

Thermo-Electrical Behavior of Al₂O₃ and SiO₂ Nanofluids in a Proton-Exchange Membrane Fuel Cell (PEMFC) Cooling Channel

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ABSTRACT

Proton Exchange Membrane Fuel Cell (PEMFC) generates electricity through the reaction of hydrogen and oxygen. PEMFC is considered clean technology since the by-products of the reaction are only electricity, water, and heat. Thermal management of PEMFC can be further improved through the adoption of nanofluids as its cooling medium. Nanofluids are fluids that contain suspensions of nanoparticles in their base fluid. Nanofluids have better heat transfer performance as compared to their base fluid due to their significant thermal conductivity improvement. However, unlike any other heat transfer application, there is a strict limit on the electrical conductivity of the nanofluids due to their electrically active environment. Therefore, there is a possible current leakage to the coolant due to the nanofluids' conductive behavior. In this study, heat transfer performance and current drop of 0.5% Al₂O₃ and 0.5% SiO₂ water were investigated. The nanofluids were forced to flow in a charged channel subjected to a heater pad of 60°C to 70°C to mimic the operating condition of a PEMFC. The performance of each nanofluid was observed and compared to distilled water. The channel temperature was reduced by 43.3% and 42.7% by Al₂O₃ and SiO₂ nanofluids, respectively, compared to base fluids at Re 700. In terms

of current drop, SiO₂ nanofluids have the highest current drop with 2.33% from the initial current value. It was further justified with the increment in electrical conductivity value of the fluids after the experiment, thus justifying the current leakage hypothesis.

Keywords: Current drop, heat transfer, nanofluids, PEM fuel cell

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INTRODUCTION

Fuel cells are electrochemical devices that generate electricity. The operation of the fuel cell consists of the reaction between hydrogen and oxygen. First, water is electrolyzed into hydrogen and oxygen by flowing an electric current. Next, two electrodes are used for the electrolysis and are immersed in an electrolyte. Once the power source is removed, the hydrogen and oxygen recombine; thus, electric current, heat, and water are produced (Larminie & Dicks, 2013).

PEMFC has many advantages, such as low operating temperature, which is within the range of 30°C to 100°C (Larminie & Dicks, 2013). It provides a quick start-up advantage to automotive applications. Furthermore, developments in recent years have allowed PEMFC to generate more power (Larminie & Dicks, 2013). The efficiency of PEMFCs is in the range of 40 to 50%, while the internal combustion engine (ICE) has an average efficiency of 20%. Moreover, the greenhouse gas emission is almost zero or very low while in operation (Islam, 2016). These traits make the PEMFC a suitable power source for vehicles. The optimum operating temperature for PEMFC is 60 to 80°C. The small temperature difference will limit the heat transfer rate (Islam, 2016). A higher power fuel cell requires a large size of radiator. Currently, cooling for PEMFCs consists of separate reactant and cooling air, using cathode air supply and water cooling (Larminie & Dicks, 2013). Alternatively, the heat transfer of PEMFC can be passively improved with the adoption of nanofluids as its alternative cooling medium or coolant.

Nanofluids are defined as the base fluid that contains suspensions of nano-sized particles (Jama et al., 2016). The nanoparticles used in nanofluids are generally metal, metal oxides, carbon nanotubes, and carbides. Aluminum Oxide (Al_2O_3) and Silicon Dioxide (SiO_2) are among the popular metal oxide nanofluids due to their enhancement in thermo-physical properties (Khalid et al., 2019). Common base fluids used are distilled water, ethylene—glycol, a mixture of water and ethylene—glycol, and engine oil (Zakaria et al., 2018). Previous studies showed an enhancement in heat transfer coefficient when nanoparticles are introduced in base fluids. For example, according to Xuan & Li, 2003, the heat transfer properties were increased as the concentration of nanoparticles is increased. Similarly, the enhancement of heat transfer properties was echoed by Xuan and Li (2003) and Sahin et al. (2015). The enhancement of heat transfer properties is due to the increase of thermal conductivity, thus increasing the convection coefficients (Islam, 2016; Sahin et al., 2015; Xuan & Li, 2003). Muhammad et al. (2019) conducted a study on the effects of water— Al_2O_3 , water— SiO_2 , and water—Cu nanofluids heat transfer and pressure drop in a mini channel heat sink. The authors concluded that heat transfer enhancement is prominent in water— Al_2O_3 followed by water— SiO_2 and water—Cu, respectively (Muhammad et al., 2019). In addition, the author notes that the high thermal conductivity compared to the water, together with the effects of Brownian diffusion, played a part in the heat transfer performance (Muhammad et al., 2019). In another study conducted by Muhammad and

Sidik (2018), the authors stated that a high surface-to-volume ratio in nanofluids had improved heat transfer performance compared to the base fluid.

Studies on the adoption of nanofluids as an alternative coolant in PEMFC were investigated by Zakaria et al. (2015b), who experimented with Al₂O₃ nanofluids' thermal behavior in a single cooling plate of PEMFC. The authors stated an enhancement in heat transfer coefficient and Nusselt number of nanofluids compared to the base fluid. Islam (2016) also reviewed nanofluids for PEMFC cooling in automotive applications. The author stated that nanofluids adoption could reduce the size of the radiator of vehicles running on PEMFC due to its improvement in heat transfer behavior.

