Summary of the PEM Fuel Cell modeling

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***Abstract*—In this paper a brief review of different static and dynamic models that have been proposed for Proton Exchange Membrane Fuel Cells is proposed. Among the different models of Proton Exchange Membrane Fuel Cell models that have been reported in publications, some of them focus on one particular Fuel Cell stack, some other take in account a part or all auxiliaries as compressor, humidifier, ..., some other take into account different phenomenon as fluidic, chemical, thermic or only electric. Each model is designed according to the modeling objectives (diagnosis, fault tolerance, parameter identification, predictive tool, automatic control, energy management, ...). Each model (author) states its simplification hypothesis according to its domain of use. For use in automotive applications, a model should be dynamic to some degree to account for the important transients that can take place. The aim of this paper is to propose an overview about FC static and dynamic models. One static and one dynamic model have been chosen among others and the simulation of these two models are presented and validated experimentally in the FCLab (Fuel Cell Lab) and in UTBM, France.**

***Index Terms*—Proton exchange membrane fuel cells, Modeling.**

I. Introduction

Fuel Cell (FC) is an electrochemical device that converts directly the chemical energy of a fuel into electricity. Since FCs can offer a highly efficient environment and friendly electrochemical transformation for energy conversion, they are widely considered as a potential alternative power source and have drawn much attention and intensive development for commercial stationary power generation, residential applications, and transportation technologies in these days. A proton exchange membrane fuel cell (PEMFC) operates typically at low temperature and can achieve many benefits, such as high power density, easy and safe operation mode, and compactness. With low-temperature operation and high power density, PEMFC is currently considered for power supply and propulsion of vehicle applications. In order to improve power quality, system performance and design optimization, the PEMFC system should be modeled and analyzed to give more insight for constructing the subsystems and interactions between them. The FC and H_2 are part of the new vectors of power generation, offering interesting perspectives to the field of renewable energy [1], [2]. The FC system is a complex multi-physics system (chemical, electrical, fluidic, thermal, mechanical phenomena are inter-acting). Thus, focusing on large-scale industrialization of this electrochemical converter, multidisciplinary studies are still required. The main areas of development can be classified into three categories:

- At the cell level: development of materials and catalysts (membrane, electrodes, bipolar plates, ...), ...
- At the stack level: design of the flow fields, design of the cooling circuit, global electrical and mechanical constrains, ...
- At the system level: design of specific power converters, coupling to electrical storage elements, air supply system, gas humidification system, control and diagnosis, ...

Whatever the application, PEMFCs require such a high level of performance and reliability. The overall system must operate with a high degree of reliability in a wide range of operating conditions, leading to a robust control system. Going through the modeling of the complete PEMFC system is a required step because it facilitates the understanding of the involved phenomena. There are a lot of models for PEMFC stacks, which their own specificities and utilities, according to the studied underlying phenomena.

One of the most important aspects in developing FC models is linked to the ability to investigate complex design geometries and different operating conditions by expending less time and effort than doing experimental investigations. Besides, validation of theoretical and numerical models requires experimental confirmation. To date, there is no complete computational model for FC stacks including all the phenomena together. Nevertheless, increasing focus on this topic has produced rudimentary attempts which will probably support later studies. Available experimental data and mathematical models have been obtained for very restricted and idealized situations, and do not take into account of phenomena other than the one investigated. Research is needed both at an experimental as well as at an analytical/numerical level; there is a need for measurements, comparisons with existing models, and development of new models. The focus of research in the future will be on the performance and systems integration of FC stacks and associated sub-systems in advanced electric and hybrid electric vehicles. The sub-systems include fuel storage, fuel reforming and processing, air delivery systems, heat exchangers and thermal integration with other vehicle sub-systems, humidification and water management.

II. PEMFC models literature review

It is essential first to define the objectives for the future use of the model. There are establishing criteria for using

the model: speed, accuracy, flexibility, graphical interface and implemented in software. Based on these criteria for use, it is possible to determine the design of model criteria (theoretical or semi-empirical model, dynamic or static, mono, bi or tri-dimensional ...). There have been many studies on modeling and simulation of the PEMFC [3][29]. In [5], the author determined an electric-equivalent circuit for the PEMFC stack. [4] introduces the Energetic Macroscopic Representation (EMR) as approach for the dynamic modeling of a PEM fuel cell stack. Using semi-empirical equations for modeling a PEMFC is proposed for providing a tool for the design and analysis of FC complete systems. The focus of [7] is to derive an empirical model including process variations to estimate the performance of FC without extensive calculations. The model takes into account not only the current density but also the process variations, such as the gas pressures, temperatures, humidities, and utilizations to cover operating processes, which are important factors in determining the real performance of FC.

