

## Electrochemical Aided Model to Study Solid Polymer Electrolyte Water Electrolysis

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**Résumé** – *L'objet de cet article est de proposer une analyse de l'électrolyse de l'eau par membrane polymère solide. L'investigation théorique s'articule autour de l'application de l'équation de Butler-Volmer et la caractérisation du transport de l'eau à travers la membrane. On aboutit aux équations de surtension cathodique et anodique dont la précision est testée par comparaison avec les données publiées.*

**Abstract** - *The aim of this study is to analyze the Proton Exchange Membrane (PEM) water electrolysis. On the basis of theoretical investigation the well know Butler-Volmer equation and water transport characteristics through the solid electrolyte membrane were employed to simulate the electrode activation over potential and membrane ohmic over potential. Then, the simulation results were compared with the published experimental data.*

**Keywords:** Hydrogen - Water Electrolysis - Proton Exchange Membrane - Butler-Volmer equation - Computer Simulation - Electrochemical Model - Over potential.

### 1. INTRODUCTION

From hydrogen a sustainable source of energy could be produced. In fact, hydrogen may play an important role as an energy carrier of the future [1]. Hydrogen can be produced from many different sources and it looks like that water electrolysis would be the easiest option and the only one currently practical [2].

The chemical process involved in the water electrolysis, split the water molecules to produce hydrogen and oxygen, can be viewed as the reverse process which the proton exchange membrane fuel cells (PEMFC) is based on. From this observation merge the idea to use a solid membrane, like perfluorosulfonic acid polymer (Nafion), as a membrane electrolyser.

The purpose of this study is to apply a simple, but useful, first-generation theoretical model to explain the current-potential characteristics of solid polymer electrolysis (SPE) cell, based on the well known Butler - Volmer kinetics on the electrode surfaces.

### 2. MODEL DESCRIPTION

A basic schematic of solid polymer electrolyte water electrolysis is presented in Fig. 1. Water undergoing adequate electrical voltage, split into hydrogen and oxygen and the following reaction occurs.

At the anode, water is introduced and dissociated into oxygen, protons and electrons.



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At the cathode, the protons are driven through the SPE to the cathode under an electric field where they combine with the electrons arriving from the external circuit to form hydrogen gas.



The net reaction of the entire cell



*Note: the preceding reactions are possible because of the net acidity influence of the solid polymer electrolyte.*

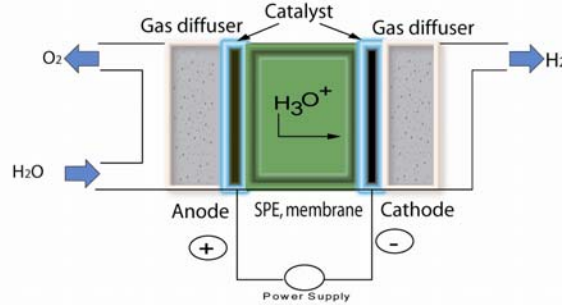


Fig. 1: Schematic representation of SPE water electrolysis

## 2.1 Steady state conservation equations

We consider an ideal electrochemical process in which all species involved are governed by Faraday's law of electrolysis. The laws state that the amount of chemical change produced by current at an electrode-electrolyte boundary is proportional to the quantity of electricity used, and the amounts of chemical changes produced by the same quantity of electricity in different substances are proportional to their equivalent weights.

The first hypothesis made is that the species are well mixed in the anodic and cathodic chambers, thus the surface concentrations do not differ appreciably from the bulk phase [4]. The mass balances of water and oxygen at the anode, and that of hydrogen at the cathode can be written as.

