

Estimating the end-of-life of PEM fuel cells: Guidelines and metrics

Marine Jouin, Mathieu Bressel, Simon Morando, Rafael Gouriveau, Daniel Hissel, Marie Péra, Noureddine Zerhouni, Samir Jemei, Mickaël Hilairet, Belkacem Bouamama

To cite this version:

Marine Jouin, Mathieu Bressel, Simon Morando, Rafael Gouriveau, Daniel Hissel, et al.. Estimating the end-of-life of PEM fuel cells: Guidelines and metrics. Applied Energy, 2016, 177, pp.87 - 97. hal-02380401

HAL Id: hal-02380401 <https://hal.science/hal-02380401v1>

Submitted on 26 Nov 2019

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Estimating the end-of-life of PEM fuel cells : guidelines and metrics

Marine Jouin^{1,2*}, Mathieu Bressel^{1,2,3}, Simon Morando^{1,2}, Rafael Gouriveau^{1,2}, Daniel Hissel^{1,2}, Marie-Cécile Péra^{1,2}, Noureddine Zerhouni^{1,2}, Samir Jemei^{1,2}, Mickael Hilairet^{1,2}, Belkacem Ould Bouamama³

¹ FEMTO-ST Institute, UMR CNRS 6174 - UBFC / ENSMM / UTBM / UFC, 24 rue Alain Savary, 25000 Besancon, France $2FC-LAB$ Research, FR CNRS 3539, Rue Thierry Mieg, 90010 Belfort, France

³CRIStAL, UMR CNRS 9189 - Université de Lille-Sciences et Technologies, Avenue Paul Langevin, 59655 Villeneuve d'Ascq, France

name.lastname@femto-st.fr

Abstract

Prognostics applications on PEMFC are developing these last years. Indeed, taking decision to extend the lifetime of a PEMFC stack based on behavior and remaining useful life predictions is seen as a promising solution to tackle the too short life's issue of PEMFCs. However, the development of prognostics shows some lacks in the literature. Indeed, performing prognostics requires health indicators that reflect the state of health of stack, while being able to interpret them in an industrial context. It is also important to propose criteria to set its end of life. Moreover, to trust any prognostics' application, one should be able to evaluate the performance of its algorithms with respect to standards. To help launching a discussion on these subjects among scientific and industrial actors, this paper addresses some of the issues encountered when performing prognostics of a PEMFC stack. After showing the link between prognostics and decision, this paper proposes guidelines to set the limits of a prognostics approach. The definitions of healthy and degraded modes are discussed as well as how to choose the time instant to perform predictions. Then, three criteria based on the power produced by the stack are proposed as indicators of the state of health of the stack. The definition of the end of life of the stack is also discussed before proposing some criteria to assess the performance of any prognostics algorithm on a PEMFC. Some perspectives of works are also discussed before concluding.

Keywords: Proton exchange membrane (PEM) fuel cell, Prognostics, Remaining useful life, PHM

1. Introduction

²³ of great interest and has started to appear in the PEMFC field through Prognostics and Health Management (PHM) $[7, 8]$.

PHM is an engineering discipline originated from the predictive maintenance. It is defined by [9] as "a maintenance and asset management approach utilizing signals, mea-²⁹ surements, models and algorithms to detect, assess and track degraded health, and to predict failure progression". In other terms, by continuously monitoring the system, it is possible to detect and anticipate, thanks to prognostics, the occurrence of failures and to take decisions at the right time to preserve the system. These actions enable the system to reach the end of its mission. Prognostics is widely accepted as a key stage of PHM. Indeed, based on the system knowledge and historical data, it allows predicting the future State of Health (SoH) of the system. Prognostics is defined as the "estimation of the operating time before failure and the risk of existence or later appearance of one or more failure modes" [10]. The operating time before failure is more commonly known as the Remaining Useful ⁴³ Life (RUL).

 During the last years, prognostics has started to be ap- plied to PEMFC. Different types of approaches are pro- posed: model-based approaches [11, 12, 13], data-driven approaches $[14, 15, 16]$ and a combination of both, hy-

In the current context of energy transition, re-³ placing fossil energies by alternative power sources such as fuel cells appears as a promising solution ⁵ [1, 2]. Fuel cells can be used in a great number of ⁶ applications such as transportation, mobile elec-⁷ tronic devices, micro-CHP (Combined Heat and ⁸ Power), etc. [3, 4].

⁹ Despite their great industrial potential and large scale-¹⁰ projects all around the world, fuel cells experience diffi-¹¹ culty to enter the energy market and this industry remains ¹² brittle [5]. Actual obstacles are, among others, high ex-¹³ ploitation cost, public acceptance and life duration that ¹⁴ remains to short [6]. Indeed, for the fuel cells of interest in ¹⁵ this paper, namely Proton Exchange Membrane Fuel Cells ¹⁶ (PEMFC), current lifetimes are around 2000-3000 hours ¹⁷ when 8000 hours are needed for transportation applica-¹⁸ tions and 100 000 hours for stationary. Different options ¹⁹ are available to tackle the lifetime issue: working on the ²⁰ material, reducing the causes of degradation, improving ²¹ the stack design, implementing new supervision and man-22 agement strategies, etc. This last solution appears to be $\frac{1}{45}$

 $\overline{\text{Corresponding author}}$, Tel.: +33 (0)3 81 40 29 04, Fax +33 (0)3 81 40 28 09

brid approaches [17]. The growing interest of the scientific community to that subject has also been shown thanks to the IEEE PHM data challenge 2014 [18] in which dif- ferent propositions for SoH predictions [19, 20] and RUL estimates [21, 22] were presented.

 Despite those recent works, a deep thinking and standards proposal on how to perform prognostics on PEMFC are needed. Although different consid- erations about prognostics standards are available [9, 23], they are too general and further question-⁵⁸ ing is required for the PEMFC case.

- ⁵⁹ 1. Which level of granularity should be chosen on the ⁶⁰ PEMFC?
- ⁶¹ 2. How to define the healthy and degraded modes for a ⁶² PEMFC?
- ⁶³ 3. When prognostics should be performed?
- ⁶⁴ 4. Which health indicator should be used to perform ⁶⁵ prognostics?
- ⁶⁶ 5. How to define the End-of-Life (EoL) of a PEMFC?
- ⁶⁷ 6. How can the prognostics' performances be evaluated?