However, unlike any other heat transfer application, coolant in PEMFC requires a strict limit of electrical conductivity property. The permissible electrical conductivity of the current cooling fluid for PEMFC is very minimum, which is at 5 μ s/cm at 20°C (Ballard, 2010). This low value of electrical conductivity requirement is important to avoid current during the PEMFC operation (Barbir, 2005). There is also a possibility of a performance drop in a PEMFC due to the current leakage to the conductive coolant. The possible current drop may occur from the generated current from the reaction of PEMFC to the nanofluids coolant flowing in the cooling channel, which is highly conductive (Zakaria et al., 2016). Chereches and Minea (2019) reported that the nanofluids experience an increase in their electrical conductivity compared to the base fluid. The electrical conductivity is also reported to increase linearly with the increment in volume concentration (Chereches & Minea, 2019).

The effects of the current drop in a PEMFC cooling channel are not widely explored. This novel experimental study has addressed the effect of the conductive cooling medium of nanofluids on the electrically charged channel to mimic the PEMFC channel. It is a fundamental study performed on a heated channel with a similar working temperature of PEMFC, which is in the range of 60°C to 70°C (Barbir, 2005; Islam, 2016). A heater pad was placed directly underneath the cooling channel to provide the supplied heat. The cooling channel and piping were insulated with EPDM (ethylene propylene diene monomer) foam to minimize heat loss to the surroundings. The nanofluids used was water as it is the most used base fluid in PEMFC. The comparison made was against the Al₂O₃ and SiO₂ nanofluids at the volume concentration of 0.5%. This experimental study has observed the relationship between the nanofluids' heat transfer improvement and the current drop effect experienced.

METHODOLOGY

Preparation of Nanofluids

The nanofluids were prepared using the two-step method. The Al₂O₃ nanoparticle comes in powder form, obtained from Sigma-Aldrich with 99.8% purity and in the size of 13nm. The

SiO₂ nanoparticle was in liquid form with a 13.67% concentration and had a size of 30nm. Both nanoparticles were prepared independently. The properties of the nanoparticles and base fluid used are summarized in Table 1. The mass of Al₂O₃ particles needed for dilution in distilled water to form a 0.5% volume concentration was determined with Equation 1. The volume % concentration was used as practiced by other researchers (Zakaria et al. 2016; Chereches & Minea, 2019; Muhammad & Sidik, 2018).

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

Where Φ is the volume concentration, m is the mass, ρ is the density. Subscripts p and bf represent the nanoparticle and base fluid, respectively.

As for SiO₂ nanofluids, since the material received was in suspension form, the nanofluids were then diluted in distilled water. The dilution was measured using Equation 2 to obtain 0.5% volume concentration.

$$\Delta V = (V_2 - V_1) = V_1 \left(\frac{\Phi_1}{\Phi_2} - 1 \right) \quad (2)$$

Where ΔV is the volume of base fluid needed to be added into the current volume of base fluid, V_1 with a concentration of Φ_1 to obtain the needed volume of nanofluid, V_2 with volume concentration, Φ_2 . Once the preparation of nanofluids is completed, they are mixed with distilled water and then stirred with an electric stirrer. Each nanofluid requires a different mixing duration. For example, Al₂O₃ was stirred for 30 minutes while SiO₂ required 15 minutes (Usri et al., 2015). It was to ensure that the nanoparticles were dispersed evenly. The final mixing process was on the particle level in which it was sonicated in a sonicator bath for two hours (Usri et al., 2015; Zakaria et al., 2019). The prepared nanofluids are shown in Figure 1. The prepared nanofluids presentation was adopted based on a common practice by researchers in nanofluids (Zakaria et al., 2016; Chereches & Minea, 2019; Muhammad & Sidik, 2018).

Table 1
Properties of nanoparticles and distilled water

Property	Al ₂ O ₃	SiO ₂	Distilled Water
Density/ Kg m ⁻³	4000	2200	996
Thermal conductivity/ W m ⁻¹ K ⁻¹	36	1.4	0.615
Specific heat/ J Kg ⁻¹ K ⁻¹	765	745	4178

Source: <https://www.innovacera.com/materials/99-9-alumina>; <https://www.makeitfrom.com/material-properties/99-Percent-Purity-Alumina-ASTM-D2442-Type-IV-IEC-60672-Type-C-799>; <https://pubchem.ncbi.nlm.nih.gov/compound/Silicon-dioxide#section=Density>

The dispersion of nanoparticles in the base fluid was then analyzed through Transmission electron microscopy (TEM), as shown in Figure 2. The TEM images for Al₂O₃ and SiO₂ nanoparticles confirmed the presence of Al₂O₃ and SiO₂ nanoparticles with particle sizes of 13 nm and 30 nm, respectively, in the suspension prepared.

