# Prognostics of PEM fuel cells under a combined heat and power profile ⋆

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**Abstract:** Prognostics have started to be applied to Proton Exchange Membrane Fuel Cells (PEMFC). Indeed, it seems an interesting solution to help taking actions that will extend their lifetime. PEMFC are promising solution for combined heat and power generation ($\mu$CHP). As power suppliers, they cannot afford running to failure. This work presents a prognostics application on a PEMFC following a $\mu$CHP profile. A critical issue with such a mission profile is to be able to model the variation of the power demand. So a key point of this work is the presentation of a model introducing the time dependency of the mission profile as well as the degradations of different inner components of the PEMFC. This model starts from a classical polarization expression transformed based on a detailed understanding of the degradation phenomena and the introduction of time-varying parameters. This model is able to follow accurately the behavior of the PEMFC during its functioning. It is then used to perform prognostics and predict the future behavior of the stack with a particle filter-based framework. The results are very encouraging as the behavior predictions are accurate, with a low uncertainty and an horizon as great as thirty days.

*Keywords:* State of Health, Prognostics, Proton exchange membrane fuel cell, micro-cogeneration.

## 1. INTRODUCTION

Proton Exchange Membrane Fuel Cells (PEMFC) have started their spreading in the industry. Their ability to produce electricity and heat while rejecting water from hydrogen and oxygen make them inescapable for the future of energy conversion. Their range of applications (Wee (2007)) goes through transportation (car, bus, boats, etc.), stationary applications (combined heat and power generation ($\mu$CHP)) or powering of portable devices.
Nevertheless, their lifetime still has to be extended to meet the current needs of industry. An approach to tackle this problem is to anticipate the failures, and more precisely thanks to Prognostics and Health Management (PHM) (Jouin et al. (2013)). PHM is a set of activities that starts from the monitoring the system, then goes to an analyzing process consisting in health assessment, diagnostic and prognostics which finally leads to decisions aiming at helping the system fulfills its mission while preserving its integrity. Within the PHM process, prognostics can be considered as a key element to manage the system. Indeed, it aims at predicting its future states and failures as well as its remaining useful life (RUL).
Various prognostics applications on PEMFC have started to appear during the last two years. They can be divided into two types of approaches: 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. (2014a,b)). However, none of them has been able yet to predict the behavior of PEMFC with great variation of power. Yet, these kind of variations are very likely to happen in all PEMFC application.
Consequently, this work proposes a prognostics approach that enable predicting the future behavior of a PEMFC used with a $\mu$CHP mission profile. The main contribution of the paper is the proposal of a behavioral model including degradation phenomena and method able to predict the behavior with a high coefficient of determination with the data. This model is transformed and used in a particle filter to make predictions of the system behavior. This allows estimating the future state of the system almost on month before with a reasonable certainty.
The present paper is organized as follows. First, an overview on PEMFC is proposed to better understand the system and also to set the context of the study. Then in Section 3, a new modeling of the system behavior including degradation is developed. Then the prognostics framework based on a particle filter is introduced in Section 4. This leads then to the model validation is Section 5 and the presentation of the prognostics results. Finally, some remarks about future developments are proposed in the conclusion.

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## 2. TOWARDS PROGNOSTICS OF PEMFC

### *2.1 An overview of PEMFC*

PEMFC is one of the fuel cell types, differing from the other by the reactants used, the materials of inner components, the operating conditions and the application targeted. Descriptions of the different types of fuel cells are presented in (Sharaf and Orhan (2014)). A 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 ultra-capacitors in a wide variety of applications as introduced earlier.
Different levels of granularity exist when PEMFC is concerned. First, a PEMFC system refers to a PEMFC stack and all the ancillaries it is interacting with such as energy collectors, reactant storages, pumps, etc. A non-exhaustive illustration is proposed on Figure 1(a). The stack is the part that converts the energy thanks to an assembly of elementary cells. Their number can vary from a single one to several hundreds depending of the expected output power. Finally, a cell is composed of different components (Figure 1(b)): 2 bipolar plates to bring the reactant and collect electrons; 2 gas diffusion layers (GDL) to diffuse reactants toward the electrodes and help to products removal; 2 electrodes (anode and cathode) where occur the oxidation and reduction reactions; a proton exchange membrane and sealing gaskets to ensure the tightness of the cell.
To provide electricity, different reactions occur within the stack (Figure 1(c)). At the anode side, hydrogen is oxidized to split into electrons and protons. The electrons go through an external circuit to provide electricity and the protons go through the membrane to the cathode side. Here they meet again the electrons, and also the oxygen provided to the stack to form water during the reduction reaction. The reactions involved at the electrodes are:

