PEMFC aging modeling for prognostics and health assessment ⋆

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Abstract: When a system suffers from a too short lifetime, applying prognostics is a good solution to help taking actions extending its life duration. This solution is applied to Proton Exchange Membrane Fuel Cell (PEMFC) stacks in this paper. An important requirement for prognostics of a PEMFC stack is a well-defined framework as well as a great understanding of the degradation mechanisms and failures occurring within the stack. These requirements are addressed here and allow building an efficient model integrating the different levels (stack - cells - components) as well as the multiple causes leading to degradation. Such a model enables then health assessment and remaining useful life predictions. This work proposes a model built based on a selection of critical degradations and to validate it for both state of health estimations and prognostics. The results show that the stack's state of health during aging can be followed accurately with coefficients of correlation greater than 0.9. Also, the behavior of the system can be assessed with a coefficient of correlation greater than 0.9 showing the great predictive capabilities of the model.

Keywords: State of Health, Prognostics, Proton exchange membrane fuel cell, Aging model.

1. INTRODUCTION

Thanks to the growing maturity of the technology, Proton Exchange Membrane Fuel Cells (PEMFC) become closer to a large scale deployment. They provide an efficient way to convert chemical energy into electricity. However, an extension of their lifetime is required to industrials needs. A possible solution to that problem is the Prognostics and Health Management (PHM). Composed of different layers of activities, PHM aims at taking decisions at the right time to preserve the integrity of the system until it fulfills its mission. PHM of PEMFC is still a new topic of research and a lot of challenges can be highlighted, particularly regarding prognostics (Jouin et al. (2013)).

Prognostics can be considered as the key process of PHM as it enables predicting the future behavior of a system as well as its remaining useful life (RUL) (Gouriveau and Zerhouni (2012)). Prognostics applications of PEMFC are still rare in literature. Two types of approaches can be identified: 1) data-driven approaches based on artificial intelligence tools such as Echo-State Networks (Morando et al. (2013)) or adaptive neuro fuzzy inference systems (Silva et al. (2014)), 2) hybrid approaches based on filtering methods such as Unscented Kalman Filter (UKF) (Zhang and Pisu (2012)) or particle filters (Jouin et al. (2014, 2015a)). By paying more attention to the hybrid approaches which need aging models, it can be seen that the major difficulty is to find a prognostics model that can include the aging and the current solicitation. In (Zhang and Pisu (2012)), the model is based on physics but is restricted to a single component of the fuel cell stack. Whereas in (Jouin et al. (2014, 2015a)), empirical models of the stack aging at constant current are developed, however, they do not include explicitly the current solicitation making their applications with variable current profiles impossible.

This paper aims at integrating the main aging mechanisms selected as critical and their impacts in a prognostics model. The main contributions of that work are the proposal of a working framework leading to the setting and the validation of a new semi-empirical prognostics model that includes both time and current dependencies. To achieve these goals, the paper is organized as follow. First, the background of PEMFC is introduced. This allows introducing the framework and the hypotheses limiting the study. Section 3 presents the setting of the new aging model. Its validation on different mission profiles for health assessment and prognostics is the purpose of Section 4 before concluding.

2. TOWARD PROGNOSTICS OF PEMFC

2.1 An overview of PEMFC

PEMFC is a fuel cell types, differing from the other by the reactants used, the materials of the inner components, its operating conditions and the applications targeted. Descriptions of the different types are presented in (Sharaf

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and Orhan (2014)).
PEMFC uses air (oxygen) and hydrogen to produce electricity, water and heat. It can be encountered alone or combined with other devices such as batteries or ultracapacitors in a wide variety of applications (Wee (2007)) such as transportation (car, bus, boats, etc.), stationary applications (combined heat and power generation (CHP)) or powering of portable devices.
For more details on the system, please refer to Jouin et al. (2015a). In this study, the focus is the stack and its subcomponents, auxiliaries are left aside. However, a great number of factors may impact its functioning and a clear framework for PHM studies has to be proposed.