 To the authors' knowledge none of these questions are answered in the literature. Consequently, this paper intends to propose solutions and guidance to start answering these questions and going to- wards standardization. The main contribution is to propose general solutions that are completely independent of prognostics tools and that can be adapted with any approach. New practitioners can use these guidelines as tutorial to start performing 77 prognostics of PEMFC.

 To adopt a logical reasoning, the paper follows the chrono- logical order of the prognostics process' stages (Fig 1). First, the time constants involved in the prognostics are discussed as well as the level of granularity recommended for prognostics in Section 2. Section 3 focuses on the data selection and their processing before defining health indi- cators in Section 4. This section also discusses the EoL definition. The SoH definition is considered in Section 5 in addition to the time instant to start predicting. Finally, some performance metrics are defined in Section 6 before concluding.

89 2. Decision levels and system granularity

⁹⁰ 2.1. Decision levels and time constants

⁹¹ Prognostics intends to provide the future SoH of a sys-92 tem and its RUL based on data, system knowledge and the 108 93 expected mission profile. This generally creates a closed109 94 loop, Fig. 2. This figure could be completed with other110 95 PHM processes but only the ones of interest for this paper₁₁₁ ⁹⁶ are kept.

97 The system initiates its mission based on an ex-113 98 pected mission profile. Data are collected and pro-114 99 cessed continuously for health assessment. This 100 provides an input for prognostics. These data are¹¹⁶

Figure 1: Stages of the prognostics' process

Figure 2: The system-prognostics-decision loop

 used, alone or combined with a model according to the prognostics' approach, to predict the future 103 SoH based on the planned mission profile. Accord- ing to the predictions, this profile can be modified and adapted to the actual capabilities of the sys- tem. This process is repeated until the system reaches its mission or fails.

Different levels of decision exist with respect to different time scales (Fig. 3). Regarding PEMFC, the time scales go from the microseconds to months (Fig. 4). Among all the phenomena occurring within the PEMFC, the accumula-¹¹² tion of degradation and ageing effects are the main factors that shorten the RUL $[24]$. Consequently, to perform prognostics, phenomena with time constants greater than the hour are considered. This enables making mid-term and long term predictions and the consequent decisions. Lower ¹¹⁷ scales are more related to adaptive control or system re-¹¹⁸ configuration.

Figure 3: Different levels of decision and link with prognostics

Figure 4: Time scales in a PEMFC adapted from [25]

 Covering all possible uses, events and configuration en- countered by a PEMFC in a single paper would be a hard task. Consequently, it is assumed that prognostics is per- formed on a single stack working in its nominal conditions. The decision is out of the scope of that paper, even if it is 161 mentioned later to argue some points.

¹²⁵ 2.2. Stack vs component level

¹²⁶ From the previous paragraphs, it seems obvious to fo- 127 cus on degradation phenomena to perform prognostics. $^{166}\,$ ¹²⁸ Degradations occur at all levels of the stack granular- 129 ity: stack/cells/components and at the interfaces between 168 $_{130}$ them [26]. All the published approaches are dealing with¹⁶⁹ 131 prognostics at the stack level (Section 5). The main reason¹⁷⁰ ¹³² is that the data acquisition is very often performed at this¹⁷⁷ ¹³³ level.

 However, working at the cell level can be an interesting op- tion. Indeed, the functioning of the stack can be suddenly stopped when a cell reaches a minimum voltage value. ¹³⁷ This value is given by the manufacturer to automatically¹⁷⁶ 138 shut down the stack when a cell seems too damaged and 177 so prevent safety issues. Performing prognostics on cells would allow anticipating such events.

¹⁴¹ An issue appears with this type of prognostics: cells are 142 not degrading in a uniform way within the stack $[27, 28]$.

Cells near the edges tend to degrade faster. As for an il- lustration, Fig. 5 depicts the individual cell voltages for a stack aged with a constant mission profile. Cell 1 refers to the cell near to the hydrogen input. This figure clearly shows the voltage heterogeneity among the cells. As an ex- ample, at the end of the experiments, cell 1 has lost 21.6% of its initial performance while cell 3 lost only 8.8%. Some information can be extracted from these measurements, such as the mean cell voltage value or the standard devi- ation. It helps isolating the cells that may come prema- turely to their EoL. Based on this information, it can be decided to perform prognostics on the most degraded cells only. It could be done on all cells but it would not be practical for stacks that have more than 10 cells. More- over, nowadays, stack reconstructions to replace only these faulty cells are not systematic due to all the assembly con- strains. Also, a stack can still convert energy even with faulty cells. So such a level of study can be questionable.

current of $60A$ [14] Figure 5: Individual cell voltages for a 5-cell stack aged at a constant

168 be predicted. Some failure thresholds have already been 0 ¹⁶⁷ sistances (or conductivities) or materials integrity could 50 ¹⁶⁶ other critical parameters such as hydrogen crossover, re-100 ¹⁶⁴ of the electrochemical active surface area (ECSA) is pre-150 $\frac{162}{162}$ formed at a smaller level, namely the component level. Another perspective for prognostics may be being per-¹⁶³ It has already been initiated in [13] in which the size dicted with a model-based prognostics framework. Many proposed for some of them. For instance, a crossover cur- 170 rent of 10 mA.cm⁻² is proposed in [29] as the EoL of the membrane subject to hydrogen crossover. However, different limitations appear when performing prognostics at the cell or components levels:

- 1. the accessibility of the parameters: this point is discussed in [8]. Accessing the inner parameters of the stack, when it is possible, almost always disturbs the stack behavior and tends to accentuate its degradation:
- ¹⁷⁹ 2. the lack of validated degradation models;
	- 3. the degradation at the interfaces between the components or with the auxiliaries may not be taken into

account.

183 According to all ahead considerations, in all the remain-²³⁶ der of the paper, the discussion focus on a prognostics²³⁷ performed at the stack level.

186 3. Data selection and processing

3.1. Data collection

 In an industrial operating environment, very few physi- cal parameters are easily accessible with current technolo- gies. It is the reason why, the existing prognostics method- ologies focus on indicators built based on voltages mea-surements such as energy indicators.

 Voltages and current are the most common measurements on PEMFC as they allow obtaining the power provided by the system whenever during its life. Even if it is moni- tored, the current is supposed to be known as it is defined by the mission profile.

