with hybrid Al_2O_3 : SiO_2 nanofluids in water behavior.

Dynamic viscosity measurement

The effect of dynamic viscosity for hybrid Al_2O_3 : SiO_2 nanofluids in different mixture ratios at different temperatures is shown in Figure 9. It was noticed that the viscosity reduced as the temperature increased. This was impacted by the molecules' increasing average kinetic energy as the temperature increased. These energetic molecules move faster and weaken the intermolecular bonds between the molecules resulting in a lower viscosity value. The lowest viscosity was measured at 1.46 cP at 10:90. at 70 °C.

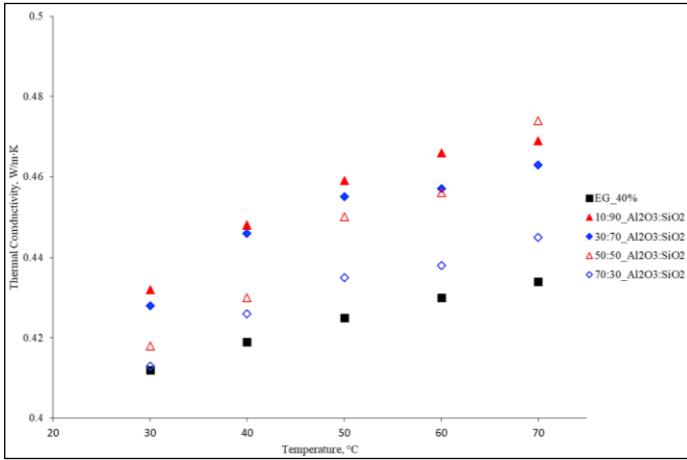


Figure 7: Thermal conductivity effect at different temperatures

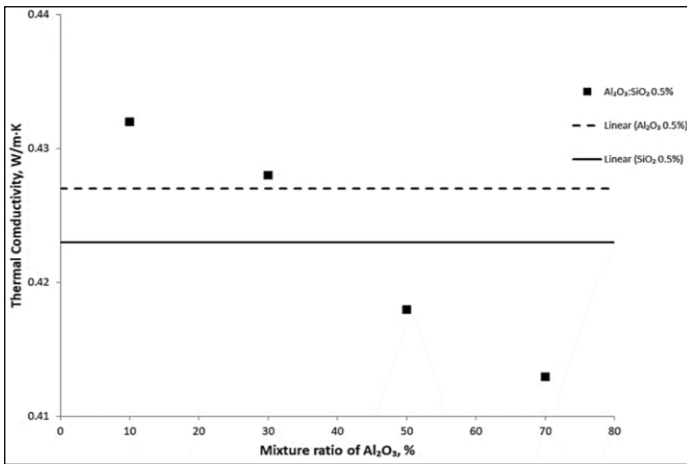


Figure 8: Comparison of thermal conductivity of hybrid Al₂O₃:SiO₂ nanofluids at 30 °C against single Al₂O₃ and SiO₂ nanofluids with 0.5% volume concentration [28]

Meanwhile, the viscosity value was observed to be increased in higher content of Al₂O₃ in the mixture ratio. This was due to the higher value of dynamic viscosity in single Al₂O₃ nanofluids which is 1.4 cP as compared to single SiO₂ nanofluids with 0.98 cP, measured at the same temperature of 60 °C [17]. At 70 °C, the hybrid Al₂O₃:SiO₂ nanofluids with the maximum viscosity were 70:30, which was 2.3 times greater than the base fluid of water:

EG. Subsequently, mixing ratios of 50:50, 30:70, and 10:90 were employed, with 1.8 times, 1.75 times, and 1.5 times higher viscosities than the base fluid. This was due to the existence of nanoparticles suspended in water:EG fluid, which increased the frictional forces between particles [12]. The ratio with the lowest viscosity value should be favorable in the actual application of hybrid Al_2O_3 : SiO_2 nanofluids in PEMFC due to the lower pumping work required to circulate the coolant throughout the cooling system. The operating temperature of PEMFC which is in the range of 60 °C to 80 °C was also favourable to this hybrid Al_2O_3 : SiO_2 nanofluids adoption as at this temperature region, a lower value of viscosity was achieved. The additional increment of viscosity of nanofluids as compared to the base fluid of water: EG will eventually increase the pumping power requirement. The additional pumping power gain is still negligible, though, considering the PEMFC's high electrical output [21].