2.2 Framework for PHM of PEMFC

To clearly fix the limits of the study, it is important to have a precise knowledge of the factors that affects the stack. The literature shows one paper describing and classifying them in an interesting way for PHM, (Kundu et al. (2006)). However, the architecture of the stack environment proposed in this work lacks of precise vocabulary definition and should be further completed to be used in PHM applications. Consequently, a general framework for PHM application is proposed in Figure 1 (upper part for the general case).
In all PEMFC applications, the critical outputs of the stack are the power delivered and the lifetime. These outputs can be impacted positively or negatively according to different causes namely: quality, maintenance, operation and monitoring. The link between the causes and the effects is made by maintenance and degradation pathways.
Quality gathers the physical properties of the stack components, the manufacturing defaults as well as the characteristics of the assembly. Maintenance includes both corrective and predictive maintenances, PHM being an extension of the second one. Then, the operation can be divided into three categories:

- the mission profile, which is limited to the current demand;
- the operating conditions that can be controlled as the stack temperature or reactant pressures, among others;
- the environmental factors that cannot be controlled such as air pollution, vibrations or environment temperature.

Finally, two parts are distinguished in the monitoring category:

(1) "disturbing measure" means creating disturbances in the behavior such as polarization curve measurements or electrochemical impedance spectrometry (EIS) which create power recovery phenomena,;
(2) "no effect measure" means that seems to have no impact on the stack behavior such as voltage, current measurements or other external measures (temperature,pressure ,etc.)along the power supply.

For further explanations on the recovery phenomena, the reader may refer to Jouin et al. (2015a).

Once, the framework defined, the hypotheses of the present study have to be included to keep only the factors of interest for the next steps.

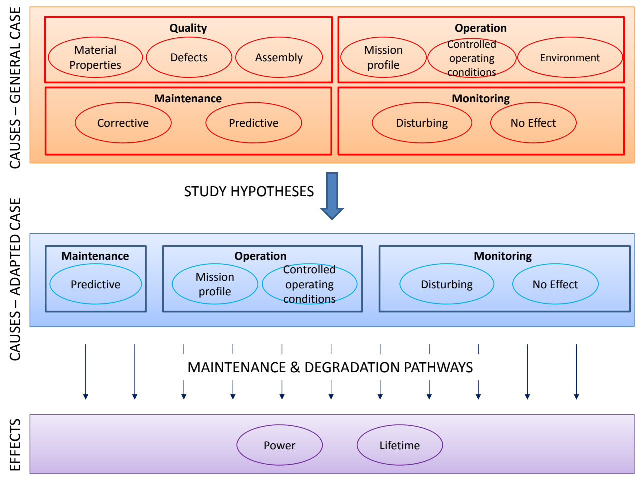

Fig. 1. General framework for PHM of PEMFC

2.3 Hypotheses of the study

Although the quality of the stack can influence the performance, here it is considered as perfect so all the corresponding category is left apart. As nowadays when a failure occurs, in most cases, the whole stack is replaced, maintenance is limited to predictive maintenance. Moreover, the experiments conducted are realized in a controlled environment: the influence of environment can be ignored. Also it is supposed that whatever the current variations, the operating conditions are automatically regulated and set to their optimal values. Other important hypotheses that do not appear on the scheme must be introduced.
The stack cannot suffer from fuel starvation. This limits the impact of operating conditions on the aging, as out of range temperatures or humidities. Then, start-up and shut-down of the system and extreme working temperatures are not considered. Moreover, only phenomena with time constants in hours are taken into account.
Finally, some limitations due to the measure capabilities have to be introduced. The measurements available and of interest for that study are stacks and individual cell voltages, reference and real currents, incoming and outgoing gases/water temperatures and pressures. Finally, punctual measurements of polarization curves are performed.

3. MODELING OF PEMFC AGING

The aging of the stack represents a huge part of PEMFC literature. However, aging models at the stack level are almost nonexistent or, as the one proposed in Laffly et al. (2009) or Franco (2012) not given with enough precision to be used. As a consequence, a new model has to be developed.
Main steps to built this model are described hereafter. They are based on a consequent literature review, but for the sake of clarity only the conclusions drawn from this literature review are presented here.

3.1 Methodology

The main idea is to start from the system degradation and, thanks to different selection processes, go to the aging model. First, all the degradations occurring within the stack are reviewed. They are divided into four categories starting from the higher level, i.e. the stack, to go lower toward its components:

- degradation of the stack;
- degradation of the cells according to their location in the stack;
- degradation of the individual components and their materials (membrane, electrodes, etc.);
- degradation of the interfaces between the components.

