

PHM of Proton-Exchange Membrane Fuel Cells - A review

Marine Jouin, Rafael Gouriveau, Daniel Hissel, Marie-Cécile Péra, Nouredine Zerhouni

FEMTO-ST Institute (UMR CNRS 6174) - UFC / ENSMM / UTBM, FCLAB (FR CNRS 3539)
24 rue Alain Savary, F-25000 Besançon, France
rafael.gouriveau@femto-st.fr

Fuel Cell (FC) systems are promising power-generation sources that are more and more presented as a good alternative to current energy converters such as internal combustion engines. They suffer however from insufficient durability for stationary and transport applications, and lifetime may be improved. A greater understanding of underlying wearing processes is needed in order to improve this technology. However, FCs are in essence multi-physics and multi-scales systems (from the cells to the whole power system), which makes a modeling step of behaviors and degradation very difficult, even impossible. Thereby, data-driven Prognostic and Health Management (PHM) principles (as defined in condition-based-maintenance scheme CBM) appear to be of great interest to face with the problems of health assessment and life prediction of FCs. According to all this, the aim of this paper is to present the current state of the art on PHM for FCs. Developments emphasize on PHM of the Proton-Exchange Membrane Fuel Cells (PEMFC) stack. The paper is organized so that important aspects like "behavior and losses FCs", "observation techniques", and "advanced PHM techniques" are addressed. Also, a taxonomy of existing works on PHM of PEMFC is given accordingly to the processing layers of CBM. The whole enables PHM practitioners as well as FCs experts to get a better understanding of remaining challenging issues.

1. Introduction

Fuel Cells (FCs) systems benefit from a growing interest and are nowadays considered as major long-term energy-conversion solutions, since they can offer high fuel economy through substantially lower emissions (than internal combustion engines). Nevertheless, even if this technology is close to being competitive, it is not yet ready to be considered for large scale industrial deployment since a major gap has to be filled: life duration of FCs is still to be improved. A greater understanding of underlying wearing processes is thereby needed in order to improve this technology. However, FCs are in essence multi-physics and multi-scales systems (Jain, 2009), which makes a modeling step of behaviors and degradation very difficult. Also, even if successful works have already been published on monitoring and diagnostics of FCs, further developments are required to accurately estimate the remaining useful life of FCs and to decide mitigation actions accordingly. Following that, Prognostics and Health Management (PHM) appears to be an enabling discipline that aims at utilizing real monitoring data to facilitate relevant indicators and trends that depict the health of the system (Pecht and Jaai, 2010). Also, PHM technology enables deciding adequate actions at the right time when needed in order to extend the system's life, and it benefits from a growing interest from FC community. The aim of this paper is to draw the current state of the art on PHM of the Proton-Exchange Membrane Fuel Cells (PEMFC), and to point out current and future challenging problems to be addressed. Developments emphasize on PHM of the stack of PEMFC.

The core of the paper is organized in three main sections. First, prognostics and PHM discipline are briefly described. This part enables distinguishing important sets of activities to be carried out to "observe", "model / analyze" and "control" a system. Then, main degradation and behavioral models of PEMFC are synthesized in order to depict the variety of aspects to be taken into account for PHM purpose. Finally, before concluding, section 4 is dedicated to the analysis of existing works on PHM of PEMFC accordingly to the processing layers defined for PHM. This enables pointing out remaining challenging issues.

2. Prognostics within Condition-Based Maintenance (CBM)

2.1 Prognostics as a key process

Global performance requirements lead industrials to strengthen their capability to anticipate degradation phenomena and failures. This is mainly achieved thanks to prognostics, that appears to be a key process to move from a "fail to fix" to a "predict to prevent" strategy, enabling the improvement of reliability, availability and safety of systems, while reducing costs and down times.

Although there are some divergences in literature, prognostics can be defined as proposed by the International Organization for Standardization: "prognostics" is the estimation of time to failure and risk for one or more existing and future failure modes" (ISO, 2004). In this acceptation, prognostics is also called the "prediction of a system's lifetime" as it is a process whose objective is to predict the Remaining Useful Life (RUL) before a failure occurs, given the current machine condition and its past operation profile. That said, prognostics forms part of a set of classical processes that all together aim at detecting, diagnosing and predicting degradation mechanisms of systems.

