Influence of Al\textsubscript{2}O\textsubscript{3}-Water Nanofluid Coolant on Thermal Performance of Hydrogen PEM Fuel Cell Stacks

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Abstract. Hydrogen polymer electrolyte membrane (PEM) fuel cells are renewable and future sustainable power generation systems that operate with zero emissions. Thermal management in fuel cells is still an important issue that must be addressed to extend their life and the possibility of their widespread commercial use. In this research, a simulation model has been developed and validated using MATLAB to investigate the cooling process and its effect on the PEM fuel cells using nanofluids as coolant. Al\textsubscript{2}O\textsubscript{3} nanoparticles have been used due to their thermal characteristics, chemical stability, and cost-effectiveness compared to other nanoparticles. The Al\textsubscript{2}O\textsubscript{3}-H\textsubscript{2}O nanofluid was used with 0.1 to 0.4 vol % concentration. The results showed that employing nanofluids as coolant increases the temperature difference between the coolant outlet and inlet. Nanoparticles concentration increases, the temperature difference increases. Similarly, the nanoparticles concentration increases, both pressure drop across the cooling channel and pumping power increases. Therefore, employing nanofluids in PEM fuel cells seems an impractical choice for the lower temperature difference criterion at a certain pumping power.

Keywords. PEM fuel cell, Nanofluid coolant, Simulation model, Thermal management, Al\textsubscript{2}O\textsubscript{3}.

1. Introduction

Great efforts have been made seeking alternative, sustainable, and clean power generators to overcome critical issues facing the global community, namely the depletion of fossil fuel and pollution of the environment [1]. As a clean power generator, the hydrogen polymer electrolyte membrane fuel cell (H\textsubscript{2}-PEMFC) technology has emerged, attracting tremendous interest from both the scientific community and industry due to the inherent advantages, namely: high volumetric power density; low operating temperature; high conversion efficiency; low weight to volume ratio; and rapid start [2]. These attractive characteristics have stimulated the car manufacturers to employ the H\textsubscript{2}-PEMFCs instead of internal combustion engines (ICEs), for example, in Toyota Mirai, Hyundai iX35, Honda Clarity Fuel Cell, and BMW [3]. Although significant progress has been made in H\textsubscript{2}-PEMFC technology, more research is needed to be conducted to increase these cells’ lifetime via thermal management techniques [4]. This measure transfers the heat generated inside the H\textsubscript{2}-PEMFC stack to the surroundings and manages the water produced by the electrochemical reaction keeping it at desirable levels (i.e., the membrane electrode assembly (MEA) dehydration or flooding affects the electrical performance of the stack). This can be achieved by employing a well-designed cooling system, which ensures a temperature distribution as uniform as possible to keep thermal stresses as minimum as possible within the stack [5, 6]. Since the
The difference between the operating temperature of H₂-PEMFCs (60-80°C) and the temperature of the surrounding (>50°C in hot climates) is lower than that in conventional ICEs, cooling the H₂-PEMFC is a real challenge [7]. This challenge is due to the amount of heat dissipation proportional to the physical properties, flow rate, and the difference between the coolant’s outlet and inlet temperatures. Therefore, the temperature difference of the coolant is limited by the low-temperature difference between the H₂-PEMFC stack and the surroundings, the effective parameters that need to be optimized are the physical properties and flow rate of the coolant.

Since increasing the coolant flow rate increases the pumping power, researchers have focused on improving the coolant’s physical properties, especially the effective thermal conductivity, to enhance the heat transfer. Consequently, the term “nanofluids” has emerged to represent the existence of nanoparticles (NPs) suspended in a fluid (usually liquid). The main reason behind suggesting nanofluids is that solids (metal, metal oxides, and ceramics) have higher thermal conductivities than water [8]. However, adding nanoparticles to a fluid does not change the thermal conductivity alone but affects other properties such as density, viscosity, and heat capacity, determinants for heat transfer processes [9, 10].