Measurement of Electrical Conductivity

The introduction of nanoparticles to a base fluid has increased its electrical conductivity property. Zakaria et al. (2015a) has conducted measurements on the electrical conductivity of Al₂O₃ nanofluids and discovered that the increase of electrical conductivity was linear to the temperature and volume concentration of nanofluids. Therefore, the electrical conductivity measurement was conducted to investigate the relationship of the electrical conductivity property of nanofluids to the current drop effect in this study.

In this study, the electrical conductivity was measured at the temperature of 30°C to 80°C to cover the operating temperature range of PEMFC. The nanofluids were placed in a water bath to provide the required temperature. The measurement was taken using Cyberscan PC-10, equipped with the ATC (automatic temperature compensation) as shown in Figure 3. The Cyberscan PC-10 was also used by Abdolbaqi et al. (2016) for the same purpose.

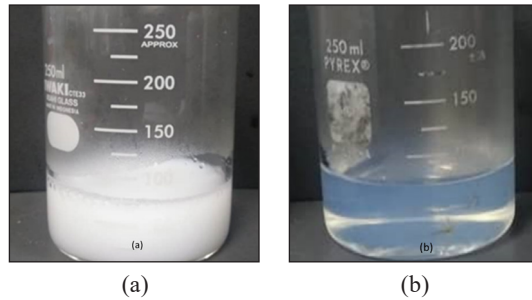


Figure 1. (a) 0.5% Al₂O₃ in distilled water; (b) 0.5% SiO₂ in distilled water

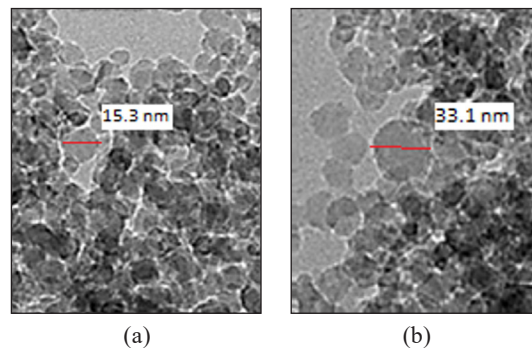


Figure 2. TEM images of: (a) Al₂O₃ (15.3nm); (b) and SiO₂ (33.1nm)

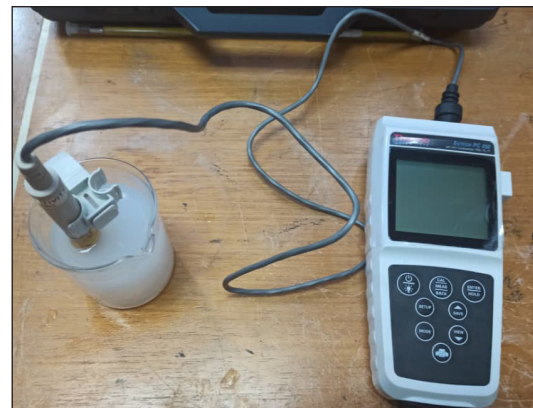


Figure 3. Measuring electrical conductivity using Cyberscan PC – 10

Experimental Set-Up

A test bench was developed to mimic the operating condition of a single channel in a PEMFC's cooling plate. A cooling channel in the test section was heated to 60°C and 70°C to simulate the optimum operating temperature of PEMFC. The channel was also charged with 3A at a voltage of 0.7 V to simulate the charged condition of cooling plates during the PEMFC operation. Nanofluids were then circulated through the system by a water pump. The required flow rate to achieve the specified Reynolds number, which was in the range of Re 300 to 700, was confirmed with the DigiFlow flow meter. The cooling channel was also heated with a silicone heater pad placed underneath the channel to the temperatures of 60°C and 70°C during the experiment. Both channel surface and fluid temperatures were measured with K – Type thermocouples then recorded to Graphtec Midi Logger. The current drop was observed and measured using MultiCom Pro digital multimeter. When nanofluids were circulated through the system, the supplied current experienced a drop. To further justify the current drop experienced, the electrical conductivity of the nanofluids was measured before and after the experiment. The experimental set-up and schematic diagram are shown in Figures 4 and 5, respectively.