2.2 PHM as a set of complementary activities

The complete aspects of failure analysis and prediction must be viewed as a set of activities, all of them must be performed. Indeed, various activities, ranging from data collection through the recommendation of specific mitigation actions, must be carried out to perform predictive control and maintenance (and thereby improve systems' performances). This aspect is highlighted within the Prognostics and Health Management concept. Generally, a PHM system is seen as the integration of seven layers (initially defined as for CBM, or Condition Based Maintenance), one of them being that of Prognostics. A general PHM architecture is proposed on Figure 1. A brief description of each layer follows.

- **Layer 1: Sensor Module.** It provides the PHM application with digitized sensor or transducer data;
- **Layer 2: Signal Processing Module.** It receives data from the sensors (or transducers or signal processors), and performs signal transformations and features extraction, reduction and selection;
- **Layer 3: Condition Monitoring Module (detection).** It compares on-line data with expected values of system's parameters. It should also be able to generate alerts based on preset operational limits;
- **Layer 4: Health Assessment Module (diagnostics).** It determines if the condition of the system has degraded. The module also generates a diagnostic record and suggests fault possibilities;
- **Layer 5: Prognostics Module.** It predicts the future condition of the monitored system, subsystem or component. The module should be able to acquire data from all previous modules;
- **Layer 6: Decision Support Module.** Its primary function is to provide recommended maintenance actions or alternatives on how to run the system until the mission is completed.
- **Layer 7: Presentation Module.** This module receives data from all previous modules. This module could be built into a regular human-machine interface.

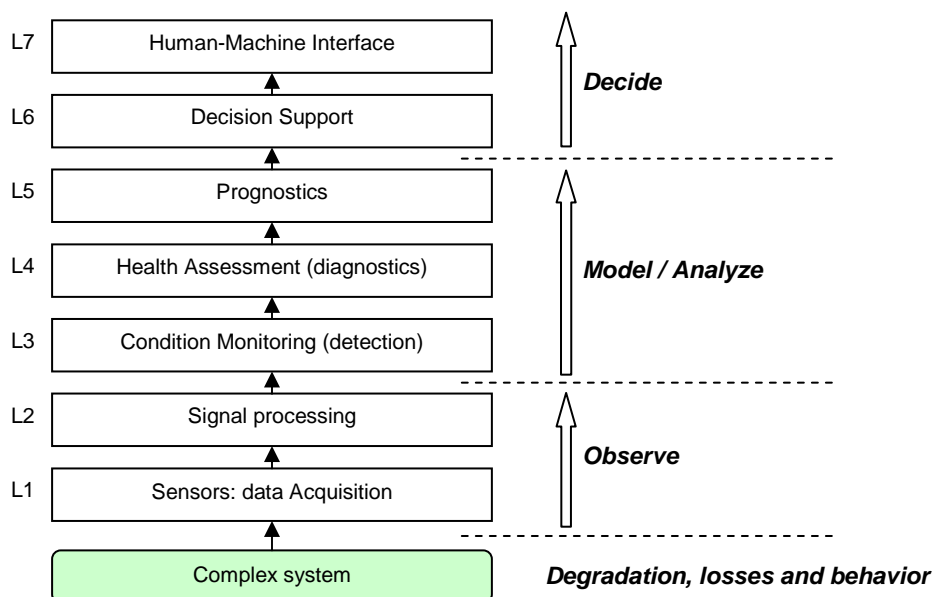


Figure 1: adaptation of the CBM / PHM architecture proposed by (Lebold et al., 2003)

2.3 Towards PHM of PEMFC

Over the past 20 years, many successful applications and implementations of PHM theory in industrial problems have been presented in the literature (it can not be detailed in this paper). Hundreds of papers in PHM (both theoretical and applied) appear every year in academic journals, conference proceedings and technical reports. In sections below, we propose to discuss those papers that address PHM of PEMFC by considering main sets of aspects depicted in Figure 1: "degradation, losses and behavior" (section 3), "observation", "modeling / analysis" and "decision" (section 5).

3. Degradation, losses and behavior of PEMFC

3.1 Degradation mechanisms

Due to its components and all the ancillaries surrounding, PEMFC implies several scales (from μ meter to meter) and multiphysical phenomena, namely electrical, mechanical, electrochemical and thermodynamic phenomena. Consequently, a great number of parameters influencing performances, degradation and durability of PEMFC can be identified. They can be attributed to operating conditions, cell design and assembly, environmental conditions and degradation mechanisms.