$$2H_2 \rightarrow 4H^+ + 4e^- \quad (1)$$

$$O_2 + 4H^+ + 4e^- \rightarrow 2H_2O \quad (2)$$

The general reaction equation of the system is:

$$2H_2 + O_2 \rightarrow 2H_2O + electricity + heat \quad (3)$$

Different output powers can be obtained from a stack according the application case. It depends on the input mission profile which is expressed in terms of current solicitation (in Amperes). The mission profile strongly impacts the lifetime of the stack. Also, the ancillaries make the operating conditions varying (temperatures, pressures, reactant flows, products evacuations, etc.) to allow the stack to stay in the best operating conditions. If not, a severe degradation may happen. In this study, the focus is the stack and its subcomponents.

### *2.2 Hypotheses of the study*

Although the quality of the stack can influence the performance, here it is considered as well manufactured. Moreover, the experiments conducted are realized in a controlled environment: the influence of environment can be ignored. And it is supposed that whatever the current variations, the operating conditions are automatically regulated and set to their optimal values.
The stack cannot suffer from fuel starvation. This limits

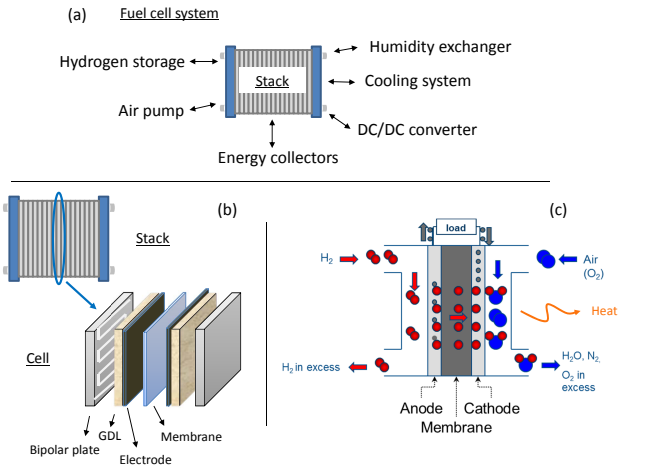


Fig. 1. (a) The stack within the fuel cell system, (b) the components of the stack and (c) its functioning

the impact of operating conditions on the aging, as out of range temperatures or humidities for example. 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 are:

- stacks and individual cell voltages;
- time;
- reference and real currents;
- incoming and outgoing gases/water temperatures;
- incoming and outgoing gases pressures;
- relative humidities of incoming and outgoing gases;
- stoichiometries.

Finally, punctual measurements of polarization curves are performed.

## 3. MODELING OF PEMFC AGING

As the focus of the paper is on prognostics, the complete setting of the behavioral model can not be fully detailed here. Only the main steps are mentioned.
The basis of the model is the polarization curve equation. That polarization equation basically models the losses that impact the reversible cell voltage $E_{rev}$, also called the Nernst voltage. This voltage would be obtained if all the energy was converted into electricity without any loss. The losses can be divided into four categories:

- activation losses ($E_{act}$);
- concentration losses ($E_{conc}$);
- ohmic losses ($E_{ohm}$);
- and crossover losses ($E_{cross}$);