Then, the components are classified according to their contribution to the loss of power of the stack and to the reduction of its lifetime. Three classes are defined:

- Class A: membrane and electrodes;
- Class B: GDL and bipolar plates;
- Class C: sealing gaskets;

It allows selecting the membrane and the electrodes as the most critical components. For both, different degradations occur with more or less intensity and more or less critical impacts. So a selection of the leading mechanisms is done for each one thanks to a failure mechanism and criticality analysis. The selected degradations will be introduced when needed for the model construction. The last step of the analysis is to find a starting point for the model. Contrary to the aging models, static models of PEMFC are widely spread in the literature. Among them the widespread polarization curve is the selected starting point for the modeling.

3.2 Modeling

Basic modeling As announced before, the starting point for the behavior modeling is the polarization curve equation. The model is built at the cell level but can easily be adapted to the stack level as it will be seen in the validation section. The main idea is to start from the traditional loss modeling, to select the parameters that age during a long-term functioning and to replace them by a time-dependent expression.

The polarization equation basically models the losses that impacts the reversible cell voltage E_{rev}, also called the Nernst voltage, which is the voltage that would be obtained if all the energy was converted into electricity without any loss. The losses can be divided into four categories: (1) activation losses (E_{act}), (2) concentration losses (E_{conc}), (3) ohmic losses (E_{ohm}) and (4) and crossover losses (E_{cross}). The combination of these losses impacts the voltage, however each one has a different prevalence zone according to the current density. The impact of the concentration and crossover losses will be gathered in a same term. To show the individual contributions of the electrodes the equation is written:

$$E = E_{rev} - E_{conc+cross,a} - E_{conc+cross,c} - E_{ohm} - E_{act,a} - E_{act,c} \quad (1)$$

where index a stands for anode and c for cathode.

As the pure hydrogen diffuses better than the oxygen in the nitrogen and water, the concentration losses at the anode can be neglected. The equation becomes:

$$E = E_{rev} - E_{act,a} - E_{act,c} - E_{ohm} - E_{conc+cross,c} \quad (2)$$

By replacing the losses by their expressions (please refer to Sharaf and Orhan (2014) for more details), the polarization equation is now written as a function of i, the current density:

$$E(i) = E_{rev} - \frac{RT}{2\alpha_a F}.ln(\frac{i_{loss}+i}{i_{0,a}}) - \frac{RT}{4\alpha_c F}.ln(\frac{i_{loss}+i}{i_{0,c}}) - i.(R_{ion} + R_{ele} + R_{cr}) + B_c.ln(1 - \frac{i}{i_{max,c}}) \quad (3)$$

where: R is the gas constant, T is the stack temperature, α_a and α_c are the charge transfer coefficients at the electrodes, F is the Faraday's constant, i_{loss} represents the internal currents within the stack, $i_{0,a}$ and $i_{0,c}$ are the exchange current densities at each electrode, R_{ion}, R_{ele} and R_{cr} are respectively, the ionic, electronic and contact resistances, B_c is an empirical parameter allowing taking into account the effect of water and gas accumulations leading to non-uniform current densities on the electrode and $i_{max,c}$ is the limiting current at the cathode.

However, it can be seen that the time does not appear in the equation, preventing from using it to describe the aging.

Introduction of the aging To select the parameters that are aging with time, the first step is to classify all the variables appearing in equation (3) into three categories:

- *constants:* R, F;
- *controlled:* T, P, E_{rev}, $i_{0,a}$, $i_{0,c}$;
- *aging:* α_a, α_c, i_{loss}, R_{ion}, R_{ele}, R_{cr}, B_c and $i_{max,c}$.

The parameters classified in the constant and controlled categories do not need to be justified but some of the other may need more explanations.

α_a and α_c, the charge transfer coefficients depend on, at least, the material of the electrode, its microstructure and the reaction mechanism (oxidation or reduction). The structure of the electrodes and its activity change with the aging. So it is logical to assume that the charge transfer coefficients vary. However, its value is very often set to make the polarization equation fit to the data, so it seems impossible with the current knowledge to guess how it varies with time.

Then regarding i_{loss}, it is assimilated to the hydrogen crossover current. Indeed, when the membrane ages, some micro-holes appear letting hydrogen passing to the cathode side. The hydrogen reacts directly with the oxygen creating an exothermic combustion reaction that can lead to a fast destruction of the membrane. Some attempts of crossover modeling can be found in the literature (Baik et al. (2013)). Nevertheless, the modeling that seems the most suitable here is the exponential modeling:

$$i_{loss}(t) = i_{loss,0}.exp(b_{loss}.t) \quad (4)$$

Indeed, this trend is shown in the great majority of the experiments reported in the literature (Liu and Case (2006); Jao et al. (2012); Wu et al. (2014)) and other models have not been fully validated until now.