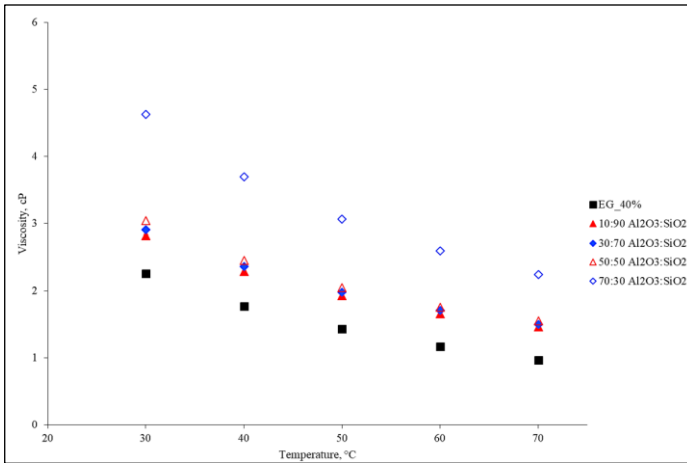


Figure 9: Viscosity effect due to variation in temperature and mixture ratio

Electrical conductivity measurement

The electrical conductivity showed an increase trend as the content of SiO_2 is increased as shown in Figure 10. This matches the single nanofluids electrical conductivity value for both Al_2O_3 and SiO_2 nanofluids as reported by [27]. It was reported that single SiO_2 nanofluids have a higher electrical conductivity value of 190 $\mu\text{S}/\text{cm}$ as compared to single Al_2O_3 nanofluids of 70 $\mu\text{S}/\text{cm}$ at the same temperature of 60 °C. It was observed that the highest electrical conductivity of 204.8 $\mu\text{S}/\text{cm}$ was observed in a mixture ratio of 10:90 (Al_2O_3 : SiO_2) at 70 °C. This is equivalent to 20.5 times higher as compared to base fluid 60:40 (water:EG) electrical conductivity [4]. Subsequently, 30:70 (Al_2O_3 : SiO_2) was observed, showing an 11-fold increase in comparison to the basic fluids, while 50:50 (Al_2O_3 : SiO_3) demonstrated a 9-fold increase. Lastly,

the mixture ratio of 70:30 with 7.5 times higher as compared to base fluid. In the case of PEMFC adoption, where there is a strict limit of electrical conductivity of as low as $5 \mu\text{S}/\text{cm}$ as specified by the fuel cell stack maker, Ballard Incorporation [37], more content of Al_2O_3 in the hybrid mixture ratio should be preferable mixture ratio as it has the lowest increase in electrical conductivity value.

However, Figure 11 shows the comparative value of hybrid $\text{Al}_2\text{O}_3:\text{SiO}_2$ nanofluids in water and hybrid $\text{Al}_2\text{O}_3:\text{SiO}_2$ nanofluids in water:EG base fluids at PEMFC working temperature of 60°C . The electrical conductivity showed that hybrid $\text{Al}_2\text{O}_3:\text{SiO}_2$ nanofluids in water were higher as compared to hybrid $\text{Al}_2\text{O}_3:\text{SiO}_2$ nanofluids in water:EG in all mixture ratios. This agrees well with the value of electrical conductivity of the base fluid which reported that water has a higher electrical conductivity value as compared to 60:40 (water:EG) [27].

