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Model identification of a Proton-Exchange Membrane Fuel-Cell from an input-output experiment : The diffusive representation approach

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Abstract— **This paper proposes a diffusive Proton-Exchange Membrane (PEM) Fuel-Cell (FC) model that reproduces the static and low-frequency dynamic characteristic of a PEMFC. Due to the nature of physical phenomena involved in a PEMFC, a fractional operator is a good candidate for describing its input-output behavior. In this paper, this operator is replaced by a well suited representation called "diffusive model" which simplifies its analysis and which leads to numerical realizations suitable for identification purposes. The diffusive model is approximated by a finite dimensional model that allows to formulate an optimization problem of least square error type to estimate the model distribution. The diffusive model is identified by means of experimental measurements of current (input) and voltage (output) in the FC. The obtained model is simple and can be used in systems that require real-time emulators or complex long-time simulations. The experimental results using the Ballard NEXA 1.2 kW fuel cell validate the advantages of the diffusive model.**

I. Introduction

Power systems based on proton-exchange membrane fuel cell (PEMFC) technology have been the object of increasing attention and extensive research over recent years. They are very interesting because they display a lot of potentials for usage in both stationary and mobile applications due, in particular, to their high-efficiency, low-operating temperatures, fast start-up, high-power density, solid electrolytes, low corrosion rate, non-polluting emissions, and longer cell and stack lifetimes in comparison with other kinds of fuel cells [1]–[3]. One of the areas addressed by researchers has been FC emulators design which has been widely promoted thanks to the advances in computing technologies. Furthermore, the use of a FC emulator is the most suitable way of testing power converters and other different devices in a safe, economical, realistic, and repetitive manner before being connected to the FC. In addition, the emulators also make possible to carry out critical tests that would otherwise be a risk for the equipment [4], [5]. An input-output FC model is enough in the case of FC emulators to be used in conjunction with power electronics devices. This is because input-output models have a low computational cost in comparation with other models that allows its implemented in real-time emulators or complex long-time simulations, however one of the important problems is how to derive this model. Some models have been developed in the literature [1], [6], [7]. For almost all of them, the combination of electrical models with chemical and physical laws are used to derive models where the values of a great number of parameters are difficult to evaluate precisely. From an input-output point of view, it is possible to exploit such models where the values of the different unknown parameters can be determined using nonlinear optimization methods and algorithms [8]. But in general, due to the important number of parameters, these approaches lead to models with a limited precision obtained by time-consuming algorithms. In this paper, due to the nature of physical phenomena involved in a PEMFC, the first step is to suppose that a fractional operator could be a good candidate for describing the input-output behavior of a PEMFC. The idea is to replace this operator by a well suited representation called "diffusive model" which simplifies the analysis and which leads to numerical realizations suitable for identification purposes. The complete theory of such models has been developed by G. Montseny in [9], [10] where all the mathematical machinery and its use in many contexts is largely motivated. It is worth to be noted that diffusive models allow to represent a wider class of operators, see [10] for details. Another way for describing fractional operators has been developed in parallel by the team of A. Oustaloup in [11] with a different approach. In [11], several applications are also proposed. In this paper, Montseny's approach is preferred because the diffusive models are defined in the setting of functional analysis, more precisely in the framework of Hilbert spaces, which are naturally well suited for deriving finite dimensional approximations. In the context of model identification problems, such a setting appears more appropriate. The paper is organized as follows. Next section presents the main principles behind diffusive representation in a concise way. In particular, it is shown how to obtain an infinite dimensional representation of a class of operators which includes the fractional ones. In this context, it is possible from an input-output experiment, to extract a model by solving a least square error optimization problem. The obtained solution is adapted to both finite-dimensional and discrete-time models. The section ends by presenting some extensions, which are important for the identification of a PEMFC model. Section III presents a circuit PEMFC model developed in the literature [1], [6], [7]. It allows a comparison with the one obtained by the

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proposed method. Section IV describes the used platform to test the potentialities of the identification technique. The Nexa PEMFC has been selected because it is commonly used by several research groups and thus constitutes a benchmark. Section V presents the obtained experimental results. The paper ends by presenting some conclusions and by discussing some research perspectives.

