

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.

23

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

1. Introduction

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

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, measurements, 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 Life (RUL).

During the last years, prognostics has started to be applied to PEMFC. Different types of approaches are proposed: 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, replacing fossil energies by alternative power sources such as fuel cells appears as a promising solution [1, 2]. Fuel cells can be used in a great number of applications such as transportation, mobile electronic devices, micro-CHP (Combined Heat and Power), etc. [3, 4].

 $[\]frac{}{^{*}\text{Corresponding author, Tel.: +33 (0)3 81 40 29 04, Fax +33 (0)3}_{47}$

⁴⁸ brid approaches [17]. The growing interest of the scientific
⁴⁹ community to that subject has also been shown thanks
⁵⁰ to the IEEE PHM data challenge 2014 [18] in which dif⁵¹ ferent propositions for SoH predictions [19, 20] and RUL
⁵² 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 considerations about prognostics standards are available [9, 23], they are too general and further questioning is required for the PEMFC case.

- ⁵⁹ 1. Which level of granularity should be chosen on the⁶⁰ PEMFC?
- ⁶¹ 2. How to define the healthy and degraded modes for a
 PEMFC?
- ⁶³ 3. When prognostics should be performed?
- 4. Which health indicator should be used to performprognostics?
- 5. How to define the End-of-Life (EoL) of a PEMFC?
- 6. How can the prognostics' performances be evaluated?

To the authors' knowledge none of these questions 68 are answered in the literature. Consequently, this 69 paper intends to propose solutions and guidance 70 to start answering these questions and going to-71 wards standardization. The main contribution is 72 to propose general solutions that are completely 73 independent of prognostics tools and that can be 74 adapted with any approach. New practitioners can 75 use these guidelines as tutorial to start performing 76 prognostics of PEMFC. 77

To adopt a logical reasoning, the paper follows the chrono-78 logical order of the prognostics process' stages (Fig 1). 79 First, the time constants involved in the prognostics are 80 discussed as well as the level of granularity recommended 81 for prognostics in Section 2. Section 3 focuses on the data 82 selection and their processing before defining health indi-83 cators in Section 4. This section also discusses the EoL 84 definition. The SoH definition is considered in Section 5 85 in addition to the time instant to start predicting. Finally, 86 some performance metrics are defined in Section 6 before 87 101 concluding. 88

⁸⁹ 2. Decision levels and system granularity

90 2.1. Decision levels and time constants

Prognostics intends to provide the future SoH of a sys-107
tem and its RUL based on data, system knowledge and the108
expected mission profile. This generally creates a closed109
loop, Fig. 2. This figure could be completed with other110
PHM processes but only the ones of interest for this paper111
are kept.

The system initiates its mission based on an ex-113
pected mission profile. Data are collected and pro-114
cessed continuously for health assessment. This115
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 SoH based on the planned mission profile. According to the predictions, this profile can be modified and adapted to the actual capabilities of the system. 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 accumulation 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

102

103

104

105

scales are more related to adaptive control or system re-143
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 encountered by a PEMFC in a single paper would be a hard
task. Consequently, it is assumed that prognostics is performed on a single stack working in its nominal conditions.
The decision is out of the scope of that paper, even if it is¹⁶¹
mentioned later to argue some points.

125 2.2. Stack vs component level

From the previous paragraphs, it seems obvious to fo-165 126 cus on degradation phenomena to perform prognostics.¹⁶⁶ 127 Degradations occur at all levels of the stack granular-¹⁶⁷ 128 ity: stack/cells/components and at the interfaces between $^{\rm 168}$ 129 them [26]. All the published approaches are dealing with¹⁶⁹ 130 prognostics at the stack level (Section 5). The main reason 170 131 is that the data acquisition is very often performed at this $^{\scriptscriptstyle 171}$ 132 172 level. 133

However, working at the cell level can be an interesting op-¹⁷³
tion. Indeed, the functioning of the stack can be suddenly¹⁷⁴
stopped when a cell reaches a minimum voltage value.¹⁷⁵
This value is given by the manufacturer to automatically¹⁷⁶
shut down the stack when a cell seems too damaged and¹⁷⁷
so prevent safety issues. Performing prognostics on cells¹⁷⁸
would allow anticipating such events.