Water is consumed at the anode and the oxygen and hydrogen are produced at the cathode.

$$\dot{N}_{\text{H}_2\text{O},\text{in}} - \dot{N}_{\text{H}_2\text{O},\text{out}} = \dot{N}_{\text{H}_2,\text{out}} - \dot{N}_{\text{H}_2,\text{in}} = 2 \left( \dot{N}_{\text{O}_2,\text{out}} - \dot{N}_{\text{O}_2,\text{in}} \right) = \frac{J}{2F} \quad (4)$$

$J$ : Total current density, ( $\text{A}\cdot\text{m}^{-2}$ ),  $J = I/A$ ;  $F$ : Faraday's constant ( $\text{C}\cdot\text{mol}^{-1}$ );  $\dot{N}_k$ : Molar flux density of species ( $\text{mol}\cdot\text{m}^{-2}\cdot\text{s}$ ),  $k = \text{H}_2\text{O}, \text{H}_2, \text{O}_2$ .

### 2.1.1 Electrode kinetics – Anode and cathode layers

The electrode kinetics is modelled incorporating the exchange current. Assuming no transport limitations, the Butler-Volmer expression is utilized for the overall electrochemical reaction at the anode [3][4]. We noticed that the Butler-Volmer equation, derived from the kinetic of water electrolysis, can be expressed differently according to the predominance of the factors that are involved in the equation, and the phenomena that researcher want to focus on.

As noticed by Barbir [2], the PEM electrolysis is a process just reverse of a PEM fuel cell process, thus for the same elements constituting the electrodes, and the SPE, the Butler-Volmer equation developed for the fuel cell can be used for the SPE electrolyser, with some precautions dictated by the nature of reactions at the cathode chambers and the anode chambers.

Here we choose to apply a simple expression [5] that can be easily compute on most of Personal computer or sophisticated one.

The anodic current density is expressed as:

$$J_a = J_a^{\text{ref}} \left( \frac{p_{\text{O}_2}}{p^0} \right)^{\beta_1} \left( \frac{p_{\text{H}_2\text{O}}}{p_{\text{H}_2\text{O}}^0} \right)^{\beta_2} \left[ \exp\left( \frac{\alpha_a v_{e^-} F \eta_a}{R T} \right) - \exp\left( \frac{\alpha_c v_{e^-} F \eta_a}{R T} \right) \right] \quad (5)$$

The cathodic current density is expressed as:

$$J_c = J_c^{\text{ref}} \left( \frac{p_{\text{H}_2}}{p^0} \right)^{\beta_3} \left[ \exp\left( -\frac{\alpha_c v_{e^-} F \eta_c}{R T} \right) - \exp\left( \frac{\alpha_a v_{e^-} F \eta_c}{R T} \right) \right] \quad (6)$$

$J_a$ ,  $J_c$ : Current density respectively of the anode and the cathode ( $\text{Am}^{-2}$ )

$p_{\text{H}_2}$ ,  $p_{\text{O}_2}$ ,  $p_{\text{H}_2\text{O}}$ : The partial pressure of the different species at gas phase (atm)

$p^0$ : The standard pressure (atm) and  $p_{\text{H}_2}^0$  the vapor pressure of water (atm)

$v_{e^-}$ : The stoichiometric coefficient of the reaction considered for the numbers of electrons involved

$\alpha_a$ ,  $\alpha_c$ : The anodic and the cathodic charge transfer coefficients

$\eta_a$ ,  $\eta_c$ : The anodic and cathodic overpotential

$J_a^{\text{ref}}$ ,  $J_c^{\text{ref}}$ : The anodic and the cathodic reference exchange current density ( $\text{Am}^{-2}$ ) expressed as Arrhenius's equation like [5]:

$$J_a^{\text{ref}} = J_{a,0}^{\text{ref}} \cdot \exp\left[ -\frac{E_{A,a}}{R} \left( \frac{1}{T} - \frac{1}{T_0} \right) \right] \quad (7)$$

$$J_c^{\text{ref}} = J_{c,0}^{\text{ref}} \cdot \exp\left[ -\frac{E_{A,c}}{R} \left( \frac{1}{T} - \frac{1}{T_0} \right) \right] \quad (8)$$

$R$ : Universal gas constant,  $R = 8.3145 \text{ (Jmol}^{-1}\text{K}^{-1}\text{)}$ .

$T_0$ : Reference temperature, (K).