The combination of these losses impacts the voltage, however each has a different prevalence zone according to the current density as can be seen on Figure 2. Consequently, the polarization equation is given by:

$$E = E_{rev} - E_{conc+cross} - E_{ohm} - E_{act} \quad (4)$$

The impact of the concentration and crossover losses are gathered in a same term. As pure hydrogen diffuses better than the oxygen in the nitrogen and water, the concentration losses at the anode can be neglected. By

differentiating the contribution of each electrodes and adding this last hypothesis, the equation becomes:

$$E = E_{rev} - E_{act,a} - E_{act,c} - E_{ohm} - E_{conc+cross,c} \quad (5)$$

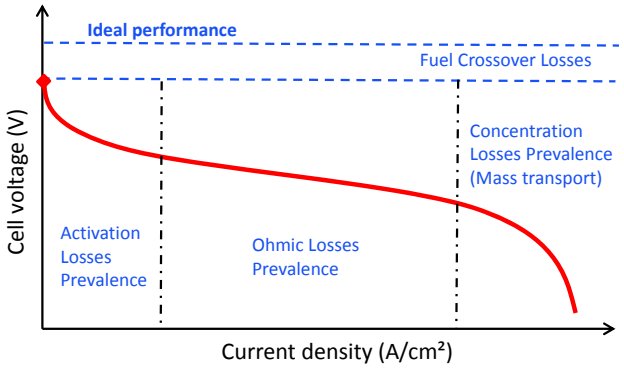


Fig. 2. Representation of the different losses on the polarization curve

By replacing the losses by their expressions (please refer to Sharaf and Orhan (2014) for more details), the polarization equation can be written as a function of $i$, the current density. The current density varies according to the output power expected from the stack.

$$E(i) = E_{rev} - \frac{RT}{2\alpha_a F}.ln(\frac{i_{loss}+i}{i_{0,a}}) - \frac{RT}{4\alpha_c F}.ln(\frac{i_{loss}+i}{i_{0,c}}) - i.(R_{ion}+R_{ele}+R_{cr}) + B_c.ln(1-\frac{i}{i_{max,c}}) \quad (6)$$

where:
- $R$ is the gas constant equal to 8.3145 $J.mol^{-1}.K^{-1}$;
- $T$ is the stack temperature;
- $\alpha_a$ and $\alpha_c$ are the charge transfer coefficients at the anode and at the cathode;
- $F$ is the Faraday's constant equal to 96 485 $C.mol^{-1}$;
- $i_{loss}$ represents the internal currents within the stack. Here we assume that it can be assimilated to the hydrogen crossover current alone and that no current caused by membrane shorting appears;
- $i_{0,a}$ 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.
- $i_{max,c}$ is the limiting current at the cathode, it is the asymptotic value of the current for which the rate of disappearance of the product equals to the rate of their transport.

The power demand might not be satisfied all along the stack lifetime as numerous degradations occur within the stack during the aging. This has to be modeled and introduced in the last equation. A literature review shows that the main phenomena to take into account are: the loss of active area at the electrodes, the crossover of hydrogen through the membrane, the variations in the ohmic and protonic resistances and the changes gas diffusion. Moreover, the model described by equation (6) is built for a single cell, so it is multiplied by the number of cells in the stack ($n$) to obtain the stack voltage. Some works tend to show that cells degrade in an heterogeneous way according to their location within the stack (Bose et al. (2013); Radev et al. (2013)). This is taken into account by introducing a corrective term $p$ that models these heterogeneities:

$$V_{stack} = n.V_{mean,cell} - p \quad (7)$$

Finally, by replacing some parameters by expressions modeling their aging, using (7) and multiplying by the current to obtain a power, the final model is:

$$\begin{aligned} P(I,t) = n.I(t).[&E_{rev} \\ &- \frac{RT}{2\alpha_a F}.ln(\frac{i_{loss,0}.exp(b_{loss}.t) + \frac{I(t)}{A_0.exp(b_{A1}.t)+A_1.exp(b_{A2}.t)}}{i_{0,a}}) \\ &- \frac{RT}{4\alpha_c F}.ln(\frac{i_{loss,0}.exp(b_{loss}.t) + \frac{I(t)}{A_0.exp(b_{A1}.t)+A_1.exp(b_{A2}.t)}}{i_{0,c}}) \\ &- \frac{I(t)}{A_0.exp(b_{A1}.t)+A_1.exp(b_{A2}.t)}.(R_{ion,0}.exp(b_{ion}.t) + R_0 + b_R.t) \\ &+ (B_{c,0}+b_B.t).ln(1 - \frac{\frac{I(t)}{A_0.exp(b_{A1}.t)+A_1.exp(b_{A2}.t)}}{\frac{4F}{RT}\left(\frac{D_{O_2,0}+b_D.t}{L_{GDL}}\right)P_{O_2}})] - p \end{aligned} \quad (8)$$

A validation of this model is proposed in Section 5.