Since the first proposition of employing nanofluids by Choi [11] as potential effective alternatives instead of the traditional heat transfer fluids, many studies have been conducted investigating the effects of suspending NPs in fluids on the thermophysical properties and heat transfer. Although some studies showed degradation in heat transfer, many studies reported anomalous heat transfer enhancement, and Brownian motion was suggested as the primary responsible factor in improving the heat transfer [12-14]. Therefore, using nanofluids to face the thermal management challenge in H₂-PEMFC has attracted researchers and scientists [9, 11, 15]. However, the feasibility, practicality, and safety of nanofluids are still controversial [8].

In this paper, we present part of a research project’s results, which investigates the feasibility, practicality, and safety of employing nanofluids in H₂-PEMFC as alternatives to conventional heat transfer fluids. Mathematical models have been developed for H₂-PEMFC and nanofluids’ properties to estimate the performance of the H₂-PEMFC using alumina aqueous nanofluids (H₂O-Al₂O₃).

2. Model description
Mathematical models of a PEM fuel stack, shown in Figure 1, and a nanofluid are described in the following subsections.

2.1. PEM fuel cell model
The mathematical model describing the performance of a PEM fuel cell is expressed as following [16]:

The voltage produced by a single cell of an active area $A$ is given by:

$$ V_{cell} = V - \eta_{act} - \eta_{ohmic} - \eta_{diff} $$  \hspace{1cm} (1)

Nernst equation expresses the reversible thermodynamic potential of the H₂+O₂ reaction in catalyst layers:

$$ V = V_o + \frac{RT}{zF} \ln \left( \frac{P_{H_2}P_{O_2}^{0.5}}{P_{H_2O}} \right) $$  \hspace{1cm} (2)

where $V_o$ is a reference potential [V], $R$ is the gas constant [8.314 J/mol K], $T$ is the cell temperature [K], $z$ is the number of electrons per reacting ion or molecule, $F$ is Faraday’s constant [96487 C/mol], $P_{H_2}$, $P_{O_2}$ and $P_{H_2O}$ are the partial pressure of hydrogen-oxygen at the anode catalyst/gas interface and cathode catalyst/gas interface, and water vapor, respectively [atm] [16, 17].
Figure 1. A PEM fuel cell stack liquid-cooling system and heat paths in a PEM fuel cell segment.

Activation overpotential is a result of the kinetics of charge transfer reaction through the electrode-electrolyte interface. Therefore, losses in the electrode potential occur to drive the electron transfer reaction. These losses directly depend on the nature of the electrochemical reaction, and their magnitude represents the activation energy when the reaction propagates at a rate demanded by the current. The total activation overpotential is the sum of the anode and cathode overpotentials. The Equation for the anode overpotential is [18, 19]:

\[
\eta_{\text{act}} = \frac{RT}{\alpha F} \ln \left( \frac{i}{i_0} \right)
\]

where \( \alpha \) is the charge transfer coefficient, and \( i \) is the current density [A/cm\(^2\)].

Owing to the electrical resistance losses (namely, the membrane ionic resistance, the electrodes ionic and electronic resistance, and the gas diffusion layers, bipolar plates and terminal connections ionic and electronic resistance), ohmic overpotential is developed and can be expressed as:

\[
\eta_{\text{ohmic}} = -iR_{\text{internal}}
\]

where \( R_{\text{internal}} \) is total internal specific resistance of the fuel cell [\( \Omega \) cm\(^2\)].
On the other hand, diffusion overpotential is related to mass transfer limitations depending on the reactants’ availability near the electrodes. The electrode reactions require a constant supply of reactants in order to sustain the current flow. Low or high availability of reactants near the electrodes causes losses in the cell’s output voltage since a part of the reaction energy is used to drive the mass transfer process. The following expression can represent the total diffusion overpotential:

\[ \eta_{\text{diff}} = \frac{R}{zF} \ln \left(1 - \frac{i}{i_L}\right) \]  

(5)

where \( i_L \) is limiting current density. 

