Investigation of Gradient Platinum-Loading and Porosity Distribution for Anion Exchange Membrane Fuel Cells

Hassan Mousa, Lei Xing, Prodip K. Das

Abstract
Anion exchange membrane fuel cells (AEMFCs) are in development as a low-cost alternative to proton exchange membrane fuel cells (PEMFCs). AEMFCs produce water at the anode side and consume it at the cathode side, resulting in no cathode water-flooding like in PEMFCs. However, it brings complexity to water transportation behaviour and requires appropriate water balance to avoid membrane drying out. In this study, a two-dimensional two-phase multi-physics model has been developed to investigate the impacts of three key electrode parameters (porosity, catalyst loading, and ionomer content) that are responsible for water production and transport as well as the performance of an AEMFC. A piecewise constant function along the x-direction (reactant diffusion direction) is used to apply the gradient on the porosity and platinum loading. The present results show that a larger porosity gradient near the cathode gas-diffusion layer (GDL)/flow-channel interface and lower near the GDL/microporous layer (MPL) interface can enhance mass transport and water removal, which is benefited the AEMFC performance. However, anode GDL porosity gradients show a lower AEMFC performance compared to the cathode porosity gradients. Moreover, it was confirmed that for both electrodes, the performance of AEMFC was significantly dependent on each electrode parameter.

Keywords: Anion exchange membrane fuel cell; Numerical modelling; Gradient porosity; Gradient platinum loading; Water transport

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Introduction

Low-temperature polymer electrolyte fuel cells (PEFCs) using anion exchange membranes (AEMs) have attracted attention as a promising alternative to proton exchange membrane fuel cells (PEMFCs) [1-7]. Since the commercialization of the PEMFC is currently hindered by the extremely high-cost hardware materials and platinum group metal (PGM) catalysts for sluggish oxygen reduction reaction (ORR), AEMFCs could provide an alternative avenue. AEMFCs can operate with economical materials for a variety of components such as catalyst layers (CLs) and bipolar plates. Moreover, in AEMFC’s alkaline circumstances, a variety of affordable non-noble metal-based ORR catalysts have catalytic activity comparable to that of platinum (Pt) [8, 9]. For this reason, AEMFCs are considered a promising approach to mitigate system costs by converting the current acidic system into an alkaline environment. Also, AEMFCs operate at high pH levels that provide a less corrosive environment compared to PEMFCs [4]. In addition, state-of-the-art cells often run at near ambient pressure; thus, it is expected that some of the balance-of-plant components will be simplified because of this [10-12]. However, by comparing AEMFC’s performance with PEMFCs, it is observed that AEMFCs are more sensitive to cell operating conditions due to complex water dynamics [13, 14]. Nevertheless, the power density of AEMFCs has grown dramatically, with power densities exceeding 2.5 W/cm² achieved using highly conductive membranes and ionomers [12, 14-16]. The other source of the AEMFC limitation that has been reported in the literature is the reactants’ kinetic processes, which can be either anode reaction, cathode reaction, or both electrode reactions [7, 17-20]. Accordingly, additional study is required to determine which function of the two electrodes contributes to reducing the AEMFC performance.

AEMFCs have made considerable progress in system performance and durability over the past year, which is linked to the development of AEM with high ionic conductivity (over 200 mS/cm²) and better alkaline stability [11, 12, 21-26]. A recent study with a poly(norbornene)-based AEMFC with a total PGM loading of 1.3 mg/cm² showed an extremely high power density (about 2.34 W/cm²) at 80°C and a steady operation for over 200 hours [22]. Conversely, AEMFC with poly(diphenyl-terphenyl piperidium)-based AEM with fairly low PGM loading (0.65 mg/cm²) showed a peak power density 2.58 W/cm² [25]. In addition, Adabi et al. have recently developed a high-performance non-PGM cathode-based AEMFC that surpasses the US Department of Energy 2022 milestone for AEMFC performance [26]. Their study used a low-loading PGM anode of 0.125 mg/cm² of platinum and ruthenium (PtRu) and a commercial Fe-N-C cathode. Moreover, a remarkable specific power output of 10.4 W per mg is reported with a high maximum power density surplus of 2 W/cm². Although these are promising results, the
majority of AEMFCs research is currently focused on the use of commercially affordable AEMs with low resistivity and chemical stability [27]. Despite the relatively high total PGM loading (> 0.6 mg/cm²), often the performance of an AEMFC with commercially affordable AEMs is significantly inferior to that of the PEMFC, in contrast to the previously reported AEMFCs using new AEMs [20, 28-33]. The inferior performance of AEMFCs with commercial AEMs might be linked with the water balance, as inappropriate water balance can lead to the membrane drying out.

There are a few studies that claimed the intrinsic water balance through AEMFC is one of the main factors that lead to inferior overall performance [34, 35]. In AEMFC, water is generated at the anode side by the hydrogen oxidation reaction (HOR) and consumed at the cathode side by the ORR. Accordingly, the mass transport mechanism in the AEMFC system can have a high impact on the water complex behaviour, resulting in cathode dehydration. The mass transport issue is exacerbated by the ionomeric component adsorption at the anode catalyst layer. Under low potential circumstances, the cation groups in the ionomer are readily adsorbed on Pt. The hydroxide co-adsorption, as well as water on the catalyst surface, is persuaded by the adsorbed cation. As a result, a significantly larger hydroxide ions concentration is developed [36, 37]. The accumulating co-adsorbed layer prevents water and hydrogen from diffusing through the anode [38]. Thus, effective water management in AEMFC is necessary to guarantee improved system performance. The anode flooding and cathode dehydration problems can be alleviated by controlling some operating parameters such as the relative humidity (RH) of inlet gases [14, 39].

As a result, a proper electrode design of AEMFC is required because an appropriate electrode design for a specific AEM is critical to minimising the water imbalance issue [10]. As the electrode is a complex multi-component system made of carbon-supported catalyst agglomerates coupled to ionomer [27], balancing electrode components may lead to an improvement of the electrode mass transport mechanism that could be more effective than just changing the components themselves [13].

Recently, several studies on the composition and design of AEMFC electrodes have been published using novel and commercial AEMs. According to Omasta et al. [10], the amount and percentage of ionomer to carbon present in the anode catalyst have a substantial impact on the AEMFC performance when utilising radiation-grafted power ionomer [13, 20, 22, 28-30, 32, 39-41]. It has been claimed that having an adequate carbon quantity in the electrode is essential for the anode’s water capacity. This is achieved by making space for water retention or removal. They also studied the influence of anode thickness and catalyst distribution on the AEMFC performance by incorporating carbon into the present thin catalyst layer or establishing a multi-
layered structure [40]. It has been shown that using a double-layered anode with a carbon-based microporous layer for managing water in the anode AEMFC was more successful than just weakening the catalyst with carbon. Two other studies also revealed that the performance and durability of AEMFCs may increase due to the water balance improvement, where both electrodes are hydrophobic using polytetrafluoroethylene (PTFE) [22, 41]. In terms of the use of commercial AEMs, several studies reported that electrode manufacturing factors such as slurry-solvent type have a substantial effect on the electrode microstructure, namely the pore structure [28, 30]. Several studies have shown that commercial AEMs can be beneficial to improving operating conditions by modifying the electrode parameters such as ionomer content and catalyst loading [20, 29, 42]. While electrode parameters have been investigated rigorously, the effect of GDL properties on AEMFC’s performance is unclear due to a lack of studies. Reducing GDL porosity results in a higher effective electronic conductivity but it reduces the reactant gases and water transport and increases liquid-water saturation inside the GDL [43]. Thus, balancing GDL properties or developing a graded GDL design is a viable pathway for improving water transport and management for AEMFCs. While there are a few studies available on GDL porosity gradient, all of them are for PEMFCs [44-50], the investigation of graded GDL on AEMFC’s performance is still elusive. Thus, the purpose of this study is to numerically investigate graded GDL and gradient Pt-loading on AEMFC’s performance. Here we have developed a two-dimensional two-phase multi-physics AEMFC model to discover the best variation of anode and cathode properties for improving the current density distribution and devolving a better water management protocol.