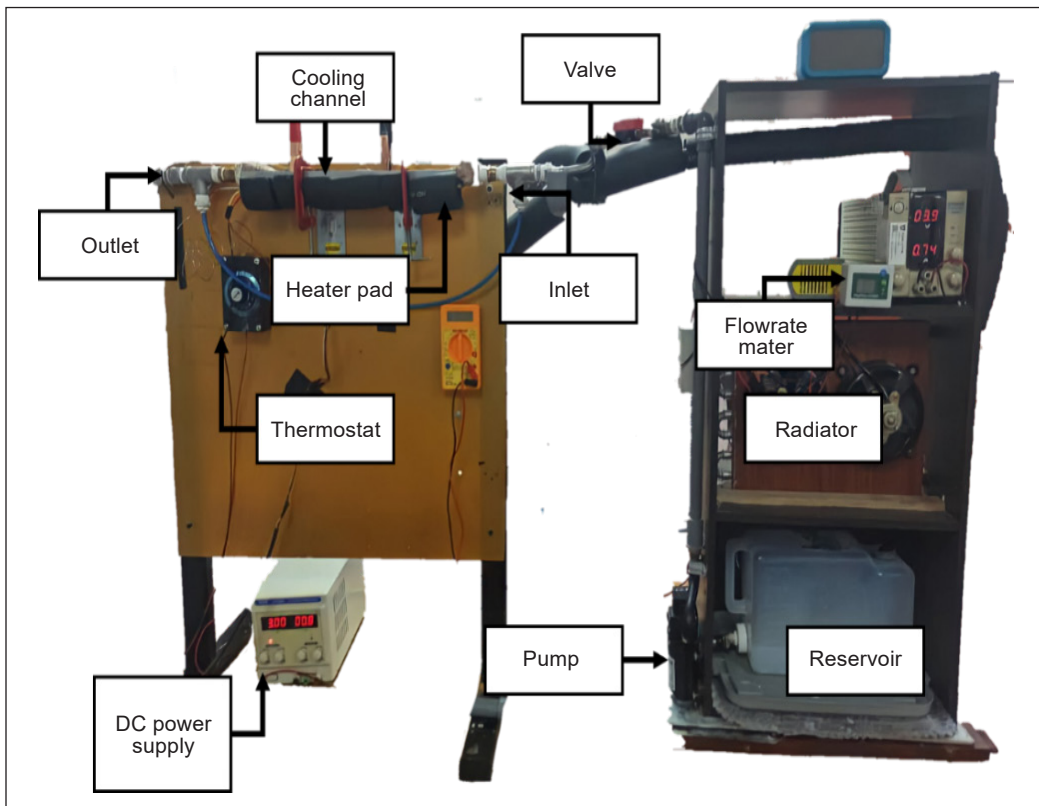


Figure 4. Experimental set-up

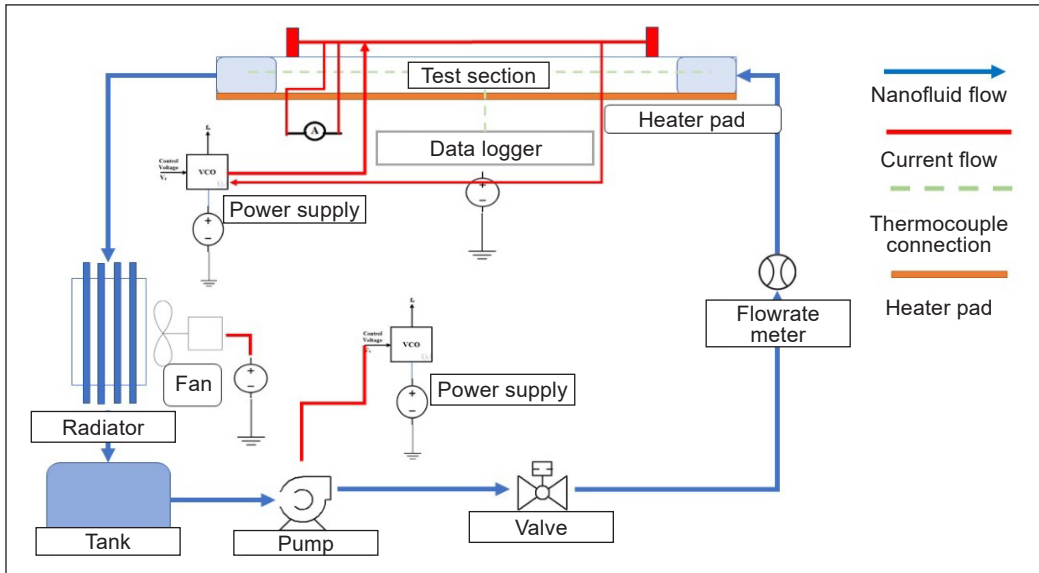


Figure 5. Schematic diagram of the experimental set-up

Uncertainty Analysis

The accuracy of the experimental set-up was validated by performing an uncertainty analysis as practiced by Taner (2018) and Coleman and Steele (1995). In this analysis, uncertainties of instruments were considered to determine the total resistance measurement of the device. The uncertainties of the experimental instrumentation are tabulated in Table 2. In summary, as shown in Table 3, the calculated maximum uncertainty was less than 1.5% which is acceptable (Beckwith et al., 2007).

Table 2
Experimental parameters and uncertainties

No	Instrument	Range of Instrument	Variable measured	Least Division in Measuring Instrument	Values measured in experiment			
					Min	Max	Max	Min
1	Thermocouple	0–300°C	Bulk temperature, T_b	$U_T = 0.1^\circ\text{C}$	33.5	35.7	0.422	0.396
2	Thermocouple	0–300°C	Average surface temperature, T_w	$U_{\Delta T_{coolant}} = \sqrt{0.1^2 + 0.1^2}$	34.0	40.1	0.658	0.558
3	Flowmeter	0.8–6 lpm	Volume Flow rate, \dot{V}	$U_T = 0.1^\circ\text{C}$ $U_{T_w} = \sqrt{5 \times (0.1^2)}$	0.23	0.36	1.786	0.634
4	Current	0–20 A	Current Drop, ΔI	0.01	2.93	2.98	0.341	0.338
5	Properties	Electrical conductivity					0.1	0.1

Table 3
Summary of uncertainty analysis

No	Variables	Uncertainty (%)
1	Channel temperature, T_{channel}	1.471 - 1.247
2	Coolant temperature drop, $\Delta T_{\text{coolant}}$	0.597 - 0.560
3	Current drop, ΔI	0.483 - 0.478

Mathematical Model

The detailed dimensions of the studied channel are shown in Figure 6.