¹⁴¹ An issue appears with this type of prognostics: cells are₁₈₀ ¹⁴² not degrading in a uniform way within the stack [27, 28].₁₈₁

Cells near the edges tend to degrade faster. As for an illustration, 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 example, 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 deviation. It helps isolating the cells that may come prematurely 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. Moreover, nowadays, stack reconstructions to replace only these faulty cells are not systematic due to all the assembly constrains. Also, a stack can still convert energy even with faulty cells. So such a level of study can be questionable.



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

Another perspective for prognostics may be being performed at a smaller level, namely the component level. It has already been initiated in [13] in which the size of the electrochemical active surface area (ECSA) is predicted with a model-based prognostics framework. Many other critical parameters such as hydrogen crossover, resistances (or conductivities) or materials integrity could be predicted. Some failure thresholds have already been proposed for some of them. For instance, a crossover current 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;
- 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

163 164

155

156

182 account. 235

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.²³⁸

¹⁸⁶ 3. Data selection and processing

187 3.1. Data collection

In an industrial operating environment, very few physical parameters are easily accessible with current technologies. It is the reason why, the existing prognostics methodologies focus on indicators built based on voltages measurements 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 monitored, the current is supposed to be known as it is defined
by the mission profile.

Also, some punctual measurements can be available. Of-198 ten, an initial polarization curve as well as some EIS (Elec-199 trochemical Impedance Spectroscopy) measurements are 200 given by the manufacturer or measured before the stack 201 starts its mission. However, as it is not sure yet that 202 these punctual measurements can be performed during the 203 whole lifetime of an embedded system, there are left aside 204 in the remaining of the paper. 205

Fig. 5 shows that limiting the measurements to the power 206 still provide a good indicator of the stack aging. Indeed, 207 constant current experiments are typically reflecting the 208 "natural" aging of the stack, i.e. an aging not influenced 209 by the mission profile if the operating conditions are kept 210 nominal. The power decreases with time indicating the²³⁹ 211 evolution of different degradation mechanisms inside the₂₄₀ 212 PEMFC. This explains why some papers use the volt- $_{241}$ 213 age/power to evaluate the degradation rate of the system₂₄₂ 214 [30, 31]. Finally, [32] shows that almost all the degrada-₂₄₃ 215 tions can be perceived in this signal. 216 244 Consequently, the focus is set on the use of voltage or, by $_{245}$ 217 proportionality with respect to the current, power mea-246 218

220 3.2. Degradation vs transient phenomena

surements.

219

Based on the previous comments, the focus is set on the₂₅₀ 221 observation of the degradation thanks to power measure-251 222 ments. However, the degradations are mixed with tran-252 223 sient phenomena when time scales in hours are consid-253 224 ered (Fig. 4). Fig. 6 represents the power measured₂₅₄ 225 during the aging of two 5-cell stacks at a constant²⁵⁵ 226 current of 60A and 70A with weekly polarization₂₅₆ 227 curves and EIS. It highlights that these punctual charac-257 228 terizations disturb the power and create transient stages.258 229 This observation could be expanded to all events bringing²⁵⁹ 230 the stack out of its nominal conditions and that can cre-260 231 ate reorganization of water and gas repartition within the₂₆₁ 232 stack [17, 33, 34]. Very often, these events appear as re-262 233 coveries in the power signal. As the transient stages have₂₆₃ 234

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 condition 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. 7), it can be observed that the main component of the signal is located in the low

247

248

frequency domain. It allows using a simple low-pass fil-²⁸⁹ 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

Different indicators can be proposed as prognostics criteria, 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 according₃₀₄ 277 to the PEMFC use. The power (or the voltage) is₃₀₅ 278 recommended for constant mission profiles while₃₀₆ 279 the cumulative energy and the efficiency are more₃₀₇ 280 adapted for variable mission profiles. Indeed, when 308 281 the current varies, the power is not yet a mono-309 282 tonic indicator preventing from fixing a failure₃₁₀ 283 threshold. Further discussion on this issue is pro-311 284 vided in Section 4.2.2. The case of the power as health₃₁₂ 285 indicator has already been evoked in the previous sections,313 286 so the emphasize is set here on the cumulative energy and₃₁₄ 287 the efficiency. 315 288