II. Circuit PEMFC model

This section provides a brief mathematical description of a PEMFC circuit model with the aim of comparison with the diffusive model which will be described in the next section. In order to simplify the analysis, the following assumptions are taken into account [1], [6], [7]:

1) One-dimensional treatment.
2) Ideal and uniformly distributed gases.
3) Constant pressures in the fuel-cell gas flow channels.
4) The fuel is humidified H_2 and the oxidant is humidified air. The effective anode water vapor pressure is 50% of the saturated vapor pressure while the effective cathode water pressure is 100%.
5) The fuel cell works under 100 °C and the reaction product is in liquid phase.
6) Thermodynamic properties are evaluated at the average stack temperature, temperature variations across the stack are neglected, and the overall specific heat capacity of the stack is assumed to be a constant.
7) Parameters for individual cells can be put together to represent a fuel-cell stack.
8) The model parameters shall be selected for the Nexa FC as far as possible.

The circuit model of the PEMFC is represented in Fig. 1. The output voltage is given [12] by

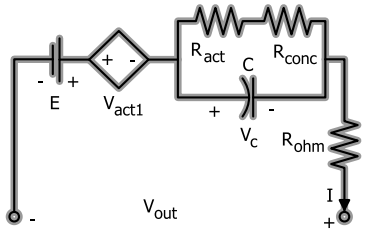


Fig. 1. Electrical model of the PEMFC used to compare with the diffusive model.

$$V_{out} = E - V_{act1} - V_C - V_{ohm} \tag{1}$$

where the reversible or Nernst potential is calculated as follows [13]

$$\begin{aligned} E &= E_0^0 + \frac{RT}{2F}\ln\left[p_{H_2}(p_{O_2}^{0.5})\right] \\ &\quad -k_E(T-298.15) - \lambda_e I(s)\frac{\tau_e s}{\tau_e s+1} \end{aligned} \tag{2}$$

where

- E_0^0 is the standard reference potential and it is equal to 1.229 V [14]–[17].
- T is the temperature of the FC [K].
- R is the constant of ideal gases [8.3143 J/(mol K)].
- F is the Faraday constant [96.487 kC/mol].
- p_{H_2} is the partial pressure of hydrogen [Pa].
- p_{O_2} is the partial pressure of oxygen [Pa].
- k_E is an empirical constant of 85 mV/K [13]–[16].
- τ_e is the constant time delay of the flow for a load transient of 80 s [13].
- λ_e is a constant factor of 69.4 $\mu\Omega$ [13].

The constants τ_e and λ_e were estimated for the PEM SR-12 fuel cell in [13] but their values are unknown for the the Nexa fuel cell. The activation losses in Fig. 1 are modeled by an independent voltage source and they are given by: $V_{act1} = \xi_1 + \xi_2 T_{pc} + \xi_3 T \ln(C_{O_2})$ while the remaining activation losses are modeled by a resistor as

$$R_{act} = -\frac{\xi_4 \cdot T \cdot \ln(I)}{I} \tag{3}$$

A generalized ξ_i parameter-based-model is proposed in [18] to be used in PEMFC, the set of parameters being: $\xi_1 = -0.948(\pm 0.004)$, $\xi_2 = 0.00286 + 0.0002\ln(A) + 4.3 \cdot 10^{-5}\ln(C_{H_2})$, $\xi_3 = (7.6 \pm 0.2)\cdot 10^{-5}$, $\xi_4 = -(1.93 \pm 0.05)\cdot 10^{-4}$ where C_{O_2} is the oxygen concentration at the cathode membrane/gas interface $[mol/cm^3]$ defined as

$$C_{O_2} = \frac{p_{O_2}}{5.08 \cdot 10^6 \exp(\frac{-498}{T})} \tag{4}$$

C_{H_2} is the liquid phase concentration of hydrogen at anode/gas interface $[mol/cm^3]$, calculated as