**Numerical Model**

**Computational Domain and Assumptions**

The numerical model is built on a two-dimensional (2D) computational domain that includes flow channels, GDLs, MPLs, CLs of anode and cathode, and an AEM sandwiched in between. A schematic diagram of the present computational domain is shown in Figure 1. The 2D computational domain includes half of the rib and channel of an AEMFC. This is due to the central point of the channel width being symmetrical on both sides of the channel width, as illustrated in part (a) of Figure 1, while part (b) shows the mesh structure used for the numerical simulations. The design and operating parameters used for the simulations are given in Table 1. The electrochemical and transport processes in the major components, including the catalyst layers and membrane, are considered in a two-phase model. The following assumptions are made: (1) all the gas species flow in this model follow the ideal gas law; (2) the flow is in the
laminar state due to the small pressure gradients as well as flow velocity; (3) steady-state condition; (4) the model is considered as an isothermal model; and (5) the anion membrane is considered as impermeable to the gas phase. The gravity force has a negligible impact on liquid water movement; thus, it is not taken into consideration in this study.

Figure 1. (a) Schematic computational domain of a 2D AEMFC and (b) computation mesh structure.

Table 1. Design and operating parameters values.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Ref.</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Operation temperature</td>
<td>60°C</td>
<td>[29, 51]</td>
<td>Typical temperature ranges are 40–80 °C</td>
</tr>
<tr>
<td>Operation pressure</td>
<td>1.5 atm</td>
<td></td>
<td>In both electrodes</td>
</tr>
<tr>
<td>Catalyst loading</td>
<td>0.45 mg/cm²</td>
<td>[29]</td>
<td>In both electrodes</td>
</tr>
<tr>
<td>Ionomer content in CLs</td>
<td>40 wt%</td>
<td>[29]</td>
<td>Fitted parameter</td>
</tr>
<tr>
<td>Ionomer weight</td>
<td>40%</td>
<td></td>
<td>Base case value (varied)</td>
</tr>
<tr>
<td>Carbon volume</td>
<td>40%</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Porosity of MPLs and GDLs</td>
<td>0.6, 0.4</td>
<td></td>
<td>In both electrodes</td>
</tr>
<tr>
<td>Porosity of CLs</td>
<td>Eq. (31)</td>
<td></td>
<td>Calculated</td>
</tr>
<tr>
<td></td>
<td>Anode</td>
<td></td>
<td>Base case</td>
</tr>
<tr>
<td>----------------------</td>
<td>-----------------</td>
<td>-----------</td>
<td>-----------</td>
</tr>
<tr>
<td>GDL thickness</td>
<td>190 µm</td>
<td>[29]</td>
<td></td>
</tr>
<tr>
<td>MPL thickness</td>
<td>20 µm</td>
<td></td>
<td>Base case</td>
</tr>
<tr>
<td>CL thickness</td>
<td>10 µm</td>
<td>[52]</td>
<td></td>
</tr>
<tr>
<td>RH</td>
<td>100%</td>
<td>[42]</td>
<td></td>
</tr>
<tr>
<td>Charge transfer coefficient</td>
<td>0.5,0.5</td>
<td>[42]</td>
<td></td>
</tr>
<tr>
<td>Reference hydrogen concentration</td>
<td>30 mol/m³</td>
<td>[53]</td>
<td>Base case</td>
</tr>
</tbody>
</table>

**Governing equations**

The mass conservation equation of the gas mixture is solved in all fluid zones including the flow fields, GDLs, MPLs, and CLs, as:

$$\nabla \cdot (\rho_g \vec{u}_g) = S_m$$  

(1)

where $g$ is the gas phase, $\rho_g$ and $\vec{u}_g$ are the gaseous phase density and the superficial velocity vector, respectively.

The momentum conservation equation of gas mixture is solved in the flow fields, GDLs, MPLs, and CLs:
\[ \nabla \left( \frac{\rho_g \vec{u}_g \cdot \vec{u}_g}{\varepsilon^2 (1 - s)^2} \right) = -\nabla P_g + \nabla \cdot \mu_g \left( \nabla \left( \frac{\vec{u}_g}{\varepsilon (1 - s)} \right) + \nabla \left( \frac{\vec{u}_g^T}{\varepsilon (1 - s)} \right) \right) \]

\[ -\frac{2}{3} \nabla \cdot \mu_g \left( \nabla \left( \frac{\vec{u}_g}{\varepsilon (1 - s)} \right) \right) + S_u \]

where the dynamic viscosity of the gaseous phase is \( \mu_g \), the porosity of the domain is \( \varepsilon \), and the liquid water volume fraction is \( s \) (defined as the volume fraction of void space occupied by liquid).

The mass conservation equation of gas species is solved in the flow fields, GDLs, MPLs, and CLs. The mass conservation for hydrogen is solved at the anode and for oxygen is solved at the cathode. While the mass conservation for water vapour is solved at both anode and cathode inlets, respectively. The mass transport of these species diffuses through the GDLs and MPLs to reach the CLs on both sides. The diffusion of the reactants in the GDLs, MPLs and CLs is described using the Stefan Maxwell equation:

\[ \rho_g (u_g \cdot \nabla) x_i^g - \nabla \left( \rho_g \sum_{j=1}^{n} D_{ij}^{\text{eff}} \left( \nabla x_j^g + \frac{\nabla p}{\rho} (x_j^g - w_j^g) \right) \right) = S_i^g \]

where \( w \) is the mass fraction, \( x \) is the molar fraction of species, and the species are donated by the subscripts \( i \) and \( j \), the \( S_i \) being the source term of the \( i^{th} \) species (production and consumption), the pressure field of the gaseous phase is \( P_g \).

The conservation equation of liquid water transport in GDLs, MPLs, and CLs, is defined from the volume-averaged continuity equation as:

\[ \nabla \cdot \left( - \frac{\rho_l k_l d p_{\text{cap}}}{\mu_l ds} \right) \nabla s + \nabla \cdot \frac{\rho_l \mu_l k_l}{\mu_g k_g} \vec{u}_l = S_l \]

where \( \rho_l \) is the liquid water density, \( s \) is the liquid water saturation, \( \mu_g \) and \( \mu_l \) are the dynamic viscosities of gas and liquid water phases, respectively, also they are functions of temperature; \( K_{rl} \) and \( k_{rg} \) are the gas and liquid relative permeability. The liquid water (capillary) diffusivity is defined by the first part on the left, which is based on the theory of capillary pressure and applying Darcy’s law for both liquid and gas phases in the porous medium.