$E_{A,a}$ ,  $E_{A,c}$ : The anodic and cathodic active energy ( $\text{J.mol}^{-1}$ ) [6].

$J_{a,0}^{\text{ref}}$ ,  $J_{c,0}^{\text{ref}}$ : The anodic and the cathodic reference exchange current density at reference temperature,  $T_0$  ( $\text{Am}^{-2}$ ).

### 2.1.2 Simplification of the Butler Volmer equation

- The parameters  $\beta_1$ ,  $\beta_2$ ,  $\beta_3$ , are empirically determined concentration. In this study the effect of pressure on the efficiency of SPE is not considered, the process of electrolysis is under the atmospheric presses, thus we can choose  $\beta_1 = \beta_2 = \beta_3 = 0$ , [5].
- The values of  $\alpha_a$  and  $\alpha_c$  can vary between 0 and 1, but for metals they are around 0.5. A value of 0.5 means that the activated complex is exactly halfway between reagents and products on the reaction coordinate, its structure reflecting reagent and product equally.
- For water electrolysis, it is easily deduced that  $\alpha_a + \alpha_c = 1$ , and  $v_{e^-} = 2$  for the anodic reaction, and  $v_{e^-} = -2$  for the cathodic reaction.

According to the above simplifications we can assume the following expressions for the activation over potential of the anode and the cathode. From (5), (6), (7) and (8) with the simplifications:

$$\text{At the anode:} \quad \eta_a = \frac{RT}{F} \sinh^{-1} \left( \frac{J_a}{2 J_a^{\text{ref}}} \right) \quad (9)$$

$$\text{At the cathode:} \quad \eta_c = \frac{RT}{F} \sinh^{-1} \left( \frac{J_c}{2 J_c^{\text{ref}}} \right) \quad (10)$$

### 2.1.3 Equation for the PEM electrolyser (SPE)

The electrolyser cell can be seen as an electrical circuit with different resistance connected in series, for the sake of simplicity an equivalent circuit for electrolysis process is admitted to well correspond to the electrolysis phenomena, (Fig. 2). The overall applied cell potential is thus composed of the cell Nernst potential  $E_0$ , anode and cathode over potentials, over potential due to membrane, and interfacial resistance [4]

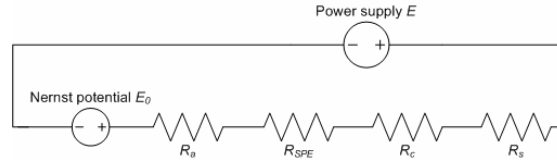


Fig. 2: Electrical equivalent circuit for water electrolyser, SPE [4]

$R_a$  = Anode resistance,  $R_c$  = Cathode resistance,

$R_{SPE}$  = Membrane resistance,  $R_s$  = Interfacial resistance

### 2.1.4 Electrochemical model description

The process considered in this paper is assumed at steady state conditions, thus the overall current density that runs through the different resistances in series is the same, we can write:

$$J_a = J_c = J \quad (11)$$

$$J = -\sigma_{PME} \cdot \frac{d\phi}{dx} \quad (12)$$

$\sigma_{PME}$  : The ionic conductivity of the solid membrane (S/m)

$\phi$  : The electrical potential of the membrane (V)

$x$  : The linear direction along the electrolysis cell beginning at the anode electrode (m)

The equivalent circuit voltage or the required voltage for the SPE (PEM electrolysis),  $E$  is expressed as the sum of the different voltage that occurs in the cell. The overall applied cell potential is thus composed of the cell Nernst potential  $E_0$ , anode and cathode over potentials,  $\eta_a$ ,  $\eta_c$ , over potential due to membrane,  $\eta_{SPE}$ , and interfacial resistance  $\eta_s$ :

$$E = E_0 + \eta_a - \eta_c + \eta_{SPE} + \eta_s \quad (13)$$

### 2.1.5 The equilibrium voltage

The equilibrium voltage is called the open circuit voltage or the reversible potential; it is determined empirically by the Nernst equation [7]:

$$E_0 = 1.23 - 0.9 \times 10^{-3} (T - 298) + \frac{RT}{4F} \ln \left( \frac{P_{H_2}^0 \cdot P_{O_2}}{P_{H_2O}} \right) \quad (14)$$

