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2D Analytical Model for Direct Ethanol Fuel Cell Performance Prediction

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ABSTRACT

Analytical models provide useful information for researchers to study fuel cell function. In this paper, it's aimed to present a 2D analytical model for direct ethanol fuel cell (DEFC) performance. The model included equations inside diffusion layer, catalyst layer, and Ethanol cross-over through membrane, which all have been solved. Analytical model has been validated by some experimental trials. The results showed that there is proper agreement between experimental and analytical curves. Furthermore, by increasing current density, cathodic over potential will remain zero but anodic over potential will increase up to certain value. The model showed that Ethanol concentration changes almost linearly inside anode channel.

Keywords*: *Analytical Model, Polarization Curve, Voltage, Current Density

1. Introduction

Fuel cells are new power sources which produce electricity without any noise or environmental contamination. Fuel cells are used nowadays for rural, military portable and station applications [1]. Direct methanol fuel cells are one of prominent fuel cells which have high current density. Unfortunately direct methanol fuel cells have some technical problems such as methanol toxicness, expensive Pt-based catalysts, and high methanol cross over, thus, there are trends to substitute methanol with other fuels. Ethanol, acid acetic and acetaldehyde are proposed alternatives [2-5]. Recent ethanol fuel cell activities have been more experimental to date and only 1D analytical model has been proposed for DEFC so far. This model has been proposed by G. Andreadis and his colleagues by considering over potential changes inside catalyst layer [6-9]. In this paper, we try to present a 2D fully analytical model with simple consideration. With regard to Fuel cell coordinates, which have been illustrated in **Figure 1**, following assumptions have been made

1) Fluid flows in the steady state.

2) Fuel cell temperature is constant in the active area and chemical reaction takes place under constant temperature.

3) Reactants diffusion and transportation in catalyst layer along y direction is not considered.

4) Due to low diffusion layer thickness before its size long channel length, reactants concentration change across diffusion layer is ignored.

5) Reactants diffusion inside diffusion layer along *y* direction has been ignored because of low diffusion layer thickness before its length.

6) Because the thickness of membrane is so smaller than its thickness, reactants diffusion along *y* direction is ignored.

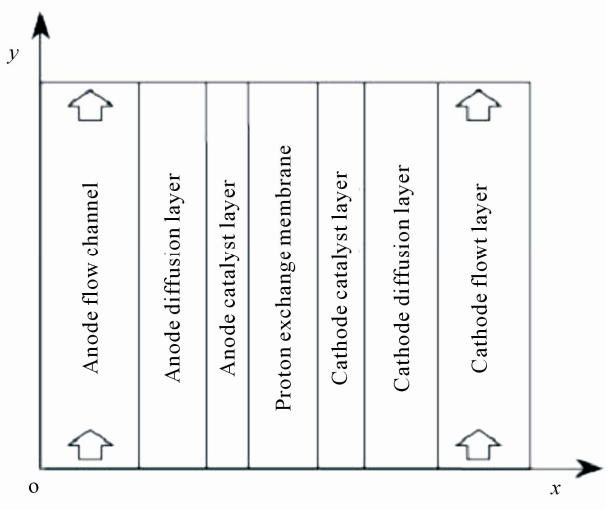

Figure 1. Coordinates of fuel cell for presented model.

7) Due to smaller depth of ethanol and water transportation channel than its length, reactants concentration change along *x* direction inside the channel has been neglected.

8) Pressure drop inside channel is neglected.

9) Fluid flows at constant velocity inside the channels.

2. Basic Equations

Anode and cathode overall reaction is as follows

$$\begin{aligned} &C_2H_5OH + 3H_2O \longrightarrow 12e^- + 12H^+ + 2CO_2 \\ &12e^- + 12H^+ + 3O_2 \longrightarrow 6H_2O \\ &+ \text{----------} \\ &C_2H_5OH + 3O_2 \longrightarrow 2CO_2 + 3H_2O \end{aligned} \quad (1)$$

Ethanol concentration inside anodic channel, c_{ah}^{ETOH} could be mentioned as below

$$\delta_{ah}\upsilon_{ah}\frac{\mathrm{d}c_{ah}^{ETOH}}{\mathrm{d}y} = -N_{ad} \quad (2)$$