$$C_{H_2} = \frac{p_{H_2}}{1.09 \cdot 10^6 \exp(\frac{77}{T})} \tag{5}$$

and A is the cell active area $[cm^2]$ and it is equal to 122 cm^2 [16]. The ohmic losses can be expressed as [13] : $R_{ohm} = R_M + R_C$, R_M being the equivalent resistance of the membrane in $[\Omega]$ and R_C represents the resistance of transfer protons across the membrane with a value of 300 $\mu\Omega$ according with [2]. R_M can be defined by

$$R_M = \frac{l}{A} \cdot \frac{181.6\left(1 + 0.03 \cdot \frac{I}{A} + 0.062 \cdot \frac{T}{303}^2 \cdot \frac{I}{A}^{2.5}\right)}{\left(\psi - 0.634 - 3 \cdot \frac{I}{A}\right)\cdot \exp\left(4.18 \cdot \frac{T-303}{T}\right)} \tag{6}$$

where l is the polymer membrane thickness that was made by Dupont using Nafion 112 with a $l = 51\ \mu m$, ψ is being considered as an adjustable parameter that considers the relative humidity, the anode gas stoichiometric ratio and the membrane age, with a possible maximum value of 23 [18]. The concentration losses are modelled as [19]

$$R_{conc} = \frac{B}{I}\ln\left(\frac{I_{limit}}{I_{limit} - I}\right) \tag{7}$$

where B is a parametric coefficient, which depends on the cell and its operation state, and it is assumed equal to 0.016 V by Ballard Mark V PEMFC stack of 500 kW and American Company BCS Technologies FC of 500 W in [2]. Finally, the capacitor of Fig. 1 models the double-layer charging effect

between the porous cathode and the membrane and can be calculated as

$$V_C = \left(I - C\frac{dV_C}{dt} \right) \cdot (R_{act} + R_{conc}) \tag{8}$$

Since the electrodes of a PEM fuel cell are porous, the capacitance is very large and can be in the order of several Farads [13], [20]. In [13] a value of 0.1 F by the SR-12 STACK is proposed while in [2] is of 3 F by the Ballard Mark V PEMFC and in [21] a capacitor of 15 F is estimated by the Nexa FC. For comparison purposes we do not require a thermal model of the fuel cell since we can use the temperature measurement given by the Nexa software to evaluate the circuit model of the Fig. 1.As presented in this section, a PEMFC circuit model based on the Larminie model [1] was selected. However, a significant number of its parameters are empirical and there are a great absence of many of them in the literature by a specific FC or even different values of certain parameters for the same FC. Therefore, it is necessary to carry out an identification of the circuital model using an optimization technique for improving the accuracy of the model with the real FC. Another alternative for improving the model accuracy, as is proposed in this article, is to do an identification by means of a diffusive model which will be described in the following section.

III. Diffusive models of fractional operators

General operators belonging to the class of pseudo-differential time operators can be realized in an infinite-dimensional state space [10]. These representations are adapted to deduce efficient numerical well suited schemes. To introduce them, consider the class of linear causal convolution operator which at any continuous function u, associates

$$\int_0^t h(t-\tau)u(\tau)d\tau$$

where $u(t)$ is the input and $h(t)$ is the impulse response. We denote by $H(s)$, the Laplace transform of $h(t)$, i.e

$$H(s) = \int_0^\infty h(t)e^{-st}dt$$

and by $H(\partial_t)$ the previous convolution operator. Among these operators, rational operators are widely used in electronics or control and are represented by a rational transfer function. Numerous non rational operators exist and lead to non-rational transfer functions. $Log(\partial_t)$ or ∂_t^{a+jb} are examples of such kind of operators, but also the fractional operators belong to this class. The idea is to replace these operators which usually are nonlocal in time by a representation which simplifies the analysis and which leads to suitable numerical realizations of integral time operators encountered in many physical problems. Here, only a simplified theory is exposed. For a deeper presentation, see the book [10]. Define the function $\mu(\xi) : R^+ \to R$ called the diffusive representation by

$$h(t) = \int_0^\infty \mu(\xi)e^{-\xi t}d\xi$$

We have the fundamental result, obtained by Laplace transform and Fubini theorem, which gives the connection between $\mu(\xi)$ and $H(s)$. *The diffusive representation* μ of an operator with symbol H, satisfies [10]