 Also, some punctual measurements can be available. Of- ten, an initial polarization curve as well as some EIS (Elec- trochemical Impedance Spectroscopy) measurements are given by the manufacturer or measured before the stack starts its mission. However, as it is not sure yet that these punctual measurements can be performed during the whole lifetime of an embedded system, there are left aside in the remaining of the paper.

 Fig. 5 shows that limiting the measurements to the power still provide a good indicator of the stack aging. Indeed, constant current experiments are typically reflecting the "natural" aging of the stack, i.e. an aging not influenced by the mission profile if the operating conditions are kept nominal. The power decreases with time indicating the $_{212}$ evolution of different degradation mechanisms inside the₂₄₀ 213 PEMFC. This explains why some papers use the volt- $_{241}$ 214 age/power to evaluate the degradation rate of the system₂₄₂ [30, 31]. Finally, [32] shows that almost all the degrada- $_{243}$ tions can be perceived in this signal. $_{217}$ Consequently, the focus is set on the use of voltage or, by₂₄₅ 218 proportionality with respect to the current, power mea- $_{246}$

3.2. Degradation vs transient phenomena

surements.

 Based on the previous comments, the focus is set on the observation of the degradation thanks to power measure- ments. However, the degradations are mixed with tran- sient phenomena when time scales in hours are consid- ered (Fig. 4). Fig. 6 represents the power measured 254 during the aging of two 5-cell stacks at a constant current of 60A and 70A with weekly polarization **curves and EIS.** It highlights that these punctual charac-257 terizations disturb the power and create transient stages. This observation could be expanded to all events bringing the stack out of its nominal conditions and that can cre- ate reorganization of water and gas repartition within the $_{233}$ stack [17, 33, 34]. Very often, these events appear as re- $_{262}$ coveries in the power signal. As the transient stages have

 a low duration with respect to the degradation, and the border between both of them is not always clearly defined, they should be taken with caution, or even removed, for prognostics.

Figure 6: Upper part: Power during the aging of a 5-cell stack at a constant current of 60A [14], Lower part: Power during the aging of a 5-cell stack at a constant current of 70A [18]

3.3. Data processing

Data can be easily processed to work only on the degradation part of the signal. To do so, the raw signal power is reduced and filtered. As seen in Fig. 4, because the effect of the aging is noticeable within a few hours, the dynamic of degradation is easily separable among the others.

The time constant of degradations is at least in hours while the data frequency acquisition is commonly in seconds. Reducing the data can be of great interest, as it often con- dition the speed of the prognostics' algorithms as long as it does not remove any useful information. One should avoid over sampling: the SoH information is flooded among the other dynamics. In addition, the computational cost for performing the prognostics might be too high making the implementation of the real-time algorithm challenging. Finally, one should consider the memory required to store the huge amount of data induced by the over sampling. Different options are proposed in the literature: $[12, 26]$ use a hourly sampling period while $[14]$ prefers a sampling period of 50 hours.

Then, to remove all the components of the signal that are not linked to degradation, simple methods can be used. By representing, the spectrum of a PEMFC power signal in the frequency domain (Fig.), it can be observed that the main component of the signal is located in the low

 frequency domain. It allows using a simple low-pass fil- ter, namely the moving average, to remove noise and fast phenomena from the signal. As it does not remove all the effects of the transient stages, the signal can also be smoothed, with a loess algorithm for example [26]. These stages of the signal processing are illustrated in Fig. 6.

Figure 7: Spectra of the data proposed in Fig. 6 and 12

²⁷⁰ 4. Health indicators and stack's EoL

²⁷¹ 4.1. Health indicators

272 Different indicators can be proposed as prognostics cri- $^{297}_{20}$ ²⁷³ teria, three of them are studied here:

- ²⁷⁴ 1. the power;
- ²⁷⁵ 2. the cumulative energy;
- ²⁷⁶ 3. the efficiency with respect of the current.

 The proper indicator should be chosen according 304 278 to the PEMFC use. The power (or the voltage) issos recommended for constant mission profiles while the cumulative energy and the efficiency are more $_{281}$ adapted for variable mission profiles. Indeed, when $_{308}$ the current varies, the power is not yet a mono- tonic indicator preventing from fixing a failure threshold. Further discussion on this issue is pro- $_{285}$ vided in Section 4.2.2. The case of the power as health $_{312}$ indicator has already been evoked in the previous sections, so the emphasize is set here on the cumulative energy and the efficiency.

$4.1.1.$ The cumulative energy

A classical approach in prognostics is to construct a monotonic indicator based on measurements. For a PEMFC stack the energy produced during the lifetime is accessible thanks to the voltage measurement U , the current profile i and the time of operation T and is written:

$$
E(t = 0 : T) = \int_0^T U(t) \dot{.} i(t) \, dt \tag{1}
$$

It provides a quantitative indicator of the PEMFC performance and as it is monotonic, fixing a failure threshold becomes easier.

The data proposed further in Fig. 12 are used for illustration purpose. Representing the measured cumulative energy is quite simple based on the measurements. However, it has to be compared to a reference and building it may not be simple.

The first solution to build the reference is based on the hypothesis that an initial polarization curve is available. It gives the voltage corresponding to a particular current solicitation when no degradation has occurred yet. Usually, only a few points are measured to obtain the polarization curve, so some voltages corresponding to the current encountered in the mission profile may be missing. A simple method to solve the problem is to fit the data to the polarization equation [6]:

$$
E = E_{rev} - E_{conc+cross,a} - E_{conc+cross,c}
$$

$$
- E_{ohm} - E_{act,a} - E_{act,c} \quad (2)
$$

 Different strategies can be found in the literature to per- form that fitting [35, 36] but using a simple algorithm of the least squares family seems sufficient. This fitting en- ables covering a larger range of current values. Once the fitted curve built, obtaining a reference for the cumulative energy is straightforward (see Fig. 8). For each current value, the corresponding voltage can easily be calculated with the fitted equation.

The second solution to build the reference is to use a be-²⁹⁹ havioural model that describes the evolution of the voltage ³⁰⁰ or the power according to the current profile as if no degra-³⁰¹ dation was occurring within the stack. As an example, the ³⁰² reference in Fig. 9 and 12 are built based on the model pro- $\frac{303}{200}$ posed in [26] making the current varying at time t=0 to remove the effect of degradations. However, such models are still scarce and have to be further validated.

