Parametric sensitivity analysis of a PEMFC physics-based model developed for Prognostics

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***Abstract*—In order to face one of the bolt for Proton Exchange Membrane Fuel Cell, PEMFC's industrial breakthrough, the development of Prognostics and Health Management (PHM) is a promising option. Indeed, the prognostics part of the PHM whose goal is to estimate the Remaining Useful Life (RUL) of the system in order to furnish the decision part of PHM the information needed for select the good actions has not been really developed for PEMFC. It is then the next step that need development and that would allow to optimize the use of the fuel cell and then extend it's too short life duration. For that purpose, the development of a behavioral model is necessary as it would be able to predict the dynamic of the fuel cell. A model of this kind is developed here, it is composed of a static and a dynamic part. The ageing in this model can be included by the parameters in each parts. However it is necessary to analyze which parameters is critical for the development of the ageing, as the number of parameter is high face to the data available. The discussion of the parametric sensitivity of the model is presented here as well as the validation of the model.**

I. Introduction

Nowadays, in the actual preoccupation about the ecology, the Proton Exchange Membrane Fuel Cell (PEMFC) is a technology full of promises. Indeed, with a low greenhouse gases emission and a high efficiency, the fuel cells appear to be a solution to face our need of creating energy alternative. An electrical generator based on a PEMFC is a complex system because of the numerous multiphysical phenomena happening inside. It isobviously an electrochemical system, because the production of energy is coming from two oxydo-reduction reactions. The oxidized dihydrogen and the reduced dioxygen trigger a= transfer of electrical charges. Unlike a battery, the fuel cell has permanent supplies of fuel and oxidant the gases. The fuel cell can then be characterized as electrochemical but also electrical, fluidic and thermic, leading to a complex multiphysical system. This technology has not been yet fully developed. Indeed even though the fuel cell has been discovered for more than a century, the development of PEMFC is not mature enough for allowing a real breakthrough in industry. Some bolts have still to be unlocked for making it technically and economically viable. The life duration which is, at this time, too short and not enough managed is one of these bolts. This issue brings unexpected failure at inconvenient timing. In order to meet the target of the life duration, there can be two possible complementary approaches: on one side the development of a more reliable and performing fuel cell stack, and on another side the development of control algorithms that would allow limiting the influence or mitigating degradations.

A method to face this kind of issues is the Prognostics and Health Management [1] which process permits to detect, diagnose, performs prognostics (estimating the Remaining Useful Life (RUL)) in order to take the decisions at the good moment for avoiding degradation and optimizing the use.

According to the litterature [2], for the PEMFC, the first layers of the PHM development has already been investigated in the literature [3], [4]. However, the first step with very few works on it is the prognostics. So, it has to be developed in order to be able to apply the complete PHM process to the PEMFC which final goal would allow managing the life time and expanding it. This is why this paper focuses on the prognostics part of PHM for PEMFC.

Three different kind of approaches can be distinguished for prognostics [5], [6]. Indeed, in order to predict the end of life of a system, its performances has to be reproduced. To achieve this aim, data-based approaches can be considered. In this case, a model is trained thanks to a learning phase, then the model is able to predict the behaviour of the considered system. This approach is also named a black-box approach as there is no physical knowledge required. It implies nevertheless that there is no physical causality with the phenomenon really taking place in the system. A second approach is the model-based ones, in which a precise knowledge of the system is needed but where only a limited number of experiments are required to tune the model. This approach allows also being easily modified regarding new evolving parameters or inputs. A link between the real ageing and the parameters can be drawn. A third approach is the hybrid one; it associates the two firsts that merges their advantages as well as their disadvantages. The frontier between these three approaches can be fuzzy. Indeed with the definition given here and for the PEMFC, a purely model-based approach can't really exist as some data are always needed to tune the parameters.

A model-based approach is here proposed as this would allow obtaining a good precision and even modeling some important internal variables of the fuel cell stack.

The aim of this paper is to present the parametric sensitivity analysis of a physics based model in order to reduce the number that has to be regressed and so minimize the chance of hitting a local minimum during the regressions. For that, the Proton Exchange Membrane Fuel Cell technology is presented in a first part in order to present, in a second part, the behavioral model. In the third part, the model with the ageing included is presented in order to allow the last part which is the parametric sensitivity analysis.

