# A new approach to empirical electrical modelling of a fuel cell, an electrolyser or a regenerative fuel cell

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## Abstract

In terms of fuel cell steady-state performance modelling, many electrical models have been developed either from a theoretical point of view or from an empirical point of view. The model described in this article is from the empirical point of view approach. This model enables to simulate both fuel cells and electrolysers $V$–$J$ curves (cell voltage versus current density) in typical conditions. This model is particularly adapted to regenerative fuel cell (RFC) simulation. It is a four degree-of-freedom model and it is convergent near zero current. It depends on the stack temperature and the oxygen partial pressure. The regions where mass transfer limitations occur have not been modelled, because they are usually avoided for efficiency or thermal reasons. The parameters have been fitted with a $4\,kW_e$ proton exchange membrane fuel cell (PEMFC) and a $3.6\,kW_e$ electrolyser. The electrical equations and the experimental data are well correlated.
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## 1. Introduction

The Centre d'Energétique of Ecole des Mines de Paris at Sophia Antipolis has gained experience in fuel cell (FC) studies for 10 years, and particularly in proton exchange membrane fuel cell (PEMFC). Since the development of the first fuel cell stack test bench, many works have been achieved in terms of fuel cell integration, hydrogen generation or storage and fuel cell modelling. For testing purposes, a dedicated fuel cell stack test bench is available, capable of testing fuel cells up to $15\,kW_e$ in various system configurations.

The laboratory is also doing research concerning hybrid systems made up of a renewable energy generator (photovoltaic field or PV), a back-up unit (diesel engine) and a storage system (batteries, fuel cell/gas storage/electrolyser).

For these projects two test facilities have been set up. The first one is a PV–battery–diesel test bench and the second one is a PV–electrolyser-fuel cell test bench.

This latter, constructed within the frame of the PVFC-SYS European project (ERK5-CT1999-00017) is an autonomous electricity generator including a PV field (3.6 kWp), an electrolyser (3.6 kW) splitting water into hydrogen and oxygen,

a gas storage unit ($4\,Nm^3$ of $H_2$ and $2\,Nm^3$ of $O_2$) and a PEMFC ($4\,kW_e$) to generate electricity during low sun shine periods.

This test bench, described in different articles [1–3] enables us to validate the model of each component and finally of the complete system.

Different approaches exist to simulate the electrical characteristics of a FC. There are roughly two kinds of fuel cell models.

The first one is the theoretical model (also named mechanistic model), which describes the electrochemical reactions occurring in the cells. In the model of Metkemeijer [4], the cell voltage is the thermodynamical voltage, minus the different overvoltages due to the ionic transfers at the anode and at the cathode, the resistive losses and the material transfer at high current density. Each term of the equation depends on the stack temperature and the partial pressure of hydrogen and oxygen. This approach requires the knowledge of nine parameters, which are difficult to determine.

The model of Amphlett [5], which is largely quoted in the literature, is also based on Nernst and Tafel equations. It considers all physical parameters in the system (effective pressure of oxygen and hydrogen, temperature, concentration of oxygen, hydrogen, water, proton). Since all these parameters cannot be identified, the authors use empirical means to estimate their values [6].

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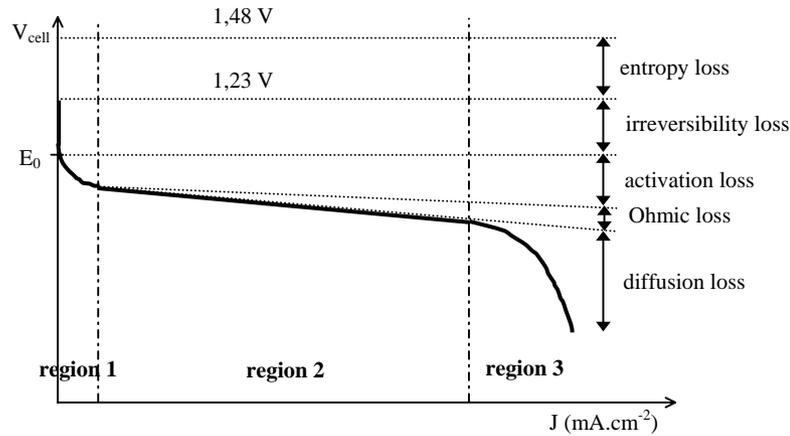
Fig. 1. Fuel cell characteristic—cell voltage–current density.