The authors in [8] proposed a dynamic model of PEMFC, the originality of which lays on the use of non-integer derivatives to model diffusion phenomena. This model has the advantage of having least number of parameters while being valid on a wide frequency range and allows simulating an accurate dynamic response of the PEMFC. [9] develops a dynamical model for single cell of the PEMFC. The model studies the phenomena of charge and mass transport through the cell elements. It integrates also the electrical aspect related to reaction kinetics by proposing an electrical equivalent circuit of the cell. The influence of gas consumption and humidification rates on water diffusion and membrane humidity are considered. Most of the current models for the cell used for an analysis of the FC system are based on the empirical polarization curve and neglect the dynamic effects of water concentration, temperature and reactant distribution on the characteristics, but [10] proposed a new model constructed upon the layers of a cell, taking into account the following factors:

1) dynamics in temperature gradient across the FC;
2) dynamics in water concentration redistribution in the membrane;
3) dynamics in proton concentration in the cathode catalyst layer;
4) dynamics in reactant concentration redistribution in the cathode gas diffusion layer.

The model integrates physical, chemical and electrochemical phenomena but don't consider the dynamic behavior in the electrical approach. Another notable work is the one given by [11]. In [12] mono-dimensional modeling presented the dynamic behavior of a 5-cell PEMFC stack operating in dead-end mode has been studied at room temperature. Some papers take into account the effect of temperature as for example [13]. Management of the water and heat as byproducts in an operating PEMFC stack are also crucial factors in their optimal design and safe operations. Models currently available for a PEMFC are based on either empirical or 3-D computational fluid dynamics (CFD). Both models do not fully meet the need to represent physical behavior of a stack because of either their simplicity or complexity. A highly dynamic PEMFC stack model, taking into account the most influential property of temperature affecting performance and dynamics is often required. [14] proposes a non-linear state-space dynamic model for planar PEMFC, the model is based on both thermodynamic and electrical aspects, proposing an equivalent circuit which integrates most of the FC components, however the obtained model is not that simple because of the number of state space order obtained. [15]-[17] present the artificial intelligence modeling techniques (based on artificial neural networks or fuzzy logic). In [16] the model predictive control mechanism was proposed by utilizing Recurrent Neural Network (RNN) optimization. [17] presents a fuzzy modeling of FC based on mutual information between variables, where the input variables selected through computation of mutual information (MIs) allowed building the predictive model with most relevant set of variables. In contrast to available traditional models of FC, Takagi-Sugeno (TS) fuzzy model provided an efficient prediction at different initial load conditions without knowledge of numerous physical, chemical, and electrochemical parameters. Other analytical model of PEMFC with the moto-compressor was recently published in [18] and [19]. This model is very interesting since it is only a 4-order state space model, but the developed reduced order model is not valid in a FC start-up period. So authors in those papers propose to start FC with another (complete order model), then switch to the reduced one. The model presented in [27] is based on a realistic electrical equivalence and the obtained model is of only three states order and is validated experimentally.

III. PEM Fuel Cell Description

The fundamental structure of a PEMFC can be described as two electrodes (anode and cathode) separated by a solid polymer membrane acting as an electrolyte (Fig. III). Hydrogen fuel flows through a network of channels to the anode, where it dissociates into protons that, in turn, flow through the membrane to the cathode. Electrons are collected as electrical current by an external circuit linking the two electrodes. The oxidant (oxygen coming from air in this study) flows through a similar network of channels to the cathode where oxygen combines with the electrons coming from the external electrical circuit and the protons flowing through the electrolyte membrane, thus producing water. The chemical reactions occurring at the anode and cathode electrode of a PEMFC are as follows:

Hydrogen oxidation and oxygen reduction:

$$H_2 \Rightarrow 2H^+ + 2e^- \qquad \text{anode} \qquad (1)$$

$$2H^+ + 2e^- + \frac{1}{2}O_2 \Rightarrow H_2O \qquad \text{cathode} \qquad (2)$$

$$H_2 + \frac{1}{2}O_2 \Rightarrow H_2O + heat + electricity \qquad \text{total cell reaction} \qquad (3)$$

In order to provide an adequate gas management, several additional auxiliary circuits are required in order to obtain a FC system like the hydrogen circuit, the cooling circuit, the air circuit and the humidification circuit as shown in Fig. 2.