II. Proton Exchange Membrane Fuel Cell

The model proposed being a physics-based development, it is necessary to describe, even succinctly the composition and functioning of a basic cell. As seen on figure 1, a single cell is composed by an anode / electrolyte / cathode assembly (called MEA for membrane electrode assembly) [7]. The electrolyte is a polymer membrane situated between two electrodes. Enough hydrated, it enables the conduction of H+ protons while preventing the conduction of electrons. The MEA is included between two Gaz Diffusion Layer (GDL) which allow the arrival of gaz to the AME and the evacuation of the water produced by the electrochemical reaction. Finally, two metal (or graphite) plates hold mechanically the layers. They are called bi-polarized plates and ensure different functions. First, the channels enhance the gaz routing on the whole surface of AME. Then its thermal properties are used for the heat evacuation, and so used in order to ensure the temperature control thanks to a cooling system. This assembly is a single cell, a PEMFC is generally a stack of cells.

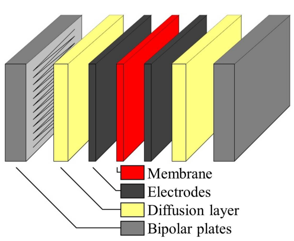

Fig. 1. Composition of a cell

As it can be seen on figure 2, on the anode side, dihydrogen is supplied, on the cathode side, it is dioxygen or air. The conversion of chemical to electrical energy is possible thanks to the reactions happening at the electrodes. At the anode, the dihydrogen is decomposed into H+ and electrons. The protons obtained cross the electrolyte while the electrons go through the external load to reach the cathode. There, the dioxygen react with the ions H+ giving water. Finally, a PEMFC product electricity, water, but heat too.

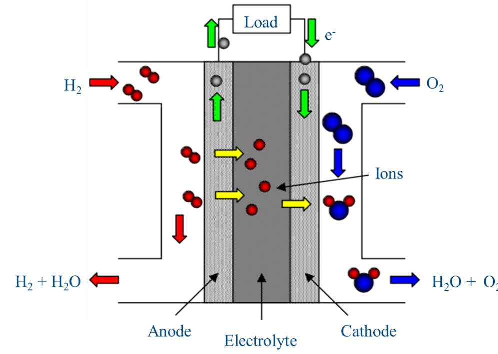

Fig. 2. Description of the operating of a single cell

There are different kind of losses of a PEMFC :

- Fuel crossover losses : results of electrons and wast of fuel passing through the electrolyte.
- Activation losses : caused by the slowness of the reaction taking place on the surface of electrodes.
- Ohmic losses : due to the resistance to the flow of electrons through the electrodes.
- Concentration losses : results from the change of concentration of the reactants through the gas diffusion layer.

III. Instantaneous behavior model

A. Behavioral model description

1) Global model: The model is here a combination of two distinct models as it can be seen on figure 3. The first input is the current and it is normalized as current density to be then decomposed on direct and alternative component. The static and dynamics model are then giving direct and alternative voltage that is finally recomposed as voltage per cell in order to provide the output of the model that is the voltage (for more details about the model refer to [8], [9]).

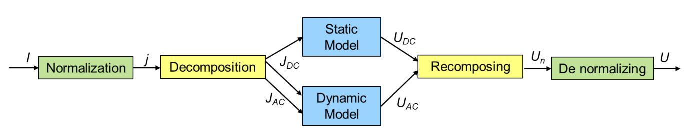

Fig. 3. Scheme of the model

2) Static part of the model: On one side, the static part of the model is based on the Butler-Volmer law with the expression of the voltage drop at the anode and the cathode (η_a and η_c).

$$U_{DC} = E_n - R_m \cdot J_{DC} - \eta_a - \eta_c \tag{1}$$

The expression developped is as follows : (eq. (2))

$$
\begin{aligned}
U_{DC} = & E_n - R_m \cdot J_{DC} \\
& - \frac{1}{b_a} \cdot asinh\left(\frac{J_{DC}}{2 \cdot j_{0a}}\right) \\
& - \frac{1}{b_c} \cdot asinh\left(\frac{J_{DC}}{2 \cdot j_{0c} \cdot \left(1 - \frac{J_{DC}}{j_{Lc}}\right)}\right)
\end{aligned} \tag{2}
$$

The parameters at the anode and at the cathode stand for :

- b_a, b_c the Tafel parameters;
- j_{0a}, j_{0c} the exchange current density, related to the activation phenomenon.
- j_{Lc} the limit current density, related to the diffusion of the oxygen through the gas diffusion layer (GDL).