Next parameters are the resistances appearing in the ohmic loss term. In the initial formulation, three resistances are distinguished: ionic, electronic and contact resistances. As electronic and contact resistance can be difficult to study separately, they are gathered in a same variable $R = R_{ele} + R_{cr}$. From the measurements reported in different studies, its aging can be defined by:

$$R(t) = R_0 + b_R.t \quad (5)$$

Regarding the ionic resistance linked to the membrane, recent results published in Collette et al. (2013) show that the conductivity as well as the water uptake and the ion exchange capacity for pieces of membrane in different Nafion decrease exponentially as function of time (in days):

$$R_{ion}(t) = R_{ion,0}.exp(b_{ion}.t) \quad (6)$$

Although this expression assumes that only time influences the conductivity and not contamination or water repartition changes, this hypothesis will be kept afterwards.

Finally, the two variables of the activation losses have to be considered. As stated before, B_c allows taking into account the effect of water and gas accumulations leading to non-uniform current densities on the electrode. Both degradation and operating events may affect these accumulations and so make B_c value change. The degradation of the GDL, mainly the loss of hydrophobicity, strongly impacts the diffusion but also the content of water and its distribution and accumulation in the electrode compartment. This should impact B_c by increasing this value during aging. So the following modeling is proposed:

$$B_c(t) = B_{c,0} + b_B.t \quad (7)$$

The same idea has to be employed to model $i_{max,c}$. Indeed, according to Morgan and Datta (2014), the limiting current density of the cathode can be written: $i_{c,L} = \frac{4F}{RT}\left(\frac{D_{O_2}}{L_{GDL}}\right)P_{O_2}$, where D_{O_2} is the diffusivity of oxygen and L_{GDL} the thickness of the GDL. In this expression, the thickness of the GDL may be affected by the carbon corrosion during the aging and the diffusivity may be influenced by degradation in the same way that of B_c. As the thickness of the GDL may not vary from more than some μm, the choice not to model its decrease during aging is made. For the diffusivity, the same modeling as for B_c is used:

$$D_{O_2}(t) = D_{O_2,0} + b_D.t \quad (8)$$

Both the equations (7) and (8) are hard to justify physically. The linear expressions are inspired by the results we had in Jouin et al. (2015a).

One last thing to do is to replace the current density value by a function of the current imposed to the stack I:

$$i(t) = \frac{I(t)}{A(t)} \quad (9)$$

where $A(t)$ is the active area of the electrode that decreases with the aging given by a combination of exponentials (Zhang and Pisu (2012); Liu and Case (2006)):

$$A(t) = A_0.exp(b_{A1}.t) + A_1.exp(b_{A2}.t) \quad (10)$$

with A_0 equals to the theoretical geometric size of the active area and A_1 must be contained in $[-1, 1]$ and reflects the error that can exist on the actual size of the active area. The model described by equation (3) is built for a single cell. It is multiplied by the number of cells (n) to obtain the stack voltage. However, some works tend to show that all cells do not degrade in the same way within the stack (Bose et al. (2013); Radev et al. (2013)). The cells next to the edges of the stack degrade faster and this impacts the global voltage. Consequently, the classical expression of power $P_{stack} = n.I.V_{cell}$ has to be modified to include this heterogeneity. For that purpose, a corrective term written p is introduced as no existing study allows to quantify the degradation differences within a stack. By replacing the parameters by their expressions, equations (4) to (10), the final expression of the power is:

$$\begin{aligned} P(I,t) = n.I(t).[E_{rev} \\ & - \frac{RT}{2\alpha_a F}.ln(\frac{i_{loss,0}.exp(b_{loss}.t) + \frac{I(t)}{A_0.exp(b_{A1}.t)+A_1.exp(b_{A2}.t)}}{i_{0,a}}) \\ & - \frac{RT}{4\alpha_c F}.ln(\frac{i_{loss,0}.exp(b_{loss}.t) + \frac{I(t)}{A_0.exp(b_{A1}.t)+A_1.exp(b_{A2}.t)}}{i_{0,c}}) \\ & - \frac{I(t)}{A_0.exp(b_{A1}.t) + A_1.exp(b_{A2}.t)}.(R_{ion,0}.exp(b_{ion}.t) + R_0 + b_R.t) \\ & + (B_{c,0} + b_B.t).ln(1 - \frac{\frac{I(t)}{A_0.exp(b_{A1}.t)+A_1.exp(b_{A2}.t)}}{\frac{4F}{RT}\left(\frac{D_{O_2,0}+b_D.t}{L_{GDL}}\right)P_{O_2}})] - p \end{aligned} \quad (11)$$

4. EXPERIMENTS AND VALIDATION

The model must now be validated. This validation comes in two steps:

(1) the first validation step proves that the model can be identified and matches to the dataset available (Section 4.1.1);
(2) then the ability of the model to predict the future behavior of a stack is demonstrated thanks to a particle filter-based prognostics (Section 4.2).

