

Modeling and Analysis the Effects of Temperature and Pressure on the Gas-Crossover in Polymer Electrolyte Membrane Electrolyzer

A. H. Abdol Rahim, Alhassan Salami Tijani

Abstract—Hydrogen produced by means of polymer electrolyte membrane electrolyzer (PEME) is one of the most promising methods due to clean and renewable energy source. In the process, some energy loss due to mass transfer through a PEM is caused by diffusion, electro-osmotic drag, and the pressure difference between the cathode channel and anode channel. In PEME, water molecules and ionic particles transferred between the electrodes from anode to cathode. Extensive mixing of the hydrogen and oxygen at anode channel due to gases cross-over must be avoided. In recent times the consciousness of safety issue in high pressure PEME where the oxygen mix with hydrogen at anode channel could create, explosive conditions have generated a lot of concern. In this paper, the steady state and simulation analysis of gases crossover in PEME on the temperature and pressure effect are presented. The simulations have been analysis in MATLAB based on the well-known Fick's Law of molecular diffusion. The simulation results indicated that as temperature increases, there is a significant decrease in operating voltage.

Keywords—Diffusion, gases cross-over, steady state.

I. INTRODUCTION

CONTINUAL use of the conventional energy sources poses threat to the environment. The climate change, pollution, and greenhouse gas emission are among the examples. Shifting to a better alternative energy such as the solar and wind energy system can be a better solution. They are renewable thus do not deplete with time. The advantage of hydrogen is that it may be used as fuel in almost every application powered by fossil fuels today and without harmful emissions [1]. Since it can be used to fuel various applications, hydrogen had been deemed as a promising future energy carrier [1], [2]. Hydrogen gases can be produced by water splitting process or known as electrolysis. Water splitting process requires electricity to flow through electrode and water in order to break their molecule into hydrogen and oxygen. During the past decade, several methods have been utilized to harvest the hydrogen. However, the only state-of-the-art technique is the PEME due to its practicality for a high quality of hydrogen production [3], [4].

The advantage of PEME over many other forms of hydrogen generation is that it is simpler, and does not require chemical reactants beyond a few specific catalysts and the PEM itself, and that it generates no harmful by-products other

than pure oxygen. In addition, it is also capable of producing hydrogen gas under pressure, thus facilitating storage [5]-[7]. Ideally, water flows only in the anode channel. In practice however, a portion of the water permeates through the MEA into the cathode channel. Sometimes it is experimentally observed that some amount of hydrogen molecules can leak out of the PEM through the porous anode to contaminate the oxygen production [8]. The gases and water can permeate through these membranes [9]-[11], leading to hydrogen at the oxygen side and vice versa. These gases have individually characteristic and this phenomenon commonly referred a gases cross-over. Extensive mixing of the product gases due to gases cross-over must be avoided, particularly at low current densities, where oxygen and hydrogen production rates are lessened [12]. The hydrogen concentration at the anode side could create dangerous and explosive conditions [13] if the permeated hydrogen concentration were to exceed 4 vol%

Grigoriev et al. [14] state the proportion of hydrogen molecules reaching the anode which are oxidized back into proton depends on operating parameters such as potential, temperature and current density. After than that the same authors developed model of gases cross-over and stated that the model relates between H_2 concentration in oxygen, temperature and pressure [15].

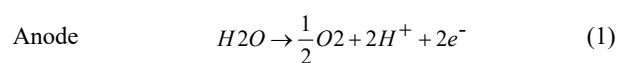
In this paper, simulation analysis for PEME cell efficiency with taking account the temperature, pressure, current density and nafion membrane thickness. Basic electrochemical and Fick's Law relation related PEME have been modeled in MATLAB. Hydrogen gas cross-over is analyzed with taking account the above parameter.

II. PEME MODEL PARAMETER

A. Polymer Electrolyte Membrane Electrolyzer

In PEME, water molecules transferred between the electrodes from anode to cathode, across the membrane thickness, where it is decomposed into oxygen, protons, and electrons. In the process, electrical energy is supplied to the system and then transformed into chemical energy. The electrons exit the cell through the external circuit. The electrons and protons are re-united at the cathode to give hydrogen gas.

The chemical reactions at the anode and the cathode are:



Abdul Hadi Abdol Rahim is with the Department of Mechanical Engineering, Universiti Teknologi MARA, UITM Shah Alam, Selangor, Malaysia (e-mail: haddierahim@yahoo.com).