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:

$$E(t = 0:T) = \int_0^T U(t).i(t).dt$$
 (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]:

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

Different strategies can be found in the literature to perform that fitting [35, 36] but using a simple algorithm of the least squares family seems sufficient. This fitting enables 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 behavioural model that describes the evolution of the voltage or the power according to the current profile as if no degradation was occurring within the stack. As an example, the reference in Fig. 9 and 12 are built based on the model proposed 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 of cumulative energy (so in the watt.hour unit) enables

290

291

292

293

294

295

296

300

301

302



Figure 8: Setting of the reference for the cumulative energy based³⁴¹ on the initial polarization curve 342



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

comparisons with other types of energy sources. 316 358 However, some issues may be encountered when building_{359} 317 the reference. First, it has to be verified that the un- $_{360}$ 318 certainty on the initial polarization curve is reasonable to $_{361}$ 319 consider it as a reference basis. Then, a comparison be- $_{362}$ 320 tween the power measured at the early stages of the stack $_{363}$ 321 life and the power calculated with the polarization $\operatorname{curve}_{364}$ 322 should be performed. If the difference between these two₃₆₅ 323 powers is too high, the reference for the cumulative energy $_{366}$ 324 should not be built based on the initial polarization curve. 325 Finally, using a model to build the reference seems to be an 326 interesting solution. However, the lack of validated models 327 should be filled to enable such a solution. 328

329 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:

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

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

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

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

4.2. EoL definition

343

357

Two thresholds can be defined for prognostics of PEMFC stack:

- 1. a threshold of conformity to a mission:
- 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).

4.2.1. Conformity to a mission threshold

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 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:

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

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

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



Figure 10: Power density for different level of aging

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

which is strictly equivalent to the power threshold for₃₉₆
constant solicitation. However, this threshold is a function₃₉₇
of the power profile and so can also work for variable₃₉₈
requested power.

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).

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

The authors states that using the fuel cell after the economic 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] 414

4.2.2. Definitive End of Life threshold

378

387

388

389

390

391

392

400

402

There is currently one definition of the EoL based on 379 the power. The US Department of Energy (DoE) defines 380 the EoL as a loss of 10% of the initial performance [39]. 381 Even if the value could be discussed, it is not the main 382 problem of this definition. Using a fixed threshold for con-383 stant current application is very convenient and does not 384 create any problem. The EoL of the stack is defined as a 385 ratio of the initial performance as illustrated in Fig. 6. 386

However, it does not work so well when the mission profile exhibits current variations. Fig. 12 shows an example of power measured under a μ -CHP (Combined Heat and Power) profile with the theoretical power that should be expected if the stack was not aging. It can be seen that according to the level of current, the loss of performance does not follow a constant percentage with respect to the reference. Indeed, at 170 A, 15% of power is lost in only 300 hours while a few hundreds of hours later, only 10% is lost at 50 A and in 700 hours. Consequently, using the 10% of power loss proposed by the DoE would have led to a EoL in the very early life of the stack showing that another proposal need to be found. In such a case, using 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 application. It refers to the inability for the PEMFC to deliver 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 constrains are quite different for stacks used in air travel applications and ones used to supply technical pylon [37]. Consequently, fixing threshold values need an important feedback from the industry.