3) Dynamic part of the model: On the other side, the dynamic part of the model is based on an electrical equivalency (figure 4).

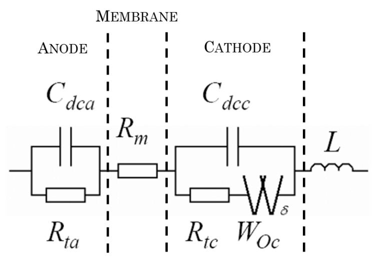

Fig. 4. Scheme of the model

It is chosen to represent the Warburg impedance W_{Oc} by its module R_{Oc} and its time constant τ_{Oc} expressed in the Laplace space(eq. (3)).

$$
W_{Oc}(p) = R_{Oc} \cdot \frac{tanh\left(\sqrt{\tau_{Oc} \cdot p}\right)}{\sqrt{\tau_{Oc} \cdot p}} \tag{3}
$$

The dipoles stand for, respectively at the anode and cathode : C_{dca} and C_{dcc} are the double layer capacities, R_{ta} and R_{tc} two transfer resistances, R_m the ionic conductance of the membrane and L the inductive behavior due to the connectors.

B. Validation of the behavioral model

For validating the model, some real experiment are realized. A 5 cells stack of 100 square centimeters of active area is experimented with a ripple current of 70 A more or less 10% at a $5kHz$ frequency. The experiment is a long term test that lasted around one thousand hours. Some measures as current and voltage are monitored during the whole experiment. Each week, an experimental characterization is realized, which is composed of polarization curves (current - voltage curves) and Electrochemical Impedance Spectroscopies (EIS) at three DC current values.

The static model is validated at each characterizations with a fitting process on a polarization curve. For the dynamic model the same step is realized with a fitting on the Nyquist plots obtained thanks to the EIS. This step shows a good efficiency as the Root Mean Square Errors are very low [9].

The global model need to be simulated entirely in order to evaluate the accuracy. At each characterizations, the model is updated as the fitting on the two parts are realized and then furnish the new set of parameters tuned to the actual state of health of the stack. However, the model then obtained is only accurate around the characterization as it can be seen on figure 5, the curve of the simulated voltage doesn't follow the trend during the 1000 hours, this demonstrates the real need for including the ageing. Nevertheless, the global model is efficient and accurate during a short period of time and after a tuning. Indeed, as it can be seen on figure 6, which represents the model and the real behaviour of the PEMFC tested under the same load; the model is well following the trend during the sixty seconds presented under the dynamic and the static part of the solicitation. The figure 6 is focused on one of the peaks that are going out of the view on figure 5. These peaks are due to the solicitation going at zero for a bit, but showing them in the figure 5 would have no interest as it would prevent us to see the real response of the model during a long amount of time as this is included between 3 V and 3.3 V.

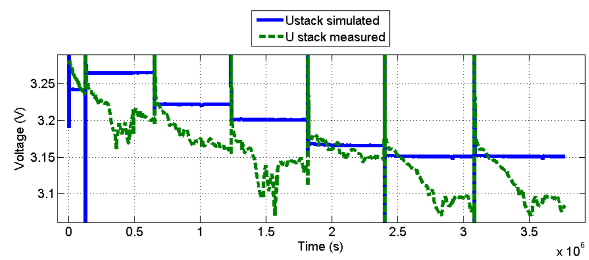

Fig. 5. Comparison of the simulation and the real data under the same solicitation during around 1000 hours

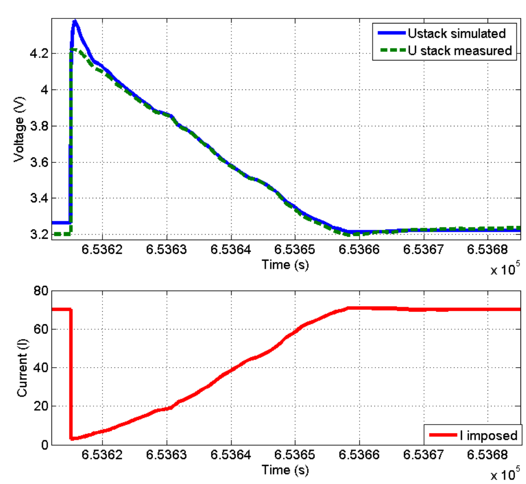

Fig. 6. Comparison of the simulation and the real data under the same solicitation during around 60 seconds

