

## PEM fuel cell fractional order modeling and identification

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**Abstract:** This paper deals with the modeling and identification of a fuel cell system using two models. It presents a comparative study between a conventional impedance model based on an equivalent electrical circuit and a fractional order model of fuel cell. In the first case, a least square method is used to identify the parameters of the model. While the second case considers a fractional order model and proposes a parameter identification approach based on the least square method adapted to fractional order models. By using these two models, this study aims to characterize the fuel cell impedance and identify the model's parameters in the objective of establishing a diagnosis method of the flooding state of the fuel cell. Experimental results, obtained from measurements made using a single PEM fuel cell, show that fractional order impedance models describe the real system behavior better than the impedance models of integer order. The goal of this work is to develop models and identification methods in view of implementing a diagnosis methodology of the fuel cell internal state in order to apply this technology to automotive sector.

*Keywords:* PEMFC, Fuel cell modeling, characteristic impedance, fractional order model, parameter identification

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### 1. INTRODUCTION

Proton Exchange Membrane Fuel Cell (PEMFC) is presently the subject of several studies and many industrial projects. One reason for the increasing interest in FC is that new environmental standards are becoming more and more stringent leading for instance to the development of electric vehicles market around the world. Research conducted in this field tends to remove some locks that prevent the system from being fully operational. Monitoring precisely the FC internal state is one way to improve the operating of a fuel cell. Diagnosis methods, which allow this, need reliable models able to accurately describe the behavior of the system. Diffusion phenomena occurring in a fuel cell are described by partial differential equations. This suggests that the most suitable model for this kind of system is a fractional order model.

A conventional method to characterize the PEMFC is the Electrochemical Impedance Spectroscopy (EIS). In this method, a small current variation is applied around the nominal operating point. Using the voltage measurement, the FC electrical impedance can be deduced over a wide range of frequencies, typically between 0.1 (Hz) to 1 (kHz). Several studies had used impedance spectroscopy as a tool for characterization of the fuel cell as in Fouquet,N.(2006), Sadli,I. (2006). Fontes,G. (2005).

The PEMFC models proposed are based on the identification of equivalent circuit parameters. Many works have already been done in this direction: Fouquet,N.(2006), Sadli,I. (2006). Fontes,G. (2005)., Philippoteau, V. (2009)., Turpin, C., Jorcin, J.B., Aglzim,E.H.,

This article deals with the fuel cell model's parameters estimation. It compares the identification results of two Fuel cell models. The first one is a conventional integer order impedance model. The second one is a fractional order fuel cell impedance model. The aim of this paper is to prove that fractional order models are more reliable than classical impedance models which use integer order transfer functions. With this objective in mind, a methodology of fractional order model's parameters estimation is proposed. The results are validated using experimental measurements, current-voltage time-series, obtained on a single fuel cell.

The second section of this paper will consider the fuel cell modeling. In the first part of this section, a brief overview of a fuel cell is introduced before presenting the fuel cell integer order impedance model. Its parameters are identified in this sub-section using the least square method. Subsequently, a fuel cell fractional order model (FOM) is developed. The first part of this sub-section explains the development which leads to a fractional order fuel cell transfer function. The third section, presents more precisely the identification method of

fractional order models. In the fourth section, the identification results are presented and compared. And finally, some conclusions and perspectives are proposed in section five.

## 2. FUEL CELL IMPEDANCE MODEL

Fig. 1 shows the structure of a fuel cell. Each unit cell is composed of an electrolyte (polymer membrane), sandwiched in between two electrodes (cathode and anode). The current  $I$  and the voltage  $V_{Cell}$  characterize the fuel cell electrical impedance.

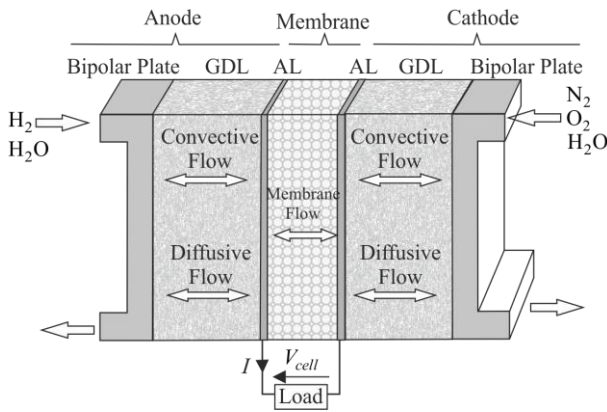


Fig. 1. Key components of a single fuel cell equivalent electrical circuit model

### 2.1 Equivalent electrical circuit model

In this type of model, every phenomenon occurring in the fuel cell is studied independently and represented by equivalent electrical circuit using active or passive components. The voltage of a fuel cell is the sum of the theoretical cell voltage and the voltage drops due to the activation phenomena, the diffusive phenomena and the membrane resistance.