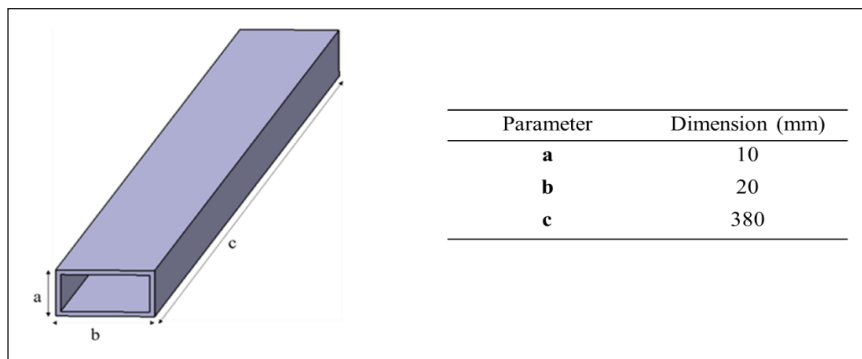


Figure 6. Geometry of cooling channel studied

The hydraulic diameter, D_h is defined as Equation 3 (Cengel & Cimbala, 2006)

$$D_h = \frac{4ab}{2(a + b)} \quad (3)$$

The Reynolds number is expressed as Equation 4:

$$Re = \frac{D_h v_m \rho}{\mu} \quad (4)$$

The rate of heat transfer, Q , W was then calculated using Equation 5 (Aghayari et al., 2014; Cengel & Afshin, 2020):

$$Q = \dot{m} C_{pnf} (T_{out} - T_{in}) \quad (5)$$

The convective heat transfer coefficient, $W K^{-1} m^{-2}$, was obtained using Equation 6 (Aghayari et al., 2014; Cengel & Afshin, 2020)

$$h = \frac{Q}{A_s (T_{avg} - T_{\infty})} \quad (6)$$

where T_{avg} was calculated using Equation 7 (Aghayari et al., 2014; Cengel & Afshin, 2020)

$$T_{avg} = \sum \frac{T_{surface}}{5} \quad (7)$$

The total surface temperature is divided by five because there are five points measured on the surface of the channel.

The T_{∞} was calculated using Equation 8 (Aghayari et al., 2014; Cengel & Afshin, 2020)

$$T_{\infty} = \frac{T_{in} + T_{out}}{2} \quad (8)$$

The surface area, A_s , was calculated using Equation 9

$$A_s = b * c \quad (9)$$

The Nusselt Number is determined by using Equation 10 (Aghayari et al., 2014; Cengel & Afshin, 2020; Pourfayaz et al., 2018)

$$u = \frac{h}{k} D_h \quad (10)$$

RESULT AND DISCUSSION

Electrical Conductivity

Electrical conductivity is a crucial thermo-physical property, which relates to the current leakage in an electrically charged channel. The electrical conductivity property of nanofluids with respect to temperature was measured and shown in Figure 7. It was observed that the electrical conductivity was increased as the temperature is increased. It was due to the higher energy supplied by the heat, which increased the energy of electrons to carry more

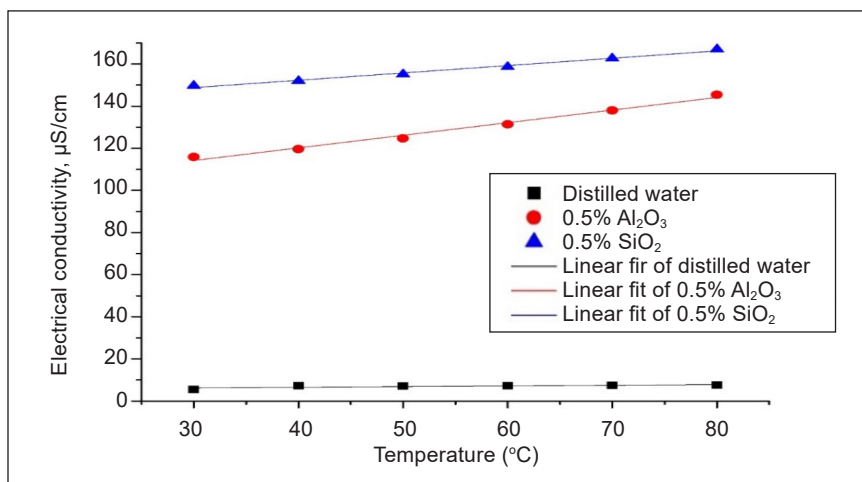


Figure 7. Temperature effect to the electrical conductivity, $\mu\text{S}/\text{cm}$

electrical current (Hermans et al., 2014). The electrical conductivity for distilled water was the lowest compared to the nanofluids. It shows that nanoparticles in distilled water increase the electrical conductivity of the fluid due to the conductivity factor of the nanoparticles. The increment of electrical conductivity for both nanofluids shown was ten times higher than the base fluid. These findings are in good agreement with the findings of Chereches and Minea (2019). Furthermore, it was observed that the electrical conductivity of SiO₂ nanofluids was higher than the Al₂O₃ nanofluids by 21.9%.