Once the cell voltage and the current of the cell are determined, the electrical power generated from it can be calculated as:

\[ P_{\text{cell}} = V_{\text{cell}} \cdot i \]  

(6)

The parameters listed in Table 1 were selected, which resulted in a realistic and validated PEM fuel cell polarization curve [16, 17].

| Property                        | Value                     |
|---------------------------------|---------------------------|
| Fuel                            | Hydrogen                  |
| Oxidant                         | Air                       |
| Temperature, \( T \)            | 353.15 (80 °C)            |
| Pressure, \( P \)               | 1 atm                     |
| Cathode charge transfer coefficient, \( \alpha_c \) | 1                         |
| Anode charge transfer coefficient, \( \alpha_a \) | 0.5                      |
| Reference exchange current density, \( i_o \) | 3E-6 A/cm²     |
| Limiting current density, \( i_L \) | 1.4 A/cm²               |
| Internal resistance, \( R_{\text{internal}} \) | 0.15 Ohm.cm² |

The thermodynamic efficiency of the fuel cell can be calculated as:

\[ E_{\text{cell}} = \frac{2V_{\text{cell}}}{M_{H_2} \cdot LHV_{H_2}} \]  

(7)

where \( M_{H_2} \) is the molecular mass of hydrogen [kg/mol] and \( LHV_{H_2} \) is the lower heating value of hydrogen [J/kg]. Heat is generated inside the fuel cell due to the reactions, phase change, and (voltage) losses, which can be expressed as:

\[ Q_{\text{gen}} = (1.23 - V_{\text{cell}}) i A \]  

(8)

Moreover, this amount of generated heat must be absorbed by the cooling fluid (coolant) and carried out in the fuel cell:

\[ Q_{\text{cool}} = m_{\text{cool}} \cdot C_{p,\text{cool}} \cdot (T_{\text{cool, out}} - T_{\text{cool, in}}) \]  

(9)

where \( m_{\text{cool}} \) is the mass flow rate of the cooling fluid [kg/s], \( C_{p,\text{cool}} \) is the specific heat of the cooling fluid [J/(kg.K)], and \( T_{\text{cool, out}} \) and \( T_{\text{cool, in}} \) are the cooling fluid outlet and inlet temperatures, respectively. The temperature difference (\( \Delta T = (T_{\text{cool, out}} - T_{\text{cool, in}}) \)) is a design variable, which must be selected carefully in conjunction with the coolant flow rate. In other words, to get a uniform distribution of temperature through the cell, \( \Delta T \) should be chosen as small as practically possible, unless larger temperature gradients are required by stack design (for example, to facilitate water management). However, small \( \Delta T \) would result in a large coolant flow rate, which leads to an increase in the parasitic power and a reduction in the system efficiency. While, larger \( \Delta T \) leads to a lower temperature to which the coolant must be cooled; therefore, practical limits associated with the ambient temperature and the characteristics and performance of the heat exchange device might emerge. Also, larger \( \Delta T \) may reduce the lifetime of the fuel cell stack due to the inherent thermal stresses.
The pressure drop through a pipe for laminar flow is expressed as follows:

\[
\frac{\Delta p}{l} = \frac{64 \rho u^2}{Re 2 D_h}
\]  

(10)

where \(\Delta p\) is the pressure difference between inlet and outlet of the pipe [Pa], \(l\) is the pipe’s length [m], \(u\) is the average speed of the coolant [m/s], \(Re\) is Reynolds number \((Re = \frac{\rho u D_h}{\mu})\), and \(D_h\) is the hydraulic diameter [m]. Finally, the pumping power can be expressed by the following equation:

\[
PP = u \cdot \nabla p \cdot A_{cs}
\]  

(11)

where \(PP\) is the pumping power [W] and \(A_{cs}\) is the cross-section area of the pipe [m²].

2.2. Thermophysical properties of the coolant

The thermophysical properties of the base fluid “bf” (i.e., water) depend on temperature and can be obtained using the following equations [12];

\[
\rho_{bf} = \frac{999.844 + 18.225(T + 273.15) - 7.92 \times 10^{-5}(T + 273.15)^2 - 5.54 \times 10^{-5}(T + 273.15)^3 + 1.498 \times 10^{-7}(T + 273.15)^4 - 3.93 \times 10^{-10}(T + 273.15)^5}{1 + 1.816 \times 10^{-2}(T + 273.15)}
\]  

(12)

\[
\mu_{bf} = 2.414 \times 10^{-5} \times 10^{247.8 \frac{T}{T-140}}
\]  

(13)

\[
C_{Pbf} = 8958.9 - 40.535T + 0.11243T^2 - 1.014 \times 10^{-4}T^3
\]  

