



A mathematical model for performance of Proton exchange membrane fuel cell as a nonlinear voltage processes

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Abstract: A mathematical model for analysis of proton exchange membrane (PEM) fuel cell is proposed. The PEMFC is mainly composed of membrane, gas diffusion layer, bipolar plates, micro porous layer and catalyst layer. The proposed model is simulated in the MATLAB environment and studied the effects of thickness of membrane, gas diffusion layer and catalyst layer on the performance of the PEM fuel cell. Theoretical model shows that the performance of PEM fuel cell improves as thickness is scaling down towards nanoscale. The model has been validated with the experimental results trends and comparisons shows there is good agreement between the experimental data trends and the proposed model.

Keywords: Proton exchange membrane fuel cell, Membrane, Gas diffusion layer, Catalyst layer, Performance of the PEM fuel cell.

INTRODUCTION

The proton exchange membrane (PEM) fuel cell converts chemical energy into electricity using an electrochemical cell, could be used as efficient power sources, offering high power density and low environmental impact. The basic structure of PEM fuel cell we used as Ref. [11]. The study of nonlinear polarization curves, nonlinear voltage loss processes and power of proton exchange membrane PEM fuel cell is presented. A thinner membrane, thinner gas diffusion layer, thinner catalyst layer, higher temperature, higher gas flow rate and higher humidity are optimum operating conditions. Here emphasis on the thinner membrane, thinner gas diffusion layer and thinner catalyst layer that means if they are scaling Determination of irreversible losses of a proton exchange membrane (PEM) fuel cell is considered to be extremely essential to assess its performance in terms of fuel cell voltage, limiting current density and power density. There are several sources that contribute to irreversible losses in a PEM fuel cell during the operation. These losses are often called overpotentials or polarizations that could be originated primarily from three sources namely activation overpotential, ohmic overpotential and concentration overpotential. Activation overpotential is associated mainly with the slowness of electrochemical reaction in the fuel cell. Ohmic overpotential is associated with the resistance of the membrane to the flow of migrating ions during an electrochemical process. And the concentration overpotential occurs due to concentration gradients established as a result of rapid consumption of the reactant (oxygen) in the electrode during the electrochemical reaction.

PEM FUEL CELL NONLINEAR MODEL

A nonlinear model is presented in order to characterize the behaviour of a PEM fuel cell. The analysis was conducted considering the following assumptions:

1. The model is one dimensional, so the gases and reactions are considered uniformly distributed in the cell.

2. The electrochemical properties are evaluated at the average stack temperature, so temperature variations across the stack are neglected.
3. The water entering to the cathode and anode is only in vapor phase.
4. The water activity is uniform across the membrane and is in equilibrium with the water activity at the cathode and anode catalyst layer.
5. The effects of liquid water accumulation are not treated.
6. Gas properties of the mixture are modeled as ideal gas.
7. Two phases of water has been modeled according to liquid gas equilibrium.
8. Water transport through the membrane is considered to be a combination of diffusion and electro-osmotic drag.
9. Electrical and thermal conductivity of the bipolar plate (BP), gas diffusion layer (GDL), micro porous layer (MPL) and catalyst layer (CL) are considered to be constant with all the range of the used operating temperature .
10. Radiation heat transfer is neglected. Only heat conduction and convection have been modeled.
11. Reactants and products temperature are assumed to be at the same temperature as the component they flow through (BP, GDL, MPL, CL, membrane).

Thermodynamic Nonlinear potential

The Nernst equation allows one to determine the voltage of an electrochemical Cell. It is derivation from thermodynamic principles. Nernst equation is shown as general form as following [1].

$$E = E^0 - \frac{RT}{NF} \ln \left(\frac{[P_{H_2}][P_{O_2}]^{\frac{1}{2}}}{[P_{H_2O}]} \right) \quad (1)$$

Where R is the universal gas constant, T is the absolute temperature, N is the charge number of the electrode reaction and P_{H_2} , P_{O_2} , P_{H_2O} are the partial pressure of hydrogen, oxygen and water respectively. While E^0 is standard electrode Potential, The standard electrode potential is actual cell potentials different under standard

condition, the standard condition which mean here, there temperature at 298 °K and pressure about 1.0 bar. It calculated as shown by Maher [2].