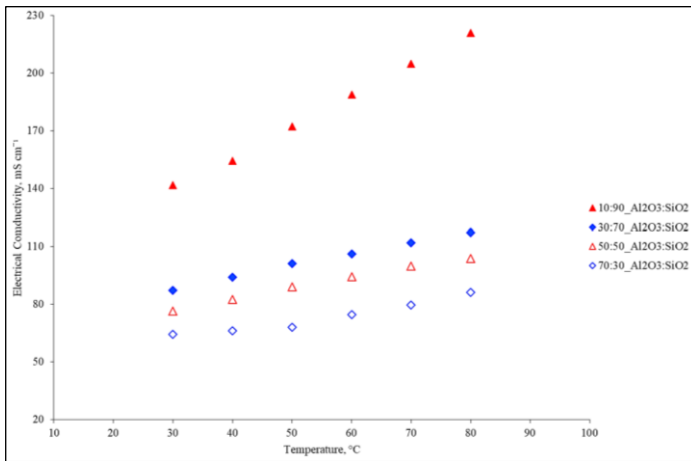


Figure 10: Variation of electrical conductivity against temperature and mixture ratios

Correlation of thermo-hydraulic-electrical properties of hybrid nanofluids

The thermo-physical properties data measured were then analyzed in order to establish a correlation between thermo-hydraulic-electrical properties. The thermal conductivity was associated with dynamic viscosity to establish the property-enhancement of thermo-hydraulic-ratio ($\text{PER}_{t/v}$), while thermal conductivity association with electrical conductivity data was used to correlate the property-enhancement of thermo-electrical-ratio ($\text{PER}_{t/e}$).

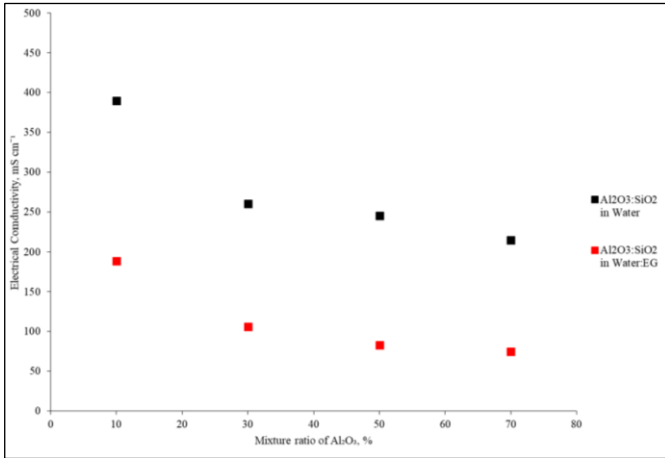
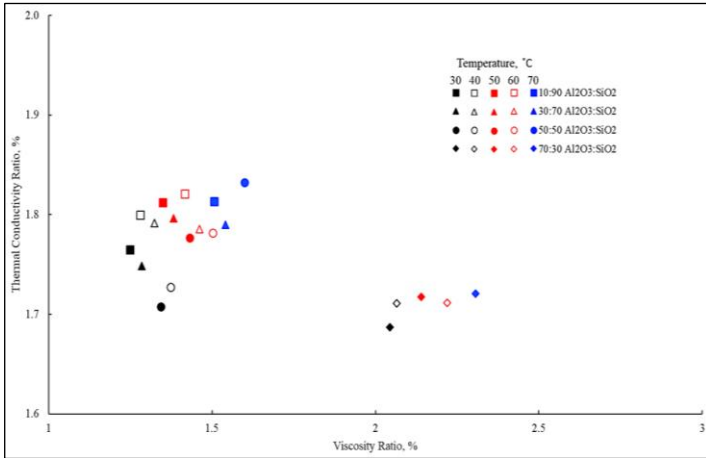