Since then, it has been improved and now allows to take into account the fuel cell parameters (such as active area and membrane thickness) and the voltage degradation with membrane ageing [7,8]. This model has to consider empirical and semi empirical parameters, like ohmic overvoltage and the equilibrium water content of the membrane.

In the second method, a semi empirical equation is used, of which parameters are calculated through an identification process with experimental data. The terms of this equation come from Nernst, Tafel and Ohm laws. This kind of model does not pretend to be universal but it is much simpler than theoretical ones.

The model of Chamberlin and Kim [9] describes the cell voltage depending on the current density, with five parameters (Eq. (1)).

$$V_{\text{cell}}(J) = E_0 - b\ln(J) - R \times J - m\exp(nJ) \qquad (1)$$

where $V_{\text{cell}}$ is the cell voltage (V), $J$ the current density ($A\,cm^{-2}$), $E_0$ the open circuit voltage (V), $b$ the Tafel's parameter for the oxygen reduction (V), $R$ the resistance ($\Omega\,cm^2$), $m$, $n$ is the diffusion's parameters (V and $A^{-1}$).

Each term of Eq. (1) is dominant in each region of the $V$–$J$ characteristic (Fig. 1). In region 1, the voltage decreases drastically due to the oxygen electrochemical activation reactions (logarithm term has the main influence). In region 2, the curve is roughly linear (resistive losses). The region 3 corresponds to the diffusion losses (exponential term).

In Kim's model, the five parameters ($E_0$, $b$, $R$, $m$, $n$) depend on the temperature and pressure in the cell and the oxygen partial pressure. They also depend on the stack itself, so that it cannot be transposed to another fuel cell without a new identification of the parameters.

This model fits very well with experimental results. Hamelin has compared the Amphlett's model and the Kim's one [12]. It appears that both predicted voltages are very close to each other and that they both differ slightly from experimental cell voltage in the same regions (low and high current densities).

However, it is a real issue to simulate the fuel cell behaviour in the first zone (i.e. for low current densities). Indeed, the Kim's model is divergent close to $J = 0$.

This problem particularly appears when modelling a regenerative fuel cell, a device that can be used either as an electrolyser or as a fuel cell, because equation (1) diverges each time the device passes continuously from one operation to the other one.

To solve this problem, we propose a new model passing through the divergence. Described in chapter II, the model is validated in chapter III for a fuel cell and an electrolyser running in regions 1 and 2.

To simulate an electrolyser, the same electrical equation is used, changing the value of the parameters.

The model is hence able to simulate a regenerative fuel cell throughout its complete behaviour.

## 2. Description of the model

The proposed model is very close to the classical semi empirical model of Kim with the advantage to be convergent for all current density. Furthermore, it is a purely empirical model. The electrical equation as well as the matlab program have been developed in the laboratory in 1999. It is a four-degree-of-freedom model.

This model does not take into account the diffusion area (region 3 in Fig. 1). Like in Kim's model, it is possible to add an exponential term to tackle this issue. The authors did not want to damage the fuel cell by using it in this region. Fuel cells are usually used in a region where no thermal or diffusion problems occur for all temperatures (normally for cell voltages > 0.5–0.6 V).

### 2.1. Electrical equation

To solve the convergence problem, we propose to change the logarithm term into (1/ln $J$), close to 0 for $J = 0$. The
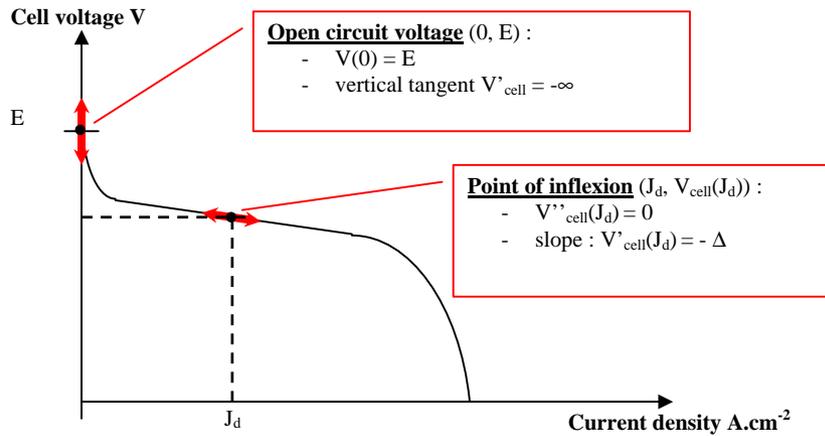
Fig. 2. Main hypothesis of the expounded model.

following equation is obtained:

$$V_{\text{cell}}(J) = E + \frac{b}{\ln(dJ)} - cJ \qquad (2)$$

where $V_{\text{cell}}$ is the cell voltage (V), $J$ the current density $(\text{A cm}^{-2})$, $E, b, c, d$ the parameters (V, V, $\Omega\,\text{cm}^{-2}$, $\text{cm}^2\,\text{A}^{-1}$).

