



Degradation prediction of PEM fuel cell based on artificial intelligence

Loïc Vichard, Fabien Harel, Alexandre Ravey, Pascal Venet, Daniel Hissel

► To cite this version:

Loïc Vichard, Fabien Harel, Alexandre Ravey, Pascal Venet, Daniel Hissel. Degradation prediction of PEM fuel cell based on artificial intelligence. International Journal of Hydrogen Energy, 2020, 45 (29), pp.14953-14963. 10.1016/j.ijhydene.2020.03.209 . hal-02617321

HAL Id: hal-02617321

<https://hal.science/hal-02617321>

Submitted on 22 Aug 2022

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.



Distributed under a Creative Commons Attribution - NonCommercial 4.0 International License

Degradation prediction of PEM fuel cell based on artificial intelligence

L. Vichard^{a,b}, F. Harel^{b,d}, A. Ravey^{a,b}, P. Venet^{b,c}, D. Hissel^{a,b}

^aFEMTO-ST Institute, Univ. Bourgogne Franche-Comté, UTBM, CNRS, Belfort, France

^bFCLAB, CNRS, Belfort, France

^cUniv Lyon, Université Claude Bernard Lyon 1, Ecole Centrale de Lyon, INSA Lyon, CNRS, Ampère, Villeurbanne, France

^dAME-Eco7, Univ Gustave Eiffel, IFSTTAR, Univ Lyon, Lyon, France

Abstract

In the last years, Proton Exchange Membrane Fuel Cells (PEMFC) became a promising energy converter for both transportation and stationary applications. However, durability of fuel cells still needs to be improved to achieve a widespread deployment. Degradation mechanisms and aging laws are not yet fully understood. Therefore, long-term durability tests are necessary to get more information. Moreover, degradation models are requested to estimate the remaining useful life of the system and take adequate corrective actions to optimize durability and availability. This paper presents in a first part the results of a long-term durability test performed on an open cathode fuel cell system operated during 5000 hours under specific operating conditions including start/stop and variable ambient temperature. Performance evolution and degradation mechanisms are then analyzed to understand influence of operating conditions and how to extend the durability. In a second part of the paper, the results are used to build a degradation model based on echo state neural network in order to predict the performance evolution. Results of the degradation prediction are very promising as the normalized root mean square error remains very low with a prediction time over 2000 hours.

Keywords: fuel cell system, fuel cell aging, long term durability test, degradation model, echo state network

1. Introduction

Today's electric vehicles are mainly powered by lithium-ion batteries and there is a long way before they become dominant in the global automotive market. Autonomy of electric vehicles is still too weak to compete with traditional gas vehicles [1]. A solution to improve the autonomy is to hybridize the powertrain by including not only batteries but also alternative electrochemical devices [2]. Proton Exchange Membrane Fuel Cell (PEMFC) is regarded to have a great potential due to the merits of high energy density, high conversion efficiency, low operating temperature and zero gas emission [3]. However, their durability is still a limitation and needs to be improved to achieve a worldwide commercialization [2]. Integrated in a vehicle, a fuel cell system is subjected to rough operating conditions [4][5], such as load variations [6], air impurities, temperature and humidity cycling, cold ambient temperatures [7][8], vibrations and shocks which accelerate their degradation [4][9]. In 2010, the US Department Of Energy (DOE) fixed a durability target at 5000 hours for transportation applications whereas current durability is around 4000 hours. For several years many researches have carried out to improve this durability. Researches focus on improving the system design and the choice of materials, and on reducing the causes of degradation by implementing optimal supervision and energy management strategies.

As part of a continuous durability improving approach, prognostic methods allow to estimate the end of life of the components and then deciding adequate actions at the right time in order to extend possibly the durability [10][11].

Based on historical data describing the performance evolution, prognostic methods model the degradation which then allows predicting the state of health of the considered system. In the case of Proton Exchange Membrane Fuel Cell (PEMFC), different approaches have been proposed in the literature which can be classified according to three categories: model-based methods, data-driven methods and hybrid methods [12][13].

To build a suitable degradation model, a good understanding of the system performance evolution and the degradation mechanisms occurring within the system is primordial. Nowadays, degradation laws and mechanisms of fuel cell are not fully understood. Then, experimental studies with a system operating on test benches or integrated in actual applications are important to get information about these degradation mechanisms. In the literature, there is a lack a long-term durability tests which are costly and time demanding. For this purpose, this study carried out a long-term durability test on a complete open cathode fuel cell system for 5000 hours (DOE target) under operating conditions close to transportation applications.

In a first part, the prognostic discipline will be introduced and existing tools in the literature will be presented. In a second part, the experimental context and the long-term durability tests will be detailed. The fuel cell system, the cycling profile and the chosen health indicators will be highlighted, then results will be drawn. Performance evolution of the system will be observed and influence of operating conditions and operating time on degradation and performance will be analyzed. Based on these analyses, a method to model performance evolution using echo state neural networks, with respect to operating time and according to operating conditions will be detailed. The structure of the network, its parameters, the learning scheme and chosen inputs will be presented. Finally, predictions provided by the model will be highlighted and discussed. The obtained degradation model can be used to estimate remaining useful life, to anticipate maintenance operation or to optimize energy management in order to extend the system durability.

2. Prognostic

2.1 Background

The objective of the prognostic discipline is to estimate the remaining useful life of a system in order to anticipate maintenance operations and to adapt energy management.

The concept of prognostic discipline is highlighted in the Figure 1. The purpose is to use historical data describing the evolution of a State-Of-Health (SOH) indicator with respect to the operating time and according to operating conditions, to build a degradation model. Once the model is built, it can be used with new inputs to predict the SOH indicator i.e. the performance evolution, with respect to the operating time and according to operating conditions.

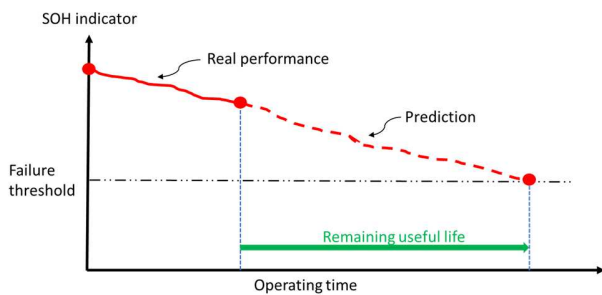


Figure 1: Prognostic concept

When the predicted SOH indicator value reaches a threshold of failure defined by the user, the remaining useful life can be quantified as the difference between the predicted time when threshold is reached and the present time. Generally the end of life is defined as 10% of performance loss, versus beginning of life and for automotive fuel cell stacks [10] but the end of life can be also defined as the inability to meet user specifications [14].