## 4. MODEL UPDATING AND PROGNOSTICS WITH PARTICLE FILTERS

This section presents the tool elected to perform prognostics, namely, the particle filter. It also introduces the model transformation necessary to use equation (8) within the prognostics framework.

### *4.1 Particle filters for prognostics*

For this prognostics application, a particle filtering framework is chosen. Particle filters have the ability to estimate a nonlinear state and to adjust the parameters of the model at the same time. Moreover, as it generates a probabilistic output to represent the state of the system, it greatly helps to deal with the uncertainty inherent to prognostics.

*Problem statement* To use particle filters, the problem has to use the Bayesian framework formulation. A state and a measurement model are defined according to the following expressions:

$$x_k = f(x_{k-1}, u_k, \omega_k) \quad (9)$$

$$z_k = h(x_k, v_k) \quad (10)$$

where $\{x_k, k \in \mathbb{N}\}$ describes the evolution of the state with time and is modeled as a Markov process of initial distribution $p(x_0)$ and a transition equation $p(x_k|x_{k-1})$ obtained from (9); $u_k$ is the input of the system (here the power demand); $\{y_k, k \in \mathbb{N}^*\}$, are measurements, assumed to be conditionally independent given the process $\{x_k, k \in \mathbb{N}\}$ and of marginal distribution $p(z_k|x_k)$; finally $\omega_k$ and $v_k$ are process and measurement noises.
In a Bayesian approach, the state is estimated recursively according to two stages:

(1) *prediction*: it uses the state model to estimate the current state at time $k$ via Chapman-Kolmogorov equation:

$$p(x_k|z_{1:k-1}) = \int p(x_k|x_{k-1})p(x_{k-1}|z_{k-1})dx_{k-1} \quad (11)$$

(2) *update*: it uses the latest measurement to modify the prediction probability density function (pdf) with the Bayes rule:

$$p(x_k|z_{1:k}) = \frac{p(z_k|x_k)p(x_k|z_{1:k})}{p(z_k|z_{1:k-1})} \quad (12)$$

This forms the optimal Bayesian solution. However, it is only a conceptual solution: in general it cannot be determined analytically. For that purpose, a whole family of filtering tools exists ranging from the Kalman filter (KF) and its variations (extended KF or unscented KF), histograms and particle filters.

*Particle filter principle* An illustration of the principle is proposed in figure 3. To solve the Bayesian problem, the particle filter performs as follow:

(1) *Initialization*: N particles are created based on the initial state of the system. According to Thrun et al. (2005), if one knows the values of the initial state $x_0$, the initial distribution should be initialized with a point mass distribution centered on the correct value of $x_0$ and 0 probability assigned anywhere else in the state space;
(2) *Prediction*: at time $k$, the prior probability distribution $p(x_k|x_{k-1})$ is evaluated thanks to the state model, the evaluated state $x_{k-1}$ at time $k-1$ and the inputs of the system $u_k$.;
(3) *Update*: a new measurement is available, weights are calculated and given to the particles according to their matching with the last measurement (likelihood).
(4) *Resampling*: the particles with low weights are eliminated and the other duplicated. Resampling is equivalent to modifying the random measure by improving the exploration of the state space at $k+1$ (Li et al. (2014)).

### *4.2 Model update*

The first step is to modify equation (8) 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. Indeed, with a good initialization and a well chosen number of particles, it will converge to the actual parameter values according to the law of large numbers. Consequently, to perform prognostics, we have a set of expressions:

- one to express the PEMFC state:

$$P(t_k, I_k) = P(t_{k-1}, I_{k-1}) + \frac{\Delta I}{I_{k-1}}(P(t_{k-1}, I_{k-1}) + p) + n.(I_{k-1} + \Delta I).(residual.terms) \quad (13)$$