Heat Transfer Characteristic

The channel temperature profile was recorded at two heater pad temperatures of 60°C and 70°C, as depicted in Figure 8. It was observed that there was a significant difference in channel temperature when compared between distilled water and nanofluids. The Al₂O₃ nanofluids had the lowest channel temperature, followed by the SiO₂ nanofluids and the base fluid, which was distilled water. The temperature reductions made by Al₂O₃ and SiO₂ nanofluids were 43.3% and 42.7%, respectively, compared to base fluids at Re 700 and heater temperature of 60°C. It was due to the presence of nanoparticles that increased the thermal conductivity of the base fluid. The values of thermal conductivity for 0.5% Al₂O₃ and 0.5% SiO₂ were 0.7 W/m.K and 0.67 W/m.K, respectively (Khalid et al., 2020). The channel temperature was also reduced as the flow rate of the coolant was increased.

Meanwhile, the channel temperature reduction was also observed to be slightly higher in 70°C as compared to 60°C heated channel, as shown in Figure 8(b). The lowest channel temperature was shown by Al₂O₃ nanofluids with 50.9% lower as compared to base fluid at Re 700. It was followed by SiO₂ nanofluid with a 50% reduction. The increase in channel temperature has eventually increased the internal energy of nanoparticles, which forced a stronger vibration at a faster speed that leads to an increase in interaction between the particles.

The channel temperature reduction was correlated to the comparative value of thermal conductivity of the nanofluids. The pattern was in good agreement with Khalid et al. (2020), who reported on the thermal conductivity of both nanofluids when dispersed in distilled water. The highest thermal conductivity was given by Al₂O₃ nanofluids, followed by SiO₂ nanofluids, and finally distilled water. The improvement of thermal conductivity was due to the enhanced Brownian motion of nanoparticles in base fluid (Zakaria et al., 2015a).

The heat transfer enhancement for both Al₂O₃ and SiO₂ nanofluids against the base fluid was then analyzed through the convective heat transfer coefficient, as illustrated in Figure 9. It was observed that heat transfer for both nanofluids was significantly higher than distilled water. In Figure 9(a), the highest recorded convective heat transfer coefficient enhancement was shown by Al₂O₃ nanofluids at Re 700, five times higher than distilled water. It was then followed by SiO₂ nanofluids with almost twice as higher as the distilled

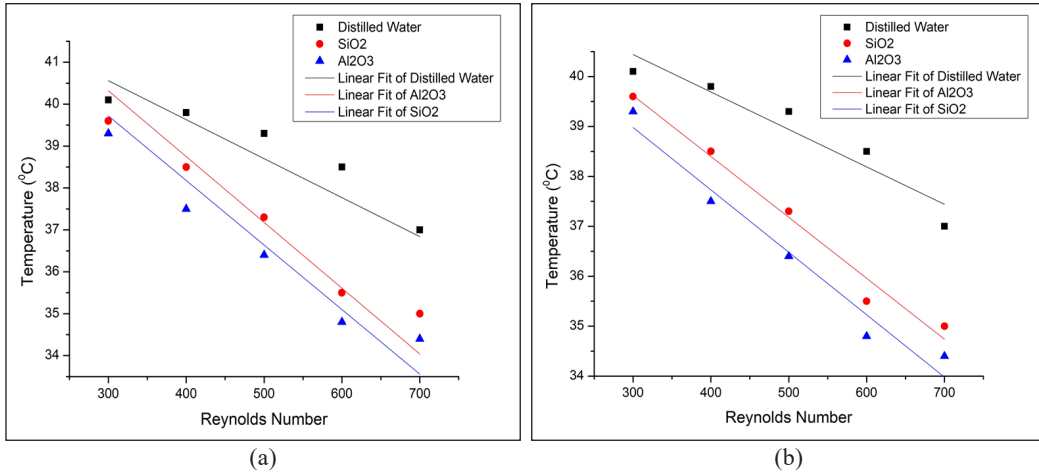


Figure 8. Channel temperature profile against Reynolds number at (a) heater pad temperature of 60°C (b) heater pad temperature of 70°C

water recorded at the same Reynolds number. Meanwhile, in a hotter channel of 70°C, as in Figure 9(b), the enhancement obtained by Al₂O₃ and SiO₂ nanofluids was further increased to five times and 1.5 times higher as compared to distilled water, respectively. It was due to the increment in their thermal conductivity as the temperature was increased. It is in good agreement with the established trending of thermal conductivity of Al₂O₃ and SiO₂ nanofluids (Khalid et al., 2020). Higher temperature is associated with a higher energy level for better heat dissipation in the cooling fluids.