$$V_{Cell} = U_{th} - \eta_{act} - |\eta_{diff}| - R_m I \quad (1)$$

Theoretical Fuel Cell voltage is deduced from the Nernst equation and given by:

$$U_{th} = U_0 + \frac{RT}{nF} \ln \left( P_{H_2} (P_{O_2})^{\frac{1}{2}} \right) \quad (2)$$

The voltage drops are given by:

$$\eta_{diff} = \frac{RT}{\beta n F} \ln \left( 1 - \frac{I}{I_{lim}} \right) [V]; \quad \eta_{act} = -\frac{RT}{\alpha n F} \ln \left( \frac{I}{I_{act}} \right) [V] \quad (3)$$

The model presented in this section is based on the work of Fontes,G. (2005) and Fontes,G and al (2004). Using these relations, Fontes proposed a FC (Fuel Cell) nonlinear dynamic model valid even in the cases of high signal variations. The schematic of Figure 2 shows the equivalent

electrical circuit of FC representation. The voltage drops due to activation and diffusion losses are represented by voltage generator controlled by current. Fontes defines a two-layer diffusion model. (Gas Diffusion Layer (GDL) and Active Layer (AL) are two different parts). Whereas, for simplicity, we only consider that diffusion is uniform in both GDL and AL.

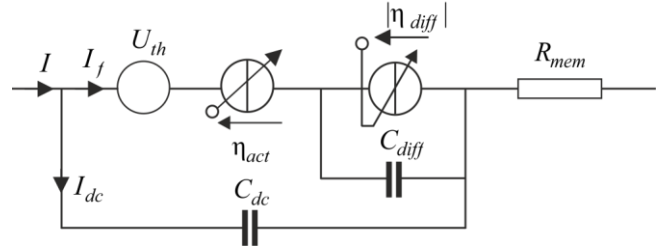


Fig. 2. One diffusion layer high signal dynamic model of Fuel Cell. Fontes.G (2006)

The linearized model around a steady state operating point is represented by the Fig3 with:

$$R_{diff} = \frac{\partial |\eta_{diff}|}{\partial I_{diff}} ; \quad R_{act} = \frac{\partial |\eta_{act}|}{\partial I_{act}} \quad (4)$$

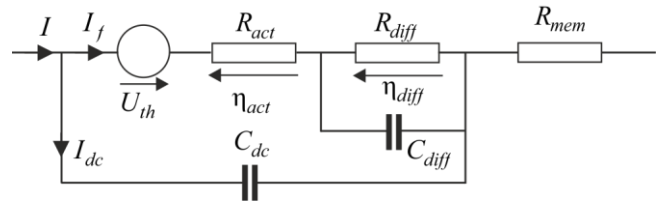


Fig. 3. Linearized one diffusion layer dynamic model of Fuel Cell. Fontes.G (2006)

This model's parameters are identified using the least square method.

$$\hat{\theta}(t) = \left[ \sum_{i=1}^t \varphi(i-1) \varphi^T(i-1) \right]^{-1} \sum_{i=1}^t y(i) \varphi(i-1) \quad (5)$$

The main incoherence of the conventional integer order models (IOM) is that the phase shift at high frequencies is multiple of 90, which doesn't match the real behavior of the impedance fuel cell. Indeed, many studies show that the fuel cell impedance at high frequencies is characterized by a phase shift of 45°, Fouquet,N.(2006), Sadli,I. (2006),, Iftikhar, M.U., and al (2006) and Sailler,S, which IOM can reproduce only on a limited frequency band. For a better representation of this property, a formulation of the impedance according to a FOM is considered in the following.