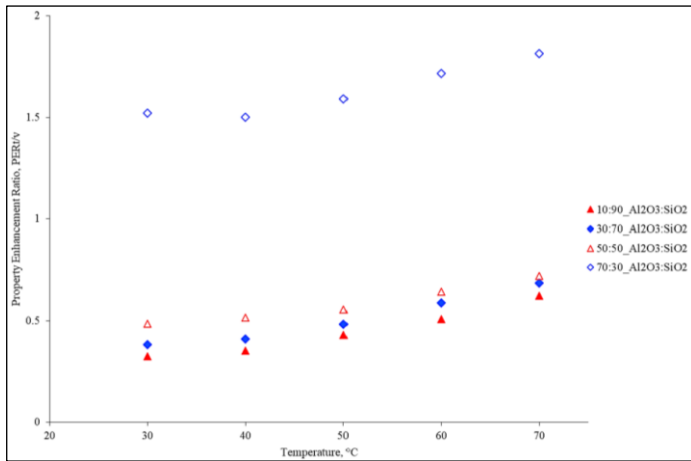
Figure 11: Comparison of electrical conductivity of hybrid Al₂O₃:SiO₂ nanofluids in different base fluids at 60 °C

Property enhancement of thermo-hydraulic Ratio ($PER_{t/v}$)

Property enhancement of thermo-hydraulic ratio ($PER_{t/v}$) as depicted in Figures 12(a) and 12(b) show the relationship of thermal conductivity and viscosity ratios for all the hybrid Al₂O₃:SiO₂ nanofluids measured. As for Figure 12(a), the lower value of the viscosity ratio at the higher value of the thermal conductivity value is preferred. Compared to other hybrid ratios, it was monitored that the 10:90 ratio had the most advantageous thermal conductivity ratio at all temperatures with a moderate impact on the dynamic viscosity ratio. Though it had a larger viscosity impact than the 10:90 ratio, the 30:70 ratio was thought to have some of the highest thermal conductivity ratios as compared to other nanofluids. The $PER_{t/v}$ curve shown in Figure 12(b) shows that 10:90 has the lowest curve at all temperatures which indicates that it has the lowest impact on additional viscosity value over the increase in thermal conductivity value. The lower $PER_{t/v}$ value is favorable since the effect of the additional pumping power will be minimized. The advantage of the 10:90 ratio is more obvious at temperature regions of 60 °C and 70 °C, which suited well with the PEMFC operating temperature. Thus, it can be concluded that the 10:90 ratio of hybrid Al₂O₃:SiO₂ nanofluids in water:EG base fluid is the most feasible ratio in terms of its thermal-hydraulic, $PER_{t/v}$ relationship. In terms of pumping power requirements, this ratio offered the greatest thermal conductivity advantage with the least amount of penalty.



(a)



(b)

Figure 12: (a) Thermal conductivity ratio effect towards dynamic viscosity ratio, and (b) $PER_{t/v}$ variation in hybrid nanofluids studied

Property enhancement of thermo-electrical ratio ($PER_{t/e}$)

To correlate the thermal conductivity of nanofluids to the electrical conductivity values, property enhancement of thermo-electrical conductivity ratio ($PER_{t/e}$) was established as shown in Figure 13. This correlation was adopted from Zakaria et al. [42] which has established the same correlation for single Al_2O_3 nanofluids in both water and water:EG base. It showed that as the content of Al_2O_3 was increased, the $PER_{t/e}$ value would reduce, which was

favourable to the adoption of nanofluids in PEMFC. A lower $PER_{t/e}$ value shows that minimal effect on the electrical conductivity value increment as the thermal conductivity value was increased. The mixture ratio of 70:30 shows the lowest $PER_{t/e}$ value due to its lowest content of SiO_2 which has a higher electrical conductivity value as compared to single Al_2O_3 nanofluids [27]. It was also observed that the $PER_{t/e}$ value reduced as the temperature was increased. This is beneficial for a PEMFC operation which is at the region of 60 °C to 80 °C.