$$H(s) = \int_0^\infty \frac{\mu(\xi)}{s+\xi}d\xi$$

with $s \in D_\mu \subset C$, the convergence domain of the integral. One main idea is that we have now three representations of the same convolution operator of a diffusive nature connected by the Laplace transform, $\mu(\xi) \to h(t) \to H(s)$. The central result is now the following [10]. *The operator $H(\partial_t)$ can be described from an input-output point of view, by the representation (diffusive representation)*

$$\left\{ \begin{array}{rcll} \partial_t\psi(\xi,t) & = & -\xi\psi(\xi,t) + u(t), & \xi \in R^+ \\ y(t) & = & \int_0^\infty \mu(\xi)\psi(\xi,t)d\xi, & \psi(\xi,0) = 0 \end{array} \right.$$

A. The infinite dimension framework [22]

We consider the case where we only dispose of measurements (input, output) obtained from an experiment on a real system. We suppose that we dispose of measurements $\hat{u}$ and $\hat{y}$ on the interval of time $[T_i, T_f]$. The objective is to identify an input/output diffusive model

$$\left\{ \begin{array}{rcl} \partial_t X(\xi,t) & = & -\xi X(\xi,t) + u(t),\ X(\xi,0) = 0 \\ y(t) & = & \int_0^\infty \mu(\xi)X(\xi,t)d\xi, \end{array} \right. \tag{9}$$

The main feature of the previous model is that the structure of the state equation is fixed. The difference between two models is captured through the distribution μ. If we express the output through the convolution product denoted by *, we obtain

$$y(t) = \int_{R^+} (e^{-t\xi} * u)\mu(\xi)d\xi$$

Associated to the previous expression, let us define the operator K_u

$$K_u : \left\{ \begin{array}{l} \mathbf{M} \to L^2(T_i, T_f) \\ \mu \to y = K_u\mu \end{array} \right. , \tag{10}$$

$$(K_u\mu)(t) = \int_{R^+} (e^{-t\xi} * u)\mu(\xi)d\xi.$$

where $\mathbf{M}$ is a convenient Hilbert space of distributions. From a practical point of view, measurement noise often affects the data and the diffusive model is not able to capture all the physical phenomena (nonlinearities,...). For these reasons, the identified model does not fit exactly the data. From a mathematical point of view, this means that it does not exist $\hat{\mu}$, solution of

$$\hat{y} = K_{\hat{u}}\hat{\mu}.$$

A way to solve the identification problem is to find a $\hat{\mu}$ which minimizes in some sense the error between $\hat{y}$ and $K_{\hat{u}}\hat{\mu}$, for example the distance

$$\|\hat{y} - K_{\hat{u}}\hat{\mu}\|_{L^2(T_i,T_f)}.$$

It is well-known that the solution of this optimisation problem is given by

$$\hat{\mu}(\xi) = (K_{\hat{u}}^* K_{\hat{u}})^{-1} K_{\hat{u}}\hat{y},$$

where $K_{\hat{u}}^*$ is the dual operator of $K_{\hat{u}}$ defined by

$$\forall w \in L^2(T_i,T_f),\ \ < K_{\hat{u}}|w >_{L^2(T_i,T_f)} = < \mu|(K_{\hat{u}}^* w >_{\mathbf{M}} .$$

where symbol $<>$ denotes scalar product. This solution is the best solution in the sense of this least square error.

B. The finite dimension framework [22]

Suppose that we approximate the model choosing as described above, a finite number of values of ξ, $\Xi = \{\xi_k\}_{k=1,\ldots,N} \subset R^+$. The approximate model reads

$$\left\{ \begin{array}{rcl} \dot{X}_i(t) & = & -\xi_i X_i(t) + u(t),\ i = 1,\ldots,N \\ Y_i(t) & = & \sum_{i=1}^N \mu_i X_i, \end{array} \right.$$