The other hypotheses are summed up in Fig. 2.

Parameter $E$ has a physical meaning: it is the open circuit voltage.

The curve of the cell potential (V) versus the current density (J) has an inflexion point for $J = J_d$ and at this point, the slope of the tangent is noted $-\Delta$; it is the sum of activation and resistive losses (see Fig. 1).

From the two equations: $V''_{\text{cell}}(J_d) = 0$ and $V'_{\text{cell}}(J_d) = -\Delta$, we obtain two new equations:

$$d = \frac{1}{J_d e^2} \quad \text{and} \quad c = \Delta - \frac{b}{4J_d}$$

Replacing this terms in Eq. (2), we finally end up with:

$$V_{\text{cell}}(J) = E + \frac{b}{\ln(J/J_d) - 2} + \left( \frac{b}{4J_d} - \Delta \right) \times J \qquad (3)$$

where $E$ is the open circuit voltage (for $J = 0$), $b$, $J_d$, $\Delta$ the other parameters of the model (V, $\text{A cm}^{-2}$, $\Omega\,\text{cm}^2$).

## 2.2. Four parameters to be determined

The four parameters ($E$, $J_d$, $b$ and $\Delta$) have to be determined, fitting the modelling curve to the experimental data. These parameters depend on the cell temperature and on the oxygen partial pressure. The influence of the hydrogen partial pressure can be neglected as long as the hydrogen fraction on the anodic FC side is superior to 20% [10].

Due to the temperature and pressure dependence of the thermo chemical potential and the Butler Volmer equation, the influence of T and $p_{O_2}$ on the four parameters has the following form: $K_1 + K_2 T + K_3 T \ln(p_{O_2})$.

Therefore, for each parameter, three constants have to be determined (Eq. (4)) in order to describe the T and P dependence.

$$\begin{bmatrix} E \\ J_d \\ b \\ \Delta \end{bmatrix} = \begin{bmatrix} E_1 & E_2 & E_3 \\ J_{d1} & J_{d2} & J_{d3} \\ b_1 & b_2 & b_3 \\ \Delta_1 & \Delta_2 & \Delta_3 \end{bmatrix} \begin{bmatrix} 1 \\ T \\ T\ln(p_{O_2}) \end{bmatrix} \qquad (4)$$

$T$ in K, $p_{O_2}$ in bar.

To do so, a minimum of 16 data is required: four couples $(J, V_{\text{cell}})$ for four pairs $\{p_{O_2}, T\}$ where $p_{O_2}$ and $T$ must be at least at two different levels.

In case of the electrolyser, the oxygen partial pressure corresponds to the operating pressure of the electrolyser (pure oxygen is produced).

In case of the fuel cell, when pure oxygen is used, the partial pressure corresponds to the system pressure. When ambient air is used, the partial pressure of the oxygen depends on the oxygen molar fraction and on the oxygen utilisation (inverse of stoichiometric factor).

## 2.3. Matlab® model

To work out the four parameters ($E$, $J_d$, $b$ and $\Delta$), the mathematical method used is the least square method for non-linear problems. Knowing the [4 × 3] matrix of parameters, the cell voltage is a function of the current density, the temperature and the oxygen partial pressure.

## 3. Experimental: fitting of empirical equation

The fuel cell and the electrolyser were tested on the last test bench described in the introduction.