Whereas N_{ad} is mass flux from anodic channel through diffusion layer. Based on Fick's law, we can write the following equation for mass flux, whereas D_{ad}^{ETOH} is diffusion coefficient of ethanol through diffusion layer

$$N_{ad} = -D_{ad}^{ETOH}\frac{\partial c_{ad}^{ETOH}}{\partial x} \quad (3)$$

It can be noticed that for 12 M electron production, 1 M ethanol is consumed. Furthermore ethanol crossover lead to a part of ethanol permeate through membrane, so

$$N_{ad} = \frac{1}{12}J_a + N_m \quad (4)$$

Current density can be written as below

$$J_a = J_{ref}^{ETOH}\left(\frac{c_{ac}^{ETOH}}{c_{ref}^{ETOH}}\right)^{\gamma_a}\exp\left(\frac{\alpha_a F}{RT}\eta_a\right) \quad (5)$$

In which η_a is anodic over potential and N_m is crossover from membrane as below

$$N_m = -D_m^{ETOH}\frac{\partial c_m^{ETOH}}{\partial x} + n_d{}^{ETOH}\frac{J}{F} \quad (6)$$

The equations for cathode are similar to those for the anode, so

$$N_{cd} = \frac{1}{4}J_c + 3N_m \quad (7)$$

J_c, current density is

$$J_c = J_{ref}^{O_2}\left(\frac{c_{cc}^{O_2}}{c_{ref}^{O_2}}\right)^{\gamma_c}\exp\left(\frac{\alpha_c F}{RT}\eta_c\right) \quad (8)$$

That η_c is cathodic over potential. Equations (2) and (3) can be written for cathode, thus

$$N_{cd} = D_{cd}^{O_2}\frac{\partial c_{cd}^{O_2}}{\partial x} \quad (9)$$

For oxygen concentration variation inside channel

$$\delta_{ch}\upsilon_{ah}\frac{\mathrm{d}c_{ch}^{O_2}}{\mathrm{d}y} = -N_{cd} \quad (10)$$

At last, for fuel cell voltage and current density, following equations is determined

$$V = E_{Cell} - \eta_c - \eta_a - \bar{J}\frac{\delta_m}{\sigma_m} - \bar{J}R_{contact} \quad (11)$$

$$E_{Cell} = E_{Cell}{}^0 + \left(T - T_0\right)\left(\frac{\partial E_{Cell}}{\partial T}\right) - \Delta n\frac{RT}{\nu F}\ln\left(\frac{P}{P_0}\right) \quad (12)$$

That $\bar{J}$ is current density, δ_m membrane thickness, σ_m membrane conductivity, E_{Cell}^0 ideal electromotive force, and $\frac{\partial E_{Cell}}{\partial T}$ is electromotive difference rate. Δn (Difference of exchanged gas moles between two sides of reaction in (1)) and ν (number of exchanged electrons) are constants which are –1 and 12 for (DEFC) respectively. Other symbols are listed in **Table 1** or Nomenclature.

3. Analytical Solution

In this solution, ethanol concentration in cathode layer in neglected (zero) and ethanol is linearly distributed, thus

$$N_m = D_m^{ETOH}\frac{c_{ac}^{ETOH}}{\delta_m} + n_d{}^{ETOH}\frac{J}{F} \quad (13)$$

c_{ac}^{ETOH} and J are substituted with $\bar{c}_{ac}^{ETOH}$, $\bar{J}$ respectively, and then will be found

$$N_m = D_m^{ETOH}\frac{\bar{c}_{ac}^{ETOH}}{\delta_m} + n_d{}^{ETOH}\frac{\bar{J}}{F} \quad (14)$$

By substituting Equation (7) in Equation (9) and integrating from 0 to δ_{cd}, oxygen concentration in catalyst layer is

$$c_{cc}^{o_2} = c_{ch}^{o_2} - \frac{1}{4}\frac{\delta_{cd}}{D_{cd}^{o_2}}J_c - 3\frac{\delta_{cd}}{D_{cd}^{o_2}}N_m \quad (15)$$

By assuming $\gamma_c = 1$ and substituting Equation (8) in Equation (15)

$$c_{cc}^{o_2} = \frac{1}{1+\xi_1}c_{ch}^{o_2} - \frac{3}{\left(1+\xi_1\right)}\frac{\delta_{cd}}{D_{cd}^{o_2}}N_m \quad (16)$$

Whereas ξ_1 is defined as follows

Table 1. Coefficients for analytical model.