The general PEME system

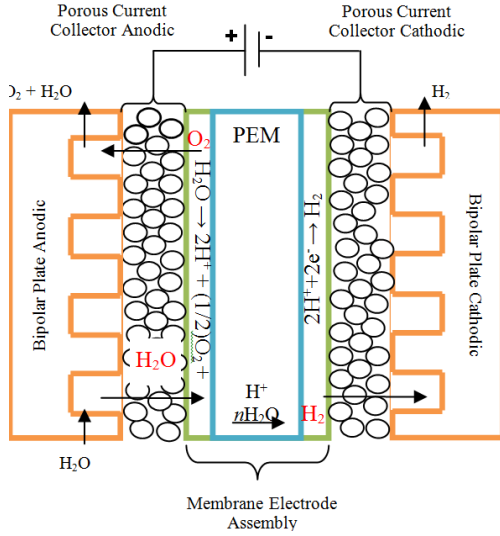
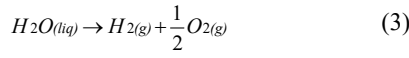


Fig. 1 Basic schematic of a PEME

B. Operating Voltage / Cell Voltage

The model was developed to determine the relationship between the cell current and cell voltage. The model is calculated based on the various subsections relationships between PEM anode, cathode and voltage. This is done by taking into account various overpotential and the open circuit voltage in the calculation of cell polarization. The real cell voltage in a cell is higher than the ideal open-circuit voltage. This single operating cell voltage is most of the commercially available electrolyzers run in current mode and the operating voltage of an electrolyzer which known as.

$$V_{cell} = V_{rev} + \eta_{act} + \eta_{ohm} \quad (4)$$

C. Reversible Cell Potential / Open Circuit Voltage

When the electrochemical cell operates in reversible conditions, the model takes into account in open-circuit conditions, based on the chemical equation its voltage can be reversible voltage and the reaction of the electrodes depends on the reactions of water at minimum electric voltage which corresponds to the reversible potential summation. This can be determined by using the Gibbs Free Energy Equation as:

$$\Delta G = nFV_{rev} \quad (5)$$

$$V_{rev} = \frac{\Delta G}{nF} \quad (6)$$

D. Activation Overpotential

The activation overpotential is described on electrode kinetic at the reaction site. The develop model of activation

overpotential can be determined in term of current density using Butler-Volmer Equation for both anode and cathode. The activation overpotentials is obtained by

$$\eta_{act} = \eta_{act,a} + \eta_{act,c} \quad (7)$$

where activation overpotential at anode side is $\eta_{act,a}$ and $\eta_{act,c}$ is activation overpotential at cathode side in PEME. $\eta_{act,a}$ and $\eta_{act,c}$ can be expressed as [16]

$$\eta_{act,a} = \frac{RT}{\alpha_a z F} \ln \left(\frac{i_a}{i_{o,a}} \right) \quad (8)$$

$$\eta_{act,c} = -\frac{RT}{\alpha_c z F} \ln \left(\frac{i_c}{i_{o,c}} \right) \quad (9)$$

where α is charge transfer coefficient, R is the universal gas constant, i is the current density, $i_{o,a}$ is the exchange current density at anode side. While $i_{o,c}$ is the exchange current density at cathode side; z is the stoichiometric coefficient refers to the number of electrons transferred in the global semi reactions (defined by Faraday's law). According to [17] exchange current densities at anode $i_{o,a} = 10^{-7} - 10^{-13} \text{ A/cm}^2$ for Pt and Pt-Ir based catalysts respectively and exchange current densities at cathode $i_{o,c} = 10^{-13} \text{ A/cm}^2$ for Pt based catalysts. Therefore the best value for exchange current densities to choose based on experimental data fitting [6]. Besides, from literature, exchange current densities for both sides can be expressed as:

Exchange current density at anode [18]

$$i_{o,a} = i_{o,a}^{ref} \exp \left(-\frac{EA_{a,a}}{R} \left(\frac{1}{T} - \frac{1}{T_0} \right) \right) \quad (10)$$

Exchange current density at cathode

$$i_{o,c} = i_{o,c}^{ref} \exp \left(-\frac{EA_{c,c}}{R} \left(\frac{1}{T} - \frac{1}{T_0} \right) \right) \quad (11)$$

where $i_{o,a}^{ref}$ and $i_{o,c}^{ref}$ are anode and the cathode reference exchange current density at reference temperature T_0 . $EA_{a,a}$, $EA_{c,c}$ is the anodic and cathodic active energy (J.mol^{-1}) [18]. For example [6] [17] suggested that the exchange current densities at anode, cathode based on Platinum electrode catalyst and platinum iridium electrode catalyst referred on the parameters given in Table I.