### 2.1.6 The ohmic over potential across the SPE (PEM)

The SPE exhibit a resistance to the movement of hydrogen proton, this cause the apparition of the over potential across the membrane. Researchers conclude an easily compressive relation between the ionic conductivity, the degree of humidification, temperature and the membrane thickness [8]. We present two equations found in the published literature about the ionic conductivity. The local ionic conductivity,  $\sigma_{PEM}$  [S/m], has been correlated empirically as a function of temperature and the water content [9], according to the correlation of Neubrand cited by Ramousse [10].

$$\sigma_{PEM} = \exp \left[ -E_{A,m} \left( \frac{1}{T} - \frac{1}{353} \right) \right] \cdot \left( 0.0013 \lambda^3(x) + 0.0298 \lambda^2(x) + 0.2658 \lambda(x) \right) \quad (15)$$

$$E_{A,m} = 1183 + 2640 \exp(-0.6 \lambda(x)) \quad (16)$$

$\lambda$ : the water content, in the case of Nafion membrane, this is the number of water molecules per sulfonate group ( $\text{mol}_{H_2O} / \text{mol}_{SO_3^-}$ ),  $E_{A,m}$  is the energy activation of the membrane ( $\text{Jmol}^{-1}$ ).

The water content,  $\lambda$ , at location  $x$  (from the anode electrode), can be expressed linearly with the empirical equation [8] as:

$$\lambda(x) = \lambda_c + (\lambda_a - \lambda_c) \frac{x}{L_{PEM}} \quad (17)$$

$L_{PEM}$ : Membrane thickness (m);  $\lambda_a$ ,  $\lambda_c$ : The water contents at the anode-membrane and cathode-membrane interface respectively.

### 2.1.7 The overall ohmic resistance

The ohmic losses are due to the resistance of the wiring and the resistance of the imperfect electrodes. The loss in the SPE or in the fuel cell is approximately linear after the activation loss levels out and before the concentration loss becomes significant [11]. The ohmic loss of the SPE is slightly nonlinear and variable due to the characteristics of the electrical conduction at different conditions. In most cases, the ohmic drop or the ohmic over potential is given by:

$$\eta_{PEM} = J R_{SPE} = J \int_0^{L_{PEM}} \frac{1}{\sigma_{PEM}(x)} dx \quad (18)$$

### 2.1.8 The interfacial over potential

The interfacial over potential  $\eta_s$ , may be written in terms of interfacial resistance  $R_s$  and current density as:

$$\eta_s = J R_s \quad (19)$$

Combining the different over potential associated with the electrolysis cell we can express the overall cell voltage as follow:

$$E = E_0 + \frac{RT}{F} \sinh^{-1} \left( \frac{J_a}{2 J_a^{ref}} \right) + \frac{RT}{F} \sinh^{-1} \left( \frac{J_c}{2 J_c^{ref}} \right) + J \cdot (R_{PEM} + R_s) \quad (20)$$

From here we can evaluate the power density (SPE water electrolysis) defined as the product of the cell voltage and the current density as:  $P(W) = J \times E$ .

### 3. MODEL VALIDATION

The preceding electrochemical model is used to simulate the voltage of the solid polymer electrolyte water electrolysis cell. The numerical values are compared with the experimental data published in the literature.

The experimental published parameters are provided on **Table 1** and **Table 2**.