2.2 Methods

Several methods to model the performance evolution can be found in the literature. These methods can be classified according to three categories: model-based, data-driven and hybrids [10][12][13].

2.2.1 Model-based methods

Model-based methods are based on analytical models to reproduce the dynamic behavior of the studied system according to the degradation mechanisms. The main advantage of the model-based methods is that they do not require a large amount of data. Lechartier et al. [15] proposed a degradation model of a fuel cell composed of a static part and a dynamic part that are independent. The static part is based on equations describing physical phenomena and Butler-Volmer law. The dynamic part is based on an electrical equivalency of the physical phenomena. Parameters of the models are updated based on characterizations: polarization curves and electrochemical impedance spectroscopy. Bressel et al. [16] used an extended Kalman filter to estimate the state of health of a fuel cell and to predict its degradation evolution. Bressel et al. [17] also proposed a model based method using the energetic macroscopic representation with time varying parameters to forecast the fuel cell remaining useful life for a given power reference. Lee et al. [18] proposed a method for estimating the state and remaining useful life of a single PEMFC cell through an equivalent impedance model based on electrochemical impedance spectroscopy (EIS). The author uses charge transfer resistance and double layer capacitance as major parameters for judging degradation. EIS is a very suitable tool to get information about state of health of a fuel cell system but for now dedicated to laboratory conditions. Kimotho et al. [19] proposed a prognostic approach based on adaptive particle filter. The proposed method lies in the introduction of a self-healing factor after each characterization and the adaptation of the model parameters to fit the degradation evolution. Results show that prognostic method is reliable with majority of the predictions falling within 5% error. Zhang et al. [20] proposed an approach based on two physical models. The first one receives a signal directly observable and related to the stack voltage. The second one is fed periodically by measurements from physical characterizations of the stack. Characterizations give good information about the SOH evolution. Then, the prognostic procedure is implemented using Particle Filtering (PF).

For a complex system whose degradation mechanisms are influenced by many factors, inferring an analytical model can be very difficult. These methods are therefore rather dedicated to simple systems whose physic laws and degradation mechanisms are well understood.

2.2.2 Data-driven methods

Data driven methods are considered as black box approaches to realize a prognostic. They do not require system knowledge and physical laws to start a prognostic. Measured data and historical data are used to learn the dynamic behavior of the system. Therefore, data-driven prognostic methods are particularly relevant for complex systems whose degradation laws are not well understood. Napoli et al. [21] used a classical neural network to predict stack voltage and cathode temperature of a 5 kW fuel cell. Results showed that the proposed method can reproduce the impact of different stoichiometric ratio on the voltage under different operating conditions. Ibrahim et al. [22] used a discrete wavelet transform to predict the power of a fuel cell. Results showed that the method is satisfying as the prediction error is less than 3%. Silva et al. [23] propose a data-driven method to predict the output voltage reduction of a fuel cell caused by degradation during nominal operating conditions. The proposed method is based on an Adaptive Neuro-Fuzzy Inference Systems (ANFIS) and use as an input the measure output voltage of the fuel cell. Results show that the proposed method is suitable to predict voltage loss. Liu et al. [24] proposed a data-based method using Long Short-term memory (LSTM) recurrent neural network (RNN) to estimate the remaining useful life of a PEMFC. Results are verified with experimental aging data and show that the prediction accuracy of the proposed method is 99.23% and the root mean square error is 0.003. Zhu et al. [25] presented a data driven method for inferring the remaining useful life of a fuel cell. They proposed a method based on the use of a Gaussian process state space model which allows to estimate the evolution of the latent states and the future behavior of the fuel cell voltage. Ma et al. [26] proposed a prognostic data-driven method based on the use of Grid Long Short Term Memory (G-LSTM). The proposed method was experimentally validated, and results indicate that the proposed Grid long short-term memory network can predict the fuel cell degradation in a precise way. Morando et al. [27] used an Echo State neural Network (ESN) for fuel cell prognostic purpose. They model and predict the fuel cell voltage evolution with a mean average percentage error lower than 5%. Moreover, the study revealed that the computational requirements for ESN are very low. Hua et al. [28] also used the ESN as a prognostic method. They also ensure that ESN structure has demonstrated better performances especially in reducing the computational requirements. The study compares the single-input structure with a multiple-input structure. Results show that the multiple-inputs has a better performance under both static and dynamic operating conditions.

2.2.3 Hybrid methods

Hybrid approaches combine data-driven approaches and model-based approaches, cumulating advantages of both approaches. Jouin et al. [29] proposed a hybrid prognostic method which aims at predicting the power losses of a fuel cell stack running under constant operating conditions and constant current. The proposed method is based on a new empirical modeling for power aging and joint particle filters framework. When enough data are available, the prediction of the behavior is promising compared to experimentation. Remaining useful life can be estimated over 500 hours with an error lower than 5%. Cheng et al. [30] proposed a hybrid prognostic method for a fuel cell. The method is based on a data-driven approach: Least Square Support Vector Machine (LSSVM), combined with a model-based approach: regularized particle filter. The results showed that the proposed hybrid method combines both advantages of data-driven and model-based approaches by providing a higher accuracy of the predicted remaining useful life. Liu et al. [31] propose an hybrid method to predict the degradation evolution and estimate the remaining useful life of a PEMFC under different current loads. In a first part the machine learning based on an Adaptive Neuro-Fuzzy Inference System (ANFIS) tool is used to predict the long-term degradation evolution. In a second part the remaining useful life is estimated by using a semi-empirical degradation model based on Unscented Kalman Filter algorithm (UKF). Zhou et al. [32] proposed an hybrid method to estimate the remaining useful life of a PEMFC. The approach is based on the combination of a physical aging model and an autoregressive and moving average (ARMA) model. The prediction accuracy and robustness are experimentally demonstrated with degradation tests performed on two types of PEMFC stack.

Hybrid methods are tough to be developed. Indeed, combining data-driven approaches and model-based approaches allow to cumulate advantages of both approaches but the drawbacks are also cumulated. Therefore, these methods require high amount of data and a good understanding of the physical laws.

Consequently, among the three categories presented previously, a data-driven method has been preferred in this study and specially a method based on the use of a neural network. Degradation laws and mechanisms of fuel cells are still not fully mastered and understood then physical models are very complex to build. In addition, in this study the operating conditions are changed all along the experimentation with a variable ambient temperature. Moreover, the studied system is a commercial system then information about components characteristics are not available. Therefore, a neural network which is independent

of the degradation laws is particularly relevant and suitable for this purpose as shown in Jemei et al. [33] and will be preferred in this study.