Parameter/Symbol(unit)	Value	ref
Temperature of the fuel cell / $T(K)$	363	exp
Pressure of the air in the cathode / $P(Pa)$	0.5	exp
Length of anode channel / $l_{ah}(cm)$	3	[10]
Length of cathode channel / $l_{ch}(cm)$	3	[10]
Number of anode channels / χ_{ah}	2	exp
Number of cathode channels / χ_{ch}	5	exp
Width of anode channels / $b_{ah}(cm)$	0.1	exp
Width of cathode channels / $b_{ch}(cm)$	0.1	exp
Depth of anode channels / $\delta_{ah}(cm)$	0.1	exp
Depth of cathode channels / $\delta_{ch}(cm)$	0.1	exp
nlet flow rate of anode / $\dot{V}_{ah}(cm^3\,\mathrm{min}^{-1})$	50	exp
IInlet flow rate of cathode / $\dot{V}_{ch}(cm^3\,\mathrm{min}^{-1})$	200	exp
Velocity of fluid in anode channel / $\upsilon_{ah}(cm\,s^{-1})$	$\upsilon_{ah}=\frac{V_{ah}}{60\chi_{ah}b_{ah}\delta_{ah}}$	–
Velocity of fluid in cathode channel / $\upsilon_{ch}(cm\,s^{-1})$	$\upsilon_{ch}=\frac{V_{ch}}{60\chi_{ch}b_{ch}\delta_{ch}}$	–
Ethanol concentration at anode inlet / $C_{ahin}^{ETOH}(mol\,cm^{-3})$	0.125 M	exp
Oxygen concentration at cathode inlet / $C_{chin}^{ETOH}(mol\,cm^{-3})$	$C_{chin}^{o_2}=0.21\times10^{-6}\times\frac{P}{RT}$	[10]
Thickness of anode diffusion layer / $\delta_{ad}(cm)$	0.03	exp
Thickness of cathode diffusion layer / $\delta_{cd}(cm)$	0.03	exp
Thickness of anode catalyst layer / $\delta_{ac}(cm)$	0.005	Assumed
Thickness of cathode catalyst layer / $\delta_{cc}(cm)$	0.003	Assumed
conductivity of membrane($S\,cm^{-1}$)	$\sigma_m=0.073\times\exp\left(1268\left(\frac{1}{298}-\frac{1}{T}\right)\right)$	[11]
Thickness of membrane / $\delta_m(cm)$	0.0175	exp

$$\xi_1=\frac{1}{4F}\frac{\delta_{cd}}{D_{cd}^{o_2}}\left(\frac{J_{ref}^{o_2}}{c_{ref}^{o_2}}\right)\exp\left(\frac{\alpha_c F}{RT}\eta_c\right) \tag{17}$$

With regard to Equations (7), (8), (10) and (16) the below equation is acquired

$$\frac{dc_{ch}^{o_2}}{dy}=\frac{\xi_2}{l_{ch}}c_{ch}^{o_2}+\frac{3}{\xi_1}\frac{\delta_{cd}}{D_{cd}^{o_2}}N_m \tag{18}$$

whereas ξ_2 is

$$\xi_2=-\frac{l_{ch}}{\delta_{ch}\upsilon_{ch}}\frac{1}{4F(1+\xi)}\frac{J_{ref}^{o_2}}{c_{ref}^{o_2}}\exp\left(\frac{\alpha_c F}{RT}\eta_c\right) \tag{19}$$

At last, by integrating Equation (18) and assuming $c_{ch}^{o_2}(y=0)=c_{chin}^{o_2}$, oxygen concentration inside anode channel will be found

$$c_{ch}^{o_2}=\left(c_{chin}^{o_2}+\frac{3}{\xi_1}\frac{\delta_{cd}}{D_{cd}^{o_2}}N_m\right)\exp\left(\xi_2\frac{y}{l_{ch}}\right)-\frac{3}{\xi_1}\frac{\delta_{cd}}{D_{cd}^{o_2}}N_m \quad (20)$$

