Effect of Membrane Thickness on Direct Methanol Fuel Cell Efficiency

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ABSTRACT

In this paper, we investigate how the membrane layer's thickness affects the cross-over methanol phenomena and, consequently, the effectiveness of fuel cells. The anode to cathode electron transfer rate is reduced as a result of the methanol cross-over phenomena, which also affects the battery's overall performance. To avoid this phenomenon, we have proposed a solution, which is the increase of the thickness of the membrane and study of the curves of each of the potential differences as a function of the current intensity, in addition to calculating the parasitic current and observing the methanol diffusion curves through the membrane's thickness. Basée sur le logiciel COMSOL 5.4, cette étude simule en deux dimensions la somme des équations de diffusion, l'intensité du courant, le courant parasite et la différence de potentiel. The results of the study demonstrate the effectiveness of the research and methodology.

Keywords: COMSOL mutiphysique, Modeling, Methanol cross-over, Membrane thickness
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1. INTRODUCTION

Providing renewable and alternative energies in the twenty-first century without affecting the environment is one of the significant challenges facing us. Renewable energies, which are considered the energy of the future, do not represent a majority of more than 21.8%, according to the report published by the International Renewable Energy (IRENA), which is equivalent to 3372 gigawatts at the end of 2022, distributed mainly on solar and wind energy.

But despite its importance, it is in separate places and is not available 24 hours a day, as is the case with solar energy and this geographical distribution poses a major problem of storage as well as transport. To solve this problem, there are two energetic factors that present themselves as a very important and effective alternative, and they can be simple exploits, namely electricity and hydrogen. However, the storage of electricity is present as a new obstacle.

So far here we have learned the importance of hydrogen as a new energy factor, but it needs conditions as a liquid at a temperature below -252.87°C degrees Celsius, as well as an atmospheric pressure below 1,013 bar, and therefore the tank which contains hydrogen inside in a liquid state must be isolated from the outside temperature and pressure in addition to that hydrogen is highly explosive.

This leads us to the use of alternative liquids that contain hydrogen in their atomic composition, such as, methane (CH4) and methanol(CH3OH), which is closest to replacing

hydrogen, which contains 4 atoms of it (hydrogen), and it also does not need to be has storage conditions such as hydrogen, because it can be placed in an ordinary plastic bottle.

A fuel cell (FC) makes it possible to directly convert chemical combustion energy (oxydo-reduction) into electrical energy, heat and water. The heart of a heat pump is made up of three elements, including two electrodes: an oxidizing anode (emitter of electrons); a reducing cathode (electron collector) separated by an electrolyte. The electrolyte has the property of conducting ionized molecules directly from one electrode to another and of blocking the electrons by forcing them to pass through the external circuit of the battery where their electromotive energy can be exploited. The supply of a fuel cell is done by continuous injection of fuel at the anode, generally hydrogen, and at the cathode, generally dioxygen (oxygen in common parlance) from the air or air himself. Continuous electrical energy is then available at the battery terminals. In common parlance, fuel cells generally using hydrogen or a hydrogenated fuel (methanol) are called “hydrogen cells”.

In the core of a hydrogen fuel cell of the PEMFC type with an acidic solid membrane, two electrochemical reactions take place successively:
At the anode**:** catalytic oxidation, in the presence of platinum, of the hydrogen which dissociates from its electrons: $H_2 \rightarrow 2H^+ + 2e^-$
At the cathode: catalytic reduction, in the presence of platinum, of oxygen which captures the H+ ions which have crossed the electrolyte membrane and the electrons arriving from the external circuit. The reaction produces heat and water: $\frac{1}{2}O_2 + 2H^+ + 2e^- \rightarrow H_2O + Q(\text{heat})$
Fuel cells are differentiated either by the nature of their electrolyte or by the fuel used, such as methanol.
There are thus 6 types of fuel cells:
Three with acid electrolytes (H+ ions migrating from the anode to the cathode)

- PEMFC (Proton Exchange Membrane Fuel Cell).
- DMFC (Direct Methanol Fuel Cell).
- PAFC (Phosphorique Acid Fuel Cell)

Three with basic electrolytes (negative ions migrating from the cathode to the anode)

- liquid potash AFCs (Alkaline Fuel Cells)
- MCFC (Molten Carbonate Fuel Cell)
- SOFCs (Solid Oxide Fuel Cell)