3. Experimental context

3.1 Mobypost project

The European project Mobypost aimed at designing and developing a fleet of ten fuel cell hydrogen electric vehicles (FCHEV)^{[16],[17]}. These compact and lightweight vehicles, illustrated in Figure 2, are dedicated to postal delivery applications. A real-time monitoring of all the components of the vehicles is performed in real time all along the driving cycles which has led to create a significant database.



Figure 2 : a vehicle of the Mobypost fleet

3.2 PEMFC in fuel cell hydrogen electric vehicles

In such vehicles, an open-cathode PEMFC system is integrated as a range extender in the powertrain architecture highlighted in Figure 3. The PEMFC system powers the DC bus with a constant current to recharge the battery pack during stop phases or to support it during driving phases. The battery pack provides the current dynamics required for the propulsion of the vehicle through two in-wheels permanent magnet synchronous machines set up with their associated inverters.

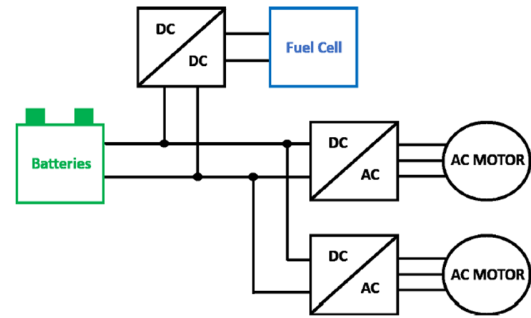


Figure 3: Mobypost vehicle architecture

3.3 PEMFC system

This PEMFC system, illustrated in Figure 4, is a commercial AIRCELLTM system from H2SYS company. The system integrates an open cathode fuel cell stack, an air supply line, a hydrogen supply line, a cooling management system, humidification system, electronics to control electrical fluxes and a controller to supervise actions and safety functions. Its nominal power is 1kW and the stack is composed of 28 cells. The open cathode stack with forced-convection and dead-end anode is air cooled by fans and self-humidified by short-circuits. The nominal operating point is 50A, 62°C (stack temperature) and the ambient operating temperature range specified by the manufacturer is +5°C to 35°C.

Once the system is started up, it is self-powered and communicates through a CAN bus network its physical operating data such as fuel cell temperature, stack current and voltage of the twenty-eight cells.



Figure 4: Fuel cell system

4. Long term durability test

4.1 Test bench

The Figure 5 proposes an illustration of the autonomous test bench used for the durability test. This test bench is composed of a climatic chamber, an active load, a fluids control and an electronic control unit dedicated to control and communications with the fuel cell system. The test bench also integrates its own sensors.

During experiments, the fuel cell system is placed in a 650L ADF type climate chamber (-20°C to +100°C, $\pm 0.5^\circ\text{C}$) from BIA climatic company. The climatic chamber controls the ambient temperature. It is worth noting that in the experimental room, the air relative hygrometry is controlled to be around 50% at 21°C. This point signifies that water content in ambient air remains identical all along the experiments. To ensure the environmental conditions inside the climatic chamber and constantly renew the air, an air circulation was set up as it can be seen in Figure 5.

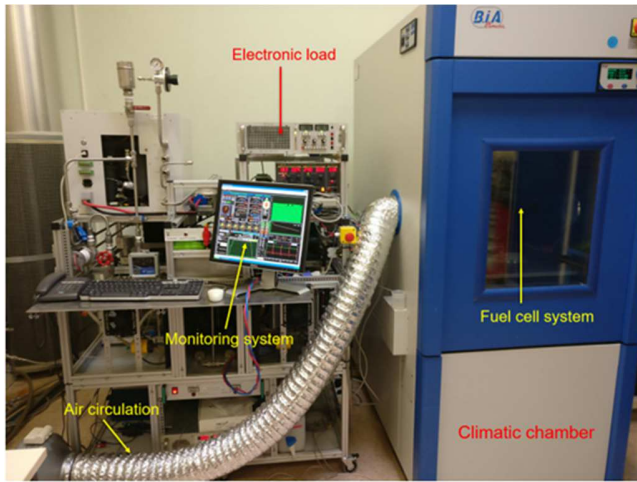


Figure 5: Test bench

4.2 Cycling profile

The purpose of the durability test was here to analyze in lab the system performance evolution when considering the same operating conditions than on the actual FC vehicle. A postal delivery lasts about four hours during which the fuel cell system provides a quite constant current to the DC bus. Based on this application, the Figure 6 highlights the cycling profile used to age the system on the laboratory test bench under the same conditions that it would operate in a range extender architecture [34]. In order to respect average postal delivery duration, the system provides a constant current at its nominal operating point (50A, 60°C) for four hours. This operating phase is followed by two hours of rest by turning off the system. This cycle of six hours was repeated continuously to allow accelerated analysis of performance degradation. Considering this cycling profile, 5000 operating hours equals to 1250 startups/shutdowns and is equivalent to approximately 4 years of vehicle service with a daily postal delivery cycle.

Table 1 : Ambient temperature evolution

Stage 1	Operating time duration	Ambient temperature
1	1000 h	20°C
2	1000 h	30°C
3	400 h	20°C
4	600 h	7°C
5	1000 h	20°C
6	1000 h	10°C

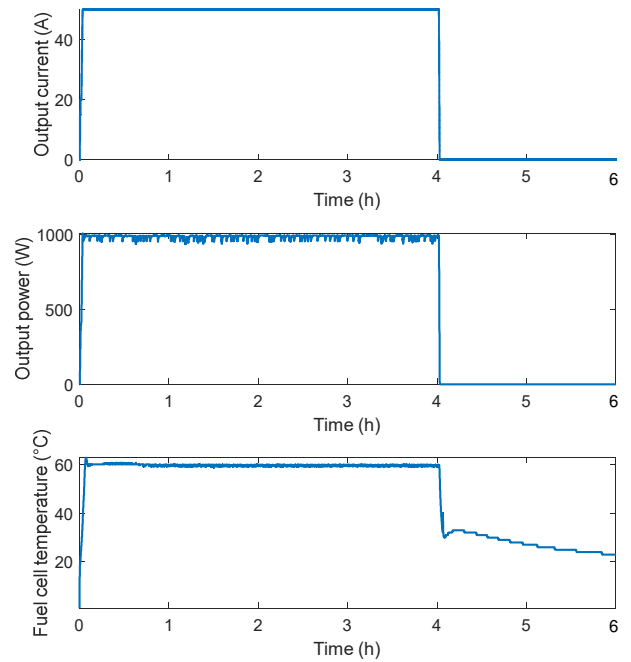


Figure 6: cycling profile

Otherwise, integrated in a vehicle means the system operates under a variable ambient temperature and this ambient temperature mainly depends on the period of the year. To consider these variations, the system was integrated into a climatic chamber. The ambient temperature value was changed per period to approach seasonal variations. The ambient temperature evolution is presented in the Table 1.