5. Health assessment and prognostics

This stage of the prognostics' process raises two main questions. The first one is "when should one start predicting?" 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

415

accurately the EoL grows and the prognostic should be₄₅₂
performed more often, above all when coming closer to₄₅₃
the sudden performance drop. But this implies to be able₄₅₄
to define the SoH of the PEMFC. So the second question
is "How can the limit between the healthy and degraded⁴⁵⁵
modes be set?" 456

423 5.1. Health assessment

Defining the healthy and degraded modes of a PEMFC is $_{458}$ 424 not trivial. The system starts degrading since its starting 425 up and the power signal does not exhibit clear degrada-459 426 tion levels. This can clearly be seen with a constant cur-460 427 rent solicitation (Fig. 6) where the power starts decreasing⁴⁶¹ 428 continuously after only a few hours. So how to define the⁴⁶² 429 different degraded modes of the PEMFC? 463 430 When representing the voltage measured during a⁴⁶⁴ 431 stack lifetime with an histogram, it can be seen⁴⁶⁵ 432 seen that different states seem to appear, see⁴⁶⁶ 433 Fig. 13. The issue is to formalize that observation⁴⁶⁷ 434 and propose a method to identify the different de-468 435 graded modes. 436



Figure 13: Histogram of the voltages for a stack aged under a con- 481 stant current profile (data from Fig. 6 lower part) 482

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

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

⁴⁴³ By looking at Fig. 6, this is not illogical as the power drop
⁴⁴⁴ faster after the 10% limit. Of course, more classes might be
⁴⁴⁵ identified before those 10%, as suggested by the histogram
⁴⁴⁶ in Fig. 13. Some unsupervised classification methods are
⁴⁴⁷ available, such as Hierarchical Ascendant Clustering [41,
⁴⁴⁸ 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 or very high current value and following its evolution;
- etc.

457

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 approaches, the prognostics is performed on datasets from finished experiments. So, the beginning of the predictions is very often chosen to get the best prediction horizon without 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:

- 1. for model-based approaches: setting a threshold on the error between theoretical and real stack power in percent. Nevertheless, the latter requires that the threshold is a function of the operating conditions (see the loss on Fig. 12). If the residual crosses a threshold, an anomaly is detected and the prognostic procedure is executed (see Fig. 14 where the prognostic should be performed at t = 38s) [43, 44].
- 2. for data-driven approaches, two solutions:
 - setting a threshold on the SoH indicator and on the gradient of the latter (to capture a performance drop or improvement);
 - using the histogram of the chosen SoH indicator to track the state evolution and to decide in consequence 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 field of 1 EMI C prognostics				
Year	Reference	Type	Health indicator	State estimation and/or prognostics tools
2012	[13]	model-based	Stack voltage (1-cell stack,	Unscented Kalman Filter
			I variable)	
2014	[17]	data-driven	Stack voltage (I constant)	Particle filter
	[14]	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
	[20]	data-driven	Impedance	Regressions
	[21]	data-driven	Stack voltage (I constant)	Regime switching vector autoregressive
				(RSVAR)
	[22]	data-driven	Stack voltage (I constant)	Particle filter
2015	[16]	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

520

521

522

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

487 6. Evaluating prognostics' performance

488 6.1. Prediction horizon

It is important to evaluate the duration needed between⁵²³ the prediction instant and the decision and/or action in-⁵²⁴ stant. The horizon may depend on the goal of prognostics.⁵²⁵ Consequently, the reasoning is proposed on an example,⁵²⁶ and can be transposed in other applications.

Let's take the example of the maintenance rescheduling for 494 a transportation or a μ -CHP PEMFC. It is assumed that 495 the mission rescheduling can be done remotely, so only the 496 maintenance constraints impact the prediction horizon ex-497 pected. To perform maintenance, different time durations 498 have to be taken into account: (1) the planning of the 499 maintenance operations, 1-2 days; (2) the delay to obtain 500 spare parts, 10-20 days; (3) the maintenance realization, 501 1-7 days. It gives a total duration between 12 and 29 days. 502 It means that the prognostics should give results with a 503 horizon between 300 and 700 hours to be considered as 504 a good performance. Obviously, this time interval should 505 be discussed with respect to industrial feedbacks in the 506 future. 507

508 6.2. Acceptable error on RUL estimates

Regarding the RUL estimates, there are two cases: (1) 509 the estimate is smaller than the actual RUL, it is an $early^{527}$ 510 prediction, or (2) the estimate is greater, it is a late predic-528 511 tion. The acceptable error cannot be the same in each case. 529 512 Indeed, by validating a prediction with a consequent delay, 530 513 one may risk a complete shutdown of its power source. It₅₃₁ 514 is intolerable in most PEMFC applications. Consequently, 532 515 according to the horizon discussed earlier, it is proposed to⁵³³ 516 allow a maximum delay of one day. It represent an error₅₃₄ 517 of 8% for a horizon of 300 hours. 518 535