$$E^0 = 1.229 - (0.83 \times 10^{-3}) \cdot (T - 298.15) + (4.3085 \times 10^{-5}) \cdot T \cdot ((\ln(P_{H_2}) + 0.5 \cdot \ln(P_{O_2})) \quad (2)$$

Where R is the universal gas constant and T is the fuel cell temperature P_{H_2} and P_{O_2} are hydrogen and oxygen partial pressures, respectively at the catalysis interface. The evaluation of the two partial pressures typically involves mass transfer calculations and normally requires averaging over a cell surface, to account for significant changes in the bulk phase partial pressures of the gaseous reactants [4]. This is the reason why in practical issues, correction factors must be considered to fit the experimental data to the governing equations. Apart from that, it is known that the cell operation voltage of a cell does not remain constant at different load conditions. This is because the current drainage from the cell produces different sorts of losses, which can be divided into three categories: activation, ohmic and concentration or diffusion.

Once of important output a source is the electrical energy. The actual cell potential V_{FC} is lower than the stranded cell potential E^0 because cell losses. Fuel cell has three types of losses are activation loss($V_{act.}$), concentration loss($V_{conc.}$) and ohmic loss(V_{ohmic}). The steady-state cell voltage is calculated by subtracting these catalytic and resistive losses from the reversible electrochemical cell voltage which can be calculated as following:

$$V_{FC} = E^0 - V_{act.} - V_{conc.} - V_{ohmic}$$

Critically obtained voltage losses in the fuel cell and the resulting polarization curve are shown below (fig.1).

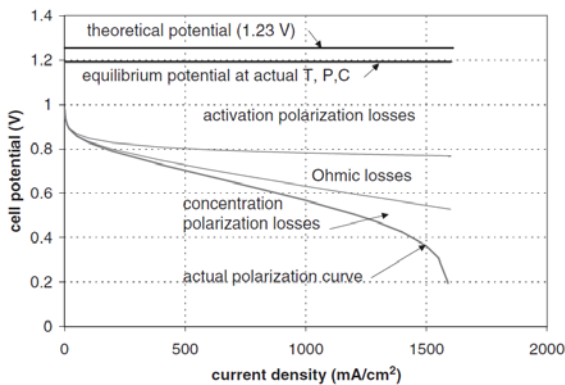


Figure: 1- Voltage losses in the fuel cell and the resulting polarization curve Ref. [11]

1. Activation losses

A certain proportion of energy is needed to start the chemical reaction. This phenomenon produces a non-linear voltage drop called activation polarization. These losses occur on both anode and cathode catalysts. The activation losses occur because the cell needs energy to produce the electrons transfer, to break and form new chemicals bonds, both in the anode and cathode [5]. This produces a voltage drop in both electrodes of the cell. The activation losses cause the voltage reduction in the low current regions of MEA operation. The steep slope of the initial portion of the

Using standard thermodynamic values of the entropy change, the open circuit voltage/reversible cell potential of a single cell (Nerst voltage) can be represented by the following expression [3]:-

curve is an indication of the activation polarization. As the current increases, the slope of the curve becomes less steep. This is a result of the activation polarization becoming less significant due to the increase in other polarization phenomena, specially concentration and ohmic effects [5]. The relationship between the activation voltage and the current density is often described through the Tafel equation[22]:

$$V_{act.} = A \ln\left(\frac{i}{i_0}\right) \quad (4)$$

where A is higher for slow electrochemical reactions, i the current density and the constant i_0 is higher if the reaction is faster. The current density i_0 can be considered as the current density at which the overvoltage begins to move from zero. It is important to stress that the Tafel equation only holds true when $i > i_0$. This current density is usually called the exchange current density and its typical values are in the range 10^{-2} – 10^{-8} A [6]. Although the Tafel was originally deduced from experimental results, it also has a theoretical basis. It can be shown that for a hydrogen fuel cell with two electrons transferred per mole, the constant A is given by[22]:

$$A = \frac{RT}{2\alpha F} \quad (5)$$

The constant α is called the charge transfer coefficient and is the portion of the electrical energy applied that is harnessed in changing the rate of an electrochemical reaction. Its vale depends on the reaction involved and the material of the electrode, but it must be in the range 0–1 [6]. The activation losses can also be expressed simply as the Tafel equation[7]:

$$V_{act.} = a + b \ln(i) \quad (6)$$

$$\text{Where } a = -\frac{RT}{\alpha F} \ln(i_0) \quad \text{and } b = -\frac{RT}{\alpha F}$$