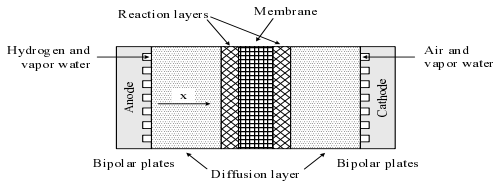

Fig. 1. Schematic of a single cell

The hydrogen circuit can be operated in three modes, open-mode, closed-mode and feedback mode. This circuit controls the pressure, mass/volume flow rates, temperature and vapor content of hydrogen. The open mode operation involves the remainder of hydrogen (i.e. the hydrogen left after passing through the anode) being released to the water condenser used by the cooling system. In the closed mode operation, the hydrogen exit valve closes; however, it is opened only to ensure hydrogen renovation by a process called "flush". In the feedback mode, the remainder hydrogen is re-injected into the cell. The air (oxidant) is humidified prior to its introduction in the FC stack. The humidification process involves heating the air and humidifying it at a high temperature and then cooling this mixture to a lower temperature so as to get a saturated mix of water vapor and air at this temperature which is measured by the sensors. The air supply circuit is probably the most important auxiliary circuit. In fact, for mid or high power PEM fuel cell stacks, this circuit relies on a air compressor, which design, control and efficiency are subject to intensive researches.

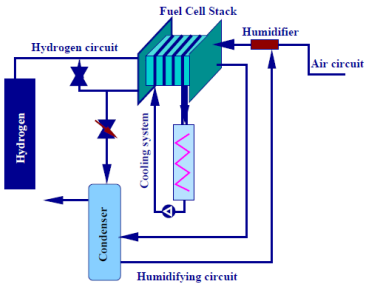

Fig. 2. Fuel Cell description

IV. Static models

A. Amplett et al. Model

A parametric model of PEMFC developed by Amphlett [20], [21] using a combination of mechanistic and empirical approach. The ideal standard potential (Nernst potential) of an H2/O2 FC is 1.229 V with liquid water product. The actual cell potential is decreased from its reference potential because of irreversible losses. The thermodynamic potential is defined via a Nernst equation in expanded form by the following equations [21]:

Therefore, the expression of the voltage of a single cell is:
The cell output voltage equation is:

$$V_{FC} = E_{th} - \eta_{act} - |\eta_{diff}| - R_{mem} I \tag{4}$$

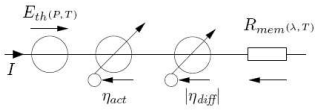

Fig. 3. Static model of FC

where[1]

E_{th}	thermodynamic voltage
η_{act}	activation losses
η_{diff}	diffusion losses
$R_{mem} I$	ohmic losses

with

$$E_{th} = E^0 + \frac{RT}{nF}(\ln(P_{H_2}) + \frac{1}{2}\ln(P_{O_2})) \tag{5}$$

E^0	Nernst voltage
R	perfect gas constant
F	Faraday constant
n	number of produced electron in the reaction
P_{H_2}, P_{0_2}	H_2 and O_2 pressures

The activation losses car be written as:

$$\eta_{act} = \frac{RT}{\alpha^* nF}\ln(\frac{I}{I_0^*}) \tag{6}$$

where

α^*	transfer coefficient
I_0^*	produced current given by:

$$I_0^* = K\exp(\frac{\Delta G}{R}(\frac{1}{T^0} - \frac{1}{T})) \tag{7}$$

K	constant defined experimentally

The diffusion losses are given by:

$$\eta_{diff} = \frac{RT}{\beta nF}\ln(1 - \frac{I}{I_{lim}}) \tag{8}$$

β	experimentally defined coefficient
I_{lim}	maximum diffusion current

B. Larminie and dicks static model

The characteristic FC voltage as a function of the FC current magnitude is presented in Fig. 4. The obtained curve is composed of three main regions corresponding to the predominance of electrochemical activation phenomena (region 1), a linear part (region 2) where the voltage drop is mainly due to electronic and ionic internal resistances and the last region where the diffusion kinetics of gases through the electrodes becomes the limiting factor (region 3). This last zone is characterized by a rapid voltage fall [22].