519 The case of the early prediction allows more flexibility.536

Anticipating a maintenance is less disadvantageous than being late, even if it can create some extra costs due to 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

6.3. Acceptable uncertainty

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

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, 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 confidence⁵⁹² 537 interval objective for prognostics. Indeed, to ensure that 593 538 the prognostics' results are reliable anywhere in the word, 594 539 the uncertainty on the SoH or RUL estimates should be 540 constrained in a $\pm 5\%$ interval. Nevertheless, a confidence 541 interval of $\pm 10\%$ allows to assert that the predictions are 542 quite satisfying but can be improved. Again, industrial₅₀₆ 543 feedbacks are necessary to discuss further on that subject.547 544

545 7. Conclusion

Developing prognostics for PEMFC can be a solution 546 to contribute to extend the system's lifetime. However, 547 the lack of standards to guide current works may lead to 601 548 the appearance of prognostics' proposals with very poor₆₀₂ 549 adaptation capacities to actual industrial situations. To⁶⁰³ 550 start a discussion on this subject, this paper proposes guid-604 551 ance on several aspects such as state definition, selection $\widetilde{_{606}}$ 552 of prognostics' criteria or performance evaluation. This is₆₀₇ 553 synthesized in Fig. 16. 554

This work first shows how important it is to set the.... 555 610 hypotheses of the prognostics application. Indeed, it \lim_{-611} 556 its the prognostics' approach to specific real life cases of 612 557 PEMFC applications. Also, it makes appear some techno-613 558 logical constrains, as the types of measurements available, $_{615}^{614}$ 559 as well as indications on the phenomena that should $\mathrm{be}_{\scriptscriptstyle 616}$ 560 taken into account (transient and/or permanent regimes).617 561 To evaluate the current and future state of health of the⁶¹⁸ 562 system, it is important to know if the stack remains in $an_{_{620}}^{^{619}}$ 563 healthy mode or already is in a degraded one. There is_{621} 564 currently no clear borders between these states. Defining622 565 623 which ones are degraded states is still to debate. 566

Performing predictions requires health indicators. Accord-567 ing to the application, the criteria might be different and 626 568 this leads to the proposal of several indicators such as the⁶²⁷ 569 power, the cumulative energy and the efficiency. All de- 628 570 pend on the mission profile and the pathway from $\mathrm{one}_{\scriptscriptstyle 630}^{--}$ 571 to another can be easily made. However, defining failure631 572 thresholds related to each criterion remains an open ques-632 573 tion. Moreover, this lead to wonder how the end of life of $^{633}_{\cdots}$ 574 the stack is defined. Two solutions are proposed related $_{635}$ 575 first to the mission in progress and then related to an in-636 576 ability of the stack to provide power in safe conditions. 577 Finally, to help validating prognostics approaches, some $^{638}_{639}$ 578 performance metrics are proposed and discussed. They₆₄₀ 579 try to take into account real life constrains such as electri-641 580 cal norms and maintenance delays. Nevertheless, the lack $^{\rm 642}$ 581 of industrial feedback in the literature prevents a $\operatorname{precise}_{_{644}}^{_{643}}$ 582 discussion on the subject. 583

To extend the proposals from this paper to short-646 584 term predictions for adaptive control, small $\mathrm{time}^{\scriptscriptstyle 647}$ 585 scales, as a minute, will have be studied and dis_{649}^{040} 586 cussed. As it may raise issue linked to the speed of_{650} 587 the algorithm or the accuracy needed to use short-651 588 term predictions for example, this stage might wait 652 589 that more prognostics' approaches are available $in_{_{654}}^{_{000}}$ 590 the literature. The field of prognostics of PEMFC has655 591

just started and a great reflection effort is needed to develop 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-TION project (contract "ANR-11-LABX-01-01") both funded by the French National Research Agency as well as the French region of Franche-Comté for their support.