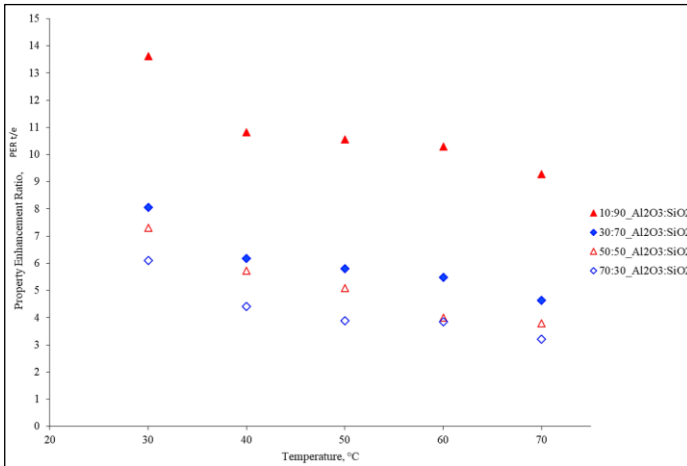


Figure 13: $PER_{t/e}$ of $Al_2O_3:SiO_2$ hybrid nanofluids

Conclusion

In this work, the thermophysical properties of hybrid $Al_2O_3:SiO_2$ nanofluids were examined concerning their possible use as an advanced cooling medium for PEMFC stacks. Thermo-physical properties of hybrid $Al_2O_3:SiO_2$ dispersed in water:EG were investigated under various temperatures in both thermal and electrical conductivity, and also their dynamic viscosity. The highest thermal conductivity improvement was monitored in a mixture ratio of 10:90 at 70 °C with a 9.8% improvement as compared to the base fluid of w:EG. Meanwhile, as one of the most critical properties for PEMFC which is electrical conductivity, the 70:30 ratio has shown the smallest increment of 63.3 $\mu S/cm$. Since the limit is 5 $\mu S/cm$, further research needs to be done on this such as applying coating to the cooling plate of PEMFC to reduce its conductive value. The smallest penalty on viscosity was shown by the 10:90 mixture ratio with 1.46 cP. To further investigate the feasibility of hybrid

$\text{Al}_2\text{O}_3:\text{SiO}_2$ in water:EG in PEMFC, the $\text{PER}_{t/v}$ and $\text{PER}_{t/e}$ were established. The findings concluded that the 10:90 mixture ratio of hybrid $\text{Al}_2\text{O}_3:\text{SiO}_2$ nanofluids can be investigated further as a cutting-edge PEMFC coolant.

Contributions of Authors

The authors confirmed the equal contribution in each part of this work. All authors reviewed and approved the final version of this work.

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Conflict of Interests

All authors declared that they have no conflicts of interest

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References

- [1] I. A. Zakaria, M. R. Mustaffa, W. A. N. W. Mohamed, and A. M. I. Mamat, "Steady - state potential energy recovery modeling of an open cathode PEM fuel cell vehicle", *Applied Mechanics and Materials*, vol. 465 - 466, pp. 114-119, 2014.
- [2] S. C. Mohapatra, "Fuel Cell and Fuel Cell Coolant Compositions," US Patent US 7,138,199 B2, 2006.
- [3] M. Saeedan, E. Afshari, and M. Ziaei-Rad, "Modeling and optimization of turbulent flow through PEM fuel cell cooling channels filled with metal foam- a comparison of water and air cooling systems", *Energy*