For our case we are interested in direct methanol fuel cell DMFC (Direct Methanol Fuel Cell). In contrast, the combustion battery that uses methanol as direct fuel (DMFC) is considered a power generator for portable applications because of the simplified system it contains that is free from the fuel handling unit (reform) and storage [2].
Methanol transporting across the membrane from the anode to the cathode side is known as methanol crossover (MCO). It is known to be the main technical obstacle affecting DMFC performance significantly. As a result, the methanol is only partially oxidized at the anode CL and the remainder diffuses across the membrane. Electro-osmotic drag (EOD) and diffusion play major roles in this transport process. On the one hand, the catalyst degrades as a result of the penetrated methanol's reaction with oxygen at the cathode, which results in a parasitic current and the activation polarization of the cathode. On the other hand, it results in a drop in anode fuel consumption [2] because parasitic current and methanol losses, which are thought to make about 30% of all losses in DMFCs [3], have a major negative impact on cell efficiency and power density .
This work is based on modeling the cell by the multiphysics COMSOL software for a biphasic and two-dimensional mixture by adopting transport equations applicable to the field studied. The results of the curves' differences potential as a function of the current intensity, parasitic current, and the diffusion curves of methanol through the thickness of the membrane are presented below [3].

2. BATTERY PRINCIPLE AND MODEL

The two-dimensional model we want to study in this work. After the aqueous methanol solution has flowed through the anodic channel (AC), it reaches the membrane (M) of the anodic diffusion layer (ADL) through convection and diffusion phenomena. Then it reaches the anodic catalytic layer (ACL) where it is oxidized, as shown in the oxidation (Eq.1) and reduction (Eq.2) equations. Still, part of this solution remains without oxidation; it penetrates the membrane, causing the phenomenon of water and methanol

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to crossover together to reach the cathode side, affecting the cathodic catalytic layer (CCL), which leads to distorted power generation, which in turn affects the efficiency of the battery as a whole [4].In the catalytic layer of the anode (ACL), the methanol solution is oxidized to produce carbon dioxide, electrons, and hydrogen ions. When the hydrogen ions are transferred to the cathode catalyst layer, they react electrically with the oxygen in this layer to produce water. Carbon dioxide is removed by reverse diffusion on the anode side from the anodic catalyst layer to the anode channel [5].

$$CH_3OH + H_2O \rightarrow CO_2 + 6H^+ + 6e^- \quad (1)$$

$$O_2 + 4H^+ + 4e^- \rightarrow 2H_2O \quad (2)$$

The model presented in this study is a two-dimensional and biphasic model for the mass transport of methanol in the DMFC.

3. PROPOSED MODEL

The model proposed for this µDMFC direct methanol fuel cell is two-dimensional with mass and heat transfer equations, includes the circulation channel of the two fluids (methanol and oxygen) for the AC anode (DC cathode), the diffusion where it diffuses these two fluids in the anode ADL (cathode CDL), the anodic catalyst layer ACL (cathode CCL) and the membrane M of pretense exchange [6]. This model makes it possible to calculate performance (voltage as a function of current) and the concentration of methanol and water in the various organs of this cell [7].

3.1 Porous anodic region

After the methanol is obtained from the anode flow channel, the latter will reach the catalyst layer after passing through the diffusion layer, where part of the methanol is chemically oxidized to form carbon dioxide CO2 on one side and the current on the other. In contrast, the rest of the methanol permeates the membrane. It reaches the catalyst layer on the cathode side, thus forming the so-called crossover phenomenon in the membrane and producing a parasitic current, which directly decreases the battery yield [8].

The mass conservation equation for the methanol in a steady state will be given as follows [9]:

$$C_{M,1} : \nabla.\left[\left(-\frac{Kk_{rl}}{\mu_l}\nabla p_l\right)C_{M,1} - D_M^{eff}\nabla C_{M,1}\right] = \dot{R}_{M,1,a} \quad (3)$$

3.2 Porous cathodic region

In the DMFC cathode, the oxygen in the air flows through the cathodic diffusion layer arriving at the catalytic layer, where the oxygen reaction (ORR) allows us to have water. The mass conservation equation for the methanol in a steady state will be given as follows [10]:

$$C_{O_2,g} : \nabla.\left[\left(-\frac{Kk_{rg}}{\mu_g}\nabla p_{g,c}\right)C_{O_2,g} - D_{O_2,g}^{eff}\nabla C_{O_2,g}\right] = \dot{R}_{O_2,g} \quad (4)$$