Once the reference is set, the collected data are used to build the observed cumulative energy and compared to the reference (Fig. 9). The difference between the observations and the reference (ΔCE) can be calculated at each time instant and converted into a percentage of energy loss to help estimating the RUL.

Working with the cumulative energy has the advantage to work with a monotonic indicator. So, it can be adaptable whatever the mission profile. Also working in terms ³¹⁵ of cumulative energy (so in the watt.hour unit) enables

Figure 8: Setting of the reference for the cumulative energy based 341 on the initial polarization curve

Figure 9: Comparison of measured cumulative energy and a reference (data from Fig. 12)

 comparisons with other types of energy sources. $_{317}$ However, some issues may be encountered when building₃₅₉ the reference. First, it has to be verified that the un- $_{360}$ certainty on the initial polarization curve is reasonable to $_{361}$ consider it as a reference basis. Then, a comparison be- tween the power measured at the early stages of the stack $_{363}$ life and the power calculated with the polarization curve should be performed. If the difference between these two₃₆₅ powers is too high, the reference for the cumulative energy $_{366}$ $\frac{1}{325}$ should not be built based on the initial polarization curve. Finally, using a model to build the reference seems to be an interesting solution. However, the lack of validated models should be filled to enable such a solution.

$329 \quad 4.1.2$. The efficiency

The efficiency is a classic measure for energetic system. For a PEMFC, the efficiency η_{stack} is defined as [37]:

$$
\eta_{stack} = \frac{P_{stack}}{P_{chemical}} \tag{3}
$$

where P_{stack} refers to the output electrical power previously defined $(U \times I)$, and $P_{chemical}$ refers to the energy flux contained in the reactants:

$$
P_{chemical} = -\dot{n}_{H2} \Delta h_f \tag{4}
$$

330 where Δh_f is the enthalpy of formation of a mole of water. ³³¹ It also corresponds to the heat released by the complete 332 combustion of a mole of hydrogen; and \dot{n}_{H2} is the molar ³³³ flowrate of hydrogen consumed by the cell to supply the ³³⁴ power.

³³⁵ As seen in these equations, the efficiency is strongly linked ³³⁶ to the mission profile and the nominal conditions set for ³³⁷ each current value I. If the nominal operating conditions 338 can be maintained each time the stack supply a current I_0 , 339 during the aging of the stack, $P_{chemical}(I_0)$ should remain ³⁴⁰ constant through time, and the efficiency should decrease with P_{stack} .

³⁴² There is not an unique criterion to perform prognostics. ³⁴³ One has to be selected according to the industrial needs ³⁴⁴ of the prognostics' applications.

³⁴⁵ 4.2. EoL definition

³⁴⁶ Two thresholds can be defined for prognostics of 347 PEMFC stack:

- ³⁴⁸ 1. a threshold of conformity to a mission:
- ³⁴⁹ 2. a definitive End of Life threshold.

 The first allows to decide if the fuel cell stack is able to perform a given mission (e.g. providing 1kW during 1 year) which does not mean that the stack is out of use once that it can be operated in degraded mode. The definitive EoL is the inability for the PEMFC to deliver power in safe conditions. It is more difficult to define with a stack scale parameter once that the EoL is usually related to the component level (e.g. massive hydrogen crossover).

³⁵⁸ 4.2.1. Conformity to a mission threshold

³⁵⁹ Defining the conformity criterion is the responsibility of the user and depends on his needs. For instance, if the fuel cell is operated at constant power, the criterion might be the ability to produce a requested power as illustrated in Fig. 10. In this example, the requested power density 364 is 0.4 W.cm⁻² which corresponds to the nominal power density given by the manufacturer. At time 500 hours, the PEMFC is no longer able to provide that power density and can not pursue the given mission.

As stated earlier, when variable mission profile is considered, a threshold on the power might not be suitable anymore. This is the reason why a threshold on the cumulative energy is proposed. Lets consider the difference between reference and real cumulative energy:

$$
\Delta CE(t) = CE_{ref}(t) - CE_{real}(t)
$$
\n(5)

One can defined a threshold of 10% of the reference cumulative energy:

$$
\Delta CE_{max}(t) = \frac{10}{100} \cdot CE_{ref}(t)
$$
\n(6)

Figure 10: Power density for different level of aging

$$
\int_{0}^{T} P_{real}(t).dt = \frac{90}{100} \int_{0}^{T} P_{ref}(t).dt
$$
\n(7)³⁹₃₉

 which is strictly equivalent to the power threshold for constant solicitation. However, this threshold is a function of the power profile and so can also work for variable requested power.

372

Chen et al [38] proposes an economical lifetime threshold₄₀₁ where the fuel cell (that powers an electrical bus) has to be replaced when its average cost is the lowest (see Fig. 11). It is a compromise between the profitability (that increase with time) and the performance (that decrease with aging). This threshold takes into account the price of the stack Q_{stack} , the consumption of hydrogen Q_{H_2} (that increase with aging) and the system efficiency Q_{ope} (because auxiliaries consume power).

$$
Q_{total}(t) = Q_{stack} + \int_{t=0}^{T} Q_{ope} dt + \int_{t=0}^{T} Q_{H_2} dt
$$
 (8)

 The authors states that using the fuel cell after the eco- nomic lifetime is reached is cost-effective for the users. Nevertheless, this approach supposes a known speed of degradation for a given bus route so the future SoH can be forecasted.

Figure 11: PEMFC average cost changes with service time repro-⁴¹³ duced from [38]

³⁷⁸ 4.2.2. Definitive End of Life threshold

 There is currently one definition of the EoL based on the power. The US Department of Energy (DoE) defines the EoL as a loss of 10% of the initial performance [39]. Even if the value could be discussed, it is not the main problem of this definition. Using a fixed threshold for con- stant current application is very convenient and does not create any problem. The EoL of the stack is defined as a ratio of the initial performance as illustrated in Fig. 6.