- Conversion and Management*, vol. 258, p. 115486, 2022. doi: <https://doi.org/10.1016/j.enconman.2022.115486>
- [4] I. A. Zakaria, W. A. N. W. Mohamed, N. H. A. Azid, M. A. Suhaimi, and W. H. Azmi, "Heat transfer and electrical discharge of hybrid nanofluid coolants in a fuel cell cooling channel application", *Applied Thermal Engineering*, vol. 210, p. 118369, 2022. doi: <https://doi.org/10.1016/j.applthermaleng.2022.118369>
- [5] M. N. I. Johari Mohamad Noor Izwan, I. A. Zakaria, and A. N. S. Mohammed, "Thermal behaviour of hybrid nanofluids in water: Bio glycol mixture in cooling plates of PEMFC", *CFD Letters*, vol. 14, no. 6, pp. 43-55, 06/26 2022. doi: 10.37934/cfdl.14.6.4355
- [6] J. Xu, C. Zhang, Z. Wan, X. Chen, S. H. Chan, and Z. Tu, "Progress and perspectives of integrated thermal management systems in PEM fuel cell vehicles: A review", *Renewable and Sustainable Energy Reviews*, vol. 155, p. 111908, 2022.
- [7] Y. Yu, M. Chen, S. Zaman, S. Xing, M. Wang, and H. Wang, "Thermal management system for liquid-cooling PEMFC stack: From primary configuration to system control strategy", *eTransportation*, vol. 12, p. 100165, 2022.
- [8] G. Liu, Y. Qin, and D. Ji, "Numerical investigation of organic fluid flow boiling for proton exchange membrane fuel cell cooling and waste heat recovery", *Applied Thermal Engineering*, p. 120564, 2023. doi: <https://doi.org/10.1016/j.applthermaleng.2023.120564>.
- [9] I. A. Zakaria, A. S. M. A. Azmin, S. Khalid, W. A. W. Hamzah, and W. A. N. W. Mohamed, "Numerical analysis of aluminium oxide and silicon dioxide nanofluids in serpentine cooling plate of PEMFC," *Journal of Advanced Research in Fluid Mechanics and Thermal Sciences*, vol. 72, no. 1, pp. 67-79, 2020.
- [10] A. I. Ramadhan, W. H. Azmi, R. Mamat, E. Diniardi, and T. Y. Hendrawati, "Experimental investigation of cooling performance in automotive radiator using Al₂O₃-TiO₂-SiO₂ nanofluids", *Automotive Experiences*, vol. 5, no. 1, pp. 28-39, 2022.
- [11] I. Muhammad Syafiq, Z. Irnie Azlin, and H. Wan Azmi Wan, "Heat transfer and pressure drop of water based hybrid Al₂O₃:SiO₂ nanofluids in cooling plate of PEMFC," *Journal of Advanced Research in Numerical Heat Transfer*, vol. 4, no. 1, pp. 1-13, 2021.
- [12] K. Saifuddin, Z. Irnie, W. H. Azmi & W. A. N. W. Mohamed "Thermal-electrical-hydraulic properties of Al₂O₃-SiO₂ hybrid nanofluids for advanced PEM fuel cell thermal management," *Journal of Thermal Analysis and Calorimetry*, vol. 143, pp. 1555-1567, 2020.
- [13] I. A. Zakaria, W. A. N. W. Mohamed, A. M. I. Mamat, K. I. Sainan, and S. F. A. Talib, "Thermal performance of Al₂O₃ in water - ethylene glycol nanofluid mixture as cooling medium in mini channel," *AIP Conference*