E. Ohmic Overpotential

The resistance caused by the PEM against the flow of electrons and electronic resistance is bringing on the ohmic overpotential. The ohmic overpotential is one the important

effect to the PEME losses. This ohmic overpotential depends on PEM, bipolar plate, and electrode material. The best selection of this material will affect the overall performance of electrolyzer. The ohmic overpotential due to membrane resistance (ionic resistance) is the resistance to the proton transport through the PEM. Meanwhile interfacial overpotential (electronic resistance) is caused by electronic materials such as bipolar plates, electrodes current collectors, etc. The ohmic overpotential is linearly proportional to the current. The ohmic overpotential due to membrane resistance can be expressed as function of the membrane thickness (cm) ϕ , conductivity of the membrane σ_{mem} and i [17];

$$\eta_{ohm,mem} = \frac{\phi}{\sigma_{mem}} i \quad (12)$$

where $R_{ion} = \frac{\phi}{\sigma_{mem}}$ is ionic resistance. The local ionic conductivity with water content and temperature function can be written as [17];

$$\sigma_{mem} = (0.005139\lambda - 0.00326) \exp \left[1268 \left(\frac{1}{303} - \frac{1}{T_{cell}} \right) \right] \quad (13)$$

where λ is the degree of membrane humidification. The interfacial overpotential can be expressed as [16];

$$\eta_{ohm,ele} = R_{ele} i \quad (14)$$

The ohmic resistance of the electronic materials as function of the material resistivity ρ in (Ωm), the length of the electrons path l , and A is the conductor cross-sectional area.

$$R_{ele} = \frac{\rho l}{A} \quad (15)$$

As a result of ionic resistance and electronic resistance, therefore the ohmic overpotential can be expressed as [16];

$$\eta_{ohm} = (R_{ele} + R_{ion}) i \quad (16)$$

The value of activation overpotential at the anode and cathode can be calculated using (12) and (13), using all parameters related in Table II.

F. Voltage Efficiency

Voltage efficiency of PEME is calculated by the value of total operating voltage over open circuit voltage [12], [19];

$$\eta_E = \frac{V_{rev}}{V_{cell}} \quad (17)$$

where

$$V_{cell} = V_{rev} + \frac{RT}{\alpha_a z F} \ln \left(\frac{i_a}{i_{o,a}} \right) + \frac{RT}{\alpha_c z F} \ln \left(\frac{i_c}{i_{o,c}} \right) + (R_{ele} + R_{ion}) i \quad (18)$$

III. MASS TRANSPORT

A. Mass Transport

The output of the electrochemical reaction is hydrogen ion (H^+) which is transported through the membrane to cathode where it combines with electron to form hydrogen molecule (H_2).

B. Electro Osmosis Drag

Water drag, or also known as electro-osmosis drag is the same as in the water transport mechanism. The hydrogen molecules that attached to hydrogen ion are dragged together as the hydrogen ion travels through the membrane. The value of oxygen and hydrogen flux density can be calculated by [12];

$$\phi_{O_2}^{drag} = \frac{i}{F} \zeta \frac{P_{O_2}^{an} S_{O_2}}{c(H_2O)} \quad (19)$$

$$\phi_{H_2}^{drag} = \frac{i}{F} \zeta \frac{P_{H_2}^{cat} S_{H_2}}{c(H_2O)} \quad (20)$$

where

$$\zeta = 0.0134 \frac{1}{K} (T) + 0.03 \quad \text{and} \quad (21)$$

$$S = \frac{\varepsilon^{dif}}{D} \quad (22)$$

C. Diffusion

Diffusion causes by the concentration difference between that of anode with the cathode. Hydrogen molecules which are produced in the cathode channel has significantly higher concentration compared to that of anode channel, hence a portion of hydrogen molecule diffuses from cathode channel into anode channel. Oxygen molecule which is produced at the anode channel is more concentrated in the anode channel and it diffuses into the cathode channel. The diffusion flux density of both molecules can be calculated by [12];

$$\phi^{dif} = D \frac{\Delta c}{\phi} = \varepsilon^{dif} \frac{\Delta p}{\phi} \quad (23)$$