IV. Modeling the ageing

This model is a behavioural one, that reproduces efficiently the fuel cell's voltage under the same solicitation. However, the purpose is here prognostics. For that, the ageing time is added : an other input of the global model is the time-lapse during the fuel cell was operated (Tageing) (figure 7).

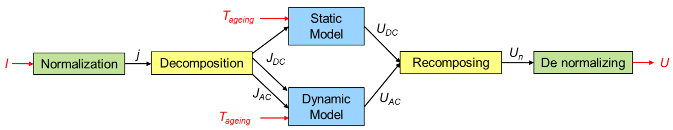

Fig. 7. Scheme of the model with the ageing

For reproducing the ageing, the parameters of the model evolve thanks to a time dependency function. It was chosen, on a first step to use an exponential function.

After each experimental characterization, the values of the models' parameters are regressed based on the experimental results. The evolution of theses parameters is then approximated by a non-linear curve in order to obtain the parameters of the exponential functions, an exemple can be seen on figure 8.

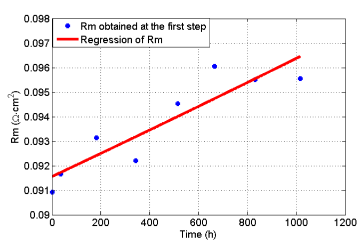

Fig. 8. Example of an exponential fitting of the model's parameters (here R_m)

The model is promising as well on a short amount of time than on the totality of the experiment on which the simulations are based as the mean error during the total experiment is around 5% (figure 9).

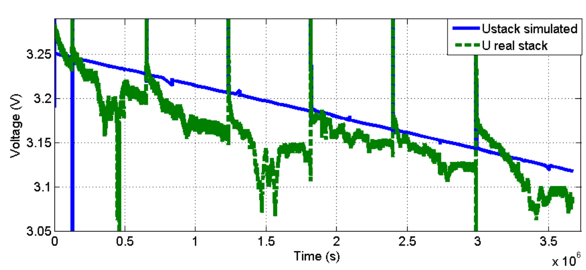

Fig. 9. Comparison of the simulation with ageing and the real data under the same solicitation during around 1000 hours

However, a certain number of parameters (C_{dca}, C_{dcc}, τ_{Oc}, b_c) doesn't seem to evolve with a clear tendency. Consequently, even if the global model with ageing is satisfying, it is not coherent to define a time dependency function for the parameters.

The necessity of having less parameters or constraining them into an interval is important also in order to have a good ageing model. Indeed, the results presented on this ageing part are satisfying but they are not reliable as all the results of the 8 characterization realized are included: the future is already known. A good ageing model rely on a certain quantity of data, even for a physics-based as the one presented here, but the necessity to shorten this amount of data as much as possible is clear. For that a clear tendency in the evolution of the parameters need to be obtain as soon as possible in the experiment.

V. Parametric sensitivity analysis

The global model and its static and dynamic parts present a large number of parameters. Too high maybe for the regressions realized : a local minimal might be found during the updating procedure. A parametric sensitivity analysis is then realized in order to find which parameters are dominant or not, in order to fix or limit the values. It was chosen to realize the evaluation of the influence of each parameters thank to the ANOVA (analysis of variance) method [10].

A. *Static model's parameters sensitivity analysis*

First the analyze is realized on the static part. This model (eq. 2) has the following parameters involved :

- R_m the resistance;
- E_n the Nernst potential;
- b_a, b_c the Tafel parameters;
- j_{0a}, j_{0c} the exchange current density;
- j_{Lc} the limit current density at the cathode only.

The parameters values are set based on previous regressions results and take three values, two extreme realistic values and the middle one (table V-A).