3.3 membranes

Our study considers the transport of liquid phases (water and methanol). However, in the case of high concentrations of methanol, the so-called crossover phenomenon, the unreacted excess is seen to have passed through the membrane and oxidized when it reached the layer catalytic in the cathode part, producing parasitic current, resulting in decreased battery efficiency. The methanol crossover flux (NM) can be given by [12]:

$$N_M = -D_M^{eff}.\nabla C_M + n_{d,M}\frac{1}{F} - \left(\frac{k}{\mu_l}\frac{\Delta p_{l,c-a}}{\delta_{mem}}\right)C_M \quad (5)$$

3.4 Electrochemical reaction equation

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In the DMFC cell, the two reactions that take place are the reaction of methanol on the anode and the reduction reaction of oxygen on the cathode side, knowing that we have several substances that are generated during this reaction. At the same time, it is well known that the mass conservation equations are coupled with the kinetics of the electrochemical reaction, which is why it is necessary to solve the formula of the electrochemical reaction. The electrochemical reaction rate for the anodic catalyst layer will be as follows [13]:

$$j_a = A_{v,a} j_{0,M}^{ref} \left(\frac{C_M}{C_M^{ref}} \right)^{\gamma} \exp\left(\frac{\alpha_a F}{RT} \eta_a \right) \quad (6)$$

The electric current arises from the cathode side of the µDMFC is come to be [14]:

$$j_c = A_{v,c} j_{0,O_2}^{ref} \left(\frac{C_{O_2}/k_{H,O_2}}{C_{O_2}^{ref}} \right) \exp\left(\frac{\alpha_c F}{RT} \eta_c \right) \quad (7)$$

In addition to the battery current (Icelle), the resulting current contains the parasitic current (Ip) resulting from the oxidation of the methanol impregnating the membrane. The stray current is given by [15]:

$$I_p = \frac{6F \iint N_M dxdy}{A_{cell}} \quad (8)$$

The total electric current (I_{celle}) generated by the battery is given by [16]:

$$I_{Cell} = \frac{\iiint_{ACL} j_a dxdydz}{A_{cell}} \quad (9)$$

The cell voltage is determined as follows [17]:

$$V_{Cell} = V_0 - \eta_a - \eta_c - I_{Cell}\left(R_{contact} + \frac{\delta_{mem}}{k} \right) \quad (10)$$

3.4 Conditions to the limits

Border 2: Entry conditions [18].

$$C_{M,1} = C_M^{in}, C_{M,W} = C_{M,V}^{in}, P_l = P_l^{in}, P_g = P_g^{in} \quad (11)$$

Border 1: The existing interface between the anodic rib collector and the anode diffusion layer.

$$\frac{\partial C_M}{\partial x} = 0, \frac{\partial C_{M,V}}{\partial x} = 0, \frac{\partial P_l}{\partial x} = 0, \frac{\partial P_g}{\partial x} = 0 \quad (12)$$

Border 5: The cathode rib, where there is no flux.

$$\frac{\partial C_{O_2}}{\partial x} = 0, \frac{\partial P_l}{\partial x} = 0, \frac{\partial P_g}{\partial x} = 0 \quad (13)$$

Border 4: The oxygen inlet and the water outlet at the cathode.

$$C_{O_2} = C_{O_2}^{in}, C_{W,V} = C_{W,V}^{in}, P_l = P_l^{in}, P_g = P_g^{in} \quad (14)$$

Border 3 and 6: Symmetric diffusion.

$$\frac{\partial \phi}{\partial y} = 0, C_{M,1} = C_{M,V} = C_{O_2}, P_l = P_g \quad (15)$$

3.6 Model parameters

The cell's geometrical and parametric operating dimensions and electrochemical properties described in the determining equations above are indicated in tables 1-3 [19].

3.7 Model validation

In our research, the power density test curve is the same as the actual µDMFC test curve [20].

4. RESULTS AND DISCUSSIONS

In this study, we checked the effect of membrane thickness on increased crossover phenomenon and stray current parasite and the optimal performance of the anode electrode. So we changed the thickness of the membranes from $0.15(mm)$ to $0.55(mm)$, with a value of $\Delta\delta_{mem} = 0.2(mm)$.