Great reviews of the main parameters have been done in (Schmittinger and Vahidi, 2008) and (Wu et al., 2008), they are relative to various fields: water management (flooding or drying out of the membrane – electrode assembly), components degradation (see Table 1), contamination (CO poisoning, presence of impurities initiating chemical attacks), reactant gas starvation or thermal management (influence of freezing or elevated temperature). These parameters, if not properly monitored and controlled, can lead to irreversible degradations and thus to the PEMFC failure. Of course performance degradation cannot be avoided in a long-term period, but mitigation strategies can be used to prevent their consequences (Wu et al., 2008). However it implies that all the phenomena previously listed are completely understood...

Table 1: Component degradation and failure modes (Wu et al., 2008)

Component	Failure modes	Causes
Membrane	Mechanical degradation	Mechanical stress due to non-uniform press pressure, inadequate humidification or penetration of the catalyst and seal material traces
	Thermal degradation	Thermal stress; thermal cycles
	Chemical/electrochemical degradation	Contamination; radical attack
Catalyst / catalyst layer	Loss of activation	Sintering or dealloying of electrocatalyst
	Conductivity loss	Corrosion of electrocatalyst support
	Decrease in mass transport rate of reactants	Mechanical stress
	Loss of reformate tolerance	Contamination
	Decrease in water management ability	Change in hydrophobicity of materials due to Nafion or PTFE dissolution
GDL	Decrease in mass transport	Degradation of backing material
	Decrease in water management ability	Mechanical stress; Change in the hydrophobicity of materials
	Conductivity loss	corrosion
Bipolar plate	Conductivity loss	Corrosion; oxidation
	Fracture/deformation	Mechanical stress
Sealing gasket	Mechanical failure	Corrosion; mechanical stress

3.2 Behavioral models

As we said, PEMFCs are multiphysics systems, which make highly difficult the establishment of a complete behavioral model. Furthermore such a model would hardly be exploitable for practical applications due to the number of parameters and the impossibility of monitoring some of them. But before elaborating a complex model, one has to think carefully of the purpose of its model. Indeed, not the same models are requested when a diagnosis is performed, when the system command is investigated or when the purpose is developing the system.

Considering that the level of the model has to be chosen (microscopic or macroscopic), dimension 0 can be enough for an elementary cell study whereas three dimensions are needed to take into account phenomena like mass transport or electrochemical interactions. Different types of model are already used for PEMFCs studies: static / dynamic models, analytical models (physical or mathematical), hybrid approaches (physical and empirical models associated), black-box models (data driven approaches such as neural networks, fuzzy inference systems, support vector machines, etc.). As for an example, a common model for an electrical approach of PEMFC is the adaptation of the Randles model proposed by (Fouquet et al., 2006), or the equivalent circuit model proposed by (Asghari et al., 2010) (see Figure 2).

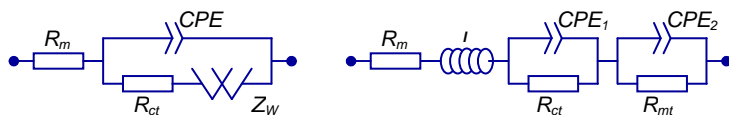


Figure 2: electrical behavioral models – left (Fouquet et al. 2006), right (Asghari et al., 2010)

3.3 Losses in PEMFC

Another efficient way to study PEMFC is to empirically characterize them, namely with polarization curves for the static behavior and Nyquist plots for the dynamical one. These curves give useful information regarding losses and internal resistances in the system. If we take a closer look at the polarization curve on figure 3 for example, we can distinguish four zones corresponding to different types of losses. The Nyquist plot, as for it, can be used to study the behavior evolution during ageing process (Hissel et al., 2007). Five parts are identified due to several phenomena: (1) polarization resistance, (2) mass transport, (3) charges transport, (4) resistance and (5) pseudo inductance due to metallic components.