The capillary pressure \( p_{\text{cap}} \) is estimated as an empirical function of liquid water saturation in the porous medium as [56, 57]:

\[ p_{\text{cap}} = p_g - p_l = \sigma \cos(\theta) \left( \frac{\varepsilon}{k} \right)^{0.5} J(s) \]  

where the gas and liquid water phase pressure, and the intrinsic permeability of porous medium are \( p_g \), \( p_l \) and \( k \), respectively. The surface tension is \( \sigma \) and the contact angle of water with the pore walls is \( \theta \). In Eq. (5), \( J(s) \) is the Leverett function, and is given as a function of water saturation as \[ \text{[43, 58-60]} \]:

\[
J(s) = \begin{cases} 
1.417(1 - s) - 2.120(1 - s)^2 + 1.263(1 - s)^3 & \theta_c < 90^\circ \\
1.417s - 2.120s^2 + 1.263s^3 & \theta_c > 90^\circ 
\end{cases}
\]  

The effective saturation should be applied to the Leverett function. The effective saturation in the hydrophilic surfaces is considered to arise within the composite volume of the overall porous electrode, including GDLs, MPLs, and CLs. It is also worth noting that the Leverett \( J \)-function may not be appropriate for diffusion media with high PTFE content due to the inaccurate prediction of capillary pressure at high non-wetting phase saturation and hysteresis of capillary pressure \[ \text{[61, 62]} \].

Note that the wettability and liquid water amount in the porous electrode calculate the capillary pressure. \( \sigma_t, \theta, \) and \( \varepsilon \) are the coefficient of surface tension between the gas and liquid water, the contact angle of the porous material (depending on the GDL, MPL, and CL wettability), and the porosity of the porous material, respectively. A linear correlation is calculated for temperatures ranging from 273.15 K to 373.15 K, \( \sigma_t \) can be expressed as:

\[
\sigma_t = -0.0001676T + 0.1218 \quad 273.15 \text{K} \leq T \leq 373.15 \text{K}
\]  

The dissolved water transport equation is solved in the CLs and membrane by the electro-osmotic drag coefficient as well as the backward and forward diffusion. Thus, the transport equation of membrane water is defined as:

\[
-\nabla \cdot \left( D_{\text{d eff}} \nabla \lambda \right) + \nabla \cdot \left( \frac{E_W}{\rho_{\text{mem}}} \frac{\eta_d}{F} k_m \right) = S_d
\]  

where the diffusivity of the membrane water content, \( D_{\text{d eff}} \), and the equivalent molecular weight of the membrane \( E_W \) (in dry conditions), the membrane density is \( \rho_{\text{mem}} \), and the ionic current density is \( k_m \), the membrane water content is \( \lambda \), which relates to the dissolved water concentration, and is given as:

\[
\lambda = \frac{E_W}{\rho_{\text{mem}}} C_d
\]  

where the dissolved water molar concentration is \( C_d \).

The diffusion coefficient of membrane water content is corrected due to the ionomer content in the CL by the widely used Bruggeman approximation \[ \text{[63]} \]:

8
\[ D_d^{\text{eff}} = \omega^{1.5} D_d \]  
\[ n_d = \frac{2.5}{22} \hat{A} \]  
(10)

where the ionomer volume fraction in the CL is donated by \( \omega \), and \( n_d \) is the electric-osmotic drag coefficient. The water flux is entered and exited from the membrane because of the sorption and desorption.

The binary diffusion coefficient \( D_{ij} \) is defined as [64]:
\[ D_{ij} = \frac{3.16 \times 10^{-8} T_0^{1.75}}{p \left[ (v_i)^{1/3} (v_j)^{1/3} \right]^2 \sqrt{\frac{1}{M_i} + \frac{1}{M_j}}} \]  
(12)

where \( v \) and \( M \) are the diffusion volume of species. Due to the porosity and the volume occupied by the liquid water in the porous medium, the diffusivity of the binary species is corrected as [63]:
\[ D_{ij}^{\text{eff}} = D_{ij} \varepsilon^{1.5} (1 - s)^{1.5} \]  
(13)

The conservation of electrical current is solved in solid and ionic phases. The two potential fields (electronic and ionic) are governed by the ohm’s low, and are expressed as:
\[ -\nabla \cdot (k_s^{\text{eff}} \nabla \Phi_s) = S_s \]  
(14)
\[ -\nabla \cdot (k_m^{\text{eff}} \nabla \Phi_m) = S_m \]  
(15)

where the effective conductivity of electrons and ions are \( k_s^{\text{eff}} \) and \( k_m^{\text{eff}} \), respectively, the source term for electronic and ionic potential conservation equations are \( S_s \) and \( S_m \), respectively. The effective conductivities \( k_s^{\text{eff}} \) and \( k_m^{\text{eff}} \) are dependent on the physical properties of the medium. Using the Bruggeman approximation, the conductivities can be evaluated and expressed as [63]:
\[ k_s^{\text{eff}} = (1 - \varepsilon - \omega)^{1.5} k_s \]  
(16)
\[ k_m^{\text{eff}} = \omega^{1.5} k_m \]  
(17)

where the electronic and ionic conductivities are \( k_s \) and \( k_m \), respectively.

The electrochemical reaction procedure of anion exchange membrane fuel cell is important to understand the relationship between current density and cell voltage, also to design and determine the overall performance of the AEMFC. Moreover, the electrochemical reaction rates in the anode and cathode CLs result in mass generation terms which are described by the conservation equation of gas species and conservation equation of mass for the ionomer water transport, Eqs. (2) and (8) respectively [42].
The electrochemical reactions in fuel cells pass through three kinetic zones [65]. In addition, the reaction rate of the fuel cell depends on the probability of reactants crossing the activation energy. The loss of voltage associated with reaction rate is known as activation loss. At low current densities, activation loss is the main source of loss. The Butler-Volmer equation is used to relate current densities and the activation voltage [42, 65-67]. Another loss is termed as ohmic loss which is generated due to ions/electrons traveling through the diffusion layers and membrane [66]. Mass transport loss is the main performance loss. This is because of the depletion of reactants in the catalyst layers. Once the mass transport in electrodes and diffusion layer is improved, the overall performance of the cell will be improved too [65].