³⁸⁷ However, it does not work so well when the mission pro-³⁸⁸ file exhibits current variations. Fig. 12 shows an example 389 of power measured under a μ -CHP (Combined Heat and ³⁹⁰ Power) profile with the theoretical power that should be ³⁹¹ expected if the stack was not aging. It can be seen that ³⁹² according to the level of current, the loss of performance 33 does not follow a constant percentage with respect to the μ reference. Indeed, at 170 A, 15\% of power is lost in only ³⁹⁵ 300 hours while a few hundreds of hours later, only 10% is lost at 50 A and in 700 hours. Consequently, using the ³⁹⁷ 10% of power loss proposed by the DoE would have led to a EoL in the very early life of the stack showing that ³⁹⁹ another proposal need to be found. In such a case, using ⁴⁰⁰ the cumulative energy or the efficiency to define the EoL seems more appropriate.

Figure 12: Power during the aging of a 8-cell stack under a μ -CHP [40] (OCV stands for Open Circuit Voltage)

 The definitive EoL threshold is closely linked to the ap- plication. It refers to the inability for the PEMFC to de- liver power in safe conditions. The safety constrains may vary from an application to another as the conditions and the norms are not the same. For example, safety con- strains are quite different for stacks used in air travel ap- plications and ones used to supply technical pylon [37]. Consequently, fixing threshold values need an important feedback from the industry.

⁴¹¹ 5. Health assessment and prognostics

 This stage of the prognostics' process raises two main questions. The first one is "when should one start predict- ing?" Indeed, when a new PEMFC starts to operate, one can expect a good SoH and the prognostic is not required. In the contrary, as the system ages, the necessity to know

417 accurately the EoL grows and the prognostic should be 452 ⁴¹⁸ performed more often, above all when coming closer to ⁴¹⁹ the sudden performance drop. But this implies to be able⁴⁵⁴ ⁴²⁰ to define the SoH of the PEMFC. So the second question ⁴²¹ is "How can the limit between the healthy and degraded⁴⁵⁵ ⁴²² modes be set?"

⁴²³ 5.1. Health assessment

 $_{424}$ Defining the healthy and degraded modes of a PEMFC is_{$_{458}$} ⁴²⁵ not trivial. The system starts degrading since its starting ⁴²⁶ up and the power signal does not exhibit clear degrada-427 tion levels. This can clearly be seen with a constant cur-460 428 rent solicitation (Fig. 6) where the power starts decreasing 461 429 continuously after only a few hours. So how to define the 462 ⁴³⁰ different degraded modes of the PEMFC? 431 When representing the voltage measured during a^{464} 432 stack lifetime with an histogram, it can be seen⁴⁶⁵ 433 seen that different states seem to appear, see⁴⁶⁶ 434 Fig. 13. The issue is to formalize that observation⁴⁶⁷ 435 and propose a method to identify the different de-468 ⁴³⁶ graded modes.

Figure 13: Histogram of the voltages for a stack aged under a constant current profile (data from Fig. 6 lower part)

⁴³⁷ In the case of a constant mission profile, the power loss⁴⁸⁴ 438 could be kept to distinguish the different SoH. As an ex^{-485} ⁴³⁹ ample, one can define:

- \bullet From 0 to 5% of power loss: Good health;
- \bullet From 5% to 10% of power loss: Acceptable:
- ⁴⁴² Over 10% of power loss: Degraded SoH.

 By looking at Fig. 6, this is not illogical as the power drop faster after the 10% limit. Of course, more classes might be identified before those 10%, as suggested by the histogram in Fig. 13. Some unsupervised classification methods are available, such as Hierarchical Ascendant Clustering [41, 42], to refine the number of classes.

⁴⁴⁹ For mission profiles with variable currents, no clear answer ⁴⁵⁰ can be given. Some work trails should be explored in the ⁴⁵¹ future:

- using the cumulative energy or the efficiency and finding different classes by analogy with the constant current example;
- using the maximum power achievable for a very low ⁴⁵⁶ or very high current value and following its evolution;
- ⁴⁵⁷ etc.

5.2. Prognostics

Let's assume in this paragraph that a satisfying definition of the SoH is available. A time instant to start performing prognostics should be decided. In all the published ⁴⁶² approaches, the prognostics is performed on datasets from ⁴⁶³ finished experiments. So, the beginning of the predictions is very often chosen to get the best prediction horizon with-⁴⁶⁵ out taking the SoH into consideration. It is important to test and validate the algorithms but in industrial applications, this practice is obviously not possible. Two possibilities appear:

- ⁴⁶⁹ 1. for model-based approaches: setting a threshold on ⁴⁷⁰ the error between theoretical and real stack power ⁴⁷¹ in percent. Nevertheless, the latter requires that the ⁴⁷² threshold is a function of the operating conditions (see $\frac{473}{473}$ the loss on Fig. 12). If the residual crosses a threshold, ⁴⁷⁴ an anomaly is detected and the prognostic procedure ⁴⁷⁵ is executed (see Fig. 14 where the prognostic should 476 be performed at $t = 38s$ [43, 44].
- ⁴⁷⁷ 2. for data-driven approaches, two solutions:
- ⁴⁷⁸ setting a threshold on the SoH indicator and on ⁴⁷⁹ the gradient of the latter (to capture a perfor-⁴⁸⁰ mance drop or improvement);
- using the histogram of the chosen SoH indicator ⁴⁸² to track the state evolution and to decide in con-⁴⁸³ sequence if the prognostic has to be performed.

Then the prognostics algorithm can be launched. The reader can refer to Table 1 for more information on different approaches currently available.

Figure 14: A residual and thresholds to trigger the prognostics procedure

Table 1. Characteristics of a few works developed in the held of I Entit C prognostics				
Year	Reference	Type	Health indicator	State estimation and/or prognostics tools
2012	$\left[13\right]$	model-based	Stack voltage (1-cell stack,	Unscented Kalman Filter
			I variable)	
2014	17	data-driven	Stack voltage (I constant)	Particle filter
	$\left[14\right]$	data-driven	Mean cell voltage (I con-	Echo State Network
			stant)	
	¹⁵	data-driven	Stack voltage (I constant)	Adaptive Neuro Fuzzy Inference Systems
	¹⁹	data-driven	Impedance	Equivalent circuit
	$\overline{[20]}$	data-driven	Impedance	Regressions
	$\overline{[21]}$	data-driven	Stack voltage (I constant)	Regime switching autoregressive vector
				(RSVAR)
	$\overline{22}$	data-driven	Stack voltage (I constant)	Particle filter
2015	$\lceil 16 \rceil$	data-driven	Stack voltage (I constant)	Summation-Wavelet Extreme Learning Ma-
				chine
	12	model-based	Stack voltage (I variable)	Kalman Filter
	[11]	model-based	Impedance and polariza-	Regressions
			tion curves	
	45	Hybrid	Stack voltage (I variable)	Particle filter

Table 1: Characteristics of a few works developed in the field of PEMFC prognostics

⁴⁸⁷ 6. Evaluating prognostics' performance

⁴⁸⁸ 6.1. Prediction horizon

⁴⁸⁹ It is important to evaluate the duration needed between⁵²³ 490 the prediction instant and the decision and/or action in- 524 491 stant. The horizon may depend on the goal of prognostics.⁵²⁵ ⁴⁹² Consequently, the reasoning is proposed on an example, ⁴⁹³ and can be transposed in other applications.