**Table 1:** Model parameters for water electrolysis for Pt based anode and cathode electrodes on Nafion<sup>TM</sup> electrolyte at 80 °C [4]

Parameters	Values	Dimension
$J_a^{\text{ref}}, \text{Pt}$	$10^{-12} - 10^{-9}$	$\text{A.cm}^{-2}$
$J_a^{\text{ref}}, \text{Pt} - \text{Ir}$	$10^{-7}$	$\text{A.cm}^{-2}$
$J_c^{\text{ref}}, \text{Pt}$	$10^{-4} - 10^{-3}$	$\text{A.cm}^{-2}$
$L_{\text{PEM}}$	178	$\mu\text{m}$

Note: The exchange densities are based on the electrochemically active surface area

The water contents at the anode-membrane and cathode-membrane are taken from Ni *et al.* [8]. The local ionic conductivity,  $\sigma_{\text{PEM}}$ , is estimated by equation (16). For the Nafion<sup>TM</sup> membrane, the interfacial resistance  $R_s$  is assumed to be negligible and hence set equal to zero in this model.

**Table 2:** Model parameters for water electrolysis for Pt based Anode and cathode electrodes on Nafion<sup>TM</sup> electrolyte at 80 °C [8]

Parameters	Values	Dimension	Reference
$p_{\text{O}_2}$	1	Atm	[8]
$p_{\text{H}_2}$	1	Atm	[8]
$E_{A,a}$	76	$\text{kJmol}^{-1}$	[8, 6]
$E_{A,c}$	18	$\text{kJmol}^{-1}$	[8, 6]
$\lambda_a$	14		[8]
$\lambda_c$	10		[8]

To compare the experimental values with the theoretical model we adopt the following parameters:

- Nafion<sup>TM</sup> 117 Membrane, the thickness of the electrolyte is taking  $L_{\text{PEM}} = 50\mu\text{m}$  [8], with water content as provided in **Table 2**.
- Temperature set  $T = 353 \text{ K}$ ,  $p = 1 \text{ atm}$ .
- Electrode catalyst is platinum based, instead to use the values of current exchange of the anode and the cathode provided in **Table 1**, we use the data giving by Meng Ni *et al.* [8], at  $T = 353 \text{ K}$ ,  $J_a^{\text{ref}} = 1.0 \times 10^{-5} \text{ Am}^{-2}$ ,  $J_c^{\text{ref}} = 10 \text{ Am}^{-2}$ .

We compare data for different contribution of over potential then for cells voltage versus published experimental data, to show how the model is accurate in simulating both over potential and overall cells voltage.

For anode overpotential, Fig. 3, presents comparison between the simulating and experimental data from [4] of anode over potential (Pt-IrO<sub>2</sub> anode based), the standard deviation = 0,04667 and the regression coefficient = 0,9540, which indicate the accuracy of the model.

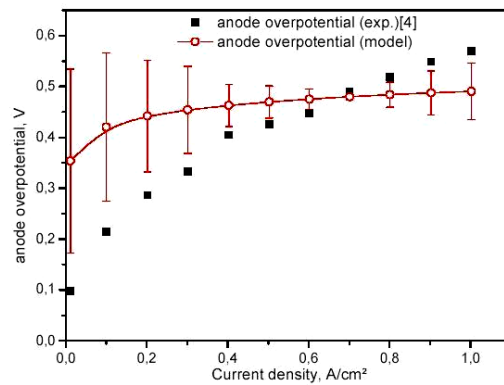


Fig. 3: Comparison between anode over potential model and experimental data. Pt-IrO<sub>2</sub> anode based

For cathode overpotential, Fig. 4, presents comparison between the simulating and experimental data from [4] of cathode over potential (Pt cathode based), the standard deviation = 0,01266 and the regression coefficient = 0,97834, which indicate the accuracy of the model.