The electrolyser is a 3.6 kW alkaline one from Hydrogen Systems, which has 16 cells of $300\,\text{cm}^2$ connected in series. The periphery has been developed and adapted to the power and to the application. The cell temperature is calculated as the average between the two inputs and two outputs of the electrolyte in the cells. The pressure is measured in the hydraulic periphery.
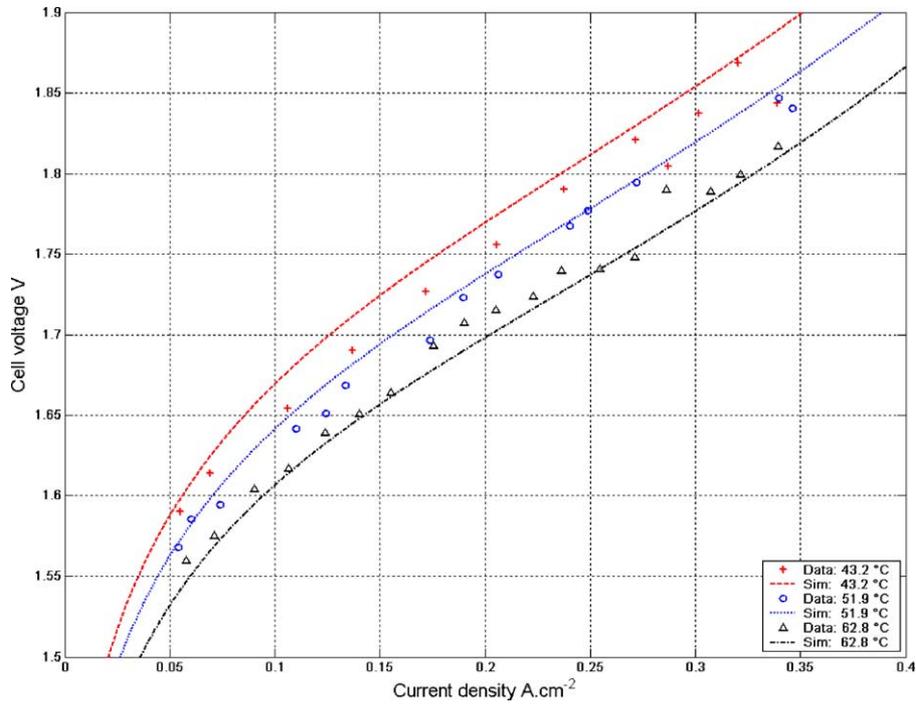
Fig. 3. Experimental data and simulation curves of the electrolyser—influence of the temperature ($P = 9.6$ bar).

The FC system (described in [11]) has been optimised to our specific application. The proton exchange membrane stack from DeNora, is made up of 18 cells of $900\,\text{cm}^2$.

The dedicated FC system enables to operate for variable oxygen (and hydrogen) partial pressures and at variable temperatures.

The system pressure can vary from 1 to 4 bar (abs). To obtain the variable oxygen and hydrogen partial pressures, oxygen and hydrogen were diluted with nitrogen. In order to avoid mismeasurements due to nitrogen migration across the membrane [10], hydrogen and oxygen partial pressures were always balanced.

In the FC, the cell temperature is not available. Usually, the temperature of the air output is selected because it represents most precisely the electrode-membrane-assembly temperature. In our approach, we try to simulate empir-
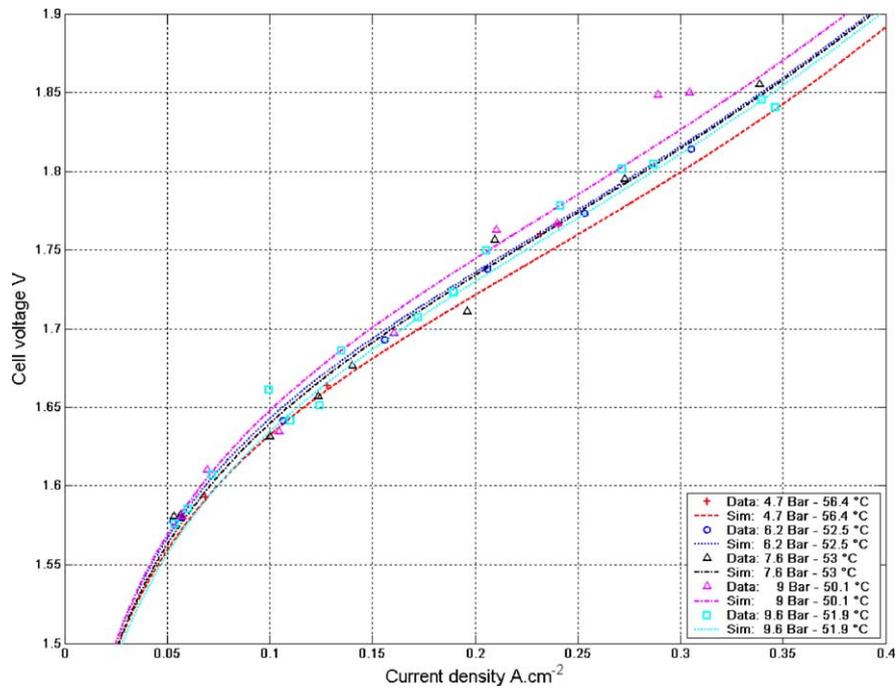