The enhancement of the convective heat transfer was then converted to a non-dimensional value, as shown in Figure 10. It showed that the highest Nusselt number was obtained by Al₂O₃ nanofluids at Re 700 with 4.5 times higher as compared to distilled

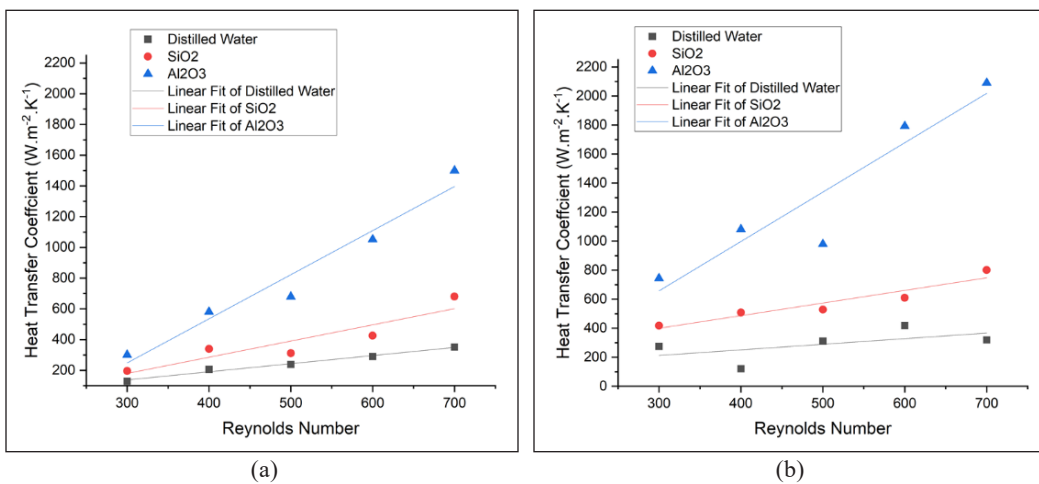


Figure 9. Heat transfer coefficient, W/ K. m² against Reynolds number at (a) heater pad 60°C (b) heater pad 70°C

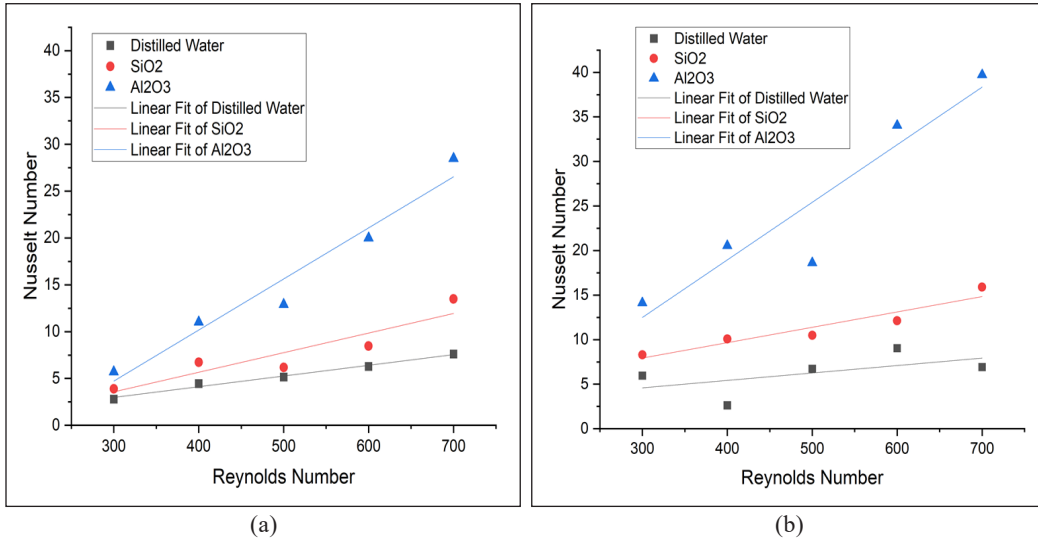


Figure 10. Nusselt number effect at different Reynolds number at (a) heater pad 60°C (b) heater pad 70°C

water. The SiO₂ nanofluids also provided an enhancement of nearly twice higher at the same Reynolds number. The effect of a higher channel temperature of 70°C was further improved in Figure 10(b), as the highest enhancement was shown by Al₂O₃ nanofluid at Re 700 with four times higher than distilled water and 1.3 times improved through SiO₂ nanofluids at the same Reynolds number. The significance of the Nusselt number is to investigate the comparative value of the convection heat transfer over the conductive heat transfer of the cooling fluid that flows through the channel. These findings were in good agreement with the findings of Zakaria et al. (2015a), Aghayari et al. (2014), and Asirvatham et al. (2009), which reported an enhancement in convective heat transfer coefficient and Nusselt number for all nanofluids studied.

Electrical Characteristics

There was a current drop observed from the nominal value supply of 3A, as shown in Figure 11. The drop was driven by the attraction of free electrons to travel to conductive coolants such as nanofluids compared to base fluids. The drop was quite small and measured in the range of 0.33% to 2.33%. It was observed that the more conductive the fluid, the higher the current drop. As presented earlier, the highest electrical conductivity property of the cooling fluids studied was shown by SiO₂ nanofluids followed by Al₂O₃ nanofluids. The least conductive fluid was the base fluid of distilled water. The current drop values were in good agreement with the electrical conductivity properties, with the highest drop shown by SiO₂ nanofluids with a 2.3% drop from the initial value. Al₂O₃ nanofluids followed with a 1% drop in the current value. As expected, the least drop was shown by the base fluid of distilled water.