An approximation of μ through a sum of Dirac masses $\{\delta_{\xi_i}(\xi),\ i = 1,\ldots,N\}$ is taken into account. For identification purposes, this approximation is the most appropriate for the construction of a finite dimensional model. It is also possible to build a ξ-continuous model by considering $\mu(\xi) = \sum \mu_i \Lambda_i(\xi)$, with Λ_i replacing δ_{ξ_i}. Define the operator $K_{\hat{u}}$

$$\begin{array}{c} K_{\hat{u}} : \mathbf{R^N} \longrightarrow L^2(T_i,T_f) \\ \hat{\mu} = \left(\ \hat{\mu}_1,\ldots,\hat{\mu}_N\ \right)' \rightarrow y(t) = \sum_{i=1}^N \hat{\mu}_i (e^{-t\xi_i} * u). \end{array} \tag{11}$$

The dual operator $K_{\hat{u}}^*$ is defined by

$$\begin{array}{l} K_{\hat{u}}^* : L^2(T_i,T_f) \longrightarrow \mathbf{R^N} \\ < K_{\hat{u}}\hat{\mu}|y(t) >_{L^2(T_i,T_f)} = \int_{T_i}^{T_f} \sum_{i=1}^N \hat{\mu}_i (e^{-t\xi_i} * u) y(t) dt \\ = \sum_{i=1}^N \hat{\mu}_i \int_{T_i}^{T_f} (e^{-t\xi_i} * u) y(t) dt = < \hat{\mu}|K_{\hat{u}}^* y >_{R^N} . \end{array} \tag{12}$$

Choosing for simplicity $\mathbf{M} = L^2(R^+)$, the induced norm in R^N is the ordinary euclidian one and

$$< \hat{\mu}|K_{\hat{u}}^* y >_{R^N} = \sum_{i=1}^N \hat{\mu}_i (K_{\hat{u}}^* y)_i$$

which leads to

$$(K_{\hat{u}}^* y)_i = \int_{T_i}^{T_f} (e^{-t\xi_i} * u) y(t) dt. \tag{13}$$

On the other hand, we have

$$\begin{array}{rcl} (K_{\hat{u}}^* K_{\hat{u}} \hat{\mu})_i & = & \int_{T_i}^{T_f} (e^{-t\xi_i} * u) \sum_{j=1}^N \hat{\mu}_j (e^{-t\xi_j} * u) dt \\ & = & \sum_{j=1}^N \int_{T_i}^{T_f} (e^{-t\xi_i} * u)(e^{-t\xi_j} * u) \hat{\mu}_j dt, \end{array}$$

$$(K_{\hat{u}}^* K_{\hat{u}})_{ij} = \int_{T_i}^{T_f} (e^{-t\xi_i} * u)(e^{-t\xi_j} * u) dt,$$

the estimated measure $\hat{\mu}$ is given by

$$\hat{\mu}(\xi) = (K_{\hat{u}}^* K_{\hat{u}})^{-1} K_{\hat{u}} \hat{y}.$$

C. The discrete-time expression [22]

In numerous practical cases, the signals are sampled. If ΔT is the sampling period, the discrete time approximated model is expressed as

$$\left\{ \begin{array}{rcl} X_i(n+1) & = & e^{-\xi_i \Delta T} X_i(n) + \dfrac{1 - e^{-\xi_i \Delta T}}{\xi_i} u(n) \\ Y(n) & = & \sum_{i=1}^N \mu_i X_i(n). \end{array} \right.$$

Some elementary calculations lead to

$$X_i(n) = \sum_{l=1}^N \frac{e^{-\xi_i (l+n)} e^{(-\xi_i \Delta T - 1)}}{\xi_i} u(l-1)$$

and

$$\begin{array}{rcl} Y(n) & = & (X_1(n),...,X_N(n)) \left(\begin{array}{c} \mu_1 \\ \vdots \\ \mu_N \end{array} \right) \\ & = & C(n)\mu, \end{array}$$

If we dispose of $M = \dfrac{T_f - T_i}{\Delta T}$ measurements for u and y, $\hat{\mu}$ is obtained by

$$\hat{\mu} = (H^* H)^{-1} H^* \hat{y},$$

where :

$$H = \left(\begin{array}{c} C(1) \\ \vdots \\ C(M) \end{array} \right).$$

The matrix H can be computed using the following recursion formula

$$\left\{ \begin{array}{rcl} C_i(0) & = & 0 \\ C_i(k) & = & e^{-\xi_i \Delta T} C_i(k-1) + \dfrac{1 - e^{-\xi_i \Delta T}}{\xi_i} u(k-1) \\ & & i = 1,...,N \qquad j = 1,...,M \end{array} \right.$$