 Let's take the example of the maintenance rescheduling for 495 a transportation or a μ -CHP PEMFC. It is assumed that the mission rescheduling can be done remotely, so only the maintenance constraints impact the prediction horizon ex- pected. To perform maintenance, different time durations have to be taken into account: (1) the planning of the maintenance operations, 1-2 days; (2) the delay to obtain spare parts, 10-20 days; (3) the maintenance realization, 1-7 days. It gives a total duration between 12 and 29 days. It means that the prognostics should give results with a horizon between 300 and 700 hours to be considered as a good performance. Obviously, this time interval should be discussed with respect to industrial feedbacks in the ⁵⁰⁷ future.

⁵⁰⁸ 6.2. Acceptable error on RUL estimates

 Regarding the RUL estimates, there are two cases: (1) 510 the estimate is smaller than the actual RUL, it is an early⁵²⁷ $_{511}$ prediction, or (2) the estimate is greater, it is a late predic- $_{528}$ tion. The acceptable error cannot be the same in each case. Indeed, by validating a prediction with a consequent delay, one may risk a complete shutdown of its power source. It ⁵¹⁵ is intolerable in most PEMFC applications. Consequently, 532 according to the horizon discussed earlier, it is proposed to allow a maximum delay of one day. It represent an error of 8% for a horizon of 300 hours.

⁵¹⁹ The case of the early prediction allows more flexibility.

⁵²⁰ Anticipating a maintenance is less disadvantageous than ⁵²¹ being late, even if it can create some extra costs due to ⁵²² the replacement of a functioning system. It is proposed to set the early prediction limit at 16% of the actual RUL, whatever the time instant. It represents 48h for a horizon of 300h and 4 days $(112h)$ for a horizon of 700 hours. The acceptable error is depicted in Fig. 15.

Figure 15: Acceptable error on RUL estimates

⁵²⁷ 6.3. Acceptable uncertainty

⁵²⁸ Defining the uncertainty allowed in the prediction is not ⁵²⁹ trivial as no standards are available for PEMFC on that subject. So let's try to reason on the current standard on electricity production.

⁵³² Nowadays, electricity suppliers in Europe have to provide a voltage of 230 V $\pm 10\%$ (at 50 Hz) [46]. This constrain evolves according to the world location. In North America, ⁵³⁵ the standard impose a nominal voltage of 120 V with a tolerance of $\pm 5\%$ (at 60 Hz) [47]. Even if, the standards are not uniform, they can be used to propose a confidence interval objective for prognostics. Indeed, to ensure that the prognostics' results are reliable anywhere in the word, the uncertainty on the SoH or RUL estimates should be $\frac{541}{541}$ constrained in a $\pm 5\%$ interval. Nevertheless, a confidence $_{542}$ interval of $\pm 10\%$ allows to assert that the predictions are quite satisfying but can be improved. Again, industrial $_{544}$ feedbacks are necessary to discuss further on that subject.

⁵⁴⁵ 7. Conclusion

 Developing prognostics for PEMFC can be a solution to contribute to extend the system's lifetime. However, $_{548}$ the lack of standards to guide current works may lead to $_{\hspace{-.1em}s\hspace{-.1em}0}^{601}$ the appearance of prognostics' proposals with very poor adaptation capacities to actual industrial situations. To start a discussion on this subject, this paper proposes guid- ance on several aspects such as state definition, selection $\frac{500}{606}$ of prognostics' criteria or performance evaluation. This is synthesized in Fig. 16.

⁵⁵⁵ This work first shows how important it is to set the ⁶⁰⁹ hypotheses of the prognostics application. Indeed, it lim- its the prognostics' approach to specific real life cases of PEMFC applications. Also, it makes appear some techno- $\frac{559}{559}$ logical constrains, as the types of measurements available, $\frac{614}{615}$ as well as indications on the phenomena that should be taken into account (transient and/or permanent regimes). To evaluate the current and future state of health of the 563 system, it is important to know if the stack remains in $\text{an}_{\text{620}}^{0.9}$ $_{564}$ healthy mode or already is in a degraded one. There $\overline{15}_{621}$ ⁵⁶⁵ currently no clear borders between these states. Defining⁶²² which ones are degraded states is still to debate. Performing predictions requires health indicators. Accord- $\frac{55}{625}$ ing to the application, the criteria might be different and this leads to the proposal of several indicators such as the power, the cumulative energy and the efficiency. All de- pend on the mission profile and the pathway from one 530 to another can be easily made. However, defining failure thresholds related to each criterion remains an open ques- tion. Moreover, this lead to wonder how the end of life of 633 the stack is defined. Two solutions are proposed related $\frac{37}{635}$ first to the mission in progress and then related to an in- ability of the stack to provide power in safe conditions. Finally, to help validating prognostics approaches, some $_{\epsilon_{20}}^{638}$ 579 performance metrics are proposed and discussed. They₆₄₀ try to take into account real life constrains such as electri-581 cal norms and maintenance delays. Nevertheless, the lack⁶⁴² of industrial feedback in the literature prevents a precise $^{643}_{644}$ discussion on the subject.

⁵⁸⁴ To extend the proposals from this paper to short-585 term predictions for adaptive control, small time⁶⁴⁷ s_{86} scales, as a minute, will have be studied and dis- $\frac{s_{66}}{649}$ 587 cussed. As it may raise issue linked to the speed of $_{650}$ 588 the algorithm or the accuracy needed to use short-651 $_{589}$ term predictions for example, this stage might wait $_{552}$ $_{590}$ that more prognostics' approaches are available in 591 the literature. The field of prognostics of PEMFC hasses just started and a great reflection effort is needed to de-⁵⁹³ velop approaches that, one day, can be transferable to the industry.