- Proceedings*, vol. 1674, no. 1, pp. 1-3, 2015. doi: <http://dx.doi.org/10.1063/1.4928831>
- [14] S. N. M. Zainon and W. H. Azmi, "Stability and thermo-physical properties of green bio-glycol based TiO₂-SiO₂ nanofluids", *International Communications in Heat and Mass Transfer*, vol. 126, pp. 1-13, 2021. doi: <https://doi.org/10.1016/j.icheatmasstransfer.2021.105402>
- [15] H. Li, L. Wang, Y. He, Y. Hu, J. Zhu, and B. Jiang, "Experimental investigation of thermal conductivity and viscosity of ethylene glycol based ZnO nanofluids", *Applied Thermal Engineering*, vol. 88, pp. 363-368, 2015.
- [16] A. I. Ramadhan, W. H. Azmi, and R. Mamat, "Experimental investigation of thermo-physical properties of tri-hybrid nanoparticles in water-ethylene glycol mixture," *Walailak Journal of Science and Technology*, vol. 18, no. 8, pp. 1-15, 2021. doi: 10.48048/wjst.2021.9335
- [17] S. Khalid, I. Zakaria, W. H. Azmi, and W. A. N. W. Mohamed, "Thermal-electrical-hydraulic properties of Al₂O₃-SiO₂ hybrid nanofluids for advanced PEM fuel cell thermal management," *Journal of Thermal Analysis and Calorimetry*, vol. 143, no. 2, pp. 1555-1567, 2021. doi: 10.1007/s10973-020-09695-8
- [18] K. Hamid, W. Azmi, M. Nabil, and R. Mamat, "Improved thermal conductivity of TiO₂-SiO₂ hybrid nanofluid in ethylene glycol and water mixture," in *IOP Conference Series: Materials Science and Engineering*, vol. 257, no. 1, pp. 1-8, 2017.
- [19] P. McMullen, S. Mohapatra, and E. Donovan, "Advances in PEM Fuel Cell Nano-Coolant," *Fuel Cell Seminar & Energy Exposition*, Curran Associates Inc Columbus, Ohio, USA, 2013.
- [20] I. A. Zakaria, Z. *et al.*, "A review of nanofluid adoption in polymer electrolyte membrane (PEM) fuel cells as an alternative coolant," *Journal of Mechanical Engineering and Sciences*, vol. 8, no. 1, pp. 1351-66, 2015.
- [21] I. Zakaria, W. A. N. W. Mohamed, W. H. Azmi, A. M. I. Mamat, R. Mamat, and W. R. W. Daud, "Thermo-electrical performance of PEM fuel cell using Al₂O₃ nanofluids," *International Journal of Heat and Mass Transfer*, vol. 119, no. 13, pp. 460-471, 2018. doi: <https://doi.org/10.1016/j.ijheatmasstransfer.2017.11.137>
- [22] R. Islam, B. Shabani, J. Andrews, and G. Rosengarten, "Experimental investigation of using ZnO nanofluids as coolants in a PEM fuel cell," *International Journal of Hydrogen Energy*, vol. 42, no. 30, pp. 19272-19286, 2017. doi: <https://doi.org/10.1016/j.ijhydene.2017.06.087>
- [23] M. R. Islam, B. Shabani, G. Rosengarten, and J. Andrews, "The potential of using nanofluids in PEM fuel cell cooling systems: A review", *Renewable and Sustainable Energy Reviews*, vol. 48, pp. 523-539, 2015. doi: <http://dx.doi.org/10.1016/j.rser.2015.04.018>

- [24] M. R. Islam, B. Shabani, and G. Rosengarten, "Nanofluids to improve the performance of PEM fuel cell cooling systems: A theoretical approach", *Applied Energy*, vol. 178, pp. 660-671, 2016. doi: <http://dx.doi.org/10.1016/j.apenergy.2016.06.090>
- [25] M. A. N. Zarizi, I. A. Zakaria, M. N. I. Johari, W. A. N. Wan Mohamed, and R. M. R. Ahsan Shah, "Thermo-electrical behavior of Al₂O₃ and SiO₂ nanofluids in a Proton-Exchange Membrane Fuel Cell (PEMFC) Cooling Channel," *Pertanika Journal of Science & Technology*, vol. 30, no. 2, 2022.
- [26] S. Aldreich, Material Safety Data Sheet, 2019.
- [27] S. F. Talib, W. H. Azmi, I. Zakaria, W. A. N. W. Mohamed, A. M. I. Mamat, H. Ismail, and W. R. W. Daud, "Thermophysical properties of silicon dioxide (SiO₂) in ethylene glycol/water mixture for proton exchange membrane fuel cell cooling application", *Energy Procedia*, vol. 79, pp. 366-371, 2015.
- [28] S. Khalid, I. A. Zakaria, and W. A. N. W. Mohamed, "Comparative analysis of thermophysical properties of Al₂O₃ and SiO₂ nanofluids," *Journal of Mechanical Engineering*, vol. 8, no. 1, pp. 153-163, 2019.
- [29] 2009 ASHRAE® Handbook Fundamentals, ASHRAE, 2009.
- [30] M. F. Ismail, W. H. Azmi, R. Mamat, K. V. Sharma, and N. N. M. Zawawi, "Stability assessment of polyvinyl-ether-based TiO₂, SiO₂, and their hybrid nanolubricants," *Lubricants*, vol. 11, no. 1, pp. 1-17, 2023.
- [31] M. F. Nabil, W. H. Azmi, K. Abdul Hamid, R. Mamat, and F. Y. Hagos, "An experimental study on the thermal conductivity and dynamic viscosity of TiO₂-SiO₂ nanofluids in water: Ethylene glycol mixture", *International Communications in Heat and Mass Transfer*, vol. 86, pp. 181-189, 2017. doi: <https://doi.org/10.1016/j.icheatmasstransfer.2017.05.024>
- [32] A. Gallego Montoya, K. Cacua, B. Herrera, D. Cabaleiro, M. Piñeiro, and L. Lugo, "Experimental evaluation of the effect in the stability and thermophysical properties of water-Al₂O₃ based nanofluids using SDBS as dispersant agent", *Advanced Powder Technology*, vol. 31, no. 2, pp. 560-570, 2019. doi: [10.1016/j.apt.2019.11.012](https://doi.org/10.1016/j.apt.2019.11.012)
- [33] K. Cacua, F. Ordoñez, C. Zapata, B. Herrera, E. Pabón, and R. Buitrago-Sierra, "Surfactant concentration and pH effects on the zeta potential values of alumina nanofluids to inspect stability", *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, vol. 583, pp. 1-6, 2019. doi: <https://doi.org/10.1016/j.colsurfa.2019.123960>
- [34] M. N. I. Johari, I. A. Zakaria, W. H. Azmi, and W. A. N. W. Mohamed, "Green bio glycol Al₂O₃-SiO₂ hybrid nanofluids for PEMFC: The thermal-electrical-hydraulic perspectives", *International Communications in Heat and Mass Transfer*, vol. 131, p. 105870, 2022. doi: <https://doi.org/10.1016/j.icheatmasstransfer.2021.105870>