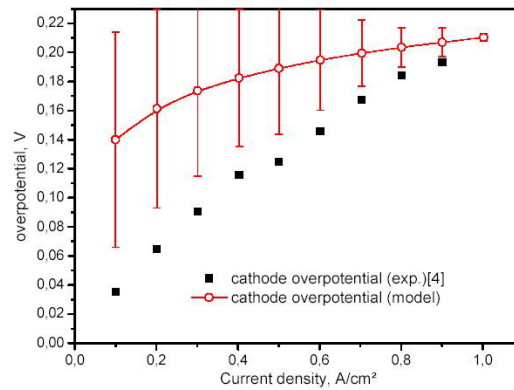


Fig. 4: Comparison between cathode over potential model and experimental data

For ohmic overpotential, Fig. 5, presents comparison between the simulating and experimental data from Ni *et al.* [8] of ohmic over potential, we introduce comparison with the model used by Choi *et al.* [4]. Data used for ohmic over potential are for  $T = 80\text{ }^{\circ}\text{C}$ ,  $L_{\text{PEM}} = 178\text{ }\mu\text{m}$ ,  $\lambda_c = 10$ ,  $\lambda_a = 10$ , the standard deviation = 0,00592 and the regression coefficient = 0,99266, this indicate the accuracy of the ohmic over potential model. We noticed that Ni *et al.* [8] is more accurate than Choi [4] model, this is because, the model used by Choi [4] is static rather than taking account of membrane thickness.

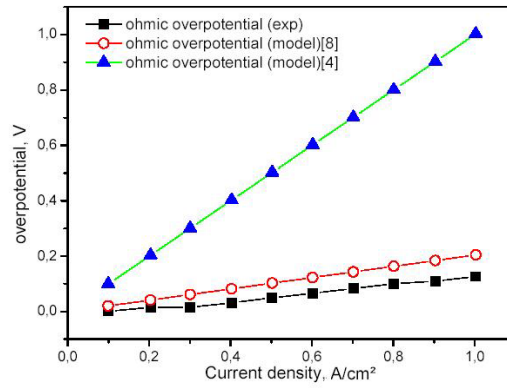


Fig. 5: Comparison between ohmic over potential model and experimental data

*For overall cell voltage*, The results of the comparison between experimental and simulated data are shown in Fig. 6. The standard deviation = 0,00496 and the regression coefficient = 0,99873.

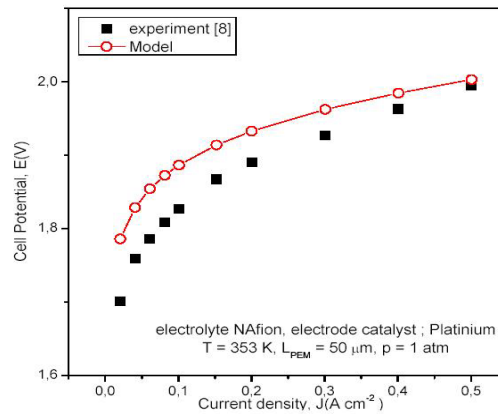


Fig. 6: Variation of electrolysis and enthalpy voltage with pressure at 298 K

We noticed that the simulated data are well correlated with the experimental results, this indicate that the model explain, somehow, the water electrolysis phenomena, the deviation depends on the hypothesis made.

#### 4. PARAMETRIC ANALYSIS

After evaluate the currency of the previous model in simulating PEMFC voltage, it is used to draw the effect of the different parameters that can have a significant effect on the cell voltage. (For the Nafion<sup>TM</sup> 117 membrane at  $T = 353 \text{ K}$  and  $p = 1 \text{ atm}$ , with Platinum electrode).

##### a- Effect of the thickness

We chose the following parameters to study the effect of the membrane thickness:

- The range of thickness choosing is  $[50 \mu\text{m}, 178 \mu\text{m}]$ .
- The water contents at the anode-membrane and cathode-membrane interface respectively are held constants as provided on **Table 2**
- Electrode catalyst is platinum based,  $T = 353 \text{ K}$ ,  $J_a^{\text{ref}} = 1.0 \times 10^{-5} \text{ Am}^{-2}$ ,  $J_c^{\text{ref}} = 10 \text{ Am}^{-2}$ .