Fig. 4. Experimental data and simulation curves of the electrolyser—influence of the pressure ($T = 52$–$56\,^{\circ}\text{C}$).
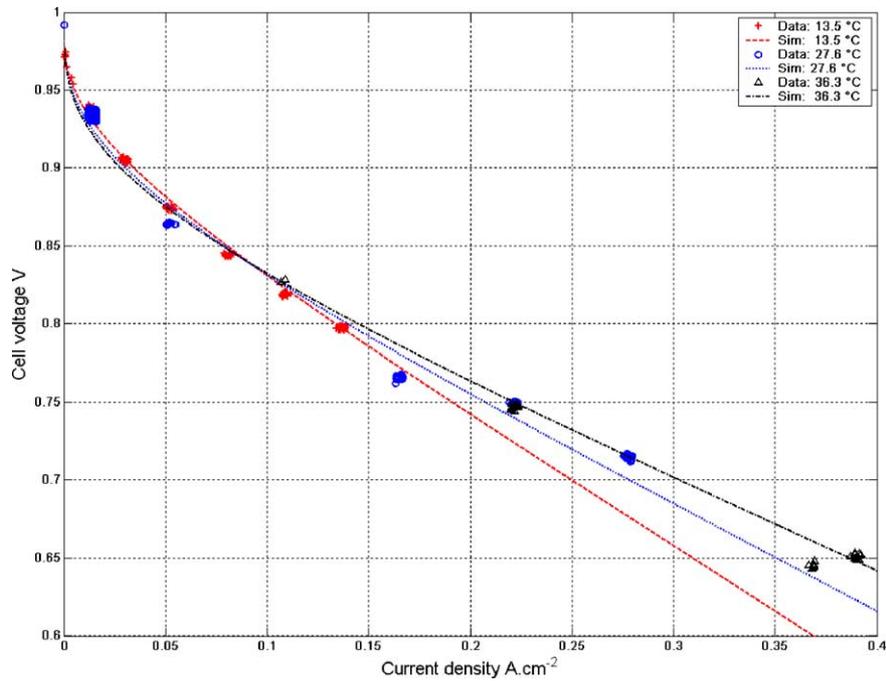
Fig. 5. Experimental data and simulation curves of the FC—influence of the temperature ($p_{O_2} = 1.4$ bar).

ically the complete FC system and then the temperature selection is not a crucial point. For technical reasons, the temperature used in the electrical equation is the temperature of the inlet cooling water (with an outlet cooling water temperature maximum 5 °C above inlet temperature). For this stack, the optimal temperature has been evaluated to 35 °C.

### 3.1. Results

Each constant has been determined for the electrolyser and for the FC as a function of temperature and oxygen partial pressure by using the experimental results obtained with the FC and the electrolyser previously mentioned (see Figs. 3–6). Although the experimental data are a bit dis-
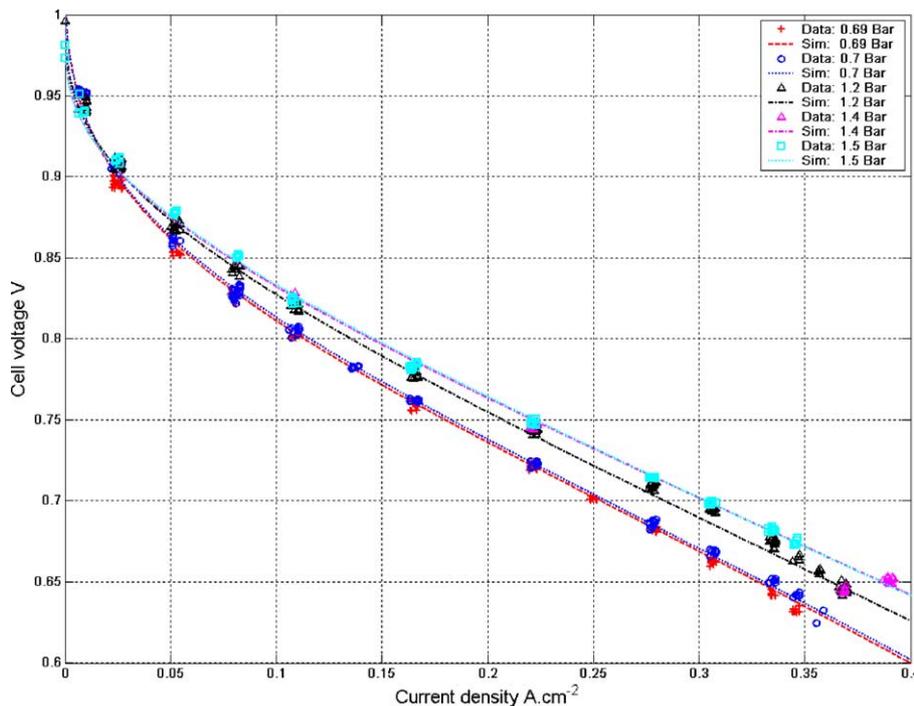