4.3 State of health indicator

To follow performance evolution and quantify degradation, a State Of Health (SOH) indicator has to be defined. The SOH of a fuel cell system refers to the quantification of the performance potential compared with a nominal reference. The SOH can be quantified by the loss of power that the system can provide for a specific current value. In the case of this study, the system output current remains unchanged then the system health evolution can be observed with the system output voltage.

- **Stage 3:** the ambient temperature was reduced and set up at 20°C. As soon as the ambient temperature was reduced a voltage increase can be observed: the

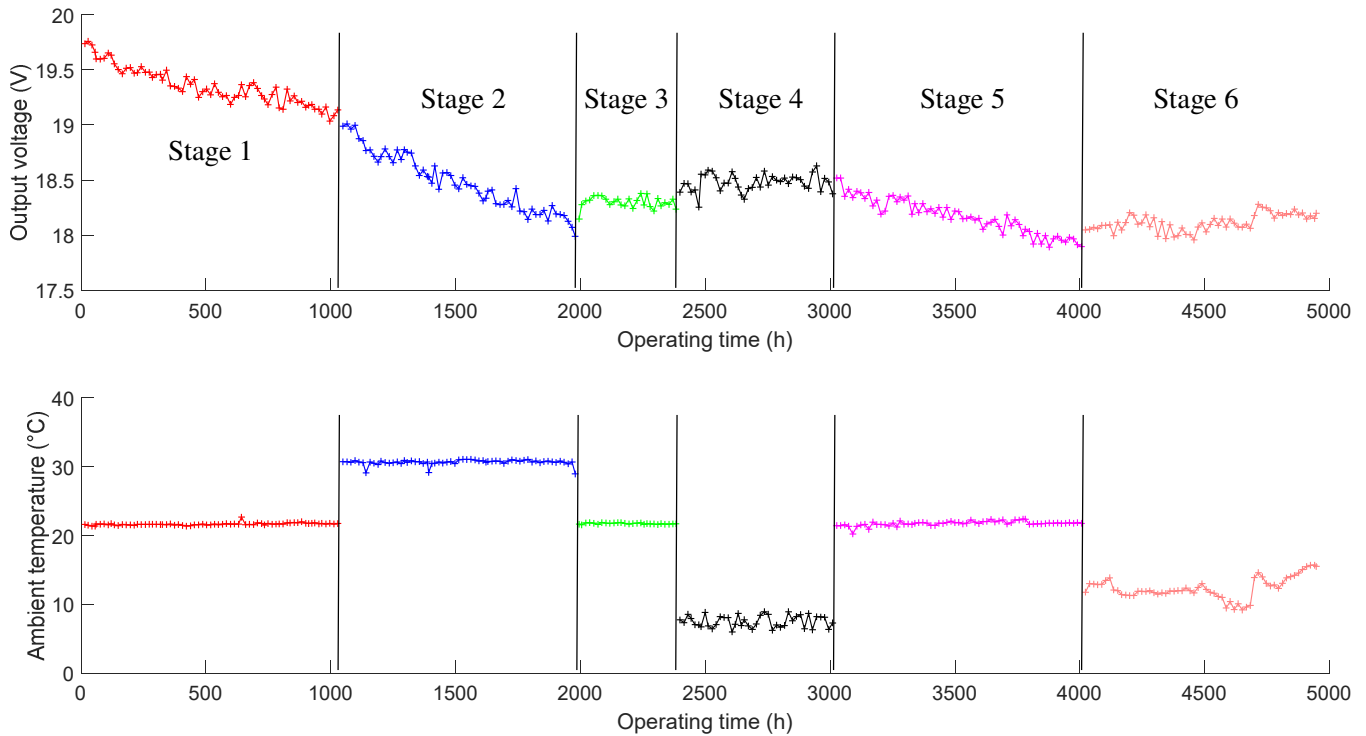


Figure 1: Evolution of system output voltage all along the experimentation

4.4 Voltage evolution

To follow system output voltage evolution during all the experimentation, the operating conditions must be identical. To respect this rule, once a day, the average output voltage is computed during a duration of 1000 seconds. In this period, the fuel cell temperature is stabilized around 60°C and nominal operating conditions are assured. The results of this durability test are presented on the Figure 7. The system output voltage is highlighted with respect to the operating time and the ambient temperature.

These results lead to the following observations:

- **Stage 1:** during the first 1000 hours the ambient temperature was set up at 20°C. It can be observed that the system output voltage decreases constantly overtime due to natural degradation of the components.
- **Stage 2:** the ambient temperature was then elevated and set up at 30°C for the next 1000 hours. As soon as the ambient temperature was elevated, the slope of the system output voltage drop is increased. Higher ambient temperatures seem to accelerate degradation of the system.

operating point seems to be changed. Moreover, the system output voltage evolution remains quite constant during the 400 hours. Degradation is no more noticeable. Reducing the ambient temperature seems to lead to a recovery of the system performance with long term phenomena.

- **Stage 4:** the ambient temperature was reduced again and set up at 7°C. Once more, as soon as the temperature was reduced, a slight variation of the operating point can be noticed. Contrary to the previous stage, here the system output voltage clearly increases during the entire stage. This verify previous observations, reducing the ambient temperature seems to lead to a recovery of the system performance with long term phenomena. Moreover, low ambient temperature seems to reduce the system degradation significantly.
- **Stage 5:** the ambient temperature was elevated and set up at 20°C for the next 1000 hours. The degradation slope returned negative as the system output voltage decreases with a constant slope quite closed to the stage 1.
- **Stage 6:** the ambient temperature was reduced and set up 10°C for the next 1000 hours. Once more, as soon

as the ambient temperature was reduced a variation of the operating point can be noticed: direct elevation of the voltage. Moreover, all along the stage, the system output voltage increases constantly. Degradation slope became positive as in stage 4 what verifies previous observations.