## 2.2 Fractional order model of fuel cell

This section develops a fractional model of the fuel cell based on the work of Fouquet, N. (2006), Sadli, I. (2006), Fontes, G. (2005), Phlippoteau, V. (2009), and some works of CRONE-IMS-Bordeaux which deal with the fractional order derivatives and identification methods of non-integer order models, like: Oustaloup, A. (1983), Malti, R., and al. (2003), Victor, S. (2010), Cois, O. (2002), Le Lay, L. (1998).

This model should describe better the diffusion phenomena than the linear model. Its representation by a transfer function can be adapted for the parameters identification.

### 2.2.1 Derivative of non-integer order

A model of non-integer order described by a fractional differential equation, where the derivation orders are real numbers, is given by:

$$y(t) + a_1 D^{n_{a1}} y(t) + \dots + a_N D^{n_{aN}} y(t) = b_0 D^{n_{b0}} u(t) + b_1 D^{n_{b1}} u(t) + \dots + b_m D^{n_{bm}} u(t) \quad (6)$$

with

$$\begin{cases} 0 < n_{a1} < n_{a2} < \dots < n_{aN} \\ 0 \leq n_{b1} < n_{b2} < \dots < n_{bm} \end{cases} \quad (7)$$

$D^\nu$  is a generalized fractional order derivative (FOD) operator, defined by Riemann and Liouville as an integer order derivative of  $m = \lfloor \nu \rfloor + 1$  order of a non integer order integral (where  $\lfloor \cdot \rfloor$  is the integer part). Cois, O. (2002).

$$D^\nu x(t) = \frac{1}{\Gamma(m-\nu)} \left( \frac{d}{dt} \right)^m \left( \int_0^t \frac{x(\tau) d\tau}{(t-\tau)^{1-(m-\nu)}} \right) \quad (8)$$

The next section will present a fractional order impedance model of fuel cell. The derivative orders of this model will be described by the generalized fractional operator defined above.

### 2.2.2 Fractional model

As mentioned before, gas diffusion phenomena are not properly represented by integer order. Indeed, the phenomenon of diffusion is described by a partial differential equation given by (9)

$$\frac{\partial C_i(x, t)}{\partial t} = D_i \frac{\partial^2 C_i(x, t)}{\partial x^2} \quad (9)$$

Using Laplace transform, diffusion phenomena can be described in small signal approach by the diffusion impedance named "Warburg impedance".

$$Z_W(s) = R_d \frac{\tanh[(\tau s)^{1/2}]}{(\tau s)^{1/2}} \quad (10)$$

A small signal model can be deduced. It is represented by an equivalent electrical circuit, where the principal characteristic element is the Warburg impedance (Fig.4).

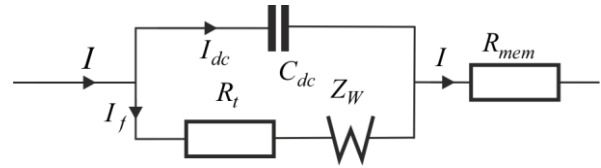


Fig. 4. Equivalent electrical circuit of Fuel Cell with Warburg impedance

Then, the transfer function which models the fuel cell is given by:

$$H(s) = \frac{1}{\frac{1}{R_t + Z_W} + sC_{dc}} + R_{mem} \quad (11)$$

The main problem in this expression resides in the Warburg impedance which is a function of a hyperbolic tangent of fractional order. This can cause numerical instability. To remedy this problem, we propose to approximate the Warburg impedance.

In Iftikhar, M.U., and al (2006). and Sailler, S., a comparison between a model type "RC circuit" of 20 cells and the fractional model previously defined is presented. For this second model, the authors propose to approximate Warburg impedance (10) by decomposing the hyperbolic tangent as a truncated Taylor series to second order to simplify the expression of the impedance. This approximation, valid in the vicinity of zero, is nevertheless valid for high frequencies. The Warburg impedance is then given by:

$$Z_W(s) = \frac{R_d}{\sqrt{1+s\tau}} \quad (12)$$

To formulate a fractional transfer function of the impedance model, the square root in the Warburg impedance expression can also be approximated by a Taylor series expansion.

Retaining the first three terms of the expansion:

$$\sqrt{1+s\tau} \approx \sqrt{s\tau} + \frac{1}{2} \left( \frac{1}{s\tau} \right)^{\frac{1}{2}} - \frac{1}{8} \left( \frac{1}{s\tau} \right)^{\frac{3}{2}} + o((s\tau)^3) \quad (13)$$

the approximation error is less than 3% and its average value is 0.09%. Figure 5 shows the error between the real expression of Warburg impedance and its approximation.