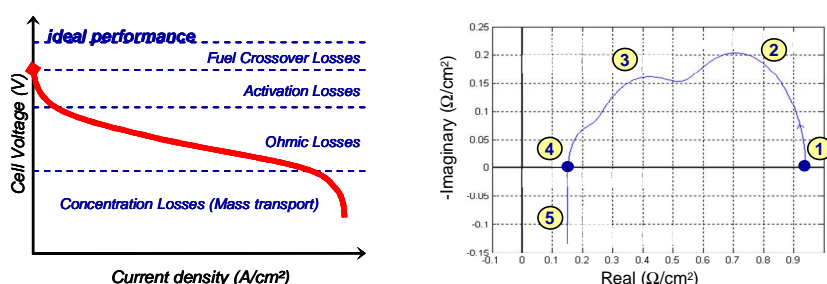


Figure 3: polarization curve and Nyquist plot showing static and dynamic behavior of a cell

4. PHM of PEMFC

Lots of research works have been published in the last twenty years regarding PEMFC but none of them have ever adopted a PHM point of view. Consequently this part aims at showing a really brief overview of PHM regarding PEMFC. The seventh layer concerning human-machine interface (HMI) is left aside: no precise paper deals with it and work teams develop homemade HMIs dedicated to their specific needs.

4.1 Observation layers

A large amount of techniques dedicated to data acquisition have been tried. Of course, usual sensors such as pressure sensors, thermocouples, flowmeter, ammeter, etc. are commonly used. But more informative techniques are also employed. Electrochemical impedance spectrometry (EIS) encounters a large success (Fennie et al., 2001), (Fouquet et al., 2006), (Hissel et al., 2007), (Onanena et al., 2010). Indeed it gives relevant information about the dynamic behavior through a large range of frequencies but it also gives static behavior data when a unique frequency is isolated. However more specific behavior-oriented means have been tested, namely, gas chromatography (Mench et al., 2003), neutron imaging (Tsukada et al., 1999), current interrupt, cyclic voltammetry (Wasterlain et al., 2010) or even transparent cells associated with a camera to record their evolution (Tüber et al., 2003). The main idea of all the methods employed is to realize non-intrusive measurements to create as less perturbations as possible.

To make raw data utilizable, a great variety of data processing tools are available. They can belong to signal processing (Fourier transform (Chen and Zhou, 2008), wavelet transform (Yousfi Steiner et al., 2010)), or to feature extraction techniques (hyperparameters extraction (Onanena et al., 2010), fuzzy model generation and comparison (Hissel et al., 2004), Nyquist diagram or polarization curve generation (Fouquet et al., 2006), (Hissel et al., 2007)).

4.2 Modeling and analysis layers

As previously said, modeling fuel cells behavior is quite difficult, making condition assessment complicated if only model-based. In simple phenomena studies, residues between model data and measurements can be analyzed (Bosco and Fronk, 2000), (Fouquet et al., 2006), (Mench et al., 2003), (Tüber et al., 2003). But to make things easier, artificial neural networks (Yousfi Steiner et al., 2011) and fuzzy logic are more and more exploited for fault detection.

Regarding diagnosis, pattern recognition algorithm (Yousfi Steiner et al., 2010) and fuzzy clustering algorithm (Hissel et al., 2007) are very useful tools as far as fault isolation is concerned. With the same idea, extracted parameters representations such as internal resistances can help to isolate the origin of bad performances (Fouquet et al., 2006).

The prognostics layer is almost absent of PEMFC studies. A few studies try to estimate the remaining lifespan of a specific component or a single cell (Lee et al., 2010). But only one paper in which a remaining useful life (RUL) is calculated has already been published until now (Zhang and Pisu, 2012). It proposes a damage tracking and RUL prediction by using an unscented Kalman filter-based PHM scheme. A physics-based prognostic-oriented catalyst degradation model is implemented in order to link the operating conditions to the degradation rate of the electro-chemical surface area.

4.3 Decision layers

Finally, as regards the decision support, there are no much more works dealing with automatic corrective actions than with prognostics. The American patent (Bosco and Fronk, 2000) is the most complete paper we can find dealing with this subject. Residues from pressure drop measurements (for H₂ and O₂) are compared to empirically defined threshold during the condition assessment and diagnosis phases, then in case of fault detection, corrective actions are launched.