Concentration of oxygen in the catalyst layer can be determined by substituting (20) into (16)

$$\begin{aligned}c_{cc}^{o_2}&=\frac{1}{1+\xi_1}\left(c_{ahin}^{o_2}+\frac{3}{\xi_1}\frac{\delta_{cd}}{D_{cd}^{o_2}}N_m\right)\\&\times\exp\left(\xi_2\frac{y}{l_{ch}}\right)-\frac{3}{\xi_1}\frac{\delta_{ad}}{D_{cd}^{o_2}}N_m\end{aligned} \quad (21)$$

Substituting Equation (21) into (8) and integrating

$$\begin{aligned}\bar{J}&=\frac{1}{l_{ch}}\int_0^{l_{ch}}J_c\mathrm{d}y=\left[\frac{J_{ref}^{o_2}}{c_{ref}^{o_2}}\exp\left(\frac{\alpha_c F}{RT}\eta_c\right)\right]\\&\left[\left(c_{chin}^{o_2}+\frac{3}{\xi_1}\frac{\delta_{cd}}{D_{cd}^{o_2}}N_m\right)\times\frac{\exp(\xi_2)-1}{(1+\xi_1)\xi_2}-\frac{3}{\xi_1}\frac{\delta_{cd}}{D_{cd}^{o_2}}N_m\right]\end{aligned} \quad (22)$$

Such trend can be implemented for anode side, thus

Ethanol concentration variation inside anode channel could be written as follows

$$\begin{aligned}c_{ah}^{ETOH}&=\left(c_{ahin}^{ETOH}+\frac{1}{\xi_3}\frac{\delta_{ad}}{D_{ad}^{ETOH}}N_m\right)\exp\left(\xi_4\frac{y}{l_{ah}}\right)\\&-\frac{1}{\xi_3}\frac{\delta_{ad}}{D_{ad}^{ETOH}}N_m\end{aligned} \quad (23)$$

Ethanol concentration distribution in the catalyst layer is

$$\begin{aligned}c_{ac}^{ETOH}&=\frac{1}{1+\xi_3}\left(c_{ahin}^{ETOH}+\frac{1}{\xi_3}\frac{\delta_{ad}}{D_{ad}^{ETOH}}N_m\right)\times\exp\left(\xi_4\frac{y}{l_{ah}}\right)\\&-\frac{1}{\xi_3}\frac{\delta_{ad}}{D_{ad}^{ETOH}}N_m\end{aligned} \quad (24)$$

Ethanol concentration average in the catalyst layer, by integrating (24) through 0 to l_{ah} is

$$\begin{aligned}\bar{c}_{ac}^{ETOH}&=\frac{\exp(\xi_4)-1}{(1+\xi_3)\xi_4\xi_5}c_{ahin}^{ETOH}\\&+\frac{1}{\xi_3\xi_5}\left[\frac{\exp(\xi_4)-1}{(1+\xi_3)\xi_4}-1\right]\times\frac{\delta_{ad}}{D_{ad}^{ETOH}}\frac{n_d^{ETOH}}{F}\bar{J}\end{aligned} \quad (25)$$

Average current density

$$\begin{aligned}\bar{J}&=\frac{1}{l_{ah}}\int_0^{l_{ah}}J_a dy=\left[\frac{J_{ref}^{ETOH}}{c_{ref}^{ETOH}}\exp\left(\frac{\alpha_a F}{RT}\eta_a\right)\right]\\&\times\left[\frac{1}{1+\xi_3}\left(c_{chin}^{ETOH}+\frac{3}{2\xi_1}\frac{\delta_{ad}}{D_{ad}^{o_2}}N_m\right)\right.\\&\left.\times\frac{\exp(\xi_4)-1}{\xi_4}-\frac{1}{\xi_3}\frac{\delta_{cd}}{D_{cd}^{o_2}}N_m\right]\end{aligned} \quad (26)$$