The equation for the anode and cathode activation overpotential can be represented by [22]:

$$V_{act_anode} + V_{act_cathode} = \left[\frac{RT}{n\alpha F} \ln\left(\frac{i}{i_0}\right)\right]_{anode} + \left[\frac{RT}{n\alpha F} \ln\left(\frac{i}{i_0}\right)\right]_{cathode} \quad (7)$$

where n is the number of exchange protons per mole of reactant, F is Faraday’s constant, and α is the charge transfer coefficient used to describe the amount of electrical energy applied to change the rate of the electrochemical reaction. The exchange current density, i_0 , is the electrode activity for a particular reaction at equilibrium. In PEM fuel cells, the anode i_0 for hydrogen oxidation is very high compared to the cathode i_0 for oxygen reduction; therefore, the cathode contribution to this polarization is often neglected. Intuitively, it seems like the activation polarization should

increase linearly with temperature based upon equation (7), but the purpose of increasing temperature is to decrease activation polarization. By increasing the temperature would cause a voltage drop within the activation polarization region.

In new model the expression for activation potential is as follows -

$$V_{act.} = \frac{RT}{2\alpha_a F} * \ln\left(\frac{i}{i_a}\right) + \frac{RT}{4\alpha_c F} * \ln\left(\frac{i}{i_c}\right) \quad (8)$$

Where R is the universal gas constant and T is the fuel cell temperature α_a, α_c are anode and cathode transfer coefficient, i_a, i_c are anode and cathode current density.

2 Ohmic losses

PEM ohmic losses are usually due to both, the resistance that presents the membrane to the protons transfer and the electrical resistance of electrodes and collectors. The voltage drop by ohmic losses remains proportional to the current density drained from the cell:

$$V_{ohmic} = iR_{ohmic} \quad (9)$$

where R_{ohmic} is the internal resistance of the cell, which has a strong dependence on the membrane humidity and on the cell temperature. Many studies [8,9] show that this resistance is a function of the conductivity of the membrane (σ_m) and the thickness of the dry membrane (t_m). In turn, the conductivity of perfluorosulphonic acid ionomer membranes is a strong function of water content (λ_m), defined as the number of water molecules per sulfonate group in the ionomer.

Ohmic overpotential depends on the electrical conductivity and thickness of every material that form the fuel cell as the bipolar plates, GDLs, CLs, MPLs and the membrane. Furthermore, the contact resistance between the GDL and the bipolar plate has been included in the ohmic overpotential due to the impact that it has over the performance of the PEM fuel cell [10]. The contact resistance has been estimated according to a well defined technique [11]. The expression to calculate the ohmic overpotential is shown in [12, 24].

$$V_{ohmic} = i \left(2 \frac{t_{BP}}{\sigma_{BP}} + 2 \frac{t_g}{\sigma_g} + 2 \frac{t_c}{\sigma_c} + 2 \frac{t_{MPL}}{\sigma_{MPL}} + \frac{t_m}{\sigma_m} + 2R_{contact} \right) \quad (10)$$

Where t_i and σ_i represent the thickness and the electrical conductivity of the BPL (bipolar plate), bipolar plate (BP),

In new model the expression for ohmic potential is as follows-

$$V_{ohmic} = i [R_{electronic} + R_{protonic}] = I \left[R_e + \left(\frac{t_m}{\sigma_m A_m} + 2 \frac{t_g}{\sigma_g A_g} + 2 \frac{t_c}{\sigma_c A_c} \right) \right] = i \left[R_e + \left(\frac{t_m}{\sigma_m} + \frac{t_g}{\sigma_g} + \frac{t_c}{\sigma_c} \right) \right] \quad (14)$$

Where t_i and σ_i represent the thickness and the electrical conductivity of gas diffusion layer (g), ,catalyst layer (c), membrane (m) , $R_{electronic}$ represents the resistance due to electron flow in the PEMFC and $R_{protonic}$ represents the resistance due to electron flow in the PEFMC.