Thus, the activities of the HOR and ORR in the electrochemical reactions of anode CL and the cathode CL, respectively, are represented using the exchange current density $i_0^\text{ref}$, which is obtained from the Butler-Volmer equation based on the mass action law as [52, 66-68]:

\begin{align}
  i_a &= (1 - s)i_a^\text{ref}\left( \frac{C_{H_2}}{C_{H_2}^\text{ref}} \right)^{0.5} \left[ \exp \left( \frac{2\alpha_a F}{RT} \eta_a \right) - \exp \left( - \frac{2\alpha_c F}{RT} \eta_a \right) \right] \\
  i_c &= (1 - s)i_c^\text{ref}\left( \frac{C_{O_2}}{C_{O_2}^\text{ref}} \right)^{-4} \left[ - \exp \left( \frac{4\alpha_a F}{RT} \eta_c \right) + \exp \left( - \frac{4\alpha_c F}{RT} \eta_c \right) \right]
\end{align}

where $i_a^\text{ref}$ and $i_c^\text{ref}$ are the reference HOR and ORR exchange current density, respectively; the term $(1-s)$ is the correction factor for the site-blockage by the liquid water saturation; $A_s$ is the surface area of the specific reactive (the specific active area of the CLs); $C_{H_2}$ and $C_{O_2}$ are the hydrogen and oxygen molar concentrations of anode and cathode, respectively; $C_{H_2}^\text{ref}$ and $C_{O_2}^\text{ref}$ are the reference hydrogen and oxygen molar concentrations; $R$ is the universal gas constant (8314 J/kmol K); $F$ is the Faraday constant (96,485 C/mol); and $T$ is the absolute temperature; $\eta_a$ and $\eta_c$ are the anode and cathode kinetic overpotential. The kinetic overpotential in both anode CL and cathode CL is defined by the difference between the solid phase potential and membrane phase potential [68]; $\alpha_a$ and $\alpha_c$ are the anodic and cathodic transfer coefficients (The parameters used for the simulation in this work are showed in Table 1). From the literature, Agar et al. [69] stated that most models assume that the charge transfer coefficient $\alpha_i$ is equal to 0.5, though this has not been validated. Because the oxidation and reduction reactions in each half cell occur at different rates, using the same reaction rate constants for both half-cell reactions is very likely to result in inaccurate predictions of the activation overpotential and cell potential.
Therefore, the cathodic transfer coefficient should be slightly higher than the anodic transfer coefficient [70-72], due to the oxygen reduction reaction on Pt/C can be described more accurate from the assumption that $\alpha_c = 1.0$ [70, 73].

The anode and cathode surface overpotentials $\eta_{a,c}$ are respectively defined as [42]:

$$
\eta_a = \phi_S^a - \phi_m \tag{20}
$$
$$
\eta_c = \phi_S^c - \phi_m - E_{eq} \tag{21}
$$

where the membrane phase (ionic) potential is defined as $\phi_m$; the solid phase potential (electronic) for anode and cathode at the end of BPs are denoted by $\phi_S^a$ and $\phi_S^c$, respectively; $E_{eq}$ is the standard theoretical reversible cell potential which is derived from the modified Nernst equation under the assumption that the overall cell reaction is in thermodynamic equilibrium as [74]:

$$
V_{rev} = 1.229 - 0.846 \times 10^{-3}(T - 298.15) + \frac{RT}{2F} \left( \ln P_{H_2} + \frac{1}{2} \ln P_{O_2} \right) \tag{22}
$$

where $P_{H_2}$ and $P_{O_2}$ are the inlets pressures of hydrogen and oxygen, respectively.

The cell voltage is calculated by the following equation as [42]:

$$
V_{cell} = \phi_S^c - \phi_S^a \tag{23}
$$
Table 2. Source terms of the conservation equations.

<table>
<thead>
<tr>
<th>Source term</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$S_m = S_{H2} + S_{O2} + S_{H2O}$</td>
<td>Kg/m$^3$.s</td>
</tr>
<tr>
<td>$S_{H2} = \begin{cases} \frac{-i_a}{2F} M_{H2} &amp; \text{(in anode CL)} \ 0 &amp; \text{(in other domains)} \end{cases}$ &amp; Kg/m$^3$.s</td>
<td></td>
</tr>
<tr>
<td>$S_{O2} = \begin{cases} \frac{-i_c}{4F} M_{O2} &amp; \text{(in cathode CL)} \ 0 &amp; \text{(in other domains)} \end{cases}$ &amp; Kg/m$^3$.s</td>
<td></td>
</tr>
<tr>
<td>$S_{H2O} = \begin{cases} S_{ec} + \frac{i_0}{F} M_{H2O} + S_{ad} M_{H2O} \rho_{mem} &amp; \text{(in CLs)} \ S_{ad} &amp; \text{(in GDLs/MPLs)} \end{cases}$</td>
<td>Kg/m$^3$.s</td>
</tr>
<tr>
<td>$S_{u} = \begin{cases} -\frac{\mu_B}{K_g} \tilde{u}_g &amp; \text{(in CLs, MPLs and GDLs)} \ 0 &amp; \text{(in all other domains)} \end{cases}$</td>
<td>Kg/m$^2$</td>
</tr>
<tr>
<td>$S_{ec} = \begin{cases} \gamma_e C (xH_2O - x_{sat}) &amp; \text{if } xH_2O \geq x_{sat} \ \gamma_e C (xH_2O - x_{sat}) &amp; \text{if } xH_2O &lt; x_{sat} \end{cases}$ (in CLs, MPLs, GDLs)</td>
<td>Kg/m$^3$.s</td>
</tr>
<tr>
<td>$S_{ad} = \begin{cases} \frac{k_a}{L_{eq} v_m} (\lambda_{eq} - \lambda) &amp; \text{if } \lambda &lt; \lambda_{eq} \text{ (absorption)} \ \frac{k_d}{L_{eq} v_m} (\lambda_{eq} - \lambda) &amp; \text{if } \lambda &gt; \lambda_{eq} \text{ (desorption)} \end{cases}$ (in CLs)</td>
<td>Kg/m$^3$.s</td>
</tr>
<tr>
<td>$S_s = \begin{cases} i_a &amp; \text{(in anode CL)} \ -i_c &amp; \text{(in cathode CL)} \end{cases}$ &amp; S/m</td>
<td></td>
</tr>
<tr>
<td>$S_m = \begin{cases} -i_a &amp; \text{(in anode CL)} \ i_c &amp; \text{(in cathode CL)} \end{cases}$ &amp; S/m</td>
<td></td>
</tr>
</tbody>
</table>
Table 3. Temperature and water content dependent parameters.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Relative permeability of gas phase, $k_{rg}$</td>
<td>$(s^3)k_0$</td>
</tr>
<tr>
<td>Relative permeability of liquid phase, $k_{rl}$</td>
<td>$(1 - s)^3k_0$</td>
</tr>
<tr>
<td>Electro-osmotic drag coefficient, $n_d$</td>
<td>$(2.5/22)\lambda$</td>
</tr>
<tr>
<td>Volume fraction of water in Ionomer, $f$</td>
<td>$\lambda V_w/(V_m + \lambda V_w)$</td>
</tr>
<tr>
<td>Absorption rate coefficient, $k_{abs}$</td>
<td>$1.14 \times 10^5 f \exp \left[ 2416 \left( \frac{1}{T_0} - \frac{1}{T_{ref}} \right) \right]$</td>
</tr>
<tr>
<td>Desorption rate coefficient, $k_{des}$</td>
<td>$4.59 \times 10^5 f \exp \left[ 2416 \left( \frac{1}{T_0} - \frac{1}{T_{ref}} \right) \right]$</td>
</tr>
<tr>
<td>Membrane intrinsic ionic conductivity [67,</td>
<td>$0.1334 - 3.882 \times 10^{-4} T_0 + (0.01148 T_0 - 3.909)\alpha_w - (0.06690 T_0 - 23.01)\alpha_w^2$</td>
</tr>
<tr>
<td>75, 76], $k_m$</td>
<td>$+ (0.1227 T_0 - 42.61)\alpha_w^3 - (0.06021 T_0 - 21.80)\alpha_w^4$</td>
</tr>
<tr>
<td>Water activity, $\alpha_w$</td>
<td>$0.8118 - 2.296x10^{-3} T_0 + (5.815x10^{-3} T_0 - 2.005)\lambda - (2.977x10^{-3} T_0 - 1.046)\lambda^2$</td>
</tr>
<tr>
<td></td>
<td>$+ (4.825x10^{-4} T_0 - 0.1676)\lambda^3 - (3.179x10^{-5} T_0 - 0.01094)\lambda^4$</td>
</tr>
<tr>
<td></td>
<td>$+ (7.427x10^{-7} T_0 - 2.539x10^{-4})\lambda^5$</td>
</tr>
<tr>
<td>Leverette function, $f(s)$</td>
<td>$\begin{cases} 1.417s - 2.120s^2 + 1.263s^3 &amp; \text{if } \theta &gt; 90^\circ \ 1.417s - 2.120(1 - s)^2 + 1.263(1 - s)^3 &amp; \text{if } \theta &lt; 90^\circ \end{cases}$</td>
</tr>
<tr>
<td>Diffusion coefficient of membrane water</td>
<td>$\begin{cases} (0.0051\lambda \times T_0 - 1.44\lambda) \times 10^{-10} &amp; 0.0 \leq \lambda \leq 14.0 \ (-23.2404 + 4.513\lambda - 0.28926\lambda^2 + 0.00613\lambda^3) \times (T_0 - 303.15) \times 10^{-10} &amp; 14.0 &lt; \lambda \leq 19.0 \ (-79.826 + 17.928\lambda - 1.3329\lambda^2 + 0.03337\lambda^3) \times 10^{-10} &amp; \lambda &gt; 19.0 \end{cases}$</td>
</tr>
<tr>
<td>content [67, 77, 78], $D_d$</td>
<td></td>
</tr>
<tr>
<td>Equilibrium membrane water content [77,</td>
<td>$\begin{cases} (-605a^3 + 0.85a^2 - 0.205a + 0.153)(T - 313.15) + 39.0a^3 - 47.7a^2 + 23.4a + 0.117 &amp; 0 \leq a \leq 1.0 \ (-0.00265a + 0.05795)(T - 313.15) + 1.5915(a - 1) + 14.817 &amp; 1.0 &lt; a \leq 3.0 \end{cases}$</td>
</tr>
<tr>
<td>78], $\lambda_e$</td>
<td></td>
</tr>
</tbody>
</table>
Porous catalyst layer properties