Using (15), (12) and (10), the equation (11) leads to the new fractional transfer function given by:

$$H(s) = \frac{b_0 + b_1 s + b_2 s^{\frac{3}{2}} + b_3 s^2 + b_4 s^{\frac{5}{2}} + b_5 s^3}{a_0 + a_1 s + a_2 s^2 + a_3 s^{\frac{5}{2}} + a_4 s^3} \quad (14)$$

where  $a_i$  and  $b_j$  are functions of the physical parameters.

$$\begin{aligned} a_i &= f_i(R_d, R_t, R_{mem}, C_{dc}, R_d, \tau), i = 0, \dots, 4 \\ b_j &= g_j(R_d, R_t, R_{mem}, C_{dc}, R_d, \tau), j = 0, \dots, 5 \end{aligned} \quad (15)$$

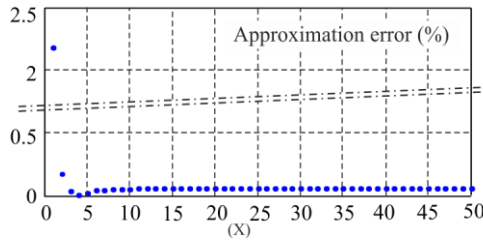


Fig. 5. Error between the Warburg impedance and its approximation

### 3. PARAMETER IDENTIFICATION OF THE FRACTIONAL ORDER IMPEDANCE MODEL

#### 3.1 Identification of fractional order model's parameters

This section deals with the parametric identification of a fractional order model. The method used is based on the least square method adapted to non-integer order models. This method was developed by CRONE- IMS-Bordeaux, in Oustaloup, A. (1983), Malti, R., and al. (2003), Victor, S. (2010), Cois, O. (2002), Le Lay, L. (1998).

In the case of IOM, derivation orders are implicitly distributed by a unit step between two successive orders. The classical identification methods, used in linear system cases, estimate only the differential equation coefficients. However, in the case of fractional order models, it is necessary to estimate the orders and coefficients of derivation. Three identification strategies can be used:

- The first one involves having a prior knowledge of derivation orders and therefore estimating only the coefficients of the derivative operators by Least Square method for example.
- The second approach consists in estimating the coefficients and orders of the derivative operators, when unknown, by non-linear algorithms.
- The third method estimates the derivative operators coefficients and only one derivation order named "commensurate fractional order", as all other orders are integer multiples of the commensurate order.

In this paper, the first case is considered and the Least Square identification method adapted to fractional order models is used to estimate the model's parameters.

#### 3.2 Equation error method: Least square method adapted to fractional order Models

This section will develop a method of parametric estimation, based on optimization approach, applied to the model

described by (18) where the derivative orders are a priori known and the coefficients are estimated.

$$\begin{aligned} D^{n_{a_0}} y(t) + a_1 D^{n_{a_1}} y(t) + \dots + a_l D^{n_{a_l}} y(t) + \dots + a_L D^{n_{a_L}} y(t) = \\ b_0 D^{n_{b_0}} u(t) + \dots + b_m D^{n_{b_m}} u(t) + \dots + b_M D^{n_{b_M}} u(t) \end{aligned} \quad (16)$$

$n_{a_l}, n_{b_m}$  are reals positives numbers, integers or no-integers (fractions) and supposed known.

$a_l, b_m$  are the coefficients of the derivative operators supposed unknown.

Relation (18) is a continuous time model but the identification method implementation is realized in discrete time. So, the optimization method is composed of four parts:

- Discretization of the continuous time fractional model
- Linear formulation of the model by a change of variables.
- Estimation of the new parameters using the least square method.
- Return to the initial parameters by reversing the variable change.

##### 3.2.1 Model discretization

Equation (18) is discretized using the Grünwald approximation of the FOD given by (19)

$$D^n f(Kh) = \frac{1}{h^n} \sum_{k=0}^K (-1)^k C_n^k f((K-k)h) \quad (17)$$

where  $h$  is the sample period and  $C_n^k$  is given for all integers  $k > 0$  by:

$$C_n^k = \frac{n(n-1)(n-2)\dots(n-k+1)}{k!} \quad (18)$$

and  $C_n^k = 1$  for  $k = 0$ .