(14)

\[
k_{bf} = -0.58166 + 6.3556 \times 10^{-3}T - 7.964 \times 10^{-6}T^2
\]  

(15)

In this study, Alumina (Al₂O₃) is used because of its good chemical stability in the base fluid (water), relatively low cost compared to other nanoparticles. The thermophysical properties of the nanofluids “nf” depend on the volume fraction \(\phi\) of the nanoparticles “np” in the suspension and can be obtained using the following equations [12];

\[
\rho_{nf} = (1 - \phi) \cdot \rho_{bf} + \phi \rho_{np}
\]  

(16)

\[
\rho_{nf} C_{Pnf} = (1 - \phi) \cdot \rho_{bf} C_{Pbf} + \phi \rho_{np} C_{Pnp}
\]  

(17)

The empirical correlation of the thermal conductivity based on the Brownian motion of nanoparticles are given as:

\[
k_{eff} = k_{static} + k_{Brownian}
\]  

(18)

The static thermal conductivity is given as

\[
k_{static} = k_{bf} \left[ \frac{k_{np} + 2 k_{bf} - 2(k_{bf} - k_{np})\phi}{k_{np} + 2 k_{bf} + (k_{bf} - k_{np})\phi} \right]
\]  

(19)

Brownian thermal conductivity is given as

\[
k_{Brownian} = 5 \times 10^4 \beta \cdot \Phi \cdot \rho_{bf} \cdot C_{Pbf} \sqrt{\frac{\sigma_{B,T}}{\rho_{np} \cdot d_{np}}} \cdot f(T, \Phi)
\]  

(20)

The empirical correlation of the dynamic viscosity is given as

\[
\mu_{eff} = \mu_{static} + \mu_{Brownian}
\]  

(21)

\[
\mu_{static} = \frac{\mu_{bf}}{(1-\phi)^{2.5}}
\]  

(22)

\[
\mu_{Brownian} = 5 \times 10^4 \beta \cdot \Phi \cdot \rho_{bf} \cdot \sqrt{\frac{\sigma_{B,T}}{\rho_{np} \cdot d_{np}}} \cdot f(T, \Phi)
\]  

(23)
where $\beta$ is the fraction of the liquid volume moving with the nanoparticles (Table 2), $\sigma_B$ is the Boltzmann constant, and $d_{np}$ is the diameter of nanofluids particles [nm].

$$f(T, \phi) = (2.8217 \times 10^{-2} \cdot \phi + 3.917 \times 10^{-3}) \left( \frac{T}{T_0} \right) + (-3.0699 \times 10^{-2} \cdot \phi - 3.91123 \times 10^{-3})$$  \hspace{1cm} (24)

Table 2. Values of $\beta$ for Al$_2$O$_3$ nanoparticles.

| Nanoparticles | $\beta$          | Volume fraction [%] | Temperature [K]  |
|---------------|------------------|---------------------|------------------|
| Al$_2$O$_3$   | $8.4407(100\%)^{-1.97304}$ | 1% ≤ $\phi$ ≤ 10% | 298 ≤ $T$ ≤ 363 K |

The thermophysical properties of the Al$_2$O$_3$ nanoparticles evaluated at 300 K are presented in Table 3.

Table 3. The thermophysical properties of Al$_2$O$_3$ nanoparticles at $T=300K$ [12].

| Thermophysical properties | Al$_2$O$_3$ |
|---------------------------|------------|
| Density $\rho$ [kg/m$^3$] | 3970       |
| Thermal conductivity $k$ [W/m.K] | 40        |
| Specific heat $C_p$ [J/kg.K] | 765       |

2.3. Validation of the model
To validate the model in this study, its results should be compared with those available in published research. Among the published results on this topic, which is very limited, the results of Nusselt number with a range of Reynolds numbers for laminar and turbulent flow.