Parameter	Minimum Value	Maximum Value	Unit
R_m	0.08	0.2	$\Omega.cm^2$
E_n	0.9	1	V
b_a	20	100	V^{-1}
b_c	20	100	V^{-1}
j_{0a}	0.001	1	A/cm^2
j_{0c}	0.001	1	A/cm^2
j_{Lc}	1.001	1.5	A/cm^2

TABLE I
STATIC PARAMETERS EXTREME VALUES FOR THE EXPERIMENTAL PLAN

The experimental plan is then realized by simulating the static model with all the combination of parameters possible and evaluating the error of the results with measurements. For this purpose the polarization curve have been taken as a reference and the model was simulated in order to furnish a polarization curve. The error taken for the study is here the Mean Absolute percent error (MAPE) calculated on each point for then calculated the mean. This is realized on the eight polarization curves available that is realized every week.

The influence of each parameter is then calculated for each characterization and is represented on figure 10. The different colors represent the influence for each parameters on the characterizations, from the left for the first to the right for the eighth.

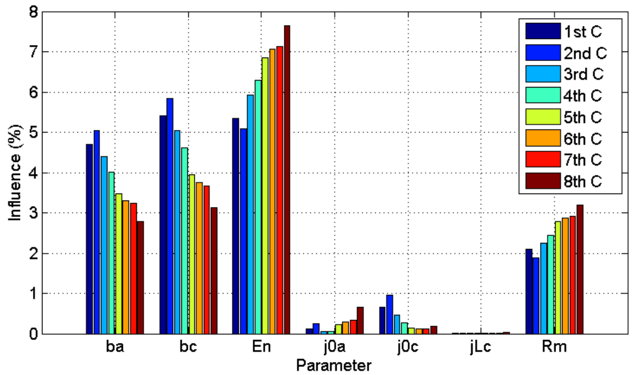

Fig. 10. Static model's parameters influence

Finally, the eight sensitivity analysis show coherent results. Indeed the range of influence of each parameter stay in the same interval. However, the total influence of the parameters on one characterization is around 18%, a very low value for a sensitivity analysis. That can certainly be explained by the short range of value followed and there might be an inter parametric influence, a point non relevant for our study.

The most predominant parameter is E_n, a crucial point as the regressions realized have never given a satisfying value, and for the results presented earlier, this parameter is set fixed. Indeed, a wrong value of this parameter implies directly an important error when the current solicitation is null. It is otherwise a parameter that can be calculated so fixing its value before realizing the regression is consistent. The parameters b_a and b_c are the next most influent parameters and closely R_m. The evolution of b_c doesn't let appear any trend with the ageing time, and seem random: a local minimum might be hit. The low influence of j_{0c} and j_{Lc}, could allow fixing their value and then having the logical evolution of j_{Lc} with the time then be on b_c.

B. Dynamic model's parameters sensitivity analysis

The dynamic model (figure 4) is then studied. The parameters in this model are :

- The Warburg impedance W_{Oc} which is decomposed in two impedances, R_{Oc} and τ_{Oc}.
- The double layer capacities C_{dca} and C_{dcc}.
- Two transfer resistances R_{ta} and R_{tc}.
- The ionic conductance of the membrane is modeled by an equivalent resistance R_m.
- The inductive behavior due to the connectors L.

Thanks to previous regression results, the range of variation for each parameter is set (table V-B) and all the simulations are realized with three level for the parameters.

Parameter	Minimum Value	Maximum Value	Unit
C_{cdca}	0.03	0.06	F/cm^2
C_{dcc}	0.02	0.05	F/cm^2
R_{Oc}	0.05	0.2	$\Omega.cm^2$
τ_{Oc}	0.1	0.6	s
L	0.8E-06	2E-06	H
R_m	0.08	0.2	$\Omega.cm^2$
R_{ta}	0.01	0.6	$\Omega.cm^2$
R_{tc}	0.01	0.4	$\Omega.cm^2$

TABLE II
DYNAMIC PARAMETERS EXTREME VALUES FOR THE EXPERIMENTAL PLAN

The data allowing to evaluate the error of the simulations is the EIS at the nominal current of the experiment ($70A$). The eighth characterization are here also all taken into account in order to confirm the accuracy of the conclusions. For the EIS, the input is the frequency and the Nyquist plot drown as the output represent the real and imaginary part of the impedance. That is why there are two errors calculated for evaluating the dispersion: the error on the real part and the error on the imaginary part. On figure 11 are represented the mean influence on the eight EIS on the real and imaginary part. Figure 12 represents the influence of each parameter on the model on the real and on the imaginary part as they are added. Indeed, this figure is really interesting with the added figures as it was noticed that some parameters have low influence on the real part but this is compensated by the influence on the imaginary part which is bigger.