The continuous time model is described by the next equation:

$$\sum_{l=0}^L a_l D^{n_{a_l}} y(t) = \sum_{m=0}^M b_m D^{n_{b_m}} u(t) \quad (19)$$

with  $a_0 = 1$ . In the next part, the sampled signals, at the sampling period  $h$ , are noted as  $x[K-k] = x((K-k)h)$ .

After discretization using the Grünwald approximation, the model becomes:

$$\begin{aligned} \sum_{l=0}^L \frac{a_l}{h^{n_{a_l}}} \sum_{k=0}^K (-1)^k C_{n_{a_l}}^k y[K-k] = \\ \sum_{m=0}^M \frac{b_m}{h^{n_{b_m}}} \sum_{k=0}^K (-1)^k C_{n_{b_m}}^k u[K-k] \end{aligned} \quad (20)$$

### 3.2.2 Linear form of the model

The objective of this step is to express the model output at  $Kh$  as a function of the previous inputs and outputs. This formulation allows us to define the FOM identification as a problem of linear model identification. Relation (22) can be written as:

$$\sum_{l=0}^L \frac{a_l}{h^{n_{a_l}}} \left[ y(Kh) + \sum_{k=1}^K (-1)^k C_{n_{a_l}}^k y[K-k] \right] = \sum_{m=0}^M \frac{b_m}{h^{n_{b_m}}} \sum_{k=0}^K (-1)^k C_{n_{b_m}}^k u[K-k] \quad (21)$$

Note that  $y(0) = 0$  and  $C_{n_{a_0}}^0 = 1$ . By isolating the term  $y[K]$  we can write as:

$$y[K] = - \frac{\sum_{l=0}^L \frac{a_l}{h^{n_{a_l}}} \left[ \sum_{k=1}^K (-1)^k C_{n_{a_l}}^k y[K-k] \right]}{\sum_{l=0}^L \frac{a_l}{h^{n_{a_l}}}} + \frac{\sum_{m=0}^M \frac{b_m}{h^{n_{b_m}}} \sum_{k=0}^K (-1)^k C_{n_{b_m}}^k u[K-k]}{\sum_{l=0}^L \frac{a_l}{h^{n_{a_l}}}} \quad (22)$$

This relation is not linear with respect to parameters. So a linear form can be obtained using a variable change and define a new parameters set  $(a'_0, \dots, a'_L, b'_0, \dots, b'_M)$ :

$$y[K] = - \sum_{l=0}^L a'_l Y_l[K] + \sum_{m=0}^M b'_m U_m[K] \quad (23)$$

where:

$$a'_l = \frac{a_l}{h^{n_{a_l}}} \quad ; \quad b'_m = \frac{b_m}{h^{n_{b_m}}} \quad (24)$$

and

$$Y_l[K] = \sum_{k=1}^K (-1)^k C_{n_{a_l}}^k y[K-k] \quad (25)$$

$$U_m[K] = \sum_{k=0}^K (-1)^k C_{n_{b_m}}^k u[K-k]$$

$$\text{with } 0 \leq l \leq L, \quad 0 \leq m \leq M \quad \text{and} \quad \sum_{l=0}^L a'_l = 1 \quad (26)$$

### 3.2.3 Parametric estimation

The new set of parameters  $\hat{\theta}_r = (\hat{a}'_0, \dots, \hat{a}'_L, \hat{b}'_0, \dots, \hat{b}'_M)$  is estimated from the constraint (28) which

become  $a'_0 = 1 - \sum_{l=1}^L a'_l$ . Using this constraint, we can express the model output linearly with respect to the vector of parameters  $\hat{\theta}_r$ .

$$\hat{y}[K, \hat{\theta}_r] = -\hat{a}'_0 Y_0[K] - \sum_{l=1}^L \hat{a}'_l Y_l[K] + \sum_{m=0}^M \hat{b}'_m U_m[K] = - \left( 1 - \sum_{l=1}^L \hat{a}'_l \right) Y_0[K] - \sum_{l=1}^L \hat{a}'_l Y_l[K] + \sum_{m=0}^M \hat{b}'_m U_m[K] = -Y_0[K] - \sum_{l=1}^L \hat{a}'_l (Y_l[K] - Y_0[K]) + \sum_{m=0}^M \hat{b}'_m U_m[K] \quad (27)$$