gas diffusion layer (g), micro porous layer (MPL) and catalyst layer (c), membrane (m) and $R_{contact}$ represents the contact resistance between the GDL and the bipolar plate .The electrical conductivity of the BP, GDLs, MPLs and CLs was obtained from the technical datasheet of each material [13 to 15]. However, the membrane proton conductivity (Nafion 117) depends strongly on the water content inside the membrane. Therefore, depending on the cell operating conditions, the membrane protonic conductivity will be different. The expression to calculate the membrane protonic conductivity as a function of the water content is shown below. The expression is widely used in PEM fuel cell models and it was developed by Springer [16].

$$\sigma_m = (0.005193\lambda_m - 0.00326)\exp\left(\frac{1268}{303 - T}\right)$$

Where λ_m is the water content inside the membrane and T is the operating temperature (K). The parameters can vary for different membranes, but the general behavior of the model was verified in different operating conditions.

Every material has an intrinsic resistance to charge flow. The material's natural resistance to charge flow causes ohmic polarization, which results in a loss in cell voltage. All fuel cell components contribute to the total electrical resistance in the fuel cell, including the electrolyte, the catalyst layer, the gas diffusion layer, bipolar plates, interface contacts, and terminal connections. The reduction in voltage is called "ohmic loss," and includes the electronic ($R_{elect.}$) and ionic (R_{ionic}) contributions to fuel cell resistance. This can be written as [7]:

$$V_{ohmic} = i(R_{elect.} + R_{ionic}) \quad (12)$$

R_{ionic} dominates the reaction because ionic transport is more difficult than electronic charge transport. R_{ionic} represents the ionic resistance of the electrolyte, and $R_{elect.}$ includes the total electrical resistance of all other conductive components, including the bipolar plates, cell interconnects, and contacts. The material's ability to support the flow of charge through the material is its conductivity. Resistance is characteristic of the size, shape, and properties of the material, as expressed

$$R = \frac{t_{cond}}{\sigma A_{cond}} \quad (13)$$

where t_{cond} is the length of the conductor, A_{cond} is the cross-sectional area of the conductor, and σ is the electrical conductivity .

3. Concentration losses

Finally, there is another kind of losses called concentration losses that occur when the reactants are rapidly consumed at the electrodes by the electrochemical reactions, and then concentration gradients are established. For instance, if the oxygen at the cathode of a fuel cell is supplied in the form of

air, it is self-evident that during fuel cell operation there will be a slight reduction in the concentration of the oxygen in the cathode region, as the oxygen is extracted [19]. Due to these losses, there is a sharp drop in voltage when the drained current is too high. However, because of non uniform conditions over the porous electrode area, the limiting current is almost never experienced in practical fuel cells [17]. To approximate this phenomenon, an empirical equation better describes the concentration losses [18, 22]:-

$$V_{conc} = m \exp(n - 1) \tag{15}$$

where m and n are empirical coefficients with typical values close to 3×10^{-5} V and $8 \text{ cm}^2/\text{A}$, respectively [19].

The concentration overpotential is caused by mass transport limitation of the reactants to the catalyst site, associated with electrochemical processes. Mass transport in fuel cell is

$$C_3 = 2, i_{max} = 2.2 \text{ and If } \frac{P_{O_2}}{0.1173} + P_{H_2O} < 2 \text{ atm (17)}$$

$$C_2 = (7.16 \times 10^{-4} - 0.622) \left(\frac{P_{O_2}}{0.1173} + P_{H_2O} \right) + (-1.45 \times 10^{-3}T + 1.68)$$

Else

$$C_2 = (8.66 \times 10^{-5}T - 0.068) \left(\frac{P_{O_2}}{0.1173} + P_{H_2O} \right) + (-1.6 \times 10^{-4}T + 0.54)$$

In new model the expression for concentration losses is as follows [23]-

$$V_{conc.} = \frac{RT}{2F} \ln \left(\frac{P_{H_2}}{P_a} \right) + \frac{RT}{4F} \ln \left(\frac{P_{O_2}}{P_c} \right) \tag{16}$$

Where R is the universal gas constant and T is the fuel cell temperature P_{H_2} and P_{O_2} are hydrogen and oxygen partial pressures, respectively at the catalysis interface. Here P_a and

mainly due to convection in channels and diffusion in electrodes. As soon as the fuel cell begins to produce current, the electrochemical reactions lead to depletion of reactants and accumulation of products at the catalyst layer. An empirical equation of the concentration overpotential is given by Guzzella [20]

$$V_{conc} = i \left(C_2 \frac{i}{i_{max.}} \right)^{C_3} \tag{16}$$

Where C_2 , C_3 and $i_{max.}$ are constants that depend on temperature and reactant partial pressure.