D. Affine output [22]

For some systems (in particular for fuel cells), the output is affine (the sum of a constant and a combination of the states), that is

$$Y(n) = \sum_{i=1}^N \mu_i X_i(n) + Y_0.$$

It is possible to determine the constant Y_0 by rewriting $Y(n)$ in the following way

$$Y(n) = (C(n) \quad 1) \left(\begin{array}{c} \mu \\ Y_0 \end{array} \right)'$$

and H becomes

$$H = \left(\begin{array}{cc} C(1) & 1 \\ \vdots & \vdots \\ C(M) & 1 \end{array} \right).$$

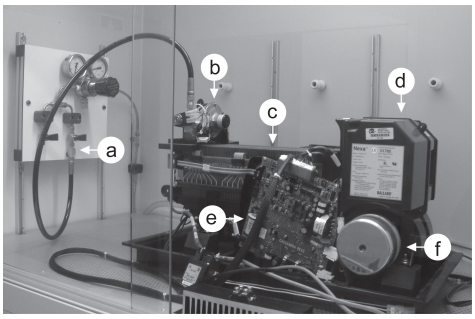


Fig. 2. Nexa power module: (a) hydrogen supply, (b) hydrogen control valve, (c) fuel cell stack, (d) air compressor, (e) control board, (f) cooling motor.

IV. Fuel cell system description

This work has been performed using the Nexa PEMFC from Ballard Manufacturer shown in Fig. 2. At the present time, this fuel cell remains a world benchmark since it has been widely used by different research groups and it represents the state of the art in terms of PEM technology [23]. However, the results presented in this paper can be extended to other types of PEM fuel cells. The Nexa fuel cell is a fully integrated system that produces unregulated DC power, until 1.2 kW, from a supply of hydrogen and air. The Nexa power module has a LabVIEW software which provides a graphical user interface [24]. This software monitors the key process parameters of the fuel cell and it can generate a data logging file with the following parameters: stack temperature, stack voltage, stack current, fuel pressure, fuel leak, fuel consumption, oxygen concentration, ambient air temperature, purge cell voltage, battery voltage (used to start the power module), process air-flow, air pump operating voltage, hydrogen concentration bridge voltage, process air pump duty cycle, and cooling air fan duty cycle. A serial port is used to communicate the mentioned variables from the fuel cell to the LabVIEW software using the RS-232 standard protocol, hence its maximum sampling time is 200 ms.

V. Experimental results

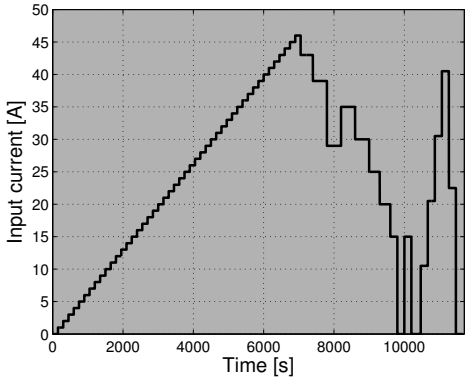


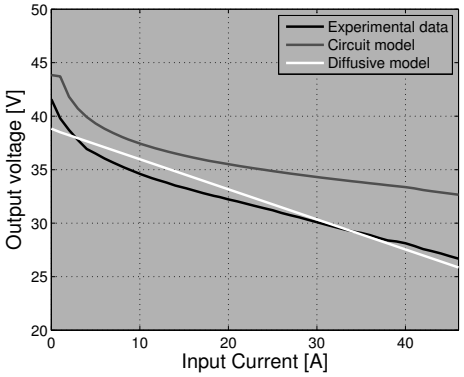
Fig. 3. Experimental current profile used for the diffusive model training.