D. Differential Pressure

The difference pressure in cathode channel and anode channel causes the hydrogen gas to permeate into the anode channel. Although this also causes the oxygen gas that has diffused due to concentration difference to be pushed back into the anode channel, however, it is being neglected. The value of differential pressure flux density can be calculated by [12]: The partial pressure of the molecule in each of their respective chamber can be calculated by [12];

$$\phi_{H_2}^{dp} = \varepsilon_{H_2}^{dp} \frac{P_{H_2}^{cat} - P_{O_2}^{an}}{\phi} \quad (24)$$

The partial pressure of the molecule in each of their respective chamber can be calculated by

$$p_{H_2}^{an} = p_{H_2}^{cat} \frac{\phi_{H_2}^{en}}{\frac{i}{2F} + \phi_{H_2}^{en}} \quad (25)$$

$$p_{O_2}^{an} = P^{an} + A_{O_2} i \quad (26)$$

$$p_{H_2}^{cat} = p^{cat} + \frac{\varepsilon_{H_2}^{dif} p_{H_2}^{cat} A_{H_2} + \varepsilon_{H_2}^{dp} p_{H_2}^{cat} A_{H_2} - \varepsilon_{H_2}^{dp} p_{O_2}^{an} A_{H_2}}{\varepsilon_{H_2}^{dp} p_{H_2}^{cat}} i \quad (27)$$

The total of hydrogen and oxygen diffusion can be calculated by summing (23), (24) and (20) respectively:

$$\phi_{H_2}^{en} = \varepsilon_{H_2}^{dif} \frac{p_{H_2}^{cat}}{\varphi} + \varepsilon_{H_2}^{dp} \frac{p_{H_2}^{cat} - p_{O_2}^{an}}{\varphi} - \frac{i}{F} \zeta \frac{p_{H_2}^{cat} S_{H_2}}{c(H_2O)} \quad (28)$$

$$\phi_{O_2}^{en} = \varepsilon_{O_2}^{dif} \frac{p_{O_2}^{an}}{\varphi} + \frac{i}{F} \zeta \frac{p_{O_2}^{an} S_{O_2}}{c(H_2O)} \quad (29)$$

E. Faraday Efficiency

The Faraday efficiency can be expressed as function of the flux densities across the membrane and production rates of the gases [12]:

$$\eta_F = 1 - \frac{\phi_{H_2}^{en}}{i} - 2 \frac{\phi_{O_2}^{en}}{i} \quad (30)$$

F. Cell Efficiency

The cell efficiency of PEME is the product of voltage efficiency and Faraday efficiency [12]:

$$\eta_{cell} = \eta_E \times \eta_F \quad (31)$$

IV. RESULTS AND DISCUSSION

In this paper, the main results related to the cell efficiency of PEME will be discussed. The effects of the operating parameters such as membrane thickness, pressure and temperature on the polarization curve have also been discussed. All model equations related to the analysis are written in MATLAB editor. Previous experimental manipulation usually focused on a different range of temperatures such as 313K and 353K. The idea of using these parameter relations is to evaluate the value of voltage drop or voltage losses in an electrochemical cell whenever the temperature is increased.