  As the recursive expression is very long, all the terms that not contain the state $P(t_{k-1}, I_{k-1})$ are gathered in the expression $residual.terms$. An important hypothesis is made during the expression transformation, we suppose that the current is never exactly equal to 0 even at the open voltage circuit;
- 8 to update the parameters:

$$param_i(t_k) = param_i(t_{k-1}) + \omega \quad (14)$$

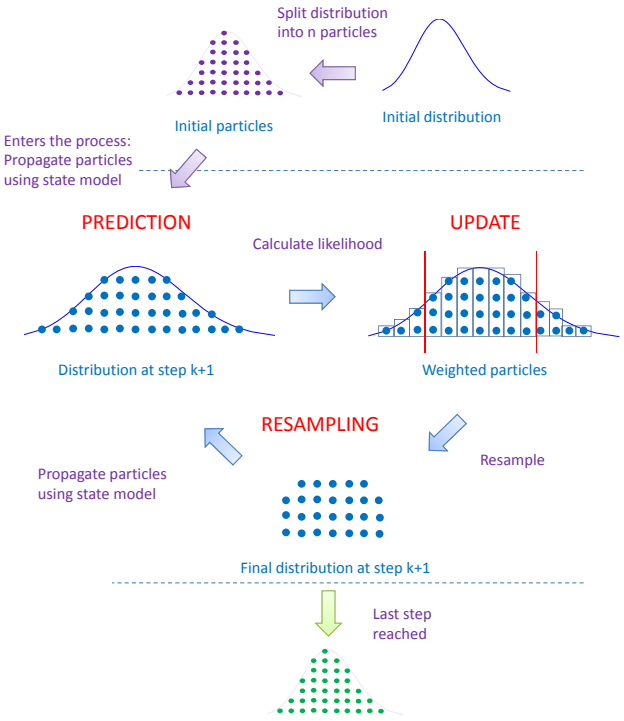


Fig. 3. Particle filter principle

where $\omega$ 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.

## 5. EXPERIMENTS AND DISCUSSION

### *5.1 Micro-cogeneration mission profile*

The data available come from stack manufactured by the CEA LITEN (Technology G). The stack is composed of 8 cells with a total active area of 220 $cm^2$. The mission profile used for the experiments is punctuated by characterizations and is proposed in Figure 4. It was made to simulate the mission profile of a combined heat and power unit all along a year. It starts with a demand that should be encountered during winters during which a building has high heating requirements, from approximately 0 to 350 hours. Then comes the spring until approximately 750 hours, the summer until 1030 hours and finally the beginning of automn.

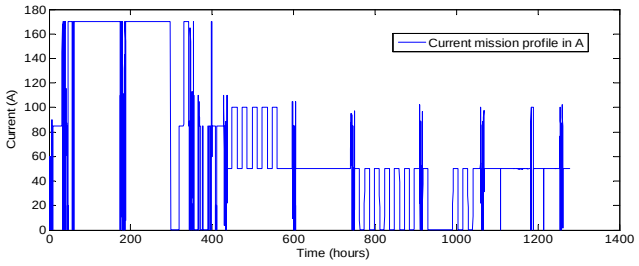


Fig. 4. Current mission profile for the 8-cell stack

### 5.2 Model validation

Before performing prognostics, the ability of the model to catch the stack behavior is checked on the whole dataset. To do so, the time is set to $t = 0$ and a first set of parameters ($\alpha_a$, $\alpha_c$, $i_{0,a}$, $i_{0,c}$, $i_{loss,0}$, $A_1$, $R_{ion,0}$, $R_0$, $B_{c,0}$, $D_{O_2,0}$) is identified by fitting the initial polarization curve thanks to a least square algorithm. Then the missing parameters ($b_{loss}$, $b_{A1}$, $b_{A2}$, $b_{ion}$, $b_R$, $b_{B,aging}$, $b_{D,aging}$, p) are identified by fitting the power using the same method. The model fits almost perfectly to the data as it can be seen in figure 5 with a coefficient of determination R2 is equal to 0.9968 and it allows validating the model for a micro-cogeneration profile.