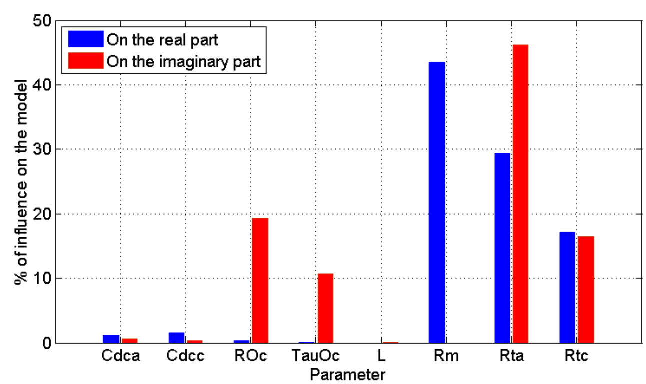

Fig. 11. Dynamic model's parameters influence (comparison of the mean influence on the imaginary and real part)

Finally, the most influent parameters are the resistances R_{ta}, R_{tc} and R_m. A coherent point as for the static parameters the global resistance R_m has a big influence too.

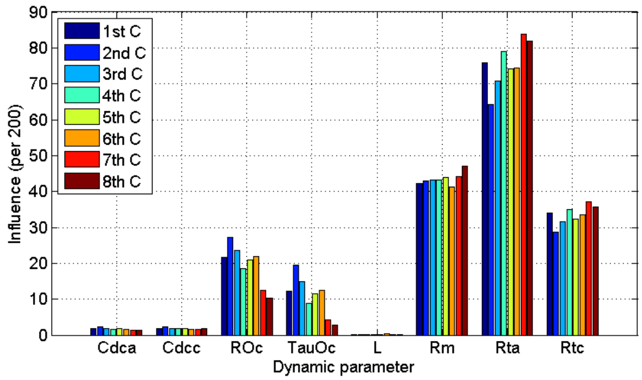

Fig. 12. Dynamic model's parameters influence (total of the influence on the imaginary and real part)

C. Global model's parameters sensitivity analysis

The global model present some dependency between the two parts of the model. Indeed, some parameters in the dynamic model are expressed thanks to static parameters. This is why evaluating the sensitivity of the global model to the parameters seems to be an unavoidable step. The parameters that are directly defined in the global model are :

- R_m the resistance;
- E_n the Nernst potential;
- b_a, b_c the Tafel parameters;
- j_{0a}, j_{0c} the exchange current density;
- j_{Lc} the limit current density at the cathode only.
- The double layer capacities C_{dca} and C_{dcc}.
- The inductive behavior due to the connectors L.
- j_{0Oc} and b_{Oc} that are sub-parameters of R_{Oc}.
- k_{Oc}, sub-parameter of τ_{Oc}.

The dynamic parameters missing here are : R_{ta}, R_{tc}, R_{Oc} and τ_{Oc}. The two last are decomposed in a current dependent function. It was not possible to decompose them for the parameter sensitivity analysis of the dynamic model, as the current is not represented on this last study. The parameters R_{ta} and R_{tc} are finally functions of the static parameters b_a, b_c, j_{0a}, j_{0c} and j_{Lc} in order to have an evolving value with the current.

The experiment plan for the simulations follows the same values than the ones developed on the two last studies for the same parameters (table V-C), three levels are taken for the parameters.