$$\begin{aligned} V_{FC} &= E - A.log\left(\frac{i_{FC} + i_n}{i_o}\right) - R_m\left(i_{FC} + i_n\right) \\ &\quad + B.log\left(1 - \frac{i_{FC} + i_n}{i_{Lim}}\right) \end{aligned} \tag{9}$$

Hence $V_{FC} = f(i_{FC})$, where E is the reversible no loss voltage of the FC, E_0 is the measured open circuit voltage,

[1]Note that in the Ohmic losses, the equivalent resistance linked to the conduction of electrons can be added to the electrolyte membrane resistance to the condution of protons. Other resistances due to the testbench can be added to the same ohmic losses.

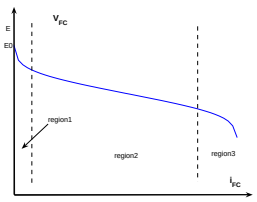

Fig. 4. Static FC characteristic.

i_{FC} is the delivered current, i_o is the exchange current, A is the slope of the Tafel line, i_{Lim} is the limiting current, B is the constant in the mass transfer, i_n is the internal current and R_m is the membrane and contact resistances.

V. Dynamic Models

A. Impedance model of fuel cell

In the case of a PEMFC, the electric energy is obtained by a redox electrochemical reaction [23]. The simplest small signal model for this type of reaction proposed by Randles can be used to model PEMFC (Fig. 5) [24]. It contains a transfer resistance R_t, which characterizes the ions transfer phenomena at the electrodes (only cathode is represented), a resistance R_m representing the membrane and contact resistances, a double layer capacitor C_{dl} and a diffusion convection impedance $Z_{W\delta}$ expressed by Nernst for a finished diffusion length by the following relation [25], [26]:

$$Z_{W\delta}(s) = R_d \frac{th(\sqrt{\tau_d s})}{\sqrt{\tau_d s}} \tag{10}$$

Where R_d, τ_d and s are the diffusion resistance, the diffusion constant time and the Laplace operator.

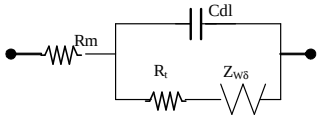

Fig. 5. Impedance model of a FC electrode

B. Dicks-Larminie Model

The Dicks-Larminie electric circuit model is shown in Figure 6. It models the activation polarization, the concentration polarization, the ohmic polarization and the Nernst voltage. R_a, the activation and concentration losses, is related to double layer capacitance. R_{ohmic} is related to the flow of hydrogen and electrons. The capacitor C is the charge double layer, which delays the dissipation of electronic charges near electrolyte/electrode interface. A voltage source models the Nernst voltage.

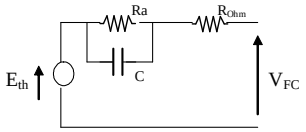

Fig. 6. Dicks and Larminie static model of FC

C. Becherif-Hissel model

The electrical equivalent model of a FC is obtained by setting an equivalent electrical component to account for the pneumatic behavior. This equivalence is based on the conservation principles of mass, charge and energy. Following the Bernoulli's laws of continuity, mass and energy flow are conserved in a pipe for an incompressible fluid, however there will be losses due to friction and geometry of the pipes. The compressibility of the fluid will add a storing capacity to the pipes. Hence, the molar flow rates in a pneumatic system can be analogous to the flow of charge i.e. current in an electrical system, based on the fact that mass and charge are conserved in both the systems. Similarly, the driving force for charge and mass is the electrical potential difference and pressure difference in the respective cases. The flows vary linearly with respect to the drive force, which in the electrical systems is given by the Ohms law. In pneumatic systems, an equivalent pneumatic resistance can be computed which will satisfy the analogous equation of Ohms Law for the pneumatic system, See [28][27] for more details.

$$ElectricalEnvironment : I = \frac{dq}{dt}$$

$$PneumaticEnvironment : Q = \frac{dn}{dt}$$

$$Q_{m,net} = Q_{m,in} - Q_{m,out}$$

The gases that are being passed through the channels are compressible. This compressibility is determined by the volume the channel can hold under given pressure and temperature conditions. This is analogous to a capacitor in an electrical system which is capable of accumulating charge under transient conditions and acts as an open circuit when steady state is achieved. The figure 7 shows the analogy used to model the capacitance of channels in our circuit [28].