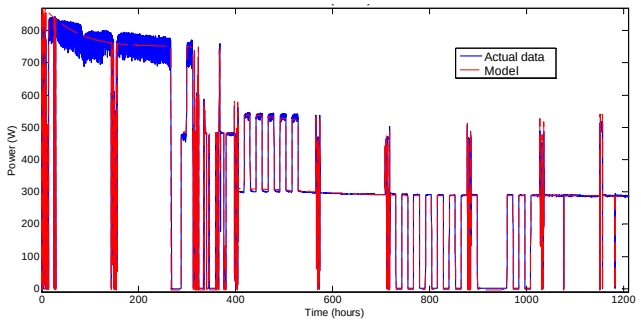


Fig. 5. Power supplied by the 8-cell stack measured experimentally versus aging time and comparison with the model

### 5.3 Power predictions

The data are split into two parts: a training set used to learn the model and a validation set that has to be predicted. To perform the predictions, we assume that the future power demand is known. In a real case application, it could be evaluated from historical data. For illustrative purpose, results with a learning of 600 hours and other for a learning of 800 are proposed in figure 6.

To evaluate the results, the coefficient of determination R2 is calculated for both learning and prediction for all the learning length tested. It is depicted in figure 7. This coefficient shows that the model learns well the behavior of the stack and also has great predictive capabilities. Indeed even with an horizon of 800 hours (i.e. a learning of 400 hours), the prediction have a R2 greater than 0.6 with data. And logically, this goes greater when the horizon decreases to reach almost 0.8 for an horizon of 300 hours. Moreover, it can clearly be seen in figure 6 that the confidence interval remains narrow around the prediction. This gives a good credibility to the results.

## 6. CONCLUSION

This work proposes a new prognostics application on a concrete industrial case, namely the combined heat and power generation with PEMFC. It uses simple voltage and current measurements to predict the future behavior of the stack expressed in terms of expected power. This is very interesting as these values are easy to access and do not imply a heavy equipment to get information of the system's state of health.

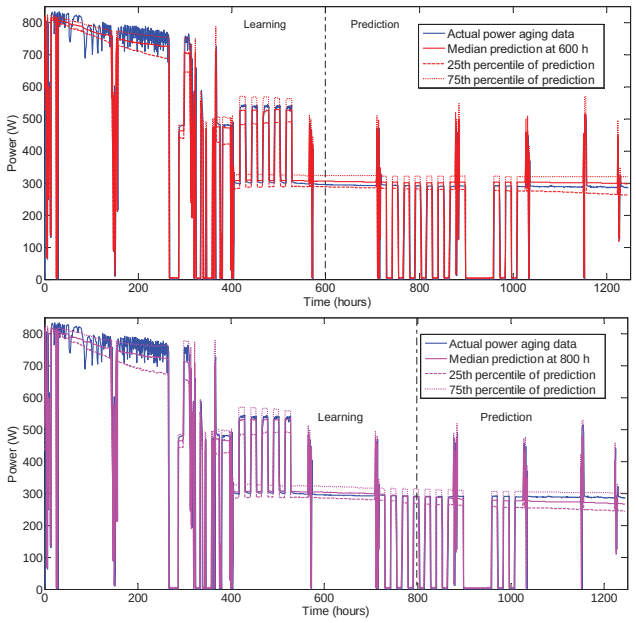


Fig. 6. Power prediction results: upper part - with a learning of 600 hours; lower part - with a learning of 800 hours

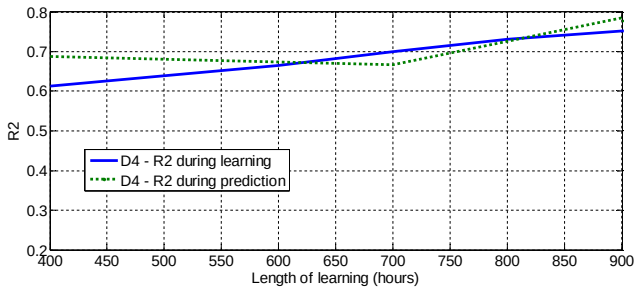


Fig. 7. Coefficient of determination during learning and prediction

A model is built based on a well-known equation but introducing the time dependency of the mission profile as well as the degradations of different components of the stack. A proof of the model ability to follow the behavior of the stack is presented. With this basis, prognostics is performed and allows to obtain very good predictions with a reduce uncertainty with an horizon of approximately 30 days.
A next step of this work is to perform prediction on data coming from stacks that ran to failure and confirm that it is well-suited to predict the RUL. Another point will be to apply such a method with other mission profiles such as transportation to validate its generic aspect.

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