Parameter	Minimum Value	Maximum Value	Unit
E_n	0.9	1	V
b_a	20	100	V^{-1}
b_c	20	100	V^{-1}
j_{0a}	0.001	1	A/cm^2
j_{0c}	0.001	1	A/cm^2
j_{Lc}	1.001	1.5	A/cm^2
R_m	0.08	0.2	$\Omega.cm^2$
C_{cdca}	0.03	0.06	F/cm^2
C_{dcc}	0.02	0.05	F/cm^2
L	0.8E-06	2E-06	H
j_{0Oc}	0.01	0.5	A/cm^2
b_{Oc}	10	30	V^{-1}
k_{Oc}	0.01	0.5	$A.s/cm^2$

TABLE III
GLOBAL MODEL PARAMETERS EXTREME VALUES FOR THE EXPERIMENTAL PLAN

The evaluation of the error was done, one more time, thanks to real data. As the aim of the global model is to reproduce the behavior of the fuel cell, this has to be the basis of the sensitivity analysis. For that experimental data (current and voltage) of around sixty second were taken around a variable solicitation as for example on figure 6. The model under the experimental solicitation was then simulated for calculating the mean error between the simulation and the experimental data.

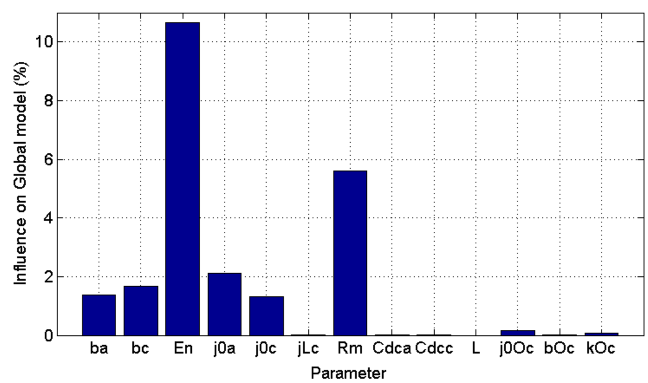

Fig. 13. Global model's parameters influence

The static parameters has more influence this could be explained by the solicitation which is current which is slightly dynamic. The dynamic and static parts has cathode and anode parameters in common in the global model : b_a, b_c, j_{0a}, j_{0c} and j_{Lc}. The fourth first has an important influence, and so evolving them with the time influence both parts, so the global model evolving with the ageing. The last one, j_{Lc}, has clearly not a big influence neither on the static part neither on the global model. However, this parameter has a clear evolution with the time, an exponential fitting is coherent (figure 14) unlike the tafel parameter at the cathode b_c. A hypothesis would be that fixing the value of the limit current density at the cathode j_{Lc} would allow the two other cathode static parameters b_c and j_{0c} to evolve less randomly.

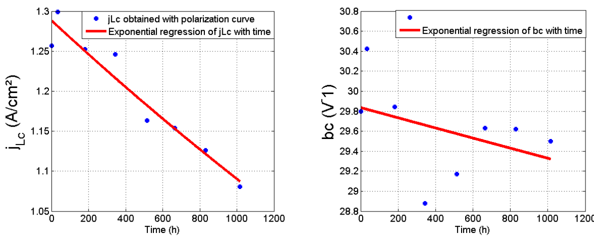

Fig. 14. Evolution of the estimated value of j_{Lc} and b_c with the time and the exponential fitting

The importance of the influence of E_n and R_m is clear. As the value of E_n is fixed, it is not an important point for the ageing modeling. However, for the accuracy of the instantaneous model the calculation of the value must be rigorous. The evolution of R_m with the time is currently coherent with an exponential fitting (less than $8e^{-4}$ Error (RMSE)), and it is important and satisfying as there is no big improvement to achieve.

The very low influence of the parameters strictly coming from the dynamic model bring a discussion. The evolution of theses parameters with the time might be unnecessary. Some study must realized in order to analyze if the evolution of the dynamic behavior with the time can only be contained in the static parameters that intervene.

VI. Conclusion

This paper addresses a model-based approach for prognostics of a PEMFC. With a validation of the behavioral model and the ageing model. In order to have a time dependency, critical point for prognostics, it was chosen to have time evolving parameters. Even though the ageing model is satisfying, the evolution of some parameters have no clear trend with the time. For that purpose, a parameter sensitivity analysis is realized. Indeed a parameter with small influence can be fixed and reduce the number of local minimum during the regressions. In a future work, the comparison between the results and the literature will allow to decide which parameters to fix. It would also allow to verify if the influence of an important parameter is not absorbed by an other. The model will, hopefully present some parameters with a clear evolution, and so a clear justification. With this steps realized, the model will be stable enough to analyze the number of characterization needed before having an accurate reproduction of the behavior with no knowledge of the future.

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