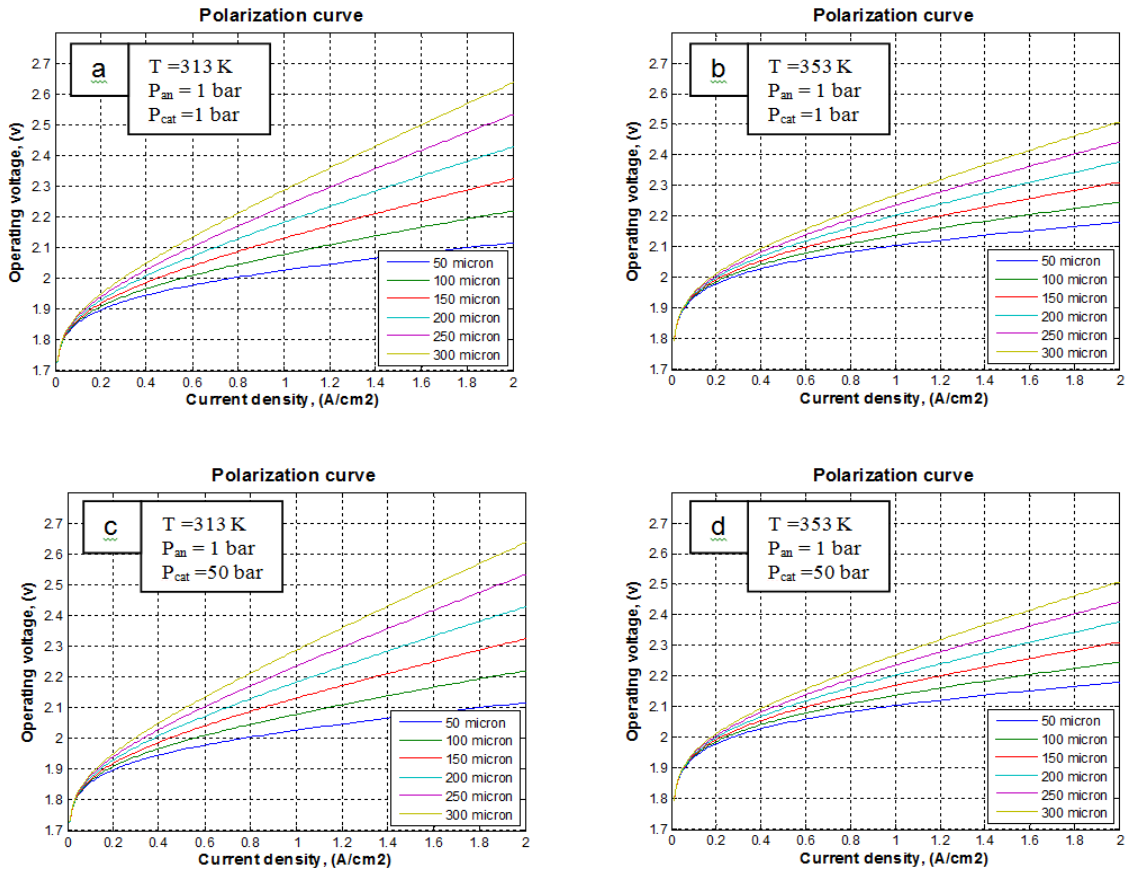


Fig. 2 Effects of temperature and membrane thickness on the operating voltage of PEME at balance pressure and different pressure

TABLE I
LIST OF PARAMETERS SELECTION FOR SIMULATION

Symbol	Parameter	Value	Reference
F	Faraday constant, (C/mol)	96485	[12]
R	Universal gas constant, (J/mol.K)	8.314	[12]
α_a, α_c	Anode & Cathode charge transfer coefficients	0.5	[17], [6]
z	Stoichiometric coefficient of electrons transferred	2	[17]
T	Temperature (K)	313 and 353	[16]
i	Current density (A/cm^2)	0.01 – 2.0	
ϕ	Membrane thickness (μm)	50 - 300	[20]
R_{ele}	Interfacial resistance (Ω)	50^{-6}	[6]
λ	Water humidification factor	25	[6]
$c(H_2O)$	Water concentration, (mol/l)	37	[12]
$i_{o,c}$	Exchange current density, Pt based catalyst, (A/cm^2)	10^{-3}	[6]
$i_{o,a}$	Exchange current density, Pt-Ir based catalyst, (A/cm^2)	10^{-9}	[6]

TABLE II
PARAMETERS SELECTION FROM [12]

Gas	A (bar cm^2/A)	ε^{diff} (10^{-11} mol/cm s bar)	ε^{dp} (10^{-11} mol/cm s bar)	S (10^{-7} mol/cm ³ bar)
H ₂	2.4	4.65	2	0.72
O ₂	2.8	2	-	0.8

Fig. 2 (a) indicates the membrane thickness has a significant effect on operating cell voltage. The operating cell voltage is increases with higher membranes thickness. The lower membrane thickness gives the lower value of operating voltage although the effects apparent start from $0.2A/cm^2$ current density. This phenomenon caused the higher membrane thickness leads the higher ohmic losses in the PEME. The comparison between Figs. 2 (a) and (b) shows effects of temperature and membrane thickness on the operating voltage of PEME at balance pressure. The result shows the operating cell voltage decreases when temperature increases at 353K. It can be observed that at a specified temperature, the operating cell voltage increases sharply at low current density and slowly thereafter with the current density at balance pressure.