The non-precious CL is comprised of a carbon-based catalyst, an ionomer electrolyte, void-space, and GDL penetration, with the total of their volume fractions yielding to one:

\[ L_m + L_{Pt/C} + L_s + \varepsilon_{CL} = 1 \]  

(24)

where the volume fraction of ionomer is \( L_m \) and platinum/carbon \( L_{Pt/C} \) and GDL penetration (solid portion) is \( L_s \), and the catalyst porosity is \( \varepsilon_{CL} \):

\[ L_{Pt/C} = L_{Pt} + L_C \]  

(25)

\[ \varepsilon_{CL} = \varepsilon_p + \varepsilon_s \]  

(26)

where \( \varepsilon_p \) is the primary pores, and \( \varepsilon_s \) is the secondary pores. The mass loading and densities of platinum/carbon, and ionomer, as well as the thickness of the CL, were used to calculate the volume fraction of each [79]:

\[ L_m = \frac{m_m}{\rho_m L_{CL}}, \quad L_{Pt/C} = \frac{m_{Pt}}{L_{Pt}} \left( \frac{1}{\rho_{Pt}} + \frac{1-f}{f} \frac{1}{\rho_C} \right) \]  

(27)

where \( f \) is the platinum mass ratio to that of carbon and the volume fraction of the CL solid portion is given as:

\[ f = \frac{m_{Pt}}{m_{Pt} + m_C} \]  

(28)

\[ L_s = L_{GDL}(1 - \varepsilon_{GDL}) \]  

(29)

where the percentage of gas diffusion layer that penetrates the catalyst layer is \( L_{GDL} \), and the porosity of the gas diffusion layer is \( \varepsilon_{GDL} \), therefore, the porosity of the catalyst layer and secondary pores are written as:

\[ \varepsilon_{CL} = 1 - L_m - L_{GDL}(1 - \varepsilon_{GDL}) - \frac{m_{Pt}}{L_{CL}} \left( \frac{1}{\rho_{Pt}} + \frac{1-f}{f} \frac{1}{\rho_C} \right) \]  

(30)

\[ \varepsilon_s = \varepsilon_{CL} - \frac{L_{Pt/C}\varepsilon_{CL}(1-L_m)}{1-\varepsilon_{CL}} \]  

(31)

Numerical methods and boundary conditions

The commercial finite element software COMSOL Multiphysics 5.4 was used to solve the above-mentioned equations simultaneously. It was used with the MUMPS solver. The complete mesh consists of 4044 domain quadrilateral elements and 556 boundary quadrilateral elements, where an increased mesh density was applied to the areas near the CL domains to improve the numerical accuracy of the model as shown in Figure 1b. In this study, hydrogen and water vapour are the two reactants on the anode side while oxygen, water vapour, and nitrogen are the three reactants on the cathode side, respectively. The boundary conditions of the anode and cathode
inlets mass flow rate, temperature, the mole fraction of reactant gases in gas mixture, saturation of liquid water, and pressure are applied and listed in Table 4:

<table>
<thead>
<tr>
<th>Variable</th>
<th>Boundary condition</th>
<th>Boundary</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pressure</td>
<td>$p = p_a$</td>
<td>Anode inlet/GDL</td>
</tr>
<tr>
<td></td>
<td>$p = p_c$</td>
<td>Cathode inlet/GDL</td>
</tr>
<tr>
<td>Temperature</td>
<td>$T = T_a = T_c$</td>
<td>Anode/Cathode outside edge of the bipolar plate</td>
</tr>
<tr>
<td>Reactant mole fractions</td>
<td>$x_{H_20,a} = \frac{p_{sat\text{RH}_a}}{p_a}$</td>
<td>Anode inlet/GDL</td>
</tr>
<tr>
<td></td>
<td>$x_{H_20,c} = \frac{p_{sat\text{RH}_c}}{p_c}$</td>
<td>Cathode inlet/GDL</td>
</tr>
<tr>
<td></td>
<td>$x_{H_2} = (1 - x_{H_20,a})$</td>
<td>Anode inlet/GDL</td>
</tr>
<tr>
<td></td>
<td>$x_{O_2} = 0.21(1 - x_{H_20,c})$</td>
<td>Cathode inlet/GDL</td>
</tr>
<tr>
<td></td>
<td>$x_{N_2} = 0.79(1 - x_{H_20,c})$</td>
<td>Cathode inlet/GDL</td>
</tr>
<tr>
<td>Electric potential</td>
<td>$\Phi_s = 0$</td>
<td>Anode outside edge of the bipolar plate</td>
</tr>
<tr>
<td></td>
<td>$\Phi_s = E_{cell}$</td>
<td>Cathode outside edge of the bipolar plate</td>
</tr>
<tr>
<td>Ionic potential</td>
<td>$\Phi_m = 0$</td>
<td>MPLs/CLs, for anode and cathode</td>
</tr>
</tbody>
</table>

**Results and Discussion**

**Model validation**

The 2D AEMFC model is verified against two sets of data (both modelling and experimental). Figure 2 shows the present 2D model of AEMFC with modelling data from Gerhardt et al. [80] and experimental data from Omasta et al. [14]. Both data focused on the electrode properties to improve the water management of the AEMFC. Omasta et al. [14] demonstrated a new benchmark for AEM fuel cells, using lowered humidities (anode and cathode platinum loading of 1.25 mg/cm² (BASF Pt/C 50%wt) at an operating temperature of 60°C. Gerhardt et al. [80] investigated how to solve the two significant operational challenges in AEMFCs such as water management and carbon-dioxide contamination from ambient air. The present model is validated by comparing the numerical results with the model and experimental data. As can be seen from Figure 2, an excellent fit was obtained between the model results, the model, and experimental
data, along the entire range of current densities and cell voltage. The parameters and operating
conditions used in validation cases are listed in Table 5.