Figure 1 shows us the methanol concentration distribution in the anode's porous region for a $0.15(mm)$ thick membrane. The methanol is transported through the porous anodic diffusion layer to the cathode catalyst layer causing the crossover phenomenon. It is found that the methanol concentration becomes low in the anodic diffusion layer due to its consumption by the methanol oxidation reaction at the anodic catalyst layer. This decrease reaches its magnitude when it reaches the catalyst layer cathodic through the membranes. In our example, the concentration of methanol which crosses the cathode catalyst layer, starts from $3.5\left(mol.m^3\right)$ to reach $3\left(mol.m^3\right)$ at the limits of the membrane. As for the membrane, the concentration of methanol in it starts from $3\left(mol.m^3\right)$ to $2.5\left(mol.m^3\right)$ to reach the cathode catalyst layer, where the concentration of methanol forms from $2.5\left(mol.m^3\right)$ to $2\left(mol.m^3\right)$.

We also notice in Figure 2 that the intensity of the current is between $260\left(A/m^2\right)$ and $300\left(A/m^2\right)$, which are smaller values compared to the results of the cases that come after when we increase the thickness of the membrane more than $0.15(mm)$, while the intensity of the parasitic current is at the most significant value between $4.48\left(A/\mathrm{m}^2\right)$ and $4.9\ \left(A/m^2\right)$ in this case (Fig. 3).

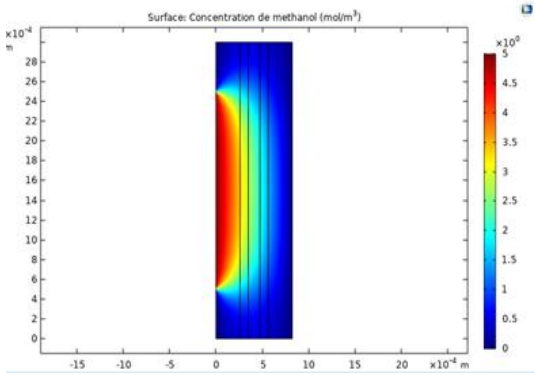

Figure 1. Distribution of methanol concentration at $\delta_{mem} = 0.15\ (mm)$ thicknesses of membrane

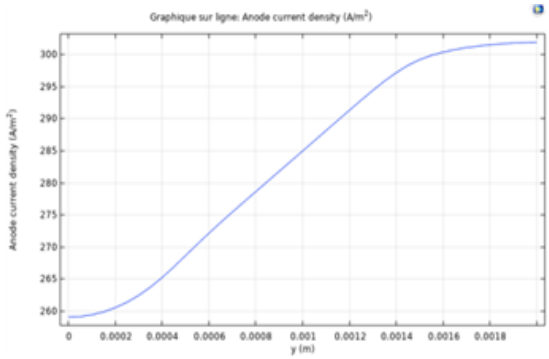

Figure 2. Effects of membrane thickness on DMFC fuel cell performance $\delta_{mem} = 0.15\ (mm)$

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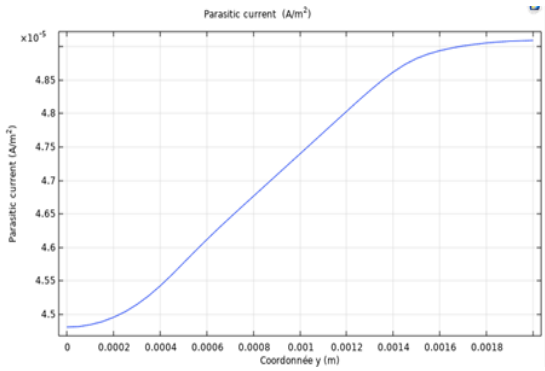

Figure 3. Parasitic current with a membrane thickness of $\delta_{mem} = 0.15\ (mm)$

suppose we increase the thickness of the membrane from $0.15(mm)$ to $0.35(mm)$, we see a significant decrease in the diffusion of methanol through the anodic diffusion layer ADL, as shown in Figure 4, and it's $5\left(mol.m^3\right)$ to $3.5\left(mol.m^3\right)$ and $3.5\left(mol.m^3\right)$ to $2.5\left(mol.m^3\right)$ for the anodic catalyst layer ACL.