The Nusselt number $N_u$ for the nanofluids coolant can be calculated from the following experimental correlations [13];

$$\begin{align*}
N_u &= 0.4328(1 + 11.285\phi^{0.754}(P_e)^{0.218}R_e^{0.333}Pr^{0.4}) \quad \text{(for laminar flow)} \\
N_u &= 0.0059(1 + 7.62868\phi^{0.6806}(P_e)^{0.001}R_e^{0.9230Pr^{0.4}}) \quad \text{(for turbulent flow)}
\end{align*}$$  \hspace{1cm} (25)

Prandtl number can be calculated as;

$$Pr = \frac{C_P n_f \mu n_f}{k n_f}$$  \hspace{1cm} (26)

and Peclet number can be calculated as;

$$Pe = \frac{u \cdot d_{np} \rho n_f C_P n_f}{k n_f}$$  \hspace{1cm} (27)

The friction factor of the nanofluids is calculated by [13];

$$f = 0.961\phi^{0.052}R_e^{-0.375}$$  \hspace{1cm} (28)

The heat transfer coefficient $h$ is calculated by;

$$h = \frac{N_u k}{2D_h}$$  \hspace{1cm} (29)

Figure 2 compares the Nusselt number of this study with the literature results [12, 13]. The comparison shows that the overall trend is very similar, and it is in good agreement with the published result with a deviation of less than 1%.
Figure 2. Comparison of the Nusselt numbers in the present study with the literature results.

3. Results and discussion

The Polarization curve, power density, and heat generation of the fuel cell working at 80°C temperature and 1 atm reactant pressure are shown in Figure 3. Clearly, the potential across the fuel cell decreases as its current increases, while the heat generation increases. However, the cell’s output power increases as its current increases until a particular value and decreases as the current increases. The maximum power of 0.32 W/cm² can be extracted from the cell when it operates at a potential of 0.4 V and a current of 0.8 A/cm². At this operation conditions, the cell generates heat of 0.66 W/cm². That means the heat generation is higher than twice of the power generation, and this needs an efficient cooling system to keep the cell at the design operating temperature and to save it from failure (the fuel cell is built from many thin layers of different materials of different mechanical and physical properties namely the polymer electrolyte membrane, catalyst, and gas diffusion layers and these thin layers are sensitive to temperature value and distribution).

Figure 3. Polarization curve, power density, and heat generation at 80 °C cell temperature and 1 atm reactant pressure.
Figure 4 shows the relationship between fuel cell efficiency and output power. Obviously, the maximum efficiency ($E_{cell} \sim 73\%$) occurs at no load on the cell (i.e., $i = 0 \text{ A/cm}^2$), and $E_{cell}$ decreases to 32% as the output power increases to its maximum value ($W_{cell} = 0.32 \text{ W/cm}^2$). After that point, $E_{cell}$ continues to decrease when $W_{cell}$ starts diminishing. This reduction in $E_{cell}$ is due to fuel cell losses, especially heat generation.

![Figure 4. Relationship between fuel cell efficiency and power output.](image)

The thermal performance of a fuel cell stack depends on the thermophysical properties of coolants used in managing its temperature. Properties of Al$_2$O$_3$-H$_2$O nanofluids, which appear in equations, govern the $Q_{cool}$ and $\Delta p$, are shown in Figure 5. Linear relationships between the nanofluid’s properties and its concentration are transparent. The nanofluid’s density and dynamic viscosity increase as the concentration increases, while the specific heat capacity decreases.