The proposed diffusive Proton-Exchange Membrane (PEM) Fuel-Cell (FC) model was tested using the current and voltage variables monitored each 200 ms by the Nexa software. It performed a load profile which reproduces different operation points and different current transients with the goal of training the diffusive model in all the fuel cell range operation as shown in Fig. 3. The diffusive model results taking 10 values of ξ in $[\frac{1}{\Delta T}, \frac{10^2}{\Delta T}]$ to cover 2 decades are listed below

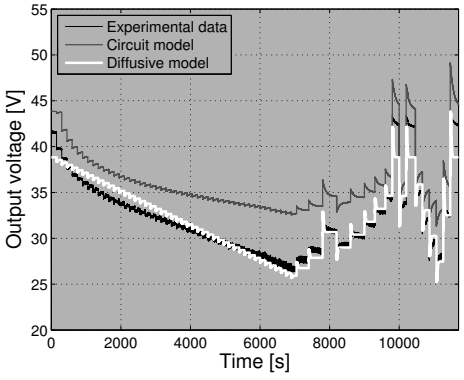
$$\hat{\mu} = (0.7215, -4.6588, 15.9586, -39.4114, 79.3927, -136.8671, 194.5954, -218.5614, 180.8790, -82.5854) \tag{14}$$

$$Y_0 = 38.8351$$

In Fig. 4 the diffusive model training results are presented



(a)



(b)

Fig. 4. Experimental and simulation, using circuit and diffusive models, data of Nexa FC by the the load profile of Fig. 3: (a) VI static characteristic, (b) dynamic characteristic.

and compared in terms of both static and dynamic characteristic with experimental data and the circuit model proposed in Section II . This figure shows how the identified model using the diffusive approach fits the data better than the circuit model in both characteristics. In order to evaluate the diffusive model validity, the model is tested with a new load profile which is different from the one used during training as shown in Fig. 5. The comparison between the experimental data, the circuit model and the diffusive model is given in Fig. 6. Once again is observed that the diffusive model

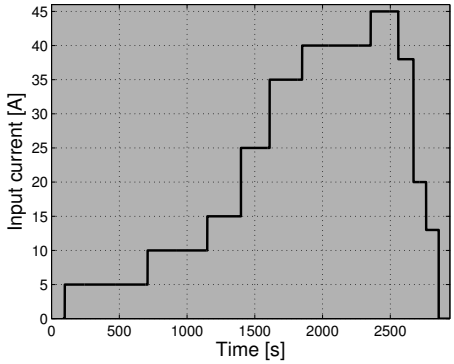


Fig. 5. Experimental current profile used for the diffusive model validation.

fits better to the experimental data than the circuit model. It is to be noted that the diffusive model is quite simple and is obtained without entering into physical or chemical laws. Therefore, the method can be directly used for other PEMFC types. Another important remark is that in real-time applications, it is possible to derive a recursive version of the proposed identification method.

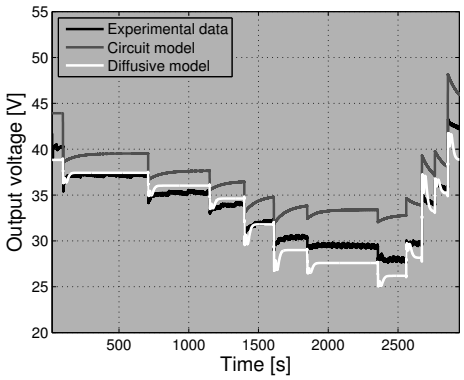


Fig. 6. Experimental and simulation, using circuit and diffusive models, data of Nexa FC by the the load profile of Fig. 5.

VI. Conclusions

In this paper, a method to identify an input-output model of a PEMFC is proposed. As it can be expected, due to the nature of physical phenomena involved, a fractional operator seems to be a good candidate model for describing appropriately its input-output behavior. By means of an infinite dimensional state space realization of the operator through a diffusive model, it is possible to extract a finite dimensional model, well suited for identification purposes using a standard least square identifier. An input-output experiment capturing the PEMFC dynamics is used to identify a model without entering into the physical and chemical laws. A comparison with a circuit model whose parametric tuning needs a specific expertise, shows that in terms of complexity and precision, the reported model is superior. Even if the opportunity is not exploited in the present paper, considering a recursive version of the method, it is possible to identify a model on-line.

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