Figure 2. Validation of present AEMFC models with literature data [14, 80]. Lines show the present
models’ results, while the symbols are literature data as indicated in the figure legends.

Table 5. Parameters and operating conditions used in validation cases.

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description (unit)</th>
<th>Omasta et al. 2018</th>
<th>Gerhardt et al. 2018</th>
<th>Validation Case 1</th>
<th>Validation Case 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>( l_{GDL} )</td>
<td>GDL thickness (m)</td>
<td>190</td>
<td>190</td>
<td>190</td>
<td>190</td>
</tr>
<tr>
<td>( l_{MPL} )</td>
<td>MPL thickness (m)</td>
<td>-</td>
<td>45</td>
<td>45</td>
<td>45</td>
</tr>
<tr>
<td>( l_{CL} )</td>
<td>CL thickness (m)</td>
<td>10</td>
<td>10</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>( l_{mem} )</td>
<td>Membrane thickness (m)</td>
<td>25</td>
<td>30</td>
<td>30</td>
<td>30</td>
</tr>
<tr>
<td>T</td>
<td>Operating temperature (°C)</td>
<td>60</td>
<td>60</td>
<td>60</td>
<td>60</td>
</tr>
<tr>
<td>p</td>
<td>Operating pressure (atm)</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>RH</td>
<td>Relative humidity (%)</td>
<td>87-79</td>
<td>86.5-78.4</td>
<td>87-79</td>
<td>87-79</td>
</tr>
<tr>
<td>( m_{Pt} )</td>
<td>Platinum loading (mg/cm²)</td>
<td>0.40±0.02</td>
<td>0.40</td>
<td>0.40</td>
<td>0.40</td>
</tr>
<tr>
<td>( \varepsilon )</td>
<td>Porosity in GDLs, MPLs and CLs</td>
<td>-</td>
<td>0.8,0.3,0.4</td>
<td>0.8,0.3,0.4</td>
<td>0.8,0.3,0.4</td>
</tr>
<tr>
<td>( A_s )</td>
<td>Specific surface area</td>
<td>-</td>
<td>( 10^5 )</td>
<td>( 10^5 )</td>
<td>( 10^5 )</td>
</tr>
<tr>
<td>( \alpha_a )</td>
<td>Anode transfer coefficient</td>
<td>-</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td>( \alpha_c )</td>
<td>Cathode transfer coefficient</td>
<td>-</td>
<td>0.5</td>
<td>0.5</td>
<td>0.5</td>
</tr>
<tr>
<td>( D_\lambda )</td>
<td>Water ionomer diffusivity (cm²/s)</td>
<td>-</td>
<td>( 1.6 \times 10^{-6} ), ( 0.8 \times 10^{-6} )</td>
<td>( 1.6 \times 10^{-6} ), ( 0.8 \times 10^{-6} )</td>
<td>( 0.8 \times 10^{-6} )</td>
</tr>
</tbody>
</table>
**Graded distribution of porosity**

A reduction of the GDL porosity occurs when carbon materials occupy more void space inside the GDL, and vice versa. A decrease in GDL porosity improves the effective electronic conductivity of the GDL. Consequently, this may obstruct the flow of reactant gases through the GDL, decreasing the effective mass transport coefficient. Increasing the GDL porosity, however, improves the gas transport while lowering the electrical conductivity. Moreover, the distributions of reactant gases, as well as current density over the CLs, are highly influenced by the GDL porosity. Thus, developing an optimum GDL porosity gradient for AEMFC for achieving a slight improvement in gas transport rate without significantly reducing the electrical conductivity, is a method worthy to be considered. The piecewise constant function along the $x$-direction (diffusion direction), as well as the anode and cathode GDLs design, are given below:

$$
\varepsilon_{\text{cath},\text{GDL}} = \begin{cases} 
\varepsilon_1, & 0 < x \leq \frac{L}{3} \\
\varepsilon_2, & \frac{L}{3} < x \leq \frac{2L}{3} \\
\varepsilon_3, & \frac{2L}{3} < x \leq L 
\end{cases} 
$$

$$
\varepsilon_{\text{an},\text{GDL}} = \begin{cases} 
\varepsilon_1, & 0 < x \leq \frac{L}{3} \\
\varepsilon_2, & \frac{L}{3} < x \leq \frac{2L}{3} \\
\varepsilon_3, & \frac{2L}{3} < x \leq L 
\end{cases} 
$$

Here $\varepsilon_1$, $\varepsilon_2$, and $\varepsilon_3$ denote three types of porosity given by a positive constant and each segment is one-third of GDL thickness, $L$ ($\mu$m). Note that on the anode side, $\varepsilon_1$ represents the channel/GDL interface, and $\varepsilon_3$ represents GDL/MPL interface while on the cathode side $\varepsilon_3$ represents the channel/GDL interface, and $\varepsilon_1$ represents the GDL/MPL interface. In this study, the influence of the GDLs porosity distribution along the reactant diffusion direction and the optimal porosity gradients are investigated, constructing six cases of the optimal porosity gradients using Eqs. (32) and (33) at the cathode and anode sides, respectively. Note that the uniform and non-uniform porosity at both sides have the same average porosity of 0.6. The findings of the GDL’s optimal porosity gradients are discussed below.
Graded GDL in cathode

Figure 3 shows the polarisation curves with various cathode GDL porosity gradients and corresponding maximum power density. It has been observed that the porosity gradient of 70% near the cathode GDL/inlet, 50% near the cathode GDL/MPL, and 60% in between ($\varepsilon = 0.5, 0.6, 0.7$) provides an improved limiting current density. The limiting current density increases by about 10% compared to the uniform porosity (from 1.94 to 2.15 A/cm$^2$). This highlights a better distribution of current density across the cathode CL and more liquid water in the cathode GDL may be achieved with the increasing of the GDL porosity near the GDL/inlet interface. By comparison, the sharp decrease of the optimal porosity gradients in the last three porosity gradient distribution cases $\varepsilon = 0.9, 0.6, 0.3$, $\varepsilon = 0.8, 0.6, 0.4$, and $\varepsilon = 0.7, 0.6, 0.5$ reduce the current density and degrading the limiting current density while reducing the liquid water saturation in the cathode GDL. In addition, the cathode GDL porosity gradients of $\varepsilon = 0.5, 0.6, 0.7$ performed a higher maximum power density of 0.644 W/cm$^2$, while the cathode GDL uniform porosity performed a lower maximum power density of 0.583 W/cm$^2$. This indicates that the optimal porosity gradient with 70% near the cathode GDL/inlet and 50% near the cathode GDL/MPL interface, is beneficial to achieving a more homogeneous current density distribution. Similar trends were observed from PEMFCs that a higher porosity gradient near the inlets leads to faster diffusion of gas transport through the porous gas-diffusion media [44, 45].