ξ_3, ξ_4, ξ_5 are variants as below

$$\xi_3=\frac{1}{4F}\frac{\delta_{ad}}{D_{ad}^{ETOH}}\left(\frac{J_{ref}^{ETOH}}{c_{ref}^{ETOH}}\right)\exp\left(\frac{\alpha_a F}{RT}\eta_a\right) \quad (27)$$

$$\xi_4=-\frac{l_{ah}}{\delta_{ah}\upsilon_{ah}}\frac{\delta_{ad}}{D_{ad}^{ETOH}}\frac{\xi_3}{1+\xi_3} \quad (28)$$

$$\xi_5=1-\frac{1}{\xi_3}\left[\frac{\exp(\xi_4)-1}{(1+\xi_3)\xi_4}-1\right]\frac{\delta_{ad}}{D_{ad}^{ETOH}}\frac{D_m^{ETOH}}{\delta_m} \quad (29)$$

Finally, by using Equations (14), (25), and (26), following equations between anode over potential and cur rent density is attained

$$\begin{aligned}&\frac{\delta_{ad}}{\xi_3 F D_{ad}^{ETOH}}\left(\frac{1}{12}-\frac{n_d^{ETOH}}{\xi_5}\left[\frac{\exp(\xi_4)-1}{(1+\xi_3)\xi_4}-1\right]\right)\bar{J}\\&-\frac{\exp(\xi_4)-1}{(1+\xi_3)\xi_4\xi_5}c_{ahin}^{ETOH}=0\end{aligned} \quad (30)$$

Using Equations (14), (22), (25), following equation between cathode over potential and current density could be presented

$$\begin{aligned}&\frac{\delta_{cd}}{\xi_1 F D_{cd}^{o_2}}\left(\frac{1}{4}-3\frac{n_d^{ETOH}}{\xi_5}\left[\frac{\exp(\xi_2)-1}{(1+\xi_1)\xi_2}-1\right]\right)\bar{J}\\&-\frac{\exp(\xi_2)-1}{(1+\xi_1)\xi_2}c_{chin}^{o_2}\\&=\frac{3}{\xi_1}\frac{\delta_{cd}}{D_{cd}^{o_2}}\left[\frac{\exp(\xi_2)-1}{(1+\xi_1)\xi_2}-1\right]\\&\times\frac{\exp(\xi_4)-1}{(1+\xi_3)\xi_4\xi_5}\frac{D_m^{ETOH}}{\delta_m}c_{ahin}^{ETOH}\end{aligned} \quad (31)$$

By using Equations (11) and (12) and combining them with Equations (30) and (31) polarization curves will be obtained. It should be mentioned that these two equations are solved by numerical methods

4. Results and Discussion

4.1. Comparison of Experimental and Analytical Results

After In this section analytical results will be compared with experiments. These experiments are performed under certain condition.

Pt/Ru/C catalyst was used for anode side and Pt/C black for cathode side. Catalyst loading on both sides was 4 mg/cm^2. and Nafion 117 was used as membrane and flow channel wide and depth was 1mm. Cathode and anode flow channel pattern was 5 parallel and 2 parallel serpentine respectively. The cell was humidified by hot water for 2hours and activated by 1 M ethanol. Active area was 10 × 10 cm^2 and back plates were made of alu-

minum.

For checking analytical model, because of some undefined coefficients, (assumed parameters in **Table 1**) first 0.125 M analytical curve is fitted to experimental curve then for other molarities analytical and experimental results will be compared (**Figures 2-5**). The results of comparison showed that at the first and second region of polarization curve, (Activation loss and Ohmic loss regions [18]) model predicts fuel cell performance well, but in the third zone (concentration loss region [18]) it seems that because of concentration loss negligence, and increase of molarity analytical somewhat model lost its accuracy.

Coefficients of analytical model are gathered in **Table 1**.

4.2. Ethanol Concentration Distribution Inside the Channel and over Potential Variation

Equation (23) foretells ethanol concentration variation inside anode channel exponentially, but based on **Figure 6** ethanol concentration inside anodic channel can be considered almost linearly.

Over potential variation both for anode and for cathode can be estimated Based on proposed analytical model. With regard to attained curve for anode over potential versus current density, by increasing current density, anodic over potential will increase, but for cathodic over potential, by increasing current density, cathodic over potential will remain approximately zero (**Figures 7** and **8**). These results are both for 0.5 M and for 0.25 M and match cathodic over potential results of G. Andreadis.