8. Acknowledgments

The authors would like to thank the ANR project PROPICE (ANR-12-PRGE-0001) and the Labex AC-⁵⁹⁸ TION project (contract "ANR-11-LABX-01-01") both ⁵⁹⁹ funded by the French National Research Agency as well 600 as the French region of Franche-Comté for their support.

References

- [1] X. Luo, J. Wang, M. Dooner, J. Clarke, Overview of current development in electrical energy storage technologies and the application potential in power system operation, Applied Energy 137 (2015) 511-536.
- [2] Y. Wanga, K. S. Chen, J. Mishler, S. C. Cho, X. C. Adroher, ⁶⁰⁷ A review of polymer electrolyte membrane fuel cells: Technol-⁶⁰⁸ ogy, applications, and needs on fundamental research, Applied Energy 88 (2011) 981-1007.
- [3] S.-D. Oh, K.-Y. Kim, S.-B. Oh, H.-Y. Kwak, Optimal operation of a 1-kw pemfc-based chp system for residential applications, ⁶¹² Applied Energy 96 (2012) 93–101.
- [4] J. Fernandez-Moreno, G. Guelbenzu, A. Martin, M. Folgado, ⁶¹⁴ P. Ferreira-Aparicio, A. Chaparro, A portable system powered with hydrogen and one single air-breathing pem fuel cell, Applied Energy 109 (2013) 60-66.
- [5] E4tech, The fuel cell industry review 2014 (Nov. 2014). URL http://www.fuelcells.org/pdfs/TheFuelCellIndustry Review2014.pdf
- [6] O. Z. Sharaf, M. F. Orhan, An overview of fuel cell technology: ⁶²¹ Fundamentals and applications, Renewable and Sustainable En-⁶²² ergy Reviews 32 (2014) 810 – 853.
- [7] G. Niu, D. Anand, M. Pecht, Prognostics and health manage-⁶²⁴ ment for energetic material systems, in: Prognostics and Health Management Conference, 2010. PHM '10., 2010, pp. 1-7.
- [8] M. Jouin, R. Gouriveau, D. Hissel, M.-C. Péra, N. Zerhouni, Prognostics and health management of PEMFC state of the art and remaining challenges, International Journal of Hydrogen ⁶²⁹ Energy 38 (35) (2013) 15307 – 15317.
- ⁶³⁰ [9] J. W. Sheppard, M. A. Kaufman, T. J. Wilmer, IEEE standards for prognostics and health management, Aerospace and ⁶³² Electronic Systems Magazine, IEEE 24 (9) (2009) 34–41.
- [10] ISO13381-1, Condition monitoring and diagnostics of machines - prognostics - Part1: General guidelines, International Standard, ISO, 2004.
- [11] E. Lechartier, E. Laffly, M.-C. Péra, R. Gouriveau, D. Hissel, ⁶³⁷ N. Zerhouni, Proton exchange membrane fuel cell behavioral model suitable for prognostics, International Journal of Hydrogen Energy 40 (26) (2015) 8384–8397.
[12] M. Bressel, M. Hilairet, D. Hissel, B. (
	- M. Bressel, M. Hilairet, D. Hissel, B. O. Bouamama, Extended kalman filter for prognostic of proton exchange membrane fuel cell, Applied Energy 164 (2016) 220 - 227.
	- [13] X. Zhang, P. Pisu, An unscented kalman filter based approach for the health-monitoring and prognostics of a polymer electrolyte membrane fuel cell, in: Proceedings of the annual conference of the prognostics and health management society, 2012.
	- [14] S. Morando, S. Jemei, R. Gouriveau, D. Hissel, N. Zerhouni, Fuel cells remaining useful lifetime forecasting using echo state network, in: Vehicle Power and Propulsion Conference (VPPC'14), 2014, pp. IS1-4.
	- [15] R. Silva, R. Gouriveau, S. Jemei, D. Hissel, L. Boulon, K. Ag-⁶⁵² bossou, N. Y. Steiner, Proton exchange membrane fuel cell degradation prediction based on adaptive neuro fuzzy inference systems, International Journal of Hydrogen Energy 39 (21) (2014) 11128 – 11144.

Figure 16: Synthesis of proposed prognostics guidelines

- ⁶⁵⁶ [16] K. Javed, R. Gouriveau, N. Zerhouni, Data-driven prognostics ⁶⁵⁷ of proton exchange membrane fuel cell stack with constraint ⁶⁵⁸ based summation-wavelet extreme learning machine., in: 6th ⁶⁵⁹ International Conference on Fundamentals and Development of ⁶⁶⁰ Fuel Cells, FDFC'15., Toulouse - France., 2015, pp. 1–8.
- 661 [17] M. Jouin, R. Gouriveau, D. Hissel, M.-C. Péra, N. Zerhouni, ⁶⁶² Joint particle filters prognostics for PEMFC power prediction ⁶⁶³ at constant current solicitation, IEEE Transactions on Reliabil-⁶⁶⁴ itydoi:10.1109/TR.2015.2454499.
- ⁶⁶⁵ [18] FCLAB research, IEEE PHM data challenge 2014 (2014).
- ⁶⁶⁶ URL http://eng.fclab.fr/ieee-phm-2014-data-challenge/
- ⁶⁶⁷ [19] T. Kim, H. Kim, J. Ha, K. Kim, J. Youn, J. Jung, B. D. Youn, ⁶⁶⁸ A degenerated equivalent circuit model and hybrid prediction ⁶⁶⁹ for state-of-health (SOH) of PEM fuel cell, in: IEEE PHM con-⁶⁷⁰ ference 2014, 2014, pp. 1–7.
- ⁶⁷¹ [20] W. O. L. Vianna, I. P. de Medeiros, B. S. Aflalo, L. R. Ro-⁶⁷² drigues, J. P. P. Malere, Proton exchange membrane fuel cells ⁶⁷³ (PEMFC) impedance estimation using regression analysis, in: ⁶⁷⁴ IEEE PHM conference 2014, 2014, pp. 1–8.
- ⁶⁷⁵ [21] A. Hochstein, H.-I. Ahn, Y. T. Leung, M. Denesuk, Switching ⁶⁷⁶ vector autoregressive models with higher-order regime dynam- 677 ics, in: IEEE PHM conference 2014, 2014, pp. 1–10.
- ⁶⁷⁸ [22] J. Kuria Kimotho, T. Meyer, W. Sextro, PEM fuel cell prog-⁶⁷⁹ nostics using particle filter with model parameter adaptation, ⁶⁸⁰ in: IEEE PHM conference 2014, 2014, pp. 1–6.
- ⁶⁸¹ [23] G. W. Vogl, B. A. Weiss, M. A. Donmez, Standards Related to ⁶⁸² Prognostics and Health Management (PHM) for Manufactur-

ing, Vol. 8012, National Institute of Standards and Technology (NIST), Gaithersburg, Maryland, USA, NISTIR, 2014.