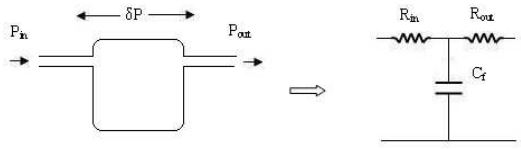

Fig. 7. Storage capacity of channel analogous to a capacitor

The overall circuit hence formulated is shown in figure 8. Each of the flow rates is denoted by the letter Q.

1) Network Analysis: This section presents the formulation of these equations based on Kirchhoff Laws, which are then used to carry out the simulations. The letter Q denotes the flow rate whereas the superscripts I, a, c and m stand for function of current, anodic, cathodic or membrane sections respectively. The subscripts in first place are the first letter of the species they denote, for instance, w for water, H for hydrogen etc., the subscript C_f denotes either the flow rate or pressure drop through the capacitor in the respective section. 'r', 'r-e', 'm', 'in', 'out' and 'P_{sat}' denote reacted, reverse electro-osmosis, membrane, inlet, outlet and property through the zener diode section.

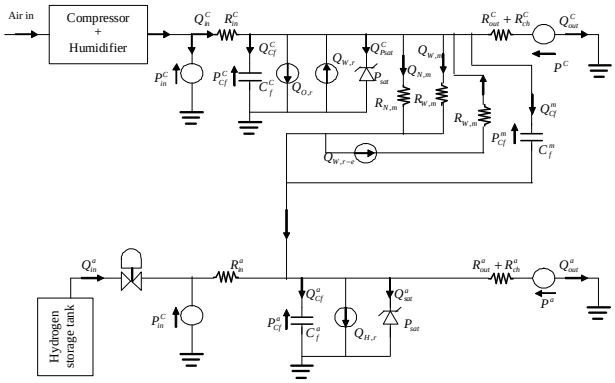

Fig. 8. Complete Circuit: Anode, Cathode and Membrane

The following equations provide the flow rates in the different branches of the circuit shown in figure 8.

$$Q^c_{in}+v_wQ^I_{w,r}+v_wQ^I_{w,r-e}=v_oQ^I_{O,r}+Q^c_{Psat}+Q_{N,m}+Q_{w,m}+Q^c_{out} \quad (11)$$

$$Q^a_{in}+Q_{N,m}+Q_{w,m}+Q^m_{Cf}=Q^c_{Cf}+v_wQ^I_{w,r-e}+v_hQ^I_{H,r}+Q^a_{Psat}+Q^a_{out} \quad (12)$$

$$P^c_{Cf}=(R^c_{out}+R^c_{ch})Q^c_{out}+P^c \quad (13)$$

$$P^a_{Cf}=(R^a_{out}+R^a_{ch})Q^a_{out}+P^a \quad (14)$$

$$P^c_{in}=(R^c_{in})Q^a_{in}+P^c_{Cf} \quad (15)$$

$$P^m_{Cf}=(0.5R_{w,m}+R^a_{out}+R^a_{ch})Q^a_{out}+P^a \quad (16)$$

$$P^m_{Cf}=(0.5R_{w,m}+R^a_{out}+R^a_{ch})Q^a_{out}+P^a \quad (17)$$

$$\frac{dP^c_{Cf}}{dt}=\frac{Q^c_{Cf}}{C^c_f} \quad (18)$$

$$\frac{dP^a_{Cf}}{dt}=\frac{Q^a_{Cf}}{C^a_f} \quad (19)$$

$$\frac{dP^m_{Cf}}{dt}=\frac{Q^m_{Cf}}{C^m_f} \quad (20)$$

$$P^{ca}=R_{N,m}Q_{N,m} \quad (21)$$

where v_i denotes the molar volume of i where i = {w for water, o for oxygen or h for hydrogen}.