4.4 Discussion: remaining challenges

As stated before, PEMFC community never adopted a PHM point of view regarding its work. That's why no paper dealing with CBM strategies for PEMFC or PHM vocabulary can be found. This brief state of the art reveals that all PHM layers are far from being complete. However, one should keep in mind that PEMFC are intended to be transportable. Thereby, some remaining challenging issues can be pointed out. They are synthetized in Figure 4 and briefly explained here after.

- **Observation.** Research on the development of data acquisition and processing techniques must be pursued so that improved observation strategies are reached (reliable, non-intrusive, and that do not induce perturbations or even damages). They should consider the transportability of the measurement means and so being easily implementable in accordance with applicative constraints (cost, volume, rapid online measurements, etc.).
- **Modeling and analysis.** Even if detection and diagnosis models exist, published works focus on specific points such as flooding, drying out or membrane degradation. Not all wearing mechanisms are taken into account within a single model. As far as prognostics is concerned, one can point out that a complete model taking into account the fuel cell behavior, the degradation processes involved, as well as environmental and operating conditions is still missing. Solid basis have to be found to ensure prognostics development.
- **Decision.** One should not forget that the decision support layer is almost empty too. Fault-tolerant, self-adaptive and reconfigurable control algorithms have to be developed so that the mission can be achieved in the best conditions. Also verification and validation of modeling techniques is an ill-problem, an ongoing issue for PHM community.

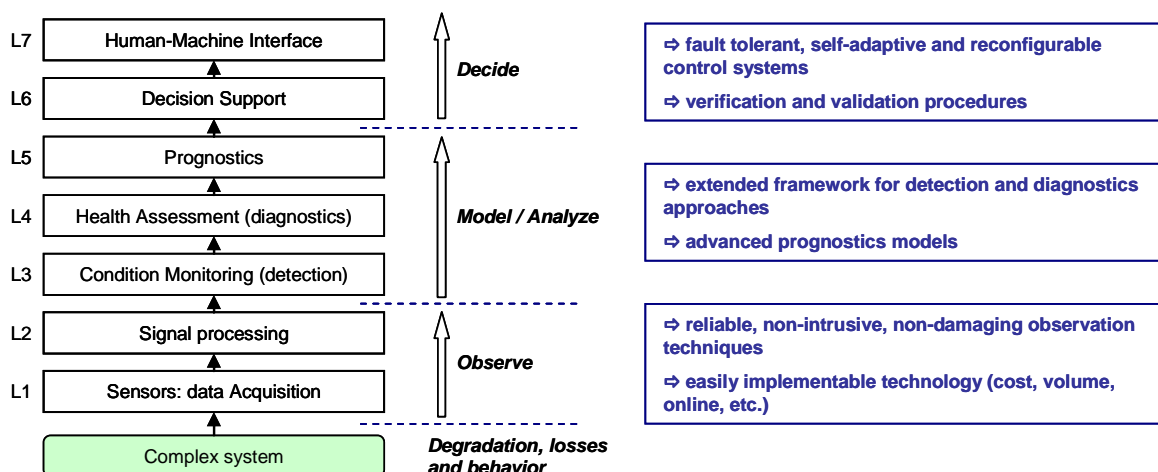


Figure 4: main remaining research issues for PHM of PEMFC

5. Conclusion

Considering benefits that can be expected, FC community has a growing interest in PHM technology. PHM approaches aim at transforming raw monitoring data into relevant information and behavior models (including the degradation) of the system. They take as inputs the current monitoring data and return as outputs predictions or trends about the health state of the system.

The aim of this paper is to present the current state of the art on PHM for FCs with an emphasis on PHM of Proton-Exchange Membrane Fuel Cells (PEMFCs). This study enables to highlight that remaining issues have to be tackled at all processing levels (from raw data to decision support). However, the main open problem is the one of prognostics.