The coefficients in the equation are determined using nonlinear regression with polarization data from automotive propulsion PEM fuel cell stack as given by Pukrushpan *et al.* [21]

P_c are anode and cathode gas pressure. The first part of the eq. due to hydrogen flow at anode and second due to oxygen flow at cathode is combined effect resulting concentration overpotential.

Hence in new improved nonlinear voltage model the complete expression of PEMFC potential is as follows:-

$$V_{FC} = E^0 - \left[\frac{RT}{2\alpha_a F} \ln \left(\frac{i}{i_a} \right) + \frac{RT}{4\alpha_c F} \ln \left(\frac{i}{i_c} \right) \right] - i \left[R_e + \left(\frac{t_m}{\sigma_m} + 2 \frac{t_g}{\sigma_g} + 2 \frac{t_c}{\sigma_c} \right) \right] - \left[\frac{RT}{2F} \ln \left(\frac{P_{H_2}}{P_a} \right) + \frac{RT}{4F} \ln \left(\frac{P_{O_2}}{P_c} \right) \right] \tag{17}$$

Power Density

Power density is the product of output voltage and current density and hence increases with the current density. After reaching the maximum value, power density starts decreasing since the rate of decrease in voltage is more than the rate of increase in current due to flooding of water at the catalyst sites and channels. The gross output power of the single cell is found as:

$$W = iV_{FC} \tag{18}$$

And power is product of power density and area of cell A:-

$$P = WA = iV_{FC}A \tag{19}$$

RESULTS AND DISCUSSIONS

The new improved model parameters for MATLAB simulation are as Ref. [2] and shown in the following tables:

Table 1-Effect of thickness (meter) of gas diffusion layer

S. No.	Thickness of catalyst layer	Thickness of membrane	Thickness of gas diffusion layer
1	56×10^{-6}	230×10^{-6}	520×10^{-6}
2	56×10^{-6}	230×10^{-6}	320×10^{-7}
3	56×10^{-6}	230×10^{-6}	120×10^{-9}

Table 2-Effect of thickness (meter) of catalyst layer

S. No.	Thickness of catalyst layer	Thickness of membrane	Thickness of gas diffusion layer
1	56×10^{-6}	230×10^{-6}	520×10^{-6}
2	26×10^{-7}	230×10^{-6}	520×10^{-6}
3	06×10^{-9}	230×10^{-6}	520×10^{-6}

Table 3-Effect of thickness (meter) of membrane

S. No.	Thickness of catalyst layer	Thickness of membrane	Thickness of gas diffusion layer
1	56×10^{-6}	230×10^{-6}	520×10^{-6}
2	56×10^{-6}	130×10^{-7}	520×10^{-6}
3	56×10^{-6}	030×10^{-9}	520×10^{-6}

Using these parameters the effects are shown below:-

1. Effect of nanoscale membrane thickness on PEM fuel cell Performance

The effect of nanoscale membrane thickness has been studied on the fuel cell performance i.e. on polarization curve and power curve. The new improved model equation shows that the major effect of thickness variation would be appeared at ohmic losses. Physically the ohmic loss is depends on the membrane thickness, current density, cross

section area, electronic conductivity and ionic conductivity. The scaling down of membrane thickness of PEM fuel cell leads to decreasing of ohmic losses .Consequently increasing the cell output voltage and power. It was found that the effect of membrane thickness on polarization curve and power has been cleared from the following results in fig 2a and 2b where we show three curves from micro to nanoscale that both are improving by scaling down the thickness of membrane.