To minimize thermal stresses, the temperature distribution within the stack must be as homogeneous as possible, and this can be achieved if the difference between the outlet and inlet temperatures ($\Delta T$) of the coolant is as minimum as possible. This temperature difference is shown in Figure 6. Clearly, $\Delta T$ decreases at high slope as Re increases (i.e., $Re < 200$), and it continues in decreasing but at lower slope when Re increases further (i.e., $Re > 200$). Obviously, $\Delta T$ of Al$_2$O$_3$-H$_2$O 4% is the biggest, and $\Delta T$ of Al$_2$O$_3$-H$_2$O 1% is more significant than $\Delta T$ of H$_2$O. Interestingly, although higher Re leads to lower $\Delta T$, still employing H$_2$O is better than Al$_2$O$_3$ aqueous nanofluids, as shown in Figure 6 (red lines). This can be interpreted as follows: increasing NPs concentration increases the coolant viscosity and density. However, the percentage increase in density is higher than the percentage increase in viscosity (as shown in the inset of Figure 5b). According to the equation of Re, lower flow velocity at the same Re leads to higher $\Delta T$. Moreover, increasing NPs concentration leads to lowering the effective heat capacity of the coolant (as shown in Figure 5a), which participates in higher $\Delta T$. Also, it is clear that the percentage increase in $\Delta T$ of a certain nanofluid concentration is independent of Re (i.e., constant over the whole Re) domain. This can be explained by returning to Equation (8), where $Q_{cool} = m_{cool} \cdot C_{p,cool} \cdot (T_{cool, out} - T_{cool, in}) = m_{cool} \cdot C_{p,cool} \cdot \Delta T$. Hence, $\Delta T = \frac{Q_{cool}}{C_{p,cool} \cdot m_{cool}}$. Since $Q_{cool}$ and $C_{p,cool}$ are constants, then $\Delta T$ is a function of $m_{cool}$, or in other words, is a function of the coolant velocity. Furthermore, since Re is a function of the coolant velocity, $\Delta T$ can be expressed as
Nanofluids properties as a function of NPs concentration. Therefore, this equation clearly shows that for the same Re for a nanofluid and water, its value depends only on the nanofluid concentration, which is why the percentage increase in temperature is constant over the domain of Re. The same procedure can be followed to show the percentage increase in the pressure drop and pumping power are independent of Re too, as seen in the next figures.

\[
\Delta T = \frac{Q_{\text{cool}}}{C_p \text{cool} \cdot Re_{\text{cool}} \cdot \mu_{\text{cool}}}.
\]

Therefore, 
\[
\frac{\Delta T_{\text{nf}}}{\Delta T_{\text{H2O}}} = \frac{\mu_{\text{H2O}} C_p_{\text{H2O}} R e_{\text{H2O}}}{\mu_{\text{nf}} C_p_{\text{nf}} R e_{\text{nf}}} - 1
\]

This equation clearly shows that for the same Re for a nanofluid and water, its value depends only on the nanofluid concentration, which is why the percentage increase in temperature is constant over the domain of Re. The same procedure can be followed to show the percentage increase in the pressure drop and pumping power are independent of Re too, as seen in the next figures.

Figure 5. Nanofluids properties as a function of NPs concentration.
To investigate the pumping power consumption, the pressure drop ($\Delta p$) of the coolants is calculated according to Equation (10), and the results are presented in Figure 7. The pressure drop ($\Delta p$) is directly proportional to Re, and as the NPs concentration increases, the $\Delta p$ increases. As the percentage of $\Delta T$, the percentage of $\Delta p$ of each nanofluid is constant over the complete domain of Re. Since the pumping power is an important parameter to compare different coolants, the pumping power ($PP$) is calculated according to Equation (11) and presented in Figure 8. This figure, in contrast to previous ones, shows a non-linear relationship between $PP$ and Re. No doubt, employing nanofluids in cooling fuel cell stacks consumes higher power than using water at the same Re, especially at high Re, and no practical gains from using nanofluids, as shown in Figure 9, where $\Delta T$ of nanofluids is higher than that of pure water.

### Figure 6. Coolants’ temperature difference and its percentage of reduction as a function of Re.

### Figure 7. Coolants’ pressure drops, and their increase percentage as a function of Re.
4. Conclusions
A simulation model of electrochemical reactions of a PEM fuel cell has been developed and validated to investigate the cooling process and its effect on the cell when using nanofluids as a coolant. The thermophysical properties of the coolant were studied. The effects of both the concentration and the velocity of the coolant on the thermal performance of the cell were examined. The results showed that using nanofluids as a coolant increases the pumping power used in circulating the coolant in the fuel cell system. Furthermore, at a particular value of pumping power, the temperature difference between the outlet and inlet temperature of the coolant increases as the nanoparticle concentration increases, which is not desirable for increasing the cells’ lifetime. Therefore, employing nanofluids seems an
impractical choice, and economic evaluation and environmental impact of using nanofluids should be addressed. The present analytical model is beneficial for estimating the performance of the PEM fuel cell stacks and optimizing the thermal performance during their operation. However, using CFD numerical model to simulate three-dimensional parts of the fuel cell is essential to provide detailed insight into the various transport mechanisms and their interaction.

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