To synthesize, the operating time leads to an irreversible degradation on the system as the global performance trend is to decrease. Otherwise, an impact of the ambient temperature on the performance and the degradation can be observed with short-term and long-term phenomena. Firstly, short-term variations of the operating point are directly linked to the ambient temperature value. Secondly, long-term phenomena acting on the degradation are depending on the ambient temperature: the higher the ambient temperature, the higher the degradation, the lower the ambient temperature the lower the degradation. Moreover, reducing ambient temperature seems to lead to a recovery of the system performance as the degradation slope (performance evolution) becomes positive.

4.5 Analysis

Previous section showed that ambient temperature variations lead to a variation of the operating point which is linked to the net output power supplied by the system. This means that ambient temperature directly affects the net output power supplied by the system. The system output current remains equal to 50A all along the experimentation, what implies that the power required by ancillaries inside the system varies according to the ambient temperature. This variation of the power required by the ancillaries can be explained by the air-cooling regulation to maintain the fuel cell stack temperature at 60°C. The higher the ambient temperature, the higher the air flow rate to cool the fuel cell stack, the higher the power required by the fans.

This link between ambient temperature and cooling air flow can explain long-term phenomena acting on system degradation. In this system, the humidification strategy remains unchanged whatever the ambient temperature. Therefore, if the air flow increases for higher ambient temperatures then the evacuation of the water created by electrochemical reactions will be accelerated. In other words, the higher the ambient temperature, the lower the water content in the fuel cell membranes. Then, reducing the ambient temperature leads to re-moisture of the membranes which can explain the long-term performance recoveries observed during the experimentation.

To conclude on this durability test, the system has operated for 5000 hours under variable ambient temperature conditions and numerous start/stop cycles and the performance degradation is lower than 10%. Global

efficiency is higher than 49% and degradation rate is 11μV/cell/h.

This kind of system: open cathode, air-cooled and self-humidified seems to be a very promising energy converter integrated in a low power range extender architecture for transportation applications. Moreover, experimentation carried out in this study showed that better humidification conditions would lead to a significant reduction of the degradation so a significant durability improvement.

Based on this durability test and the presented aging data the purpose of this study is to propose a method to model the degradation according to the operating time and ambient temperature variations which have shown a significant influence on the system performance.

5. Degradation model

Several structures of neural network have been presented in the literature. Recurrent Neural Networks (RNN) are particularly suitable to model non-linear temporal signals. Morando et al. [36] proposed a comparative study of different neural network architectures used for fuel cell prognostic purpose. They showed that RNN and more specially Echo State Networks (ESN) provide very good results and very interesting computing times. Consequently, ESN with multiple inputs will be preferred as a tool to model the performance evolution of the studied system.

5.1 Echo state network

5.1.1 Background & structure

Echo state network was introduced by Jaeger et al. [37] at the beginning of the 2000s. This type of recurrent neural network reproduces more faithfully the functioning of the human brain as the hidden layers are replaced by a reservoir of neurons. The structure is highlighted in the Figure 8.

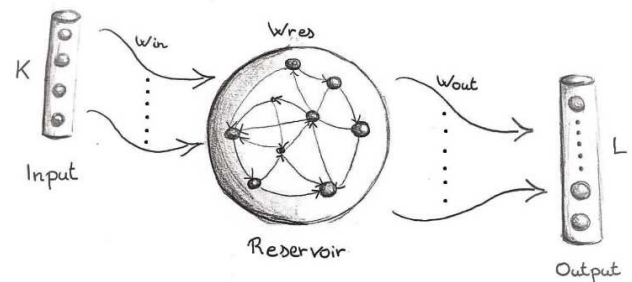


Figure 8: ESN architecture

with:

- N_{res} the number of neurons in the reservoir.
- K the number of inputs.
- L the number of outputs.
- W_{in} the input weight matrix.

- W_{out} the output weight matrix.
- W_{res} the reservoir weight matrix.

The input weight matrix and the weight matrix of the reservoir are created randomly. Concerning the optimization of the weights, only the output weight matrix is optimized by a multi linear regression. Therefore, computing times to build the model are much lower compared to feedforward neural network or again Long Short-Term Memory neural networks (LSTM).

5.1.2 ESN parameters

Some parameters configure an ESN. They can be listed as following:

- The number N_{res} of neurons in the reservoir. Theoretically, a high number of neurons leads to a better accuracy of the model. However, it is important to pay attention to not define a too important number of neurons since computation times are very closely related.
- The leaking rate α . Between 0 and 1, the leaking rate can be defined as the ability to forget the previous states. In other words, the leaking rate refers to the influence of previous states on the current state.
- The spectral radius of the reservoir weight matrix, between 0 and 1, refers to the effective time constant of the ESN. Higher spectral radius leads to a slower decay of impulse response. The spectral radius value also determines the amount of nonlinear interaction of input components through the time. This parameter corresponds to the maximum eigenvalues of the reservoir matrix. The reservoir matrix is created randomly then its spectral radius value is also random. In order to obtain a reservoir matrix with the desired spectral radius, the reservoir matrix is divided by its spectral radius and then multiplied by the desired spectral radius.

5.1.3 Learning scheme

The learning scheme of an ESN is governed by the following steps. In a first time, the reservoir update $\tilde{x}(n)$ is computed as in the equation (1).

$$\tilde{x}(n) = f(W_{in} \cdot u(n) + W_{res} \cdot x(n-1)) \quad (1)$$

with $\tilde{x}(n)$ the reservoir update, $u(n)$ the ESN input and $x(n-1)$ the output value of the reservoir according to the previous state. W_{res} is normalized by dividing it by its spectral radius value and then multiplied by the desired spectral radius value. Based on the previous equation, the reservoir output can be express as following:

$$x(n) = (1 - \alpha) \cdot x(n-1) + \alpha \cdot \tilde{x}(n) \quad (2)$$

with α the leaking rate.

Then can be determined the output of the ESN $y(n)$ from the previous equations:

$$y(n) = f(W_{out} \cdot x(n)) \quad (3)$$

where f is a nonlinear function as a sigmoid, tangent, etc. The purpose of the learning scheme is to optimize the output matrix W_{out} in order to estimate the output $y(n)$ of the ESN as close as possible to the real value y_{target} by minimizing the quadratic error (4).

$$\varepsilon(y, y_{target}) = \sqrt{\frac{1}{T} \sum_{n=1}^T (y_i(n) - y_i^{target}(n))^2} \quad (4)$$

with T the number of samples.