For  $N$  measurement points between  $Kh$  and  $(K+N)h$ , linear matrix equation is given:

$$\hat{Y}(\hat{\theta}_r) = -Y_0 + \phi \hat{\theta}_r, \quad \text{where} \quad \phi = \begin{bmatrix} -Y_1[K] + Y_0[K] & \dots & -Y_L[K] + Y_0[K] & U_0[K] & \dots & U_M[K] \\ \vdots & & \vdots & \vdots & & \vdots \\ -Y_1[K+N] + Y_0[K+N] & \dots & -Y_L[K+N] + Y_0[K+N] & U_0[K+N] & \dots & U_M[K+N] \end{bmatrix} \quad (28)$$

The estimation problem consists to search an optimal vector of parameters,  $\hat{\theta}_r$  using the least square method, where the solution is given by:

$$\hat{\theta}_{r,opt} = (\phi^T \phi)^{-1} \phi^T (Y + Y_0) \quad (29)$$

### 3.2.4 Inversion of the variable change

Initial coefficients,  $a_l$  of the differential equation (18) are obtained from the estimated parameters by the linear system resolution (32):

$$\begin{bmatrix} (a'_1 - 1)h^{-n_{a_1}} & a'_1 h^{-n_{a_2}} & \dots & a'_1 h^{-n_{a_L}} \\ a'_2 h^{-n_{a_1}} & (a'_2 - 1)h^{-n_{a_2}} & \dots & a'_2 h^{-n_{a_L}} \\ \vdots & \vdots & \vdots & \vdots \\ a'_L h^{-n_{a_1}} & a'_L h^{-n_{a_2}} & \dots & (a'_L - 1)h^{-n_{a_L}} \end{bmatrix} \begin{bmatrix} a_1 \\ a_2 \\ \vdots \\ a_L \end{bmatrix} = \begin{bmatrix} -a'_1 h^{-n_{a_0}} \\ -a'_2 h^{-n_{a_0}} \\ \vdots \\ -a'_L h^{-n_{a_0}} \end{bmatrix} \quad (30)$$

while the coefficients  $b_m$  are expressed by:

$$b_m = b'_m \sum_{l=0}^L a_l h^{n_{b_m} - n_{a_l}} \quad (31)$$

Knowing the derivative orders of the transfer function, we can estimate its coefficients using the previous method.

## 4. EXPERIMENTATION AND RESULTS

This section presents the results of the FOM identification. These results will be compared to the identification of an IOM using the classical least square method

The measures used to identify the model's parameters were made at LGEP (Laboratoire de Genie Electrique de Paris) on a single PEM fuel cell of a  $100(\text{cm}^2)$  active area with a capacity of  $0.5(\text{A}/\text{cm}^2)$ . The fuel cell operates with open anode and cathode. It is powered with a constant air and hydrogen flow.

The data used to identify the two models are current-voltage time-series. Input signal used is a Pseudo-Random Binary Sequence (PRBS) sequence of length 1023 with amplitude  $\delta I = 1(A)$ , applied around a nominal current  $I_{avr} = 7(A)$ . The measurements are the cell output voltage.

The fuel cell voltage variations within 30-35 millivolts are the response to the current variations type PRBS of 1A around an operating point of 7A.

The parameters vector to be identified is  $(1 \times 11)$  dimension, while  $L = 5$  and  $M = 6$

Figures 6 and 7 show results obtained with respect using the identified IOM and the identified fractional order model. These results are compared to experimental measurements.

Figure 8 shows the estimation errors of the identified IOM and the identified fractional order model. The FOM error is about half less than the integer order model error.

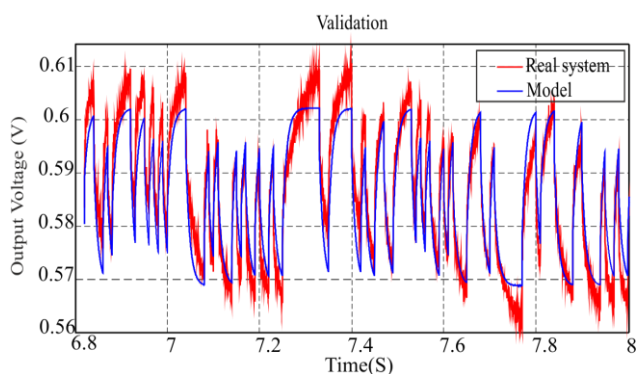


Fig. 6. Simulation results of the identified integer order model by the least square method: with input PRBS, nominal current  $I_{avr} = 7(A)$  and perturbation  $\delta I = 1(A)$ .