- [35] S. N. M. Zainon and W. H. Azmi, "Heat transfer performance of green bioglycol-based TiO₂-SiO₂ nanofluids," *Journal of Heat Transfer*, vol. 143, no. 11, 2021. doi: 10.1115/1.4051763
- [36] X. Mei, X. Sha, D. Jing, and L. Ma, "Thermal conductivity and rheology of graphene oxide nanofluids and a modified predication model", *Applied Sciences*, vol. 12, no. 7, pp. 1-10, 2022.
- [37] FCgen®-1310 Fuel Cell Stack, B. P. S. Inc., Canada, 2012.
- [38] T. G. Beckwith, R. D. Marangoni, and J. H. Lienhard, *Mechanical Measurements*, 6th Edition, Pearson Prentice, 2007.
- [39] S. Lee, J. Lee, and J. T. Ault, "The role of variable zeta potential on diffusiophoretic and diffusioosmotic transport", *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, vol. 659, p. 130775, 2023. doi: <https://doi.org/10.1016/j.colsurfa.2022.130775>
- [40] F. Mohd Amiruddin, F. Wan Mohd, A. Hasyiya Karimah, M. Rizalman, A. Wan Hamzah, and R. Anwar Ilmar, "Experimental study on thermo-physical properties of TiO₂-SiO₂ nanofluids (70:30) in water/ethylene glycol mixture," *Journal of Advanced Research in Fluid Mechanics and Thermal Sciences*, vol. 108, no. 2, pp. 200-214, 2023. doi: 10.37934/arfmnts.108.2.200214
- [41] L. S. Sundar and F. Shaik, "Heat transfer and exergy efficiency analysis of 60% water and 40% ethylene glycol mixture diamond nanofluids flow through a shell and helical coil heat exchanger," *International Journal of Thermal Sciences*, vol. 184, p. 107901, 2023.
- [42] I. Zakaria, W. H. Azmi, W. A. N. W. Mohamed, R. Mamat, and G. Najafi, " Experimental investigation of thermal conductivity and electrical conductivity of Al₂O₃ nanofluid in water - ethylene glycol mixture for proton exchange membrane fuel cell application", *International Communications in Heat and Mass Transfer*, vol. 61, pp. 61-68, 2015. doi: <https://doi.org/10.1016/j.icheatmasstransfer.2014.12.015>