![Graph showing polarisation curves with various cathode GDL porosity gradients and corresponding maximum power density.](image)

Figure 3. Polarisation curves with various cathode GDL porosity gradients ($\varepsilon = \varepsilon_1, \varepsilon_2, \varepsilon_3$, where $\varepsilon_1$ is on the MPL side and $\varepsilon_3$ is on the channel side) and corresponding maximum power density.

A noticeable drop in the GDL porosity near the cathode GDL/inlet interface (from $\varepsilon = 0.5$ to 0.3), resulted in a decrease in the overall electrochemical rate owing to an increase in the oxygen transport resistance. Therefore, the rate of water production at the cathode GDL/inlet interface is unbalanced with the rate of water removal, causing an apparent increase in water volume. By
comparing the decrease and increase of the porosity gradients, the decrease of the porosity gradients of \( \varepsilon = 0.3, 0.6, 0.9 \) and \( \varepsilon = 0.9, 0.6, 0.3 \) is less noticeable. This is because the improvements in the water removal rate are more significant. Furthermore, as compared to the case of \( \varepsilon = 0.4, 0.6, 0.8 \) with uniform porosity, the current density distribution increased by 6.5%.

To investigate the effect of cathode GDL porosity gradients more deeply, the effects on the oxygen mole fraction of the cathode electrode with the porosity gradients along the diffusion direction are presented in Figure 4. Five cases of the cathode GDL porosity gradients are compared with the uniform porosity case. The designed porosity changes could enhance the current density distribution, particularly in the areas near the cathode GDL/MPL interface as of \( \varepsilon = 0.5, 0.6, 0.7 \) and \( \varepsilon = 0.4, 0.6, 0.8 \) to a limited extent. The porosity gradient of \( \varepsilon = 0.5, 0.6, 0.7 \) showed the maximum improvement of the current density distribution. While the porosity gradient of \( \varepsilon = 0.8, 0.6, 0.4 \) showed the minimum improvement of current density distribution along the reactant diffusion direction, lower than the uniform porosity of \( (\varepsilon = 0.6) \). It is noticed that applying higher cathode GDL porosity gradients at the interface of cathode GDL/MPL, causes a decrease in the middle and inlet of the cathode GDL. Resulting in a higher decrease of the electric conductivity near the GDL/MPL interfaces. However, it accelerates the decrease near the cathode GDL/MPL interface. On the other hand, the drop in the electric conductivity near the cathode GDL/inlet is advantageous for obtaining a more uniform electrochemical reaction rate along the direction of reactants. The oxygen mass fraction case with the designed porosity gradient of \( \varepsilon = 0.4, 0.6, 0.8 \) showed that the decrease in the electrical conductivity is more severe near the GDL/inlet interface. This is because of the high GDL porosity gradients applied, compared to the uniform and porosity gradients of \( \varepsilon = 0.8, 0.6, 0.4 \). In addition, the flow

![Figure 4. Oxygen mole fraction distribution profile of the cathode GDL with high and low porosity gradients in base case condition at 0.3V.](image-url)
resistance of gas electrochemical reaction is increased by the relatively low porosity close to the cathode MPL, resulting in a significant reduction in the gas electrochemical reaction rate as well as a much more uneven current density distribution. Thus, the effects of the cathode GDL porosity gradients for the oxygen mass fraction distribution are obvious in high and low porosity gradient cases.

**Graded GDL in anode**

Figure 5 shows the influences of the anode GDL optimal porosity gradient along the diffusion direction. Likewise, on the cathode side, the anode GDL optimal porosity gradients are compared with the uniform GDL porosity. It is found that there is no apparent improvement in the current density across the cathode CL in all cases. However, the limiting current density of the case $\varepsilon = 0.9, 0.6, 0.3$ decreased by 1.8% compared with the uniform porosity ($\varepsilon = 0.6$). The current density dropped from 1.87 to 1.84 A/cm$^2$. Moreover, its maximum power density is lower than that of the uniform porosity. The maximum power density for $\varepsilon = 0.9, 0.6, 0.3$ is 0.55 W/cm$^2$, compared to 0.562 W/cm$^2$ for the uniform porosity. However, the anode GDL uniform porosity, $\varepsilon = 0.5, 0.6, 0.7$, and $\varepsilon = 0.4, 0.6, 0.8$ cases showed almost similar peak power density. This identical output current density of the anode GDL optimal porosity gradients with that of the uniform porosity case implies that achieving a homogeneous current density distribution could hardly be reached with the anode GDL optimal porosity gradient.

![Graph showing polarisation curve and maximum power density](image)

Figure 5. Polarisation curve with various anode GDL porosity gradients ($\varepsilon = \varepsilon_1, \varepsilon_2, \varepsilon_3$, where $\varepsilon_1$ is on the channel side and $\varepsilon_3$ is on the MPL side).

**Graded platinum loading in cathode catalyst layer**

Studying the changes in the AEMFC performance with cathode and anode parameters can give essential information in enhancing the overall output current density as well as mitigating the transport of mass-related issues. Therefore, the design of the graded Pt loading is seen as a viable
technique for lowering Pt loading while maintaining cell performance. The cathode CL Pt design is shown in Figure 6. The polarisation curve and the cell performance with various cathode CL graded Pt loadings of 0.1 to 0.5 mg/cm² are shown in Figure 6 including the uniform graded Pt loading of (0.3 mg/cm²). The higher Pt loading cases near the CL/MPL interface were compared to the lower Pt loading cases near the CL/mem interface. Comparing the two cases of gradient Pt loading of 0.5,0.3,0.1 mg/cm² and 0.4,0.3,0.2 mg/cm² with the uniform Pt loading. It is found that there is a slight improvement in the current density over the CL. The limiting current of the gradient Pt loading of 0.4,0.3,0.2 mg/cm² increased compared with the uniform Pt loadings. The current density slightly increased from 1.9187 to 1.9223 A/cm². However, the limiting current of the gradient Pt loading of 0.1,0.3,0.5 mg/cm² is lower than the uniform Pt loading case. This is simply because of the higher Pt loadings near the CL/MPL interface and lowers near the CL/mem interface. Thus, requires greater and quicker water and oxygen transport to reach the active site of the CL. Accordingly, an electrode with a high density of active sites at the CL/mem interface would be helpful for the cathode performance. It permits a bigger reactive water amount to quickly reach the catalyst active sites.