5.2 Model inputs

The purpose of a degradation model is to reproduce and predict the performance evolution of a system according to the operating conditions. Results of the durability tests highlighted that the performance and the degradation of the fuel cell system show relationships with the ambient temperature and the operating time. In our case, the operating time variable is meaningful as the system operates at nominal operating point during the entirety of the durability test. In the case of the current profile is different, the operating time variable would not be relevant as aging image. To tackle this issue, the total electric charge (Ah) supplied by the system would be much more meaningful as input variable referring to the aging. Accordingly, in this study ambient temperature and operating time are defined as inputs of the proposed model. Moreover, a fuel cell system is a dynamic system. It implies that the output system voltage at previous states is linked to the output voltage at present state through a non-linear differential relationship. Consequently, for the proposed model, output voltage at previous state is defined as an input.

Obviously, adding more input variables leads to a better model generality. In our case, the load current is constant then it is not relevant as an input variable. In the case of a different current profile, considering this current as an input variable would have been required. Moreover, in our case and as already explained, the water content in the ambient air is unchanged all along the test, then the relative humidity would not be relevant as an input variable. Facing another situation, considering the relative humidity as an input variable would have been relevant.

Nevertheless, adding more input variables would lead to higher computational requirements and higher equipment requirements such as sensors, wiring, data logging, etc. This is a reason why a good compromise between model accuracy and measured variables must be reached.

To summarize, the proposed model includes three inputs: ambient temperature, operating time and output voltage at previous state. It is worth noting that once the model is built, the output voltage at previous state becomes the predicted output voltage at previous state.

5.3 Results

The model has been developed with the Anaconda™ environment and the used programming language is python which is suitable for machine learning purpose. The learning rate is set at 0.5 and the spectral radius is set at 0.5. These parameters were identified empirically. The computing time to build the model is about two seconds. The computer used to build the model runs with Windows 10 and integrates a CPU Intel core I7-6800k 3.40GHz, a GPU NVIDIA TITAN X and 64 giga of memory.

Firstly, the learning rate was set at 60%. The learning rate is defined as the ratio between the quantity of data used for the training and the total quantity of data in the database. With a learning rate set at 60%, the model has been built and trained with the entire ambient temperature range, then performance recoveries observed previously are learnt. Results are highlighted in the Figure 9. It can be observed that the model is able to predict the voltage evolution with a very good reliability whatever the ambient temperature. The Normalized Root Mean Square Error (NRMSE) is equal to 0.098 with a prediction over 2000 hours.

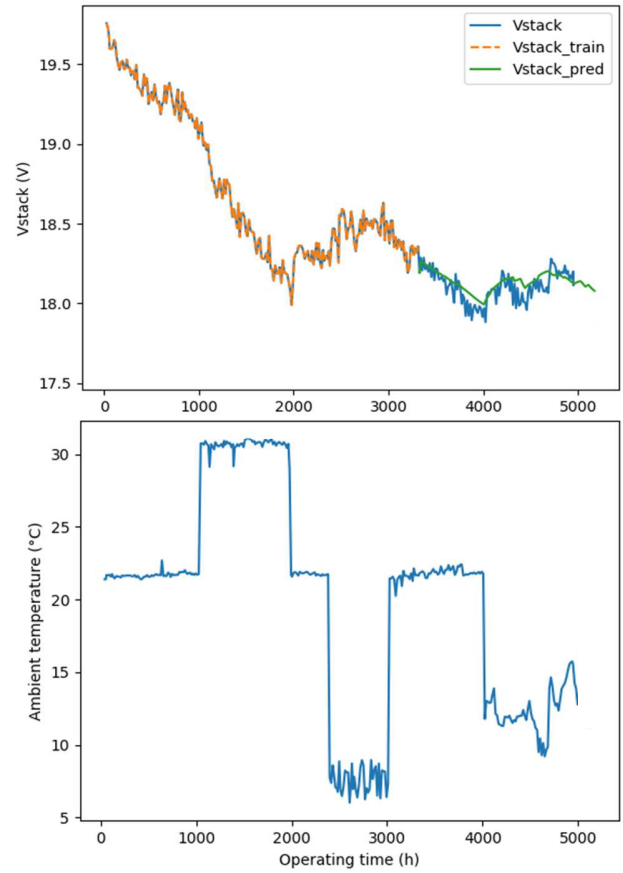


Figure 9: Results with learning rate = 60%

The learning rate value is really important, and users must be very carefully according to their application. In a second time, the learning rate was set at 33%. A learning rate set at 33% refers to an operating duration about 1600 hours and an ambient temperature range between 20°C and 30°C used to build the model. The results of the performance prediction are presented in the Figure 10.

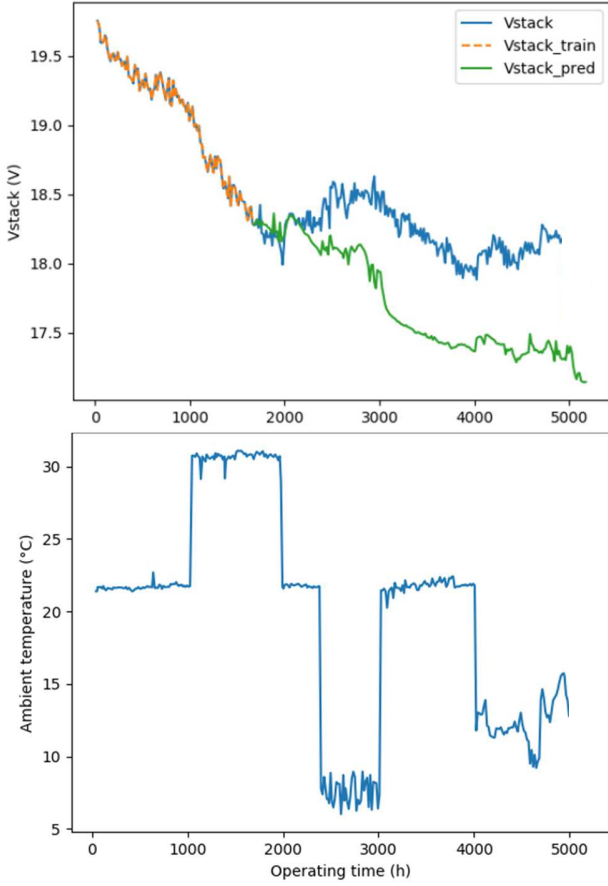


Figure 10: Results with learning rate = 33%

In this figure, it can be observed that the results are not satisfying. The prediction, is close to the reality from 1600 hours to 2000h where the ambient temperature is 30°C. Once the ambient temperature is reduced to 20°C (stage 3) the predicted degradation is attenuated in the image of the stage 1 but the performance recovery is not reproduced by the model. The same thing can be observed for stages 4 and 6 when ambient temperature is 7°C and 10°C. The predicted voltage evolution presents a negative slope, lower than the other stages but not positive as expected. To summarize with a learning rate set at 33% the performance recovery phenomena are not reproduced as the model did not learn them.