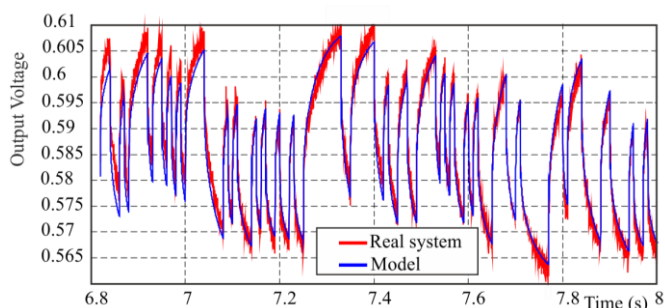


Fig. 7. Simulation of the identified fractional order model by the least square method adapted to fractional order models: with input PRBS, nominal current  $I_{avr} = 7(A)$  and perturbation  $\delta I = 1(A)$ .

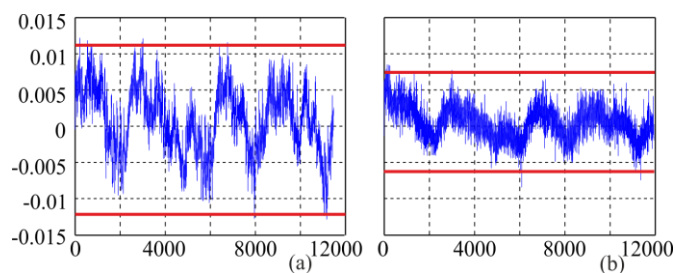


Fig. 8. Estimation error: (a)-with integer order model (b)-with fractional order model

## 5. CONCLUSIONS

In order to exploit the fuel cell on a large scale, it is necessary to be able to monitor its operating state. Diagnosis methods allowing this need reliable model capable of an accurate characterization of the system behavior. The analysis of the fuel cell impedance compared to its nominal value allows to characterize the fuel cell hydration state and to diagnose its possibly flooding or drying. Indeed, impedance general shape in the Nyquist plan and its deformations allow detection and isolation of malfunctions of the fuel cell. In this paper, using Warburg impedance, a fractional order fuel cell transfer function formulation was proposed. This model was transformed to implement FOM identification. The method used to identify the model's parameters was based on the least square method extended to fractional order models.

The proposed approach was validated using experimental measurements on a single PEM fuel cell. The obtained results had shown that FOM better characterize the real system behavior than conventional IOM. This can be explained by the diffusion phenomena which are described by a partial differential equation. Thereafter, the experimental measurements will be performed on a fuel cell stack of 500(W).

In this paper, to simplify the identification, the estimated parameters are functions of the real physical parameters. The next step will be dedicated to finding these physical parameters and implementing identification methodology of fuel cell FOM in real time to diagnose the system state: flooding, drying..., particularly in automotive transport applications.

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## Appendix A. FIRST APPENDIX

Table 1. Definition of variables

$P_{H_2}, P_{O_2}$ [Pascal]	Partial pressure of oxygen and hydrogen
$R = 8.314$ [J/K/mol]	Molar gas constant
$T$ (K)	Temperature
$n = 2$	Number of exchanged electrons
$F = 96485$ (C/mol)	Faradays constant
$I_{act}$ [A]	Activation current
$I_{lim}$ [A]	The limit current of diffusion
$\phi(i-1)$	Measurement vector
$D^{n_{a1}}$	Derivative operator
$\Gamma(m-\nu)$	Gamma distribution
$R_t$ [Ohm]	Charge transfer resistance
$R_{mem}$ [Ohm]	Membrane resistance
$C_{dc}$ [F]	Double layer capacitors
$Z_w$	Warburg impedance
$C_i(x, t)$ [Mole/L]	Gas concentration
$D_i$	Diffusion constant
$I_{dc}$ [A]	Double layer current
$I_f$ [A]	Faradic current
$\alpha$	Transfer coefficient
$\beta$	Diffusion coefficient