As for the membrane layer, a decrease in the rate of diffusion of methanol is observed, the value of the concentration of the latter (methanol) being estimated between $3\left(mol.m^3\right)$ to $2\left(mol.m^3\right)$ and from $2\left(mol.m^3\right)$ to $1.5\left(mol.m^3\right)$ for the cathodic catalytic layer CCL. the decrease in the diffusion percentage of methanol in the membrane layer was significant given of the previous case, where the thickness of the membrane layer was $0.15(mm)$ and the methanol concentration was between $3\left(mol.m^3\right)$ and $2.5\left(mol.m^3\right)$, and this explains the increase in the intensity of the current produced from $355\left(A/m^2\right)$ to $397\left(A/m^2\right)$ and therefore, the good performance of the battery, as well as the decrease in the intensity of the parasitic current from $4.24\left(A/m^2\right)$ to $4.669\left(A/m^2\right)$, which indicates the importance of increasing the thickness of the membrane concerning the efficiency of the battery to direct methanol (DMFC).

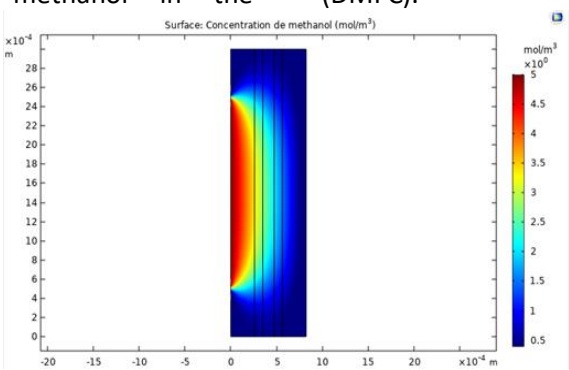

Figure 4. Distribution of methanol concentration at $\delta_{mem} = 0.35\ (mm)$ thicknesses of membrane

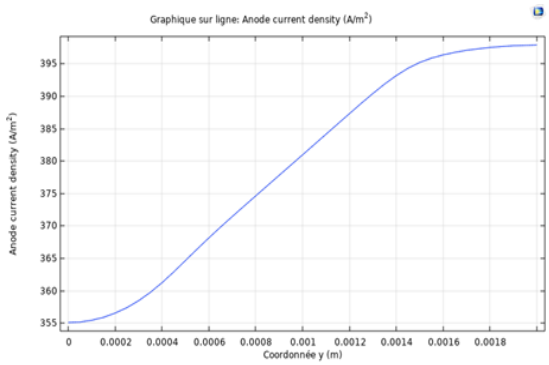

Figure 5. Effects of membrane thickness on DMFC fuel cell performance $\delta_{mem} = 0.35\ (mm)$

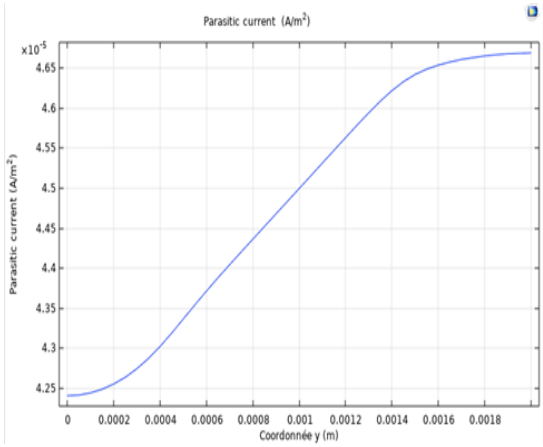

Figure 6. Parasitic current with a membrane thickness of $\delta_{mem} = 0.35\ (mm)$

In this last case, we increased the thickness of the membrane from $0.35(mm)$ to $0.55(mm)$, and as in the previous case, the increase in the diffusion of methanol in the two layers is significant; it is between $5\left(mol.m^3\right)$ and $3.5\left(mol.m^3\right)$ for the anodic diffusion layer (ADL) and between $3.5\left(mol.m^3\right)$ and $3\left(mol.m^3\right)$ for the anodic catalyst layer (ACL).