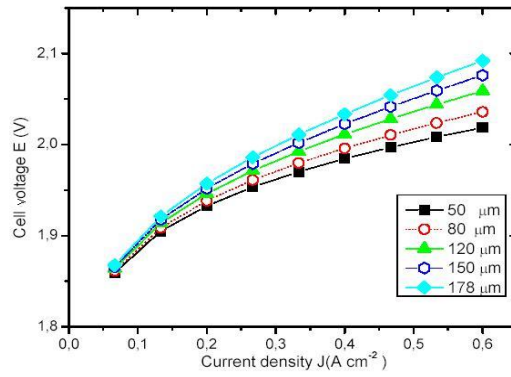


Fig. 7: The effect of membrane thickness the cell voltage SPE water electrolysis

The result shown in Fig. 7, for current density under the value of 0.1 A.cm<sup>-2</sup>, the thickness had no effect on the cell voltage, values greater than 0.1 A.cm<sup>-2</sup>, the cell voltage increases as the thickness increases, this is well understood considering that the resistance to ionic current decrease as the thinness increases, thinner membrane exhibit more resistance and thicker less resistance.

**b- Effect of the current exchange density**

The current exchange density introduced on the equation of Butler-Volmer have a great impact on the over potentials and then on the cell voltage. We study the effect of the current exchange density on the cell voltage the same way as done for the thickness effect.

We chose the following parameters to study the effect of exchange current density:

- Electrode catalyst is platinum based, the temperature is set at T = 353 K . The water contents at the anode-membrane and cathode-membrane interfaces respectively are held constants as provided on **Table 2**. The thickness choosing is 50 μm.
- The range of exchange current densities is provided on **Table 1**.
- For the effect of the anodic exchange current density, we choose  $J_c^{ref} = 10^{-3} \text{ A.cm}^{-2}$  .
- For the effect of the anodic exchange current density, we choose  $J_a^{ref} = 10^{-9} \text{ A.cm}^{-2}$  .

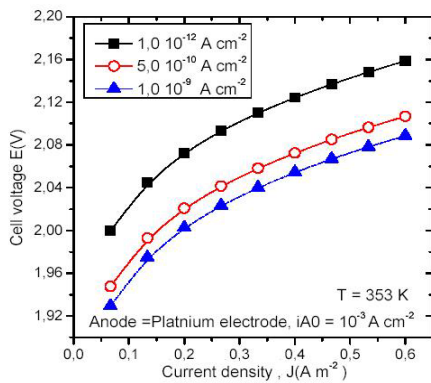


Fig. 8: The effect of anode exchange current density on the cell voltage of SPE water electrolysis

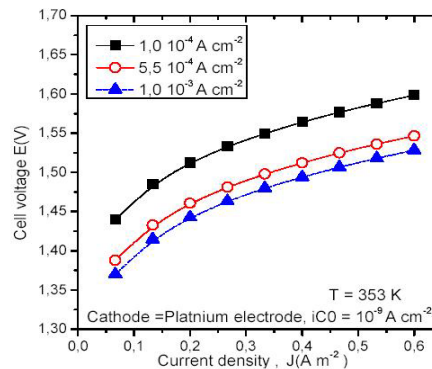


Fig. 9: The effect of cathode exchange current density on the cell voltage of SPE water electrolysis

As shown in Fig. 8, as the anodic current exchange density increases, the cell voltage decreases, the same observation is made in Fig. 9, as the cathodic current density increases, the cell voltage decreases. However, since the cathode reaction is relatively fast compared with anode reaction, the anodic current exchange have an additional impact (electrode activity and the reaction kinetic), thus the variation of electrolysis voltage cell with current density is mainly attributable to the slow kinetics of water dissociation [4].

## 5. CONCLUSION

This study deals with water electrolysis using a cell with Platinum electrodes and Solid Membrane Electrolyte, Nafion 117. After investigating theoretical aspect of water electrolysis a model based on the well known Butler–Volmer equation has been applied to test the concordance (the model) with the experiment published data. The model was used to understand the effect of some parameters on cell water electrolysis as the membrane thickness and the density exchange of the electrode and the anode, on which the water oxidation occurs, and the cathode, on which hydrogen reduction occurs. The simulation lead to numerical values that indicates the importance of the current exchange at the anode chambers, the phenomena at the anode-membrane interface is the key point of the entire efficiency of water electrolysis, this is show as over potential. Finally the result encourage our endeavour to develop a mathematical tool that serve as an expert system to optimize the different parts of SPE water electrolysis

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