Fig. 6. Experimental data and simulation curves of the FC—influence of the pressure ($T = 35$ °C).

Table 1
Constants of the electrical model: set of values for the electrolyser (left) and for the fuel cell (right)

|         | Electrolyser |          |          | Fuel cell |          |          |
|---------|--------------|----------|----------|-----------|----------|----------|
| $J_d$   | 0.355        | −1.078E−3 | 5.232E−4 | −0.868    | −1.762E−3 | 4.9E−4   |
| $E$     | 1.025        | 1.24E−3  | −1.05E−4 | 0.796     | 4.661E−3 | −3.22E−4 |
| $b$     | −6.125       | 1.57E−2  | −3.245E−4 | −1.926   | 2.616E−2 | −1.489E−3 |
| $\Delta$ | −1.819      | 4.83E−3  | −1.248E−4 | 3.892    | −7.447E−3 | −2.696E−4 |

$J_d$: $\mathrm{A\,m^{-2}}$; $E$: V; $b$: V; $\Delta$: $\Omega\,\mathrm{m^2}$.

persed due to the noise in the measurement, the fitting protocol enables to select the most representative curve. The identification process leads to the two following [4 × 3] matrixes (Table 1).

### 3.2. Discussion

To validate the model, we can observe the influence of the temperature and the oxygen partial pressure. We will point out the limits of the model validity, but before it should be born in mind that the model prediction is only valid for the specific equipment and inside the temperature and pressure range used to fit the parameters (usually real operating limits). For the electrolyser, the experimental pressure is between 1.7 and 9.9 bar (abs) and the temperature varies between 31 and 63 °C. For the fuel cell, the oxygen and hydrogen partial pressures range from 0.7 to 1.5 bar. The temperature range is 15–36 °C.

#### 3.2.1. Temperature influence
The Figs. 3 and 5 plot the *V–J* characteristics of the electrolyser and the FC at fixed pressure for different temperatures. As expected, the temperature increase induces a better performance of both the electrolyser and the FC in terms of voltage efficiency.

#### 3.2.2. Pressure influence
The pressure has an important effect on the FC performance (Fig. 6) but not on the electrolyser's one (Fig. 4). More precisely, the high pressure has an influence only on the purity of gases produced by electrolysis, but not on the reaction efficiency. A model of the gas purity will be added later, considering the flux crossing through the membrane depending on the electrolyser pressure. As predictable, the oxygen partial pressure increase induces a better performance of the FC. As the system pressure is 3 bar absolute, the oxygen concentration varies between 25% (0.7 bar) and 50% (1.5 bar). For safety reasons, we did not measure in regions higher than 50% of oxygen.

### 3.3. Conclusions concerning the electrical model

The proposed model is a first step model. It is a satisfactory model to simulate an electrolyser or a FC for low and medium current density. This model is innovative due to its convergence at zero current density and its mathematical coherence.

The model shows a good fit of the empirical equation to the experimental data and the influence of the pressure and temperature parameters is correct compared to the physical behaviour of the two electrochemical components.

## 4. System modelling

The electrical model is only a part of the complete electrolyser or fuel cell model. As seen above, the electrical model needs two inputs: the temperature and the oxygen partial pressure. Two sub models are essential to entirely simulate the electrochemical devices. The first one is the thermal model, allowing to simulate the evolution of the temperature during the component operation. The second one is the gas management model calculating particularly the oxygen partial pressure.

The Fig. 7 shows the electrolyser model, programmed with Matlab/Simulink®, made of three main modules allowing to calculate the electrical operating points, the temperature and the pressure of the electrolyser system. The fuel cell system uses the same program architecture. The electrolyser model has two inputs (power and ambient temperature) whereas the FC model has three inputs (power, ambient temperature and oxygen partial pressure).