- [24] P. Pei, H. Chen, Main factors affecting the lifetime of proton exchange membrane fuel cells in vehicle applications: A review, ⁶⁸⁷ Applied Energy 125 (2014) 60–75.
- [25] N. Wagner, A. Bauder, K. Friedrich, Diagnostics of PEM fuel cells (2011).

URL http://iet.jrc.ec.europa.eu/fuel-cells/sites/fuel-⁶⁹¹ cells/files/files/documents/events/diagnostics_of_pem_ ⁶⁹² fuel_cells_-_n.wagner.pdf

- [26] M. Jouin, R. Gouriveau, D. Hissel, M.-C. Pra, N. Zerhouni, Degradations analysis and aging modeling for health assessment and prognostics of PEMFC, Reliability Engineering $&$ System Safety 148 (2016) $78 - 95$.
- [27] I. Radev, K. Koutzarov, E. Lefterova, G. Tsotridis, Influence of failure modes on PEFC stack and single cell performance and durability, International Journal of Hydrogen Energy 38 (17) (2013) 7133 – 7139.
- ⁷⁰¹ [28] A. Bose, P. Babburi, R. Kumar, D. Myers, J. Mawdsley, J. Milhuff, Performance of individual cells in polymer electrolyte membrane fuel cell stack under-load cycling conditions, Jour- $\frac{1}{704}$ nal of Power Sources 243 (0) (2013) 964 – 972.
	- [29] F. De Bruijn, V. Dam, G. Janssen, Review: durability and degradation issues of PEM fuel cell components, Fuel Cells 8 (1) (2008) 3–22.
	- [30] L. Placca, R. Kouta, Fault tree analysis for PEM fuel cell degradation process modelling, International Journal of Hydrogen
- Energy 36 (19) (2011) 12393 12405.
- [31] X. Zhang, Y. Rui, Z. Tong, X. Sichuan, S. Yong, N. Huaisheng, The characteristics of voltage degradation of a proton exchange membrane fuel cell under a road operating environment, In- ternational Journal of Hydrogen Energy 39 (17) (2014) 9420 – 9429.
- [32] A. Franco, Modelling and analysis of degradation phenomena in pemfc, Woodhead, 2012.
- [33] S. Kundu, M. Fowler, L. C. Simon, R. Abouatallah, Reversible and irreversible degradation in fuel cells during open circuit voltage durability testing, Journal of Power Sources 182 (1) (2008) $254 - 258$.
- [34] J. Wu, X.-Z. Yuan, J. J. Martin, H. Wang, D. Yang, J. Qiao, J. Ma, Proton exchange membrane fuel cell degradation under close to open-circuit conditions: Part I: In situ diagnosis, Jour-nal of Power Sources 195 (4) (2010) 1171 – 1176.
- [35] A. Askarzadeh, Parameter estimation of fuel cell polarization curve using BMO algorithm, International Journal of Hydrogen Energy 38 (35) (2013) 15405 – 15413.
- [36] M. Santarelli, M. Torchio, P. Cochis, Parameters estimation of a PEM fuel cell polarization curve and analysis of their behavior with temperature, Journal of Power Sources 159 (2) (2006) 824 – 835.
- 733 [37] M.-C. Péra, D. Hissel, H. Gualous, C. Turpin, Fuel Cells, John Wiley & Sons, Inc., 2013, pp. 151–207. doi:10.1002/9781118576892.ch3.
- [38] H. Chen, P. Pei, M. Song, Lifetime prediction and the economic lifetime of proton exchange membrane fuel cells, Applied Energy 142 (2015) 154–163.
- [39] U. D. of Energy, The department of energy hydrogen and fuel cells program plan (2011).
- URL http://www.hydrogen.energy.gov/roadmaps_vision.h 742 tml
743 [40] E. I
- [40] E. Pahon, S. Morando, R. Petrone, al., Long-term tests duration r_{44} reduction for pemfc μ -chp application, in: ICREGA16, 2016, pp. 1–6.
- [41] J. H. Ward Jr, Hierarchical grouping to optimize an objec- tive function, Journal of the American statistical association 58 (301) (1963) 236–244.
- [42] T. Aroui, Y. Koubaa, A. Toumi, Self-organizing maps for di- agnosing induction motors supplied by a variable speed drive, European journal of electrical engineering 14 (6) (2011) 697– 752 717.
753 [43] B.
- [43] B. O. Bouamama, M. Bressel, D. Hissel, M. Hilairet, Robust diagnosability of PEMFC based on bond graph LFT, ICECE 2015: International Conference on Electrical and Control Engi-neering 2 (6) (2015) 1197.
- [44] M. Djeziri, B. O. Bouamama, R. Merzouki, Modelling and ro- bust FDI of steam generator using uncertain bond graph model, Journal of Process Control 19 (1) (2009) 149–162.
- [45] M. Jouin, R. Gouriveau, D. Hissel, M.-C. P´era, N. Zerhouni, Prognostics of PEM fuel cells under a combined heat and power profile, in: Proceedings of the 2015 IFAC Symposium on Infor-mation Control in Manufacturing (INCOM 2015), 2015.
- [46] CENELEC, European committee for electrotechnical standard-ization, http://www.cenelec.eu/index.html.
- [47] NEMA, American national standard for electric power systems and equipment voltage ratings (60 hz), http://www.nema.org/Standards/Pages/American-National-Standard-for-Electric-Power-Systems-and-Equipment-
- Voltage-Ratings.aspx.