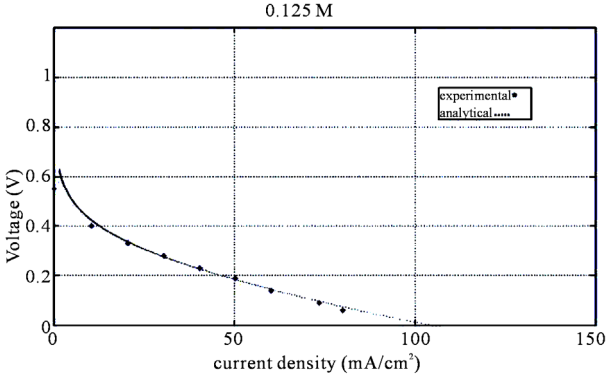

Figure 2. Comparison of experimental and analytical polarization curves for 0.125 M.

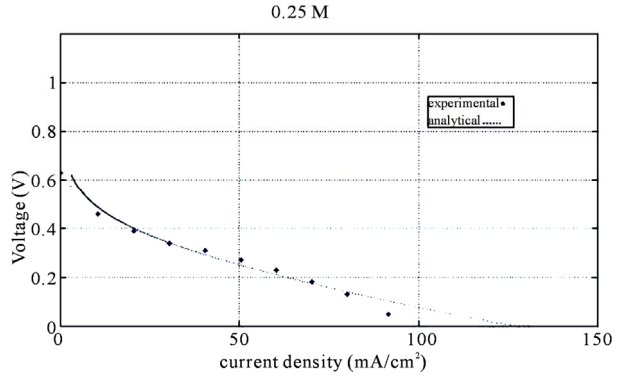

Figure 3. Comparison of experimental and analytical polarization curves for 0.25 M.

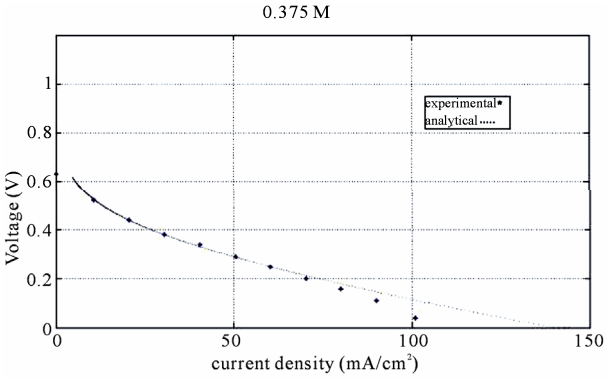

Figure 4. Comparison of experimental and analytical polarization curves for 0.375 M.

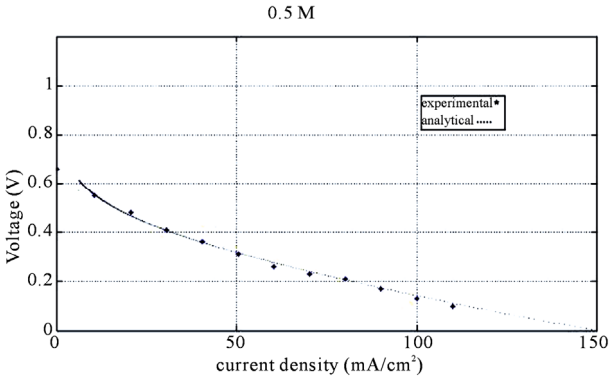

Figure 5. Comparison of experimental and analytical polarization curves for 0.5 M.

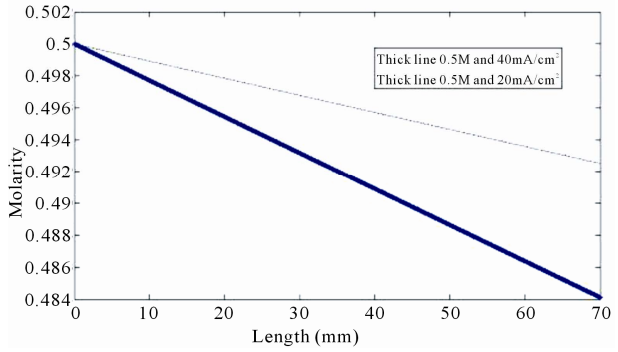

Figure 6. Ethanol concentration distribution inside anodic channel for 0.5 M; thick line 40 mA/cm2 and thin line 20 mA/cm2.