Other sub modules could be of great importance to complete the simulation depending on its application. Indeed, the simulation of the intrinsic consumption of the peripheries is required to perform the evaluation of the complete system efficiency, which can be done by a dedicated sub module. In addition, in some particular applications, transient phenomena could occur, which can be managed by another sub module.

### 4.1. Thermal model

To estimate the temperature evolution during operation, the following equation can be used.

$$C_p \left( \frac{\mathrm{d}\theta}{\mathrm{d}t} \right) = P_{\mathrm{th}} - \varphi_{\mathrm{ext}} - \varphi_{\mathrm{ech}} - \varphi_{\mathrm{gaz}}$$

$$\theta = T - T_a$$

$$P_{\mathrm{th}} = \pm n_c (U - U_{\mathrm{tn}})I \qquad \text{(thermal dissipative power)}$$

$$\varphi_{\mathrm{ext}} = h\theta \qquad \text{(heat flow lost in the atmosphere)}$$
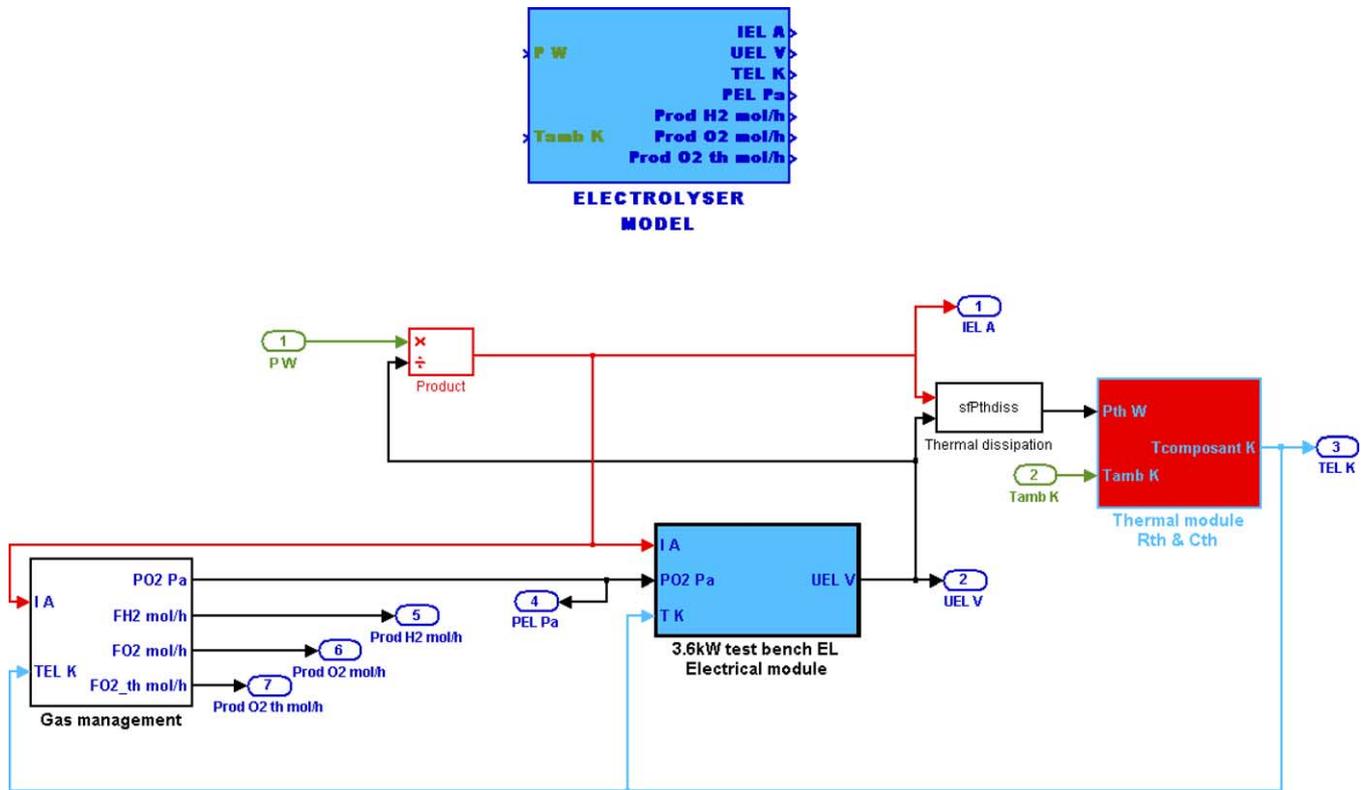
Fig. 7. The complete electrolyser model (without transient module) with Matlab/Simulink®.