While the decrease in the diffusion of methanol in the membrane layer is $3\left(mol.m^3\right)$ to $1.7\left(mol.m^3\right)$, as well as the cathode catalyst layer (CCL) is to $1.7\left(mol.m^3\right)$ to $1.2\left(mol.m^3\right)$, which leads to a noticeable increase in the intensity of electric current from $450(A/m^2)$-$490(A/m^2)$, as well as a decrease in parasitic current intensity from $3.5(A/m^2)$ to $3.9(A/m^2)$, and therefore good performance for a methanol fuel cell (DMFC).

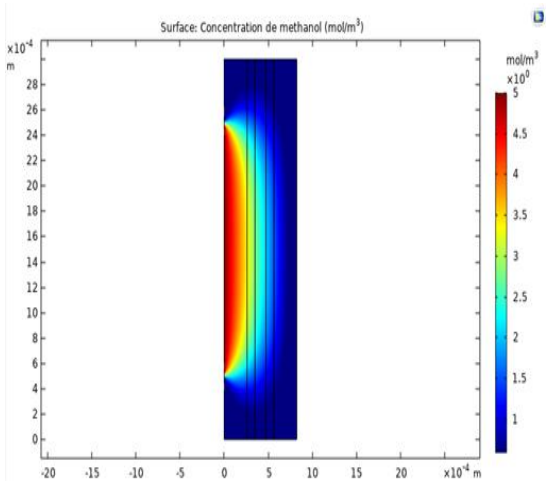

Figure 7. Distribution of methanol concentration at $\delta_{mem} = 0.55\ (mm)$ thicknesses of membrane

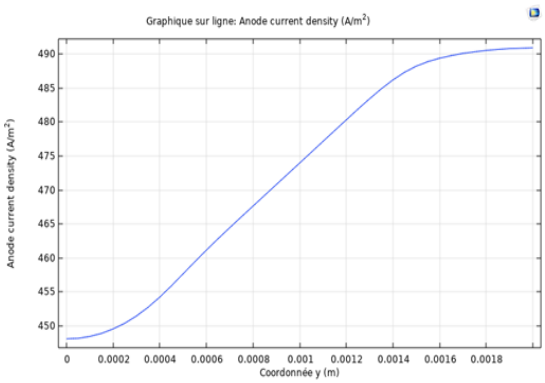

Figure 8. Effects of membrane thickness on DMFC fuel cell performance $\delta_{mem} = 0.55\ (mm)$

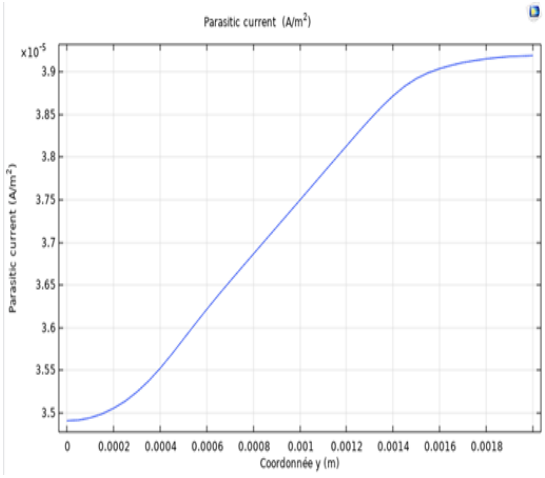

Figure 9. Parasitic current with a membrane thickness of $\delta_{mem} = 0.55\ (mm)$

5. CONCLUSIONS

Thanks to this simulation of the methanol battery and after increasing its efficiency by changing the thickness of the membrane from $\delta_{mem} = 0.15(mm)$ to $\delta_{mem} = 0.55(mm)$, we noticed that this idea was good for improving the performance of this type of battery by increasing the intensity of the current produced from $55(A/m^2)$ to $564(A/m^2)$, as well as decreasing the parasitic current from the value $568(A/m^2)$ to the value $265(A/m^2)$.

However, in practical experiments, another factor may appear and negatively affect the excellent performance of the battery, namely the percentage of humidity, which directly affects the transmission of protons through the membrane.

For this we try to examine the influence of the humidification rate (which varies between 40% and 70%) of the membrane on the polarization curve of a cell of the same model and with the same surface.

We note that the decrease in methanol crossover accompany the increase in water which also crosses the membrane.

Therefore the studies of these two factors (the humidification factor and the thickness of the membrane) are necessary to improve the performance of this type of battery.

But in general, the idea of increasing the thickness of the membrane is good and effective

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