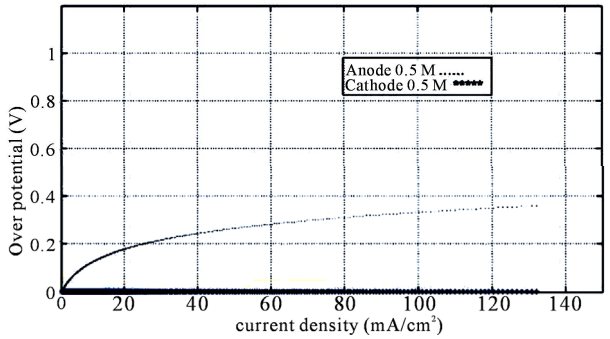

Figure 7. Anodic and cathodic over potential variation versus current density for 0.25 M.

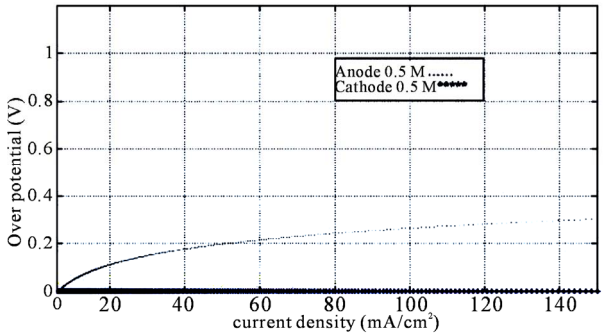

Figure 8. Anodic and cathodic over potential variation versus current density for 0.5 M.

5. Conclusions

In this paper by an analytical 2D model, (DEFC) performance was predicted. This model is capable of estimating polarization curves up to 0.5 M. This model is precise in the first and second zone (Activation and Ohmic loss region), but in the third zone (Concentration loss region) because of neglecting concentration loss and increasing inlet ethanol concentration, model error will increase and it will have more difference with experimental curves. Based on model, ethanol concentration varies almost linearly inside anodic channel. By increasing current density cathodic over potential remains zero but anodic over potential will increase up to certain value.

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Nomenclature

b	width (cm)
c	molar concentration ($\mathrm{mol\cdot cm^{-3}}$)
D	diffusion coefficient ($\mathrm{cm^2\cdot s^{-1}}$)
E	electromotive force (V)
F	Faraday's constant (96487 $\mathrm{A\,s\cdot mol^{-1}}$)
J	current density ($\mathrm{A\cdot cm^{-2}}$)
j	current density per unit volume ($\mathrm{A\cdot cm^{-3}}$)
l	length (cm)
M	molecular weight ($\mathrm{g\cdot mol^{-1}}$)
n_d	electro-osmotic drag coefficient
P	pressure (Pa)
T	temperature (K)
V	cell voltage (V)
$\dot{V}$	volume flow rate ($\mathrm{cm^3\cdot min^{-1}}$)
v	velocity ($\mathrm{cm\cdot s^{-1}}$)
x	molar fraction ($\mathrm{mol\cdot mol^{-1}}$)
	horizontal coordinate (mm)
y	longitudinal coordinate (mm)

Greek Symble

α	transfer coefficient
χ	number of flow channels
δ	thickness or depth (cm)
η	overpotential (V)
σ	proton conductivity ($\mathrm{S\cdot cm^{-1}}$)

Superscript

H_2O	water
ETOH	ethanol
O_2	oxygen
0	standard condition

Subscript

a	anode
ac	anode catalyst layer
ad	anode diffusion layer
ah	anode channel layer
ahin	inlet of anode flow channel
c	cathode
cc	cathode catalyst layer
cd	cathode diffusion layer
cell	fuel cell
ch	cathode flow channel
chin	inlet of cathode flow channel
m	proton exchange membrane
ref	reference state