$$\varphi_{\text{gaz}} = \pm(C_{p(\text{H}_2)}F_{(\text{H}_2)} + C_{p(\text{O}_2)}F_{(\text{O}_2)}) \times \theta$$

(heat flow lost in gas) (+for the electrolyser,

− for the fuel cell)

where $C_p$ is the thermal capacity of the device ($\text{J K}^{-1}$), $T$ the temperature (K), $T_a$ the ambient temperature (K), $n_c$ the number of cells, $U$ the cell voltage (V), $U_{\text{tn}}$ the thermoneutral cell voltage (=1.48 V) based on the higher heating value of hydrogen, $I$ the current (A), $h$ the heat transfer coefficient ($\text{W K}^{-1}$), $\varphi_{\text{ech}}$: energy flow lost in a heat exchanger ($\text{W K}^{-1}$), $C_{p(i)}$ the molar heat capacity of the gas $i$ ($\text{J mol}^{-1}\,\text{K}^{-1}$), $F_{(i)}$ the molar flow of the gas $i$ ($\text{mol s}^{-1}$).

The thermal dissipative power $P_{\text{TH}}$ is calculated knowing the electrical operating point (current–voltage). The two coefficients, $Cp$ and $h$, are fitted with experimental data. Depending on the system component, the heat transfer coefficient $h$ can depend on the current (usually for the alkaline electrolyser due to the electrolyte circulation proportional to the current).

This thermal module can contain a heat exchanger ($\varphi_{\text{ech}}$) model allowing to regulate the device temperature and to calculate the heat production of the system.

### 4.2. Pressure model

The gas management module enables to calculate gas production or consumption and the periphery pressure level during operation. The gas purity of the electrolyser products will be simulated shortly.

To obtain the gas production or consumption, the two next equations are used:

$$F_{\text{gas}} = \frac{n_c I}{nF} \times \eta_F(\text{EL}) \qquad F_{\text{gas}} = \frac{n_c I}{nF} \times \frac{1}{\eta_F}(\text{FC})$$

where $F_{\text{gas}}$ is the gas consumption for the fuel cell and gas production for the electrolyser ($\text{mol s}^{-1}$), $n_c$ the number of cells, $I$ the component current (A), $\eta_F$ the faraday efficiency (%), $F$ the faraday constant (96485 $\text{C mol}^{-1}$), $n$ the number of moles of electrons transferred per mole of water ($n = 2$ for hydrogen, $n = 4$ for oxygen).

The pressure calculation of the electrolyser can be interesting according to the electrolyser system and its application, although it has few influence on its electrical behaviour. During the start up of the device, the periphery pressure increases slightly until the maximal operating pressure. For our system (operating pressure at 10 bar), it usually lasts about 20 min but it could be longer for a high-pressure electrolyser. Moreover, pressure has an influence on the gas purity. Especially, hydrogen tends to accumulate in the oxygen circuit, which may lead to explosive mixtures. Electrolyser pressure calculation is therefore essential.

As for the fuel cell system, the oxygen partial pressure is fixed at the start up of the device and remains constant during

operation, the gas management module calculates only gas consumption.

To calculate the gas management unit results, some parameters have to be determined experimentally: faraday efficiency, gas flux depending on the periphery pressure, etc.

## 5. General conclusions

The model presented here is a novel empirical model to precisely calculate the major part of the *V–J* characteristic of a fuel cell, an electrolyser or a reversible fuel cell. With a set of experimental data, the parametric electrical equation can be easily fitted with any kind of fuel cell or electrolyser. This model, which is mathematically consistent and convergent near zero, allows the operator to interpolate experimental results of his studied device running in regions where current density does not cross the mass transfer limitations. This model will be soon used to study the influence of the hydrogen partial pressure on the FC performance. In case of validation, the model would enable to simulate FC behaviour supplied with different fuel inputs.

The electrical model is an essential module of an electrochemical device simulation but many other points are important during the operation. Each module proposed in the fourth part can be useful depending on the use of the electrolyser or the fuel cell. Nevertheless, in our application all these models are needed to perfectly simulate the complete system.

## Acknowledgements

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