VI. Simulation and Experimental Validation

A number of approaches have been used to model PEMFC behavior. The list of the presented PEMFC static and dynamic models is far to be exhaustive. We chose for the simulation and experimentation one static model given in section IV-B and one dynamic given in section V-C.

A. Static model

The Larminie FC static model given by (9) was validated by author using the Nexa $1.2kW$ FC from Ballard (Fig. 9)

The Larminie & Dicks model parameters were tuned to match with the experimental results and the simulation of this model (polarization curve) was validated with the experimental test as shown in figure 10.

The three regions of Fig. 3 can be seen in Fig. 10. But, for the experimental result the third region was not tested because working in this region can cause irreversible damages on the FC stack.

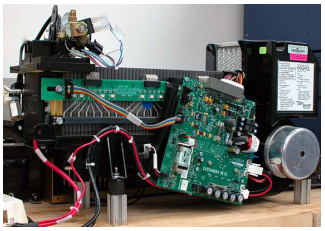

Fig. 9. 1.2 kW Nexa Fuel Cell stack

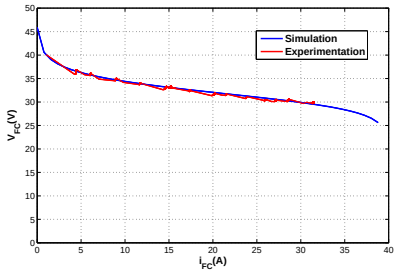

Fig. 10. Simulation and experimental results of the static FC model

B. Dynamic model

Different sets of experimental data to check the robustness of the model (as described in section V-C) were chosen. The simulations have been carried out in Matlab-Simulink. Figure 11 shows the simulations for a 400W PEMFC stack having 20 single cells. The simulated voltage is very close to the voltage obtained experimentally in both the cases. The subplot represents the FC current imposed by the load and playing the role of a perturbation for the FC stack.

The experimental test done for a 400W (electrical power) PEMFC with step changes in the load current is also provided.

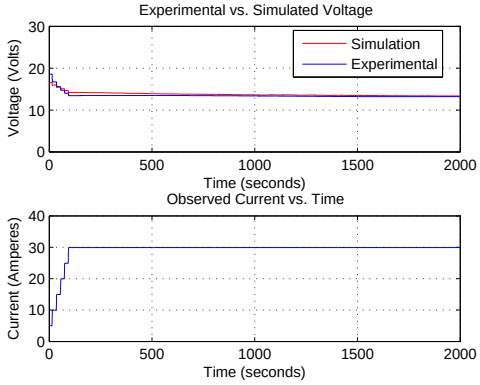

Fig. 11. Simulated and Experimental Voltages obtained for a 400W PEMFC stack

The simulated voltage is considerably close to the voltage obtained experimentally. The error is less than 10%. Note that the maximum error is generally observed for a very small time particularly at the initial time up to 100 seconds of operation. The error in remaining part is observed to be less than 3% which also tends to reduce further with time. However, we see a slight offset in the values, which is due to the non steady pneumatic resistances. The reader may be inclined to think if there could be a correction factor which can be introduced to reduce the offset. Such an approach has been considered by the authors. The use of a correction factor although, would

bring the offset values (within 3% error limits) even more close to desirable values with lesser error, but would result in higher error value during the initial phase of operation. The parameters calculated are under the specific zones of operation and the deviation can be accounted for by the migration of the operating zones away from the zones that have been used for parameter calculations. Such a migration would render these parameters unfit and hence require the calculation of these parameters again.

VII. Conclusion

A number of approaches have been used in the literature to model PEMFC behavior. This paper presents a brief overview about some selected PEMFC static and dynamic models. The list of the presented PEMFC static and dynamic models is obviously far to be exhaustive. The static and dynamics presented models have been selected according to their frequently use in the literature or to their simplicity or the ease of application. We have chosen for the simulation and experimentation one static model given by Larminie and Dicks and one dynamic model given by Becherif and Hissel.

The parameters of these two models have been tuned according to experimental results. The obtained simulation and experimentation results are very close regarding to number of unknown FC parameters.

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