References

- Asghari S, Mokmeli A, Samavati M., 2010, Study of PEM fuel cell performance by electrochemical impedance spectroscopy. *International Journal of Hydrogen Energy*, 35-17, 9283-9290.
- Bosco A.D., Fronk M.H., 2000, Fuel cell flooding detection and correction, US Patent 6,103,409.
- Chen J., Zhou B., 2008, Diagnosis of PEM fuel cell stack dynamic behaviors, *J. Power Sou.*, 177-1, 83-95.
- Fennie C., Reisner D., Barbeta J., Singh P., 2001, Fuzzy Logic- Based State-of-Health Determination of PEM Fuel Cells, in *Proceedings of EVS-18, Berlin*.
- Fouquet N., Doulet C., Nouillant C., Dauphin-Tanguy G., Ould-Bouamama B., 2006, Model based PEM fuel cell state-of-health monitoring via ac impedance measurements, *J. Power Sour.*, 159-2, 905-913.
- Hissel D., Péra M.C., Kauffmann J.M., 2004, Diagnosis of automotive fuel cell power generators, *Journal of Power Sources*, Volume 128-2, 239-246.
- Hissel D., Candusso D., Harel F., 2007, Fuzzy-Clustering Durability Diagnosis of Polymer Electrolyte Fuel Cells Dedicated to Transportation Applications, *IEEE Tr. on Vehicular Technology*, 56-5, 2414-2420.
- ISO, 2004, ISO13381-1, Condition monitoring and diagnostics of machines - prognostics - Part1: General guidelines, International Standard.
- Jain P., 2009, Multi-scale Modeling and Optimization of Polymer Electrolyte Fuel Cells, PhD Dissertation, Carnegie Mellon University, Pittsburgh, Pennsylvania
- Lebold M., Reichard K., Boylan D., 2003, Utilizing dcom in an open system architecture framework for machinery monitoring and diagnostics, *IEEE Aerospace Conference Proceedings*, Big Sky, MT, USA.
- Lee J.H., Lee J.H., Choi W., Park K.W., Sun H.Y., Oh J.H., 2010, Development of a method to estimate the lifespan of proton exchange membrane fuel cell using electrochemical impedance spectroscopy, *Journal of Power Sources*, 195-18, 6001-6007.
- Mench M.M., Dong Q.L., Wang C.Y., 2003, In situ water distribution measurements in a polymer electrolyte fuel cell, *Journal of Power Sources*, 124-1, 90-98.
- Onanena R., Oukhellou L., Candusso D., Harel F., Hissel D., Aknin P., 2010, Fuel cells static and dynamic characterizations as tools for the estimation of their ageing time, *Int. Jour. Hydr. Ene.*, 36-2, 1730-1739.
- Pecht M., Jaai R., 2010, A prognostics and health management roadmap for information and electronics-rich systems. *Microelectronics Reliability*, 50, 317-323.
- Schmittinger W., Vahidi A., 2008, A review of the main parameters influencing long-term performance and durability of PEM fuel cells, *Journal of Power Sources*, 180-1, 1-14.
- Tsukada A., Lehmann E., Vontobel P., Scherer G., 1999, In situ observation of water condensation in an operating polymer electrolyte fuel cell by means of neutron imaging at the spallation neutron source (SINQ), *Tech Report Paul Scherrer Inst.* 5, 84.
- Tüber K., Pózcza D., Hebling C., 2003, Visualisation of water build up in the cathode of a transparent PEM fuel cell, *Journal of Power Sources*, 124-2, 403-414.
- Yousfi Steiner N., Hissel D., Moçotéguy P., Candusso D., 2010, Non intrusive diagnosis of polymer electrolyte fuel cells by wavelet packet transform, *Int. Journal of Hydrogen Energy*, 36-1, 740-746.
- Yousfi Steiner N., Hissel D., Moçotéguy P., Candusso D., 2011, Diagnosis of polymer electrolyte fuel cells failure modes (flooding & drying out) by neural networks modeling, *International Journal of Hydrogen Energy*, 36-4, 3067-3075.
- Wasterlain S., Candusso D., Harel F., Hissel D., François X., 2011, Development of new test instruments and protocols for the diagnostic of fuel cell stacks, *Journal of Power Sources*, 196-12, 5325-5333.
- Wu J., Yuan X.Z, Martin J.J, Wang H., Zhang J., Shen J., Wu S., Merida W., 2008, A review of PEM fuel cell durability: Degradation mechanisms and mitigation strategies, *J. Power Sources*, 184-1, 104-119.
- Zhang X., Pisu P., 2012, An Unscented Kalman Filter Based Approach for the Health-Monitoring and Prognostics of a Polymer Electrolyte Membrane Fuel Cell, *Annual Conference of Prognostics and Health Management Society 2012*.