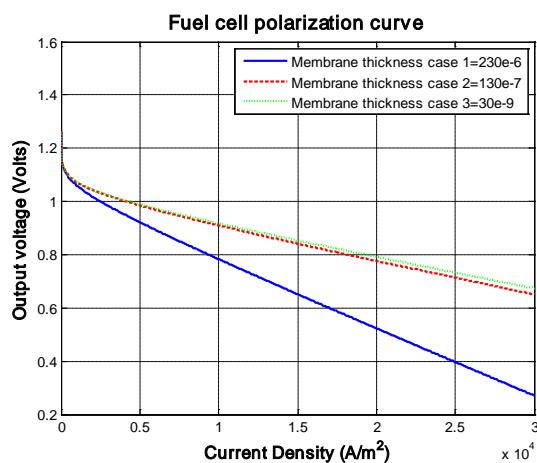


Fig. 2 (a) Fuel cell polarization curve for different membrane thickness

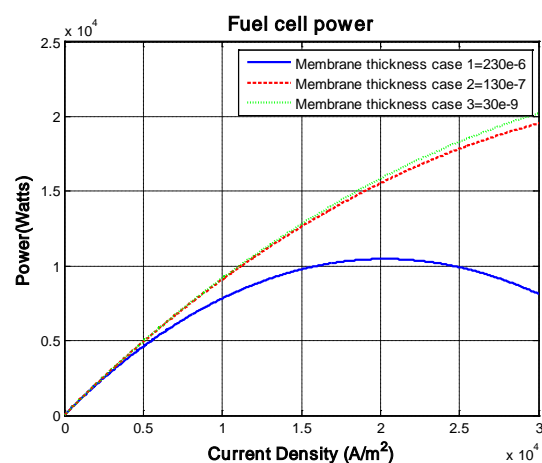


Fig. 2 (b) Fuel cell power curve for different membrane thickness

2. Effect of nanoscale gas diffusion layer thickness on PEM fuel cell Performance

Nanoscale gas diffusion layer (GDL) is also affected the fuel cell performance. i.e. on polarization curve and power curve. The new improved model equation shows that the major effect of thickness variation would be appeared at ohmic losses .The major effect of gas diffusion layer has been appeared in ohmic losses. Because, ohmic losses is depends on gas diffusion layer. Fig. 3a and 3b despite the result of variation of gas diffusion layer thickness on polarization curve and power has been cleared where we show three curves from micro to nanoscale that both are

improving by scaling down the thickness of membrane. It is clear from the divergence in fig 3a and 3b at ohmic losses region on polarization. Concentration losses have been affected by variation in gas diffusion layer, because concentration losses are losses of proton transport which depends on gas diffusion layer. A thinner GDL results in a larger oxygen transfer from the gas channel to the catalyst layer, and thus a larger potential is generated. The effect of the GDL thickness on the fuel cell performance is again mostly on the mass transport, as the ohmic losses of the electrons inside the GDL can be neglected due to the high conductivity of the carbon fiber paper.

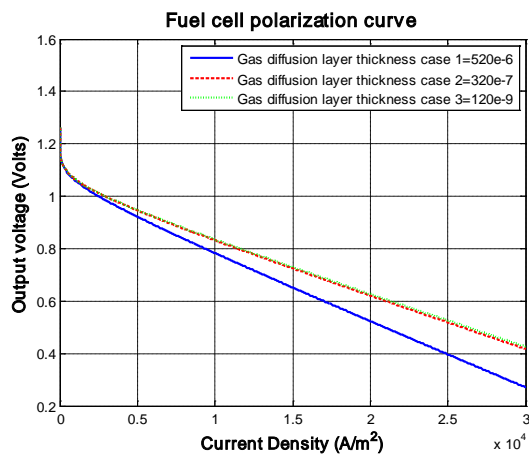


Fig. 3 (a) Fuel cell polarization curve for different gas diffusion layer thickness