4.1 Model validation for health assessment

For this first validation step, the behavioral model is tested by fitting the model to data. Both power aging and polarization curves made all along the aging are modeled. To limit the length of the paper, the results are illustrated only with a constant current mission profile. However, the model has a generic nature and its validation with a micro-CHP profile is proposed in Jouin et al. (2015b).

4.1.1. Constant current solicitation The dataset available for this case is referred as D1. It comes from a 5-cell stacks with an active area A_0 of $100cm^2$ aged during 1150 hours. More details on D1 are available in FCLAB (2014). It was produced by the manufacturer UBzM. 8 polarization curves are available for D1. To keep only the part that aged at constant current, the data are cut at 985 hours.

To start the model identification, the initial polarization curve made before the aging is estimated so i varies and some unknown parameters called Set 1 (α_a, α_c, A_1, $i_{0,a}$, $i_{0,c}$, $i_{loss,0}$, $R_{ion,0}$, R_0, $B_{c,0}$, $D_{O_2,0}$) are initialized. The identification is performed using a least square algorithm thanks to the fitting toolbox of Matlab software. Then i is fixed, the time varies and the power is estimated giving the parameters of Set 2 (b_{loss}, b_{A1}, b_{A2}, b_{ion}, b_R, b_B, b_D, p) with the same method.

4.1.2. Initial polarization curve estimate To initialize the test procedure, distributions of possible values for the parameters of all sets are built thanks to the literature and adjusted thanks to the data. Indeed, to obtain a convincing fitting, all the values should reflect the reality and respect some constrains. As an example, α_a and α_c should be in the interval $[0, 1]$ and their sum equal or close to 1.

A first polarization curve is estimated at t=0 hours (Figure 2). This eliminates time-depending terms in equation (11) allows initializing the coefficients from Set 1.

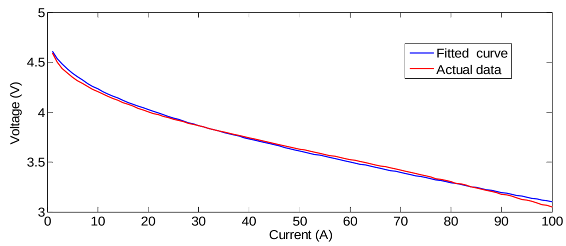

Fig. 2. Initial polarization curve of the 5-cell stack D1

4.1.3. Power behavior estimate Set 2 is identified by fitting the equation 11 to the power data. By comparing the model and the data, as shown on Figure 3, it can be seen that the global aging trend is well-followed by the model. To help evaluating the model, the coefficient of determination R2 is calculated: R2 = 0.9616. It is higher than 0.9 and validates the model for constant current solicitation.

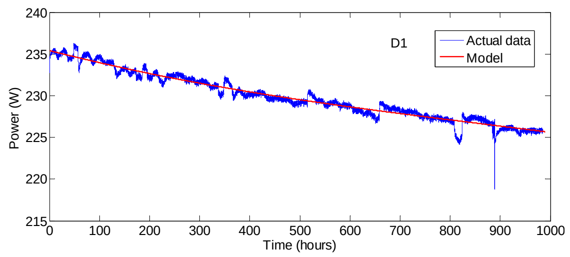

Fig. 3. Power supplied by the 5-cell stack D1 measured experimentally versus aging time at 0.7 A/cm^2 and comparison with the model