To further justify the findings on the current leakage to the cooling fluid, the electrical conductivity values of each fluid were measured before and after the experiment. It was analyzed to prove that the current was flowing to the cooling fluid of the charged channel. In Figure 12, it was shown that the electrical conductivity value of SiO₂ nanofluids increased the most compared to other fluids. It was then followed by Al₂O₃ nanofluids and, lastly, the base fluid. The increment of electrical conductivity values for SiO₂ nanofluids, Al₂O₃ nanofluids, and distilled water was recorded at 128.4, 27.0, and 32.5%, respectively. It shows that fluids with higher electrical conductivity properties will enable larger current drop through the cooling channel in an electrically active environment. The current drop also showed a decreasing trend as the Re was increased. However, there was not much difference detected between 60°C and 70°C.

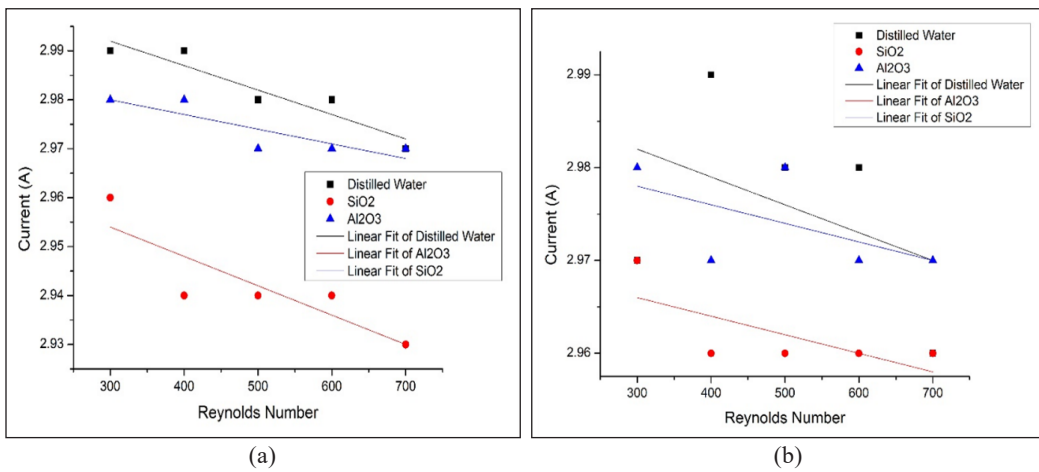


Figure 11. Current drop against Reynolds number at (a) heater pad 60°C (b) heater pad 70°C

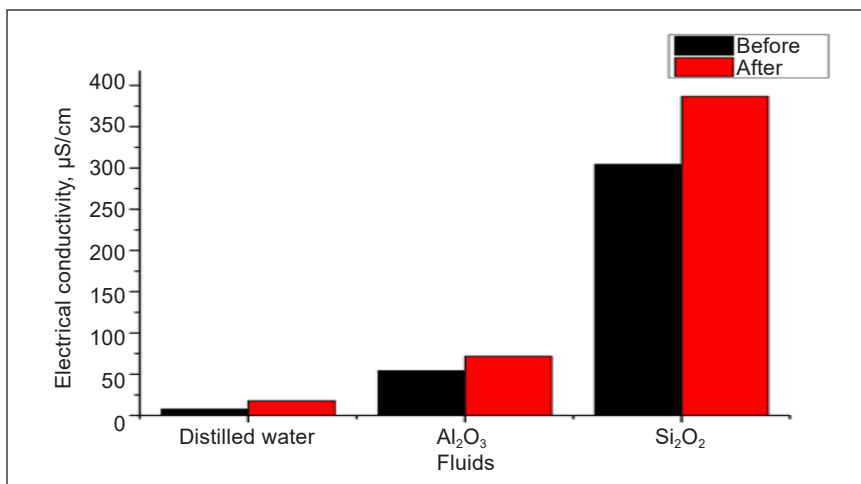


Figure 12. Electrical conductivity measurement before and after experiment

CONCLUSION

In general, nanofluids have improved the heat transfer performance in the channel. The highest heat transfer improvement was shown by Al_2O_3 nanofluids, followed by SiO_2 nanofluids, and finally, base fluid of distilled water. It was due to the significant improvement in terms of the thermal conductivity properties compared to the base fluid. The other factors affecting the heat transfer enhancement were the flow rate and the temperature of the heater pad. As both parameters were increased, the heat transfer increased as well.

The electrical characteristic, on the other hand, showed the largest current drop in the highest conductive fluids of SiO_2 nanofluids, followed by the Al_2O_3 nanofluids and finally the base fluid. The pattern of current leakage matched the electrical conductivity properties measured. It was then justified with the increment of electrical conductivity value of the fluids after the experiment. It denotes that the current leakage from the channel was due to the conductive coolant used in the charged channel. This charged channel serves as a fundamental study on PEMFC cooling plates. However, further studies at the stack level must be conducted to obtain a more accurate result.

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