In a same way, the Figure 11 highlights a prediction with a learning rate set at 20% which refers to the first 1000 operating hours. In this case, only the first stage has been used to train and build the model. This signifies that the model has been trained with an ambient temperature value always equal to 20°C. Therefore, in this case the model does not know that the ambient temperature affects the system performance and degradation, not even higher ambient temperature (30°C). This is the reason why the performance evolution is quite constant as a continuity of the first 1000

hours. In the Figure 10, despite the performance recovery phenomena are not considered, the ambient temperature effects are considered in some ways as the performance evolution slopes varies with the ambient temperature. Contrary, as shown in Figure 11, with a learning rate of 20% the ambient temperature value does not affect prediction at all.

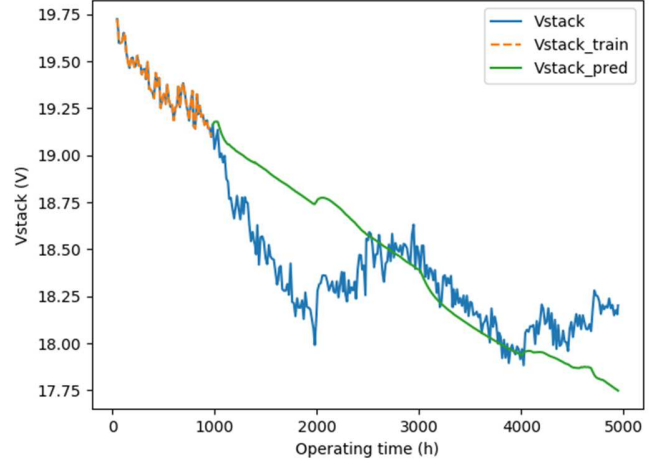


Figure 11: Results with learning rate = 20%

Depending on the application and the operating conditions, the influence of the learning rate on the model reliability can be very significative. In the case of the threshold of failure is defined at 17.5V then with a learning rate set at 33% the estimated end of life of the system would be about 3000 hours. With a learning rate set at 60% then the estimated end of life seems to be higher than 6000 hours.

To conclude on this prognostic tool, the reliability is very promising as the NRMSE is equal to 0.098 with a prediction over 2000 hours and 3000 hours of learning under variable operating conditions. It could be integrated online to model performance evolution and estimate in real time the remaining useful life of the system based on historical measured data. The higher the number of measured data, the higher the accuracy of the model.

6. Conclusion

This study presents a method to model the degradation of a fuel cell system in order to estimate its remaining useful lifetime. Firstly, a long-term durability test of 5000 hours was presented. This test was carried out to observe performance evolution of a complete open cathode fuel cell system under transportation conditions: start/stop and variable ambient temperature. Then, results were analyzed in order to understand performance evolution. Analysis showed that the ambient temperature has an influence on the performance and the degradation of the system with short and long term phenomena. Low ambient temperature leads to

better humidification conditions and significantly reduces the degradation rate. This system seems very reliable for the application as the average degradation rate is $11\mu\text{V}/\text{cell}/\text{h}$ after 5000 operating hours and the performance loss is 9% versus the beginning of life. It must be underlined that these experimental results are better than the durability target performances set by the US DOE for automotive applications. Finally, measured data were used to build a degradation model based on echo state neural network. Results show that the proposed prognostic tool is very suitable as the degradation prediction is very accurate with a prediction time over 2000 hours whatever the ambient temperature and computing times about 2 seconds.

7. Perspectives

Analysis of the aging data and the understanding of the degradation mechanisms and causes following the long-term durability test will be used to develop an intelligent energy management and humidification strategy in order to extend the fuel cell system durability.

In addition, the proposed prognostic tools can be integrated in future systems in order to estimate in real time the remaining useful life and to anticipate the maintenance operations. It can be also used to adapt energy management strategy to optimize the system durability.

8. References

- [1] Saw LH, Ye Y, Tay AAO. Integration issues of lithium-ion battery into electric vehicles battery pack. *J. Clean. Prod.* 2016; 113: 1032–1045.
- [2] Lü X, Qu Y, Wang Y, Qin C, Liu G. A comprehensive review on hybrid power system for PEMFC-HEV: Issues and strategies. *Energy Convers. Manag.* 2018; 171: 1273–1291.
- [3] Vadiée A, Yaghoubi M, Sardella M, Farjam P. Energy analysis of fuel cell system for commercial greenhouse application – A feasibility study. *Energy Convers. Manag.* 2015; 89: 925–932.
- [4] Pei P, Chen H. Main factors affecting the lifetime of Proton Exchange Membrane fuel cells in vehicle applications: A review. *Appl. Energy* 2014; 125: 60–75.
- [5] Zhao J, Li X. A review of polymer electrolyte membrane fuel cell durability for vehicular applications: Degradation modes and experimental techniques. *Energy Convers. Manag.* 2019; 199: 112022.
- [6] Wahdame B, Candusso D, François X, Harel F, Péra M-C, Hissel D, Kauffmann J-M. Comparison between two PEM fuel cell durability tests performed at constant current and under solicitations linked to transport mission profile. *Int. J. Hydrogen Energy* 2007; 32: 4523–4536.
- [7] Oszcipok M, Riemann D, Kronenwett U, Kreideweis M, Zedda M. Statistic analysis of operational influences on the cold start behaviour of PEM fuel cells. *J. Power Sources* 2005; 145: 407–415.
- [8] Jia L, Tan Z, Kang M, Zhang Z. Experimental investigation on dynamic characteristics of proton exchange membrane fuel cells at subzero temperatures. *Int. J. Hydrogen Energy* 2014; 39: 11120–11127.
- [9] Zhang T, Wang P, Chen H, Pei P. A review of automotive proton exchange membrane fuel cell degradation under start-stop operating condition. *Appl. Energy* 2018; 223: 249–262.
- [10] Jouin M, Gouriveau R, Hissel D, Péra M-C, Zerhouni N. Prognostics and Health Management of PEMFC – State of the art and remaining challenges. *Int. J. Hydrogen Energy* 2013; 38: 15307–15317.
- [11] Sutharssan T, Montalvao D, Chen YK, Wang W-C, Pisac C, Elemara H. A review on prognostics and health monitoring of proton exchange membrane fuel cell. *Renew. Sustain. Energy Rev.* 2017; 75: 440–450.
- [12] Sutharssan T, Montalvao D, Chen YK, Wang W-C, Pisac C, Elemara H. A review on prognostics and health monitoring of proton exchange membrane fuel cell. *Renew. Sustain. Energy Rev.* 2017; 75: 440–450.
- [13] Jouin M, Bressel M, Morando S, Gouriveau R, Hissel D, Péra M-C, Zerhouni N, Jemei S, Hilairret M, Ould Bouamama B. Estimating the end-of-life of PEM fuel cells: Guidelines and metrics. *Appl. Energy* 2016; 177: 87–97.
- [14] Lechartier E. Contribution au pronostic de pile à combustible PEMFC basé sur modèle semi-analytique. PhD thesis, Université de Franche Comté 2017.
- [15] Lechartier E, Laffly E, Péra M-C, Gouriveau R, Hissel D, Zerhouni N. Proton exchange membrane fuel cell behavioral model suitable for prognostics. *Int. J. Hydrogen Energy* 2015; 40: 8384–8397.
- [16] Bressel M, Hilairret M, Hissel D, Ould Bouamama B. Extended Kalman Filter for prognostic of Proton Exchange Membrane Fuel Cell. *Appl. Energy* 2016; 164: 220–227.
- [17] Bressel M, Hilairret M, Hissel D, Ould Bouamama B. Model-based aging tolerant control with power loss prediction of Proton Exchange Membrane Fuel Cell. *Int. J. Hydrogen Energy* 2018.