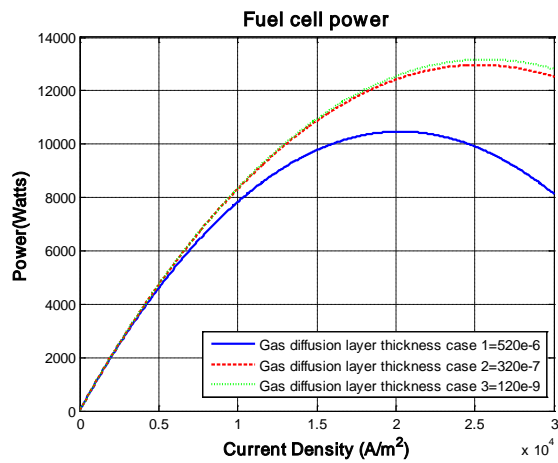


Fig. 3 (b) Fuel cell power curve for different gas diffusion layer thickness

3. Effect of nanoscale Catalysts layer thickness on PEM fuel cell Performance

The effect of nanoscale catalyst layer thickness on performance of the fuel cell i.e. on polarization curve and power curve are shown in figs.4a and 4b. The new improved model equation shows that the major effect of thickness variation would be appeared at ohmic losses. It is observed from Figs. 4a and 4b that the performance is increases by an decrease with catalyst layer thickness. It is also observed that the performance is superior for nanoscale compared to

the microscale. This is due to the fact that the reactant gases do not have as many reaction sites in the nanoscale as in the microscale. The performance is decreases beyond certain thickness due to the catalyst layer's electric and ionic resistance and to the increase in mass transport limitation. Due to increased mass transport resistance in higher catalyst thickness, at higher current densities, the reactant gases are unable to reach all the reaction sites and the catalyst at the inner half of the catalyst layer (i.e., adjacent to the membrane) largely remain underutilized.

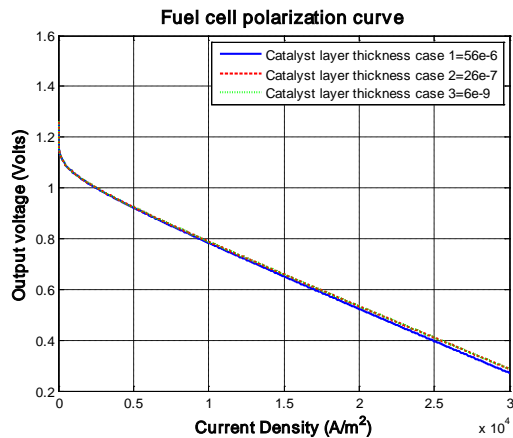


Fig. 4 (a) Fuel cell polarization curve for different Catalysts layer thickness

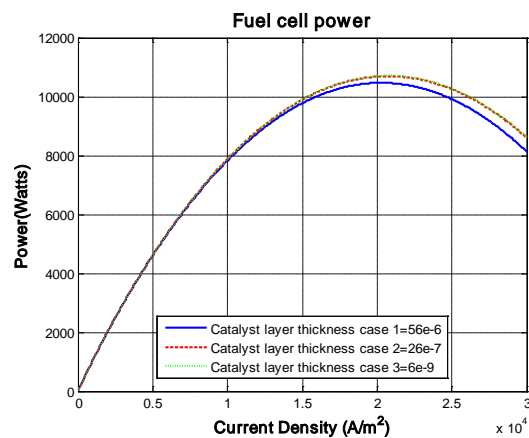


Fig. 4 (b) Fuel cell power curve for different Catalysts layer thickness

CONCLUSIONS

The theoretical model developed for single PEM fuel cell simulates effectively the various conditions that exist in a real fuel cell, which is evident from the comparison of the theoretical results with experimental data trends. The fuel cell performance is improved with a decrease in thickness from microscale to nanoscale. This is due to increase of gas diffusivity, exchange current density and membrane conductivity at nanoscale. A thinner GDL results in a larger

oxygen transfer from the gas channel to the catalyst layer, and thus a larger potential is generated. As the catalyst layer thickness decreases there is an improvement in the performance was observed. However the performance is decreases beyond certain thickness due to in catalyst layer's electric and ionic resistance and to the increase in mass transport limitation. Due to decreasing membrane thickness, the voltage of cell and its power were increasing. Thinner membranes will enhance back diffusion from cathode to anode preventing anode dehydration at higher current

densities. Hence, with the decrease in the thickness of the membrane there would be a decrease in ohmic over potential resulting in the increase of the output voltage. To reduce the value of the ohmic resistance it is necessary to use electrodes with extremely high conductivity, or reducing the distance that the electrons must travel resistance is proportional to distance. Another way to reduce the resistance is to use well-designed bipolar plates, which have high conductivities and short lengths.

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