4.2 Prognostics of a PEMFC stack

4.2.1. Particle filters for prognostics For this application, we choose a particle filter as a prognostics tool as in our previous works Jouin et al. (2014, 2015a). Indeed, particle filter is used to estimate a future nonlinear state and to adjust the parameters of the model at the same time. Moreover, such a tool has a great ability to deal with uncertainty as it generates a probabilistic output to represent the state of the system. To emphasize more on the results and the consequent discussion, the functioning of particle filters is not presented here and the reader is invited to refer to our previous papers Jouin et al. (2014, 2015a). However, the prognostics principle is quickly explained.
First the dataset is split into a training set and a set to predict. For illustration purpose, a training set of 500 hours is chosen. Then a first identification of the model is made by fitting the model to the training set. The obtained coefficients might not be the true values obtained when the whole dataset is used. However, they are good starting points to initialize the particle filter. The next step is to modify equation (11) to obtain a recursive form of the model that expresses the state at a time k from the state at time $k-1$ and the current control at time k. As the model coefficients have also to be evaluated, equations for their update must also be defined. A classical strategy is to use a random walk process:

$$param_i(t_k) = param_i(t_k) + \omega \qquad (12)$$

where ω is a white Gaussian noise with a zero-mean and a well chosen variance that is small enough to allows a sufficiently fast convergence to the actual parameter while being large enough to offer a great diversity of pathways.

4.2.2. Power prediction To illustrate the prognostics, a power prediction with the previously proposed training length of 500 hours is performed on D1. As the model parameters are estimated by random walks, the predictions are repeated 100 times to decrease the influence of the random processes. This allows keeping the median trajectory as the behavior prediction and using the distribution of predictions as confidence intervals. The results are illustrated on Figure 4.

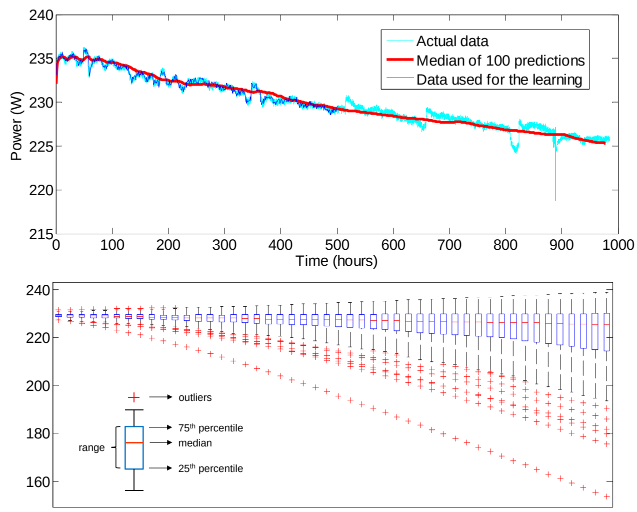

Fig. 4. Upper part - Median power prediction for a learning of 500 hours; lower part - dispersions of the 100 tries

4.3 Discussion

The first point to discuss is the parameter identified values. As the initialization intervals were all built to give plausible results, the coherence regarding the order of magnitude of estimated parameters and real measurements is considered as correct. A question harder to discuss is: can these values be the real values ? For the great majority of the parameters in Set 1 such as $i_{loss,0}$, $R_{ion,0}$, R_0 or $D_{O_2,0}$ with no measurements on the cells prior to the aging, no answer can be given. Same for Set 2 with precise measurements during the aging, the question is hard to answer. However, we decide to consider that approximated values are acceptable in the modeling as long as they can catch the behavior of the stack during aging.
Then the prognostics results proposed here are quite convincing for a first application of the aging model built in this paper. Indeed, the behavior prediction follows quite well the trend of the actual data. The correlation coefficient is 0.9578 which is very satisfying. However, the uncertainty of the prediction remains very high: around 15 W between the 25^{th} and the 75^{th} percentiles for the last hours of the predictions. One way to reduce this uncertainty is to reduce the part of random processes in the prognostics. It means that better solutions must be found to update the parameters than the random walk process and it will be the next step of this work.

5. CONCLUSION

This paper proposes a new solution to perform health assessment and prognostics of PEMFC stacks. First, a suited framework is proposed to perform PHM. It describes precisely the factors influencing the major outputs of the stacks, namely its power and its lifetime.

Then a new aging model is built starting from a classical static modeling of the stack and adding time dependency based on a deep degradation study. This model allows performing both health assessment and prognostics. Indeed, it fits to the dataset with a correlation coefficient greater than 0.9. Also, a first prognostics application shows that this model possesses predictive capabilities when used with a particle filters.

Even if the prognostics shows some weaknesses regarding the uncertainty coming with the predictions, the behavior is estimated with a correlation coefficient greater than 0.95. A next step of this work will focus on reducing the uncertainty to offer more precise predictions.

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