The simulation result shows in Fig. 2 (a) at balance pressure and 2 (c) at different pressure where no different change in temperature apparently pressure has no effect on polarization curve characteristics of the PEME as well as Figs. 2 (b) and (d). On the other hand, Figs. 2 (a)-(d) show that as the current density increased, the cell voltage is increased. The reason is that, at higher current density more water molecules dissociate into hydrogen ion and oxygen and this process occurs only at higher voltage.

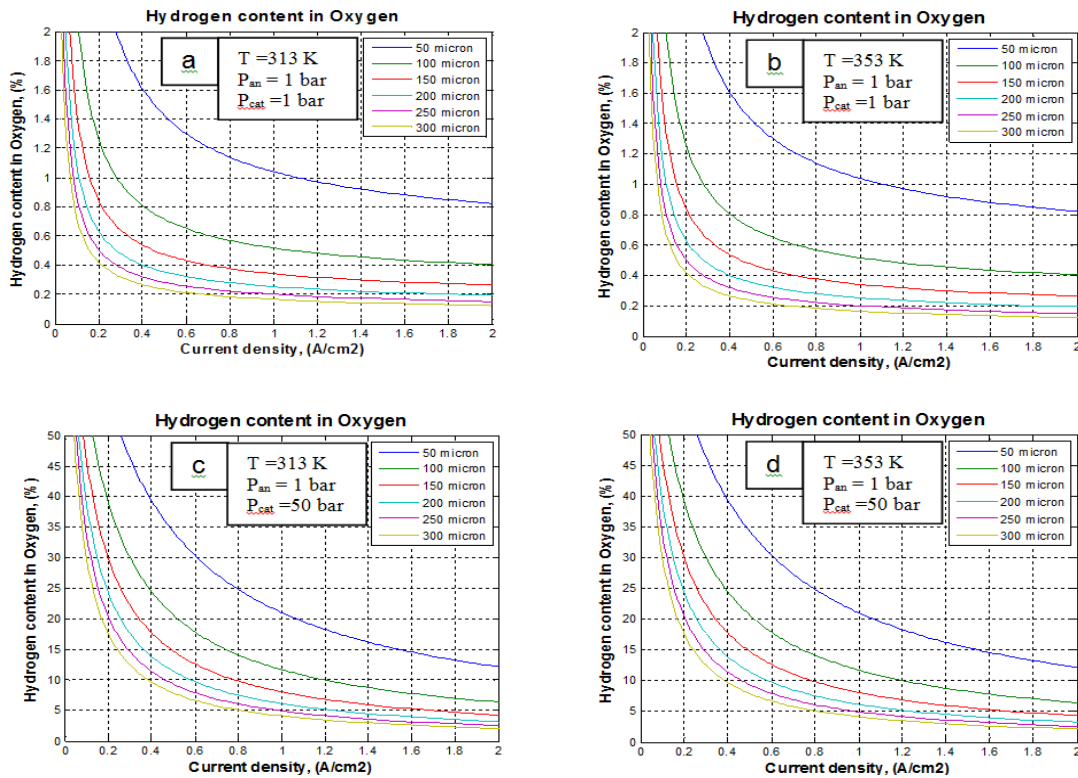


Fig. 3 Effects of different temperature and membrane thickness on hydrogen content in oxygen in PEME at balance pressure and different pressure