References

599

- 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, A review of polymer electrolyte membrane fuel cells: Technology, 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, Applied Energy 96 (2012) 93–101.
- [4] J. Fernandez-Moreno, G. Guelbenzu, A. Martin, M. Folgado, 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: Fundamentals and applications, Renewable and Sustainable Energy Reviews 32 (2014) 810 – 853.
- [7] G. Niu, D. Anand, M. Pecht, Prognostics and health management 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 Energy 38 (35) (2013) 15307 – 15317.
- [9] J. W. Sheppard, M. A. Kaufman, T. J. Wilmer, IEEE standards for prognostics and health management, Aerospace and 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, 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. 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. Agbossou, 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

692

- [16] K. Javed, R. Gouriveau, N. Zerhouni, Data-driven prognostics683
 of proton exchange membrane fuel cell stack with constraint684
 based summation-wavelet extreme learning machine., in: 6th685
 International Conference on Fundamentals and Development of686
 Fuel Cells, FDFC'15., Toulouse France., 2015, pp. 1–8.
- [17] M. Jouin, R. Gouriveau, D. Hissel, M.-C. Péra, N. Zerhouni,688
 Joint particle filters prognostics for PEMFC power prediction689
 at constant current solicitation, IEEE Transactions on Reliabil-690
 itydoi:10.1109/TR.2015.2454499.
- [18] FCLAB research, IEEE PHM data challenge 2014 (2014).
- 666 URL http://eng.fclab.fr/ieee-phm-2014-data-challenge/ 693
- T. Kim, H. Kim, J. Ha, K. Kim, J. Youn, J. Jung, B. D. Youn,694
 A degenerated equivalent circuit model and hybrid prediction695
 for state-of-health (SOH) of PEM fuel cell, in: IEEE PHM con-696
 ference 2014, 2014, pp. 1–7.
- [20] W. O. L. Vianna, I. P. de Medeiros, B. S. Aflalo, L. R. Ro-698
 drigues, J. P. P. Malere, Proton exchange membrane fuel cells699
 (PEMFC) impedance estimation using regression analysis, in:700
 IEEE PHM conference 2014, 2014, pp. 1–8.
- 675[21]A. Hochstein, H.-I. Ahn, Y. T. Leung, M. Denesuk, Switching702676vector autoregressive models with higher-order regime dynam-703677ics, in: IEEE PHM conference 2014, 2014, pp. 1–10.704
- [22] J. Kuria Kimotho, T. Meyer, W. Sextro, PEM fuel cell prog-705
 nostics using particle filter with model parameter adaptation,706
 in: IEEE PHM conference 2014, 2014, pp. 1–6.
- [23] G. W. Vogl, B. A. Weiss, M. A. Donmez, Standards Related tore
 Prognostics and Health Management (PHM) for Manufactur-709

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, 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/fuelcells/files/files/documents/events/diagnostics_of_pem_ 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.
- [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, Journal 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

- ⁷¹⁰ 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, International Journal of Hydrogen Energy 39 (17) (2014) 9420 – 9429.
- [32] A. Franco, Modelling and analysis of degradation phenomenain 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, Journal 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.
- [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).
- 741 URL http://www.hydrogen.energy.gov/roadmaps_vision.h 742 tml
- 743[40]E. Pahon, S. Morando, R. Petrone, al., Long-term tests duration744reduction for pemfc μ -chp application, in: ICREGA16, 2016,745pp. 1–6.
- [41] J. H. Ward Jr, Hierarchical grouping to optimize an objective function, Journal of the American statistical association 58 (301) (1963) 236-244.
- [42] T. Aroui, Y. Koubaa, A. Toumi, Self-organizing maps for diagnosing induction motors supplied by a variable speed drive, European journal of electrical engineering 14 (6) (2011) 697– 752 717.
- [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 Engineering 2 (6) (2015) 1197.
- [44] M. Djeziri, B. O. Bouamama, R. Merzouki, Modelling and robust 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. Péra, 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-
- 770 Voltage-Ratings.aspx.