Figure 6. Effects of various cathode catalyst platinum loadings on the polarisation curve along with the platinum loading design of the cathode CL.
According to the polarisation curves in Figure 6., higher graded Pt loading yields better performance at the reaction site of CL, although it is not proportional. In contrast to the graded Pt loadings at the cathode side with the uniform Pt loading, the highest-graded Pt loading of 0.5,0.3,0.1 mg/cm² shows a faster voltage drop at higher current densities. This drop of voltage at high current densities is usually caused by the resistance of mass transport within the CL and ionomer domains. Thus, the lower graded Pt loading near the CL/MPL interface with the higher Pt loading near the CL/mem at high current density areas may be explained by presuming additional mass transport limitations caused by the catalyst layer properties such as thickness. At low voltage, the graded Pt loading case of 0.4,0.3,0.2 mg/cm² shows the highest current density, which might be due to the combination of fast reaction kinetics and effective mass transfer caused by the optimum Pt loading.

**Graded platinum loading in anode catalyst layer**

As discussed in the literature review, the water capacity of the anode is critical for reducing the flooding problem. To develop a high-performance anode in the AEMFC, it is necessary to improve its water management by controlling the catalytic material loading as well as the active material to carbon ratio. Thus, the performance of the AEMFC is studied in accordance with the amount of active material that is loaded into the anode. The AEMFC model with varied anode active material graded Pt loadings (0.1,0.2,0.3,0.4,0.5 mg/cm²) design is shown and investigated in Figure 7.

Despite the reduction in the cases of anode catalyst graded Pt loadings, the high-voltage performance of the AEMFC continued almost unchanged. This is explained by the improved kinetics of HOR at anode CL compared to the kinetics of ORR at cathode CL. The higher Pt loadings at the CL/MPL interface, the greater the mass performance (0.5,0.3,0.1 mg/cm²). However, the first case of (0.1,0.3,0.5 mg/cm²) exhibits the lowest limiting current compared to the uniform Pt loading (0.3 mg/cm²). This is due to more Pt loadings applied near the CL/mem interface. Thus, the performance of this case is insufficient to be used in real-world applications. Therefore, for the low-graded Pt loadings near the CL/mem interface, the graded Pt loading of (0.5, 0.3, 0.1 mg/cm²) is the most suitable. Due to the improved species reaction kinetics and effective mass transfer.
Figure 7. Effects of various anode catalyst platinum loadings on the polarisation curve along with the platinum and carbon mass loading design of the anode CL.

**Ionomer loading**

The polarisation curves of the AEMFC with varied ionomer loadings of 10%, 20%, 30%, 40%, and 50% for both anode and cathode are shown in Figure 8. The cell results indicate that the best performance is when the cell operated with an ionomer loading of 40% with the highest maximum power density of 0.91 W/cm² at an operating temperature of 60°C while the lowest performance is shown with the ionomer loading of 10% with the lowest maximum power density of 0.75 W/cm². Lower ionomer loading applied leads to an increase in the anion resistance and limits the catalyst efficiency owing to a lack of ion conductivity. However, higher ionomer loading applied such as 50% causes heavy surface pore-clogging in the CLs and a decrease in electrical conductivity. Consequently, the electrocatalysts at the CLs will be shielded. These two problems reduce the AEMFC performance. Therefore, it is proven that to create a continuous network, an appropriate content in alkaline ionomer must be selected. Resulting in better hydroxide transport and improving catalyst utilization.
Liquid water saturation distribution

The liquid water saturations in both electrodes, including CLs, MPLs and GDLs at different operating temperatures from 60 °C to 80 °C, are shown in Figure 9. It is obvious that the liquid water saturation has increased as the current density increases. This is because increasing the operating temperature increases the reaction rate of the electrochemical reactions. Therefore, the liquid water saturation is particularly high at the CLs in both electrodes, suggesting that there is more severe water flooding in these regions than elsewhere. The liquid water saturation gradient is higher in the anode electrode than in the cathode, particularly along the flow diffusion direction of species. This may explain by the reduction in water saturation pressure, which is temperature-dependent. Thus, the local saturation profile is shown in Figure 9, which is higher in the domain near the CLs/mem interface. Water vapour evaporation/condensation and membrane absorption/desorption are both used to produce liquid water \((S_{ec} + S_{ad})\), as shown in Table 2. The membrane absorption/desorption process is more critical than the water vapour evaporation/condensation in terms of yielding liquid water. Due to the higher effect of EOD, the membrane dissolved water first reaches saturation level. As a result, more liquid water is produced under the lands than under the inlets.
Figure 9. Liquid water saturation profiles at various operating temperatures of (a) 60, (b) 70, and (c) 80°C at a fixed cell voltage of 0.3 V with inlet anode and cathode gases with relative humidities of (RH=100%) and (RH=90%), respectively. The initial liquid water saturation is set to 0.05 for both electrodes.

Conclusion

A two-dimensional 2D two-phase mathematical model of anion exchange membrane fuel cell AEMFC has been developed to investigate the influences of three key electrode parameters (porosity, catalyst loading, and ionomer content) that are responsible for water production and transport as well as the performance of an AEMFC. According to modelling results, the present model showed a good match with the modelling and experimental data. The AEMFC performance was highly reliant on the parameters of each side of the electrode. The cathode optimal porosity gradient of $\varepsilon = 0.5, 0.6, 0.7$ performed the best performance due to the high GDL porosity applied at the GDL/inlet interface. However, the cathode graded GDL porosities of $\varepsilon = 0.7, 0.6, 0.5, \varepsilon = 0.8, 0.6, 0.4$, and $\varepsilon = 0.9, 0.6, 0.3$ exhibited the lowest mass power density. Furthermore, the allocation gradient of the oxygen mole fraction seemed to have a distinct effect on the cathode graded porosity. The greater the porosity graded at the GDL/MPL interface, the greater the improvement of the current density at the GDL/MPL interface. Resulting in more homogeneity of the current density distribution. Thus, it is clear that the cathode graded-GDL porosity can provide more and new options for enhancing the AEMFC performance and durability. On the other hand, the GDL’s graded porosity at the anode side has no apparent improvement in the current density. The anode graded-GDL porosity of $\varepsilon = 0.7, 0.6, 0.5$ performed the lowest limiting current density, however, its mass power density was much higher compared with $\varepsilon = 0.8, 0.6, 0.4$, and $\varepsilon = 0.9, 0.6, 0.3$, which was the lowest power density. The cathode CL graded Pt loading of 0.4,0.3,0.2 mg/cm² was slightly higher than the uniform Pt loading of 0.3 mg/cm², while the graded Pt loading of 0.1,0.3,0.5 mg/cm² indicated the lowest
limiting current achieved, most likely due to the higher graded Pt loadings applied near the inlet CL/MPL interface and lower near the outlet CL/mem interface. However, the anode CL graded Pt loadings of 0.5, 0.3, 0.1 mg/cm\(^2\) showed the highest gradient, while the 0.1, 0.3, 0.5 mg/cm\(^2\) showed the lowest performance. Furthermore, the results of ionomer loadings on the 2D AEMFC model revealed that the ionomer loading of 10\% performed the lowest limiting current and lowest peak power density. In addition, the ionomer loading of 40\% was the optimal cell performance achieved with the highest peak power density for both the anode and cathode sides. Increasing the operating temperature enhanced the overall performance of the AEMFC. This is because of increasing the water content of the membrane while decreasing the liquid water saturation. In addition, higher temperatures applied to the cathode enhance the ORR kinetics, which in turn improves the cell's overall performance at lower cell voltages and higher current densities.

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References