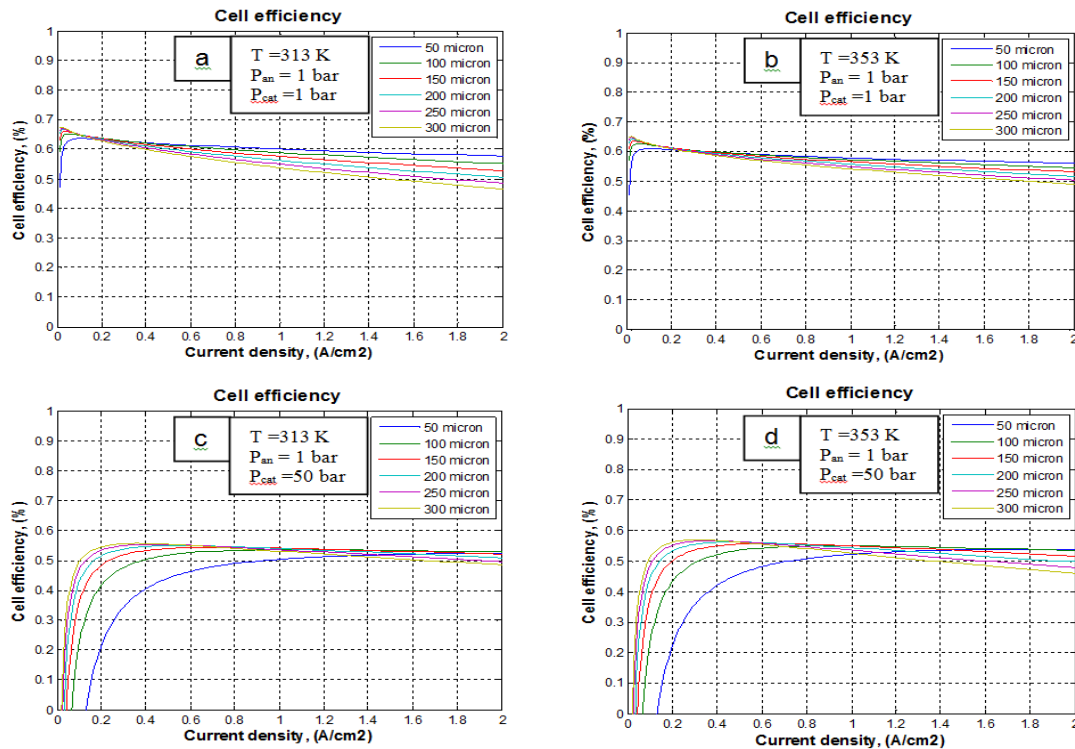


Fig. 4 Effects of different temperature and membrane thickness on cell efficiency at balance pressure and different pressure

Figs. 3 (a)-(d) represent the result of hydrogen content in anode chamber for different temperature and operating pressure of anode and cathode chamber. Figs. 3 (a) and (b) illustrate same trend in hydrogen content at anode chamber at different temperature effect. Meanwhile, Figs. 3 (a) and (c) show that hydrogen content in oxygen is increased at different pressure. In addition, the results show that as current density increases, the hydrogen content decreases. It shows PEME is ideal when operate at higher current density. Based on Figs. 3 (a)-(d) higher membrane thickness has lower hydrogen content as well as give a good result in operating PEME.

The simulation results of the effect of temperature and pressure on total cell efficiency are shown in Figs. 4 (a)-(d). Figs. 4 (a) and (b) show the effect of different temperature at balance pressure on cell efficiency. They have no significant effect on cell efficiency. Both of the results show the higher membrane thickness indicates the higher cell efficiency at current density $2\text{A}/\text{cm}^2$. In term of pressure effects, Figs. 3 (c) and (d) show the cell efficiency drop when higher pressure at cathode side. In addition, the results show that PEME with higher membrane thickness has higher cell efficiency than lower membrane thickness at low current density. However, as the current density increases, the cell efficiency of PEME with higher membrane thickness slowly decreases.

V. CONCLUSION

In this paper, the mathematical model for PEME has been presented based on a combination of fundamental thermodynamic and electrochemical relationship. Different

operating conditions such as temperature, pressure, membrane thickness and current density have been analyzed for optimized cell efficiency of PEME system. It can be concluded from this paper that when PEME operating at high temperature, the operating voltage decreases. In addition, higher pressure different between anode and cathode chamber also contribute to higher rate of hydrogen cross-over. Besides that, the rate of hydrogen gas cross-over is also affected by the thickness of membrane where higher membrane thickness contributes the low rate of hydrogen gas cross-over. The simulation results for cell efficiency shows that for lower current density, higher membrane thickness yields better cell efficiency compared to thin membrane thickness at lower temperature.

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NOMENCLATURE

ΔH	Enthalpy change
ΔG	Gibbs free energy
T	Temperature
ΔS	Entropy change
ϕ	Gas crossover flux density
p	Partial pressure
S	Solubility
ξ	Drag coefficient
Δc	Concentration difference

Δp	Partial pressure difference
ε	Permeability constant
A	Partial pressure enhancement factor

Subscripts

<i>mem</i>	Membrane
O_2	Oxygen
H_2	Hydrogen

Superscripts

<i>dif</i>	Diffusion
<i>dp</i>	Differential pressure
<i>cat</i>	Cathode
<i>an</i>	Anode
<i>en</i>	Total

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