- [18] Lee J, Lee J-H, Choi W, Park K-W, Sun H-Y, Oh J-H. Development of a method to estimate the lifespan of proton exchange membrane fuel cell using electrochemical impedance spectroscopy. *J. Power Sources* 2010; 195: 6001–6007.
- [19] Kimotho JK, Meyer T, Sextro W. PEM fuel cell prognostics using particle filter with model parameter adaptation. 2014 Int. Conf. Progn. Heal. Manag. PHM 2014 2015; 1–6.
- [20] Zhang D, Baraldi P, Cadet C, Yousfi-Steiner N, Béranger C, Zio E. An ensemble of models for integrating dependent sources of information for the prognosis of the remaining useful life of Proton Exchange Membrane Fuel Cells. *Mech. Syst. Signal Process.* 2019; 124: 479–501.
- [21] Napoli G, Ferraro M, Sergi F, Brunaccini G, Antonucci V. Data driven models for a PEM fuel cell stack performance prediction. *Int. J. Hydrogen Energy* 2013; 38: 11628–11638.
- [22] Ibrahim M, Steiner NY, Jemei S, Hissel D. Wavelet-Based Approach for Online Fuel Cell Remaining Useful Lifetime Prediction. *IEEE Trans. Ind. Electron.* 2016; 63: 5057–5068.
- [23] Silva RE, Gouriveau R, Jemei S, Hissel D, Boulon L, Agbossou K, Yousfi Steiner N. Proton exchange membrane fuel cell degradation prediction based on Adaptive Neuro-Fuzzy Inference Systems. *Int. J. Hydrogen Energy* 2014; 39: 11128–11144.
- [24] Liu H, Chen J, Hou M, Shao Z, Su H. Data-based short-term prognostics for proton exchange membrane fuel cells. *Int. J. Hydrogen Energy* 2017; 42: 20791–20808.
- [25] Zhu L, Chen J. Prognostics of PEM fuel cells based on Gaussian process state space models. *Energy* 2018; 149: 63–73.
- [26] Ma R, Yang T, Breaz E, Li Z, Briois P, Gao F. Data-driven proton exchange membrane fuel cell degradation predication through deep learning method. *Appl. Energy* 2018; 231: 102–115.
- [27] Morando S, Jemei S, Hissel D, Gouriveau R, Zerhouni N. Proton exchange membrane fuel cell ageing forecasting algorithm based on Echo State Network. *Int. J. Hydrogen Energy* 2017; 42: 1472–1480.
- [28] Hua Z, Zheng Z, Péra M-C, Gao F. Remaining useful life prediction of PEMFC systems based on the multi-input echo state network. *Appl. Energy* 2020; 265: 114-791.
- [29] Jouin M, Gouriveau R, Hissel D, Péra M-C, Zerhouni N. Prognostics of PEM fuel cell in a particle filtering framework. *Int. J. Hydrogen Energy* 2014; 39: 481–494.
- [30] Cheng Y, Zerhouni N, Lu C. A hybrid remaining useful life prognostic method for proton exchange membrane fuel cell. *Int. J. Hydrogen Energy* 2018; 43: 12314–12327.
- [31] Liu H, Chen J, Hissel D, Su H. Remaining useful life estimation for proton exchange membrane fuel cells using a hybrid method. *Appl. Energy* 2019; 237: 910–919.
- [32] Zhou D, Al-Durra A, Zhang K, Ravey A, Gao F. Online remaining useful lifetime prediction of proton exchange membrane fuel cells using a novel robust methodology. *J. Power Sources* 2018; 399: 314–328.
- [33] Jemei S. par Réseaux de Neurones To cite this version : Modélisation d ’ une pile à combustible de type PEM par réseaux de neurones. PhD thesis, Université de Technologie de Belfort Montbéliard 2004.
- [34] Ravey A, Faivre S, Higel C, Harel F, Djerdir A. Energy management of fuel cell electric vehicle with hybrid tanks. *IECON Proc. Industrial Electron. Conf.* 2014; 3962–3967.
- [35] Vichard L, Ravey A, Morando S, Harel F, Venet P, Pelissier S, Hissel D. Battery aging study using field use data, in: *IEEE Veh. Power Propuls. Conf.*, 2017.
- [36] Morando S. Contribution au Pronostic de Durée de Vie d’une Pile à Combustible à Membrane Echangeuse de Protons. PhD thesis, Université de Franche Comté 2015.
- [37] Jaeger H. The “echo state” approach to analysing and training recurrent neural networks-with an erratum note. *Bonn, Ger. Ger. Natl. Res. Cent. Inf. Technol. GMD Tech. Rep.* 2001; 148:.



**PEMFC system under
actual operating
condition**



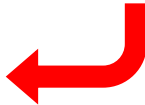
**5000h durability test:
dynamic operation &
variable ambient
temperature**



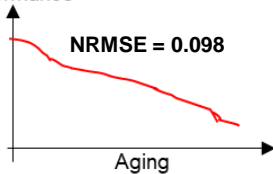
Performance loss <10%



**Degradation model
based on Echo State
Network**



Performance



Aging