

Fuzzy-Clustering Durability Diagnosis of Polymer Electrolyte Fuel Cells Dedicated to Transportation Applications

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Abstract—Polymer electrolyte fuel cells (FCs) are often considered as the most promising power-generation sources for next-generation electrical or hybrid electrical vehicles. However, areas needing further development are the improvements of the efficiency, durability, and reliability of the whole powertrain. Moreover, freeze start of the FC system is also a major issue. To reach these aims, diagnosis solutions of FC stacks and systems can speed up the development cycle of this new technology. The aim of this paper is, thus, to propose some guidelines within the framework of embedded FC system durability diagnosis.

Index Terms—Diagnosis, fuel cell, fuzzy clustering, transportation applications.

I. INTRODUCTION

ELECTRICAL energy is now accepted as a relatively clean and universally available vector of energy in almost all areas of life. One exception is road traffic. Despite the fact that electrical vehicles dominated the early development of motor cars, the internal combustion engine (ICE) has prevailed because of the high-energy density of petrol. Nevertheless, in the last decade, automanufacturers around the world have launched important research programs on fuel cells (FCs) as major long-term energy-conversion solutions because they can offer high fuel economy, through higher efficiency from the stack to the wheel, and substantially lower emissions, particularly of CO₂, or even no emission at all, if pure hydrogen is used as a fuel. Considering the different types of FCs that can be encountered, the polymer electrolyte FC (PEFC) seems to be the most promising power-generation source for FC mobile applications [1], [2]. Of course, most car manufacturers have already demonstrated buses or cars powered by FCs: most of them by PEFC. In association with the FC suppliers, these car manufacturers have made great efforts to improve stack performances [3].

However, areas needing further development are the improvements in the efficiency and the durability of the whole powertrain. Indeed, on the one hand, improvement in efficiency can still reduce the pollutant emissions from well to wheel and, therefore, increase the pertinence of FC powertrains versus classical ICE powertrains. On the other hand, improvement of FC system durability will decrease the relative cost of such technical solutions and go further to meet the requirements of the automotive industry (for example, a durability of more than 5000 h is expected for personal car applications). To reach these aims, diagnosis solutions of FC stacks and systems can speed up the development cycle of new technologies, such as FC vehicles, and improve user support and acceptance by reducing down time [4].

Different diagnosis methodologies for FC stacks or electrochemical energy sources have already been presented. Some of them are model-based [5], but efficient identification of FC stack inner parameters is often difficult to obtain, unless the FC stack internal design is modified. For instance, Burford *et al.* [6] propose to perform FC stack temperature distribution measurements using nanosized thermocouples embedded within the membrane electrode assembly. Other FC diagnosis methodologies are based on gray or black-box models. These behavioral models can be obtained owing to the fuzzy logic [7], neural networks [4], or nonparametric identification using Markov parameters [8]. Then, derivation from the “standard” behavioral conditions is detected to evaluate the state-of-health of the FC stack [4]–[8]. Moreover, in [5], the authors propose the fusion of data coming from monitored and virtual sensors to evaluate this derivation. A last FC stack diagnosis methodology is based on electrochemical impedance spectrometry (EIS) [9]. This methodology is particularly suitable for a better understanding of physicochemical phenomena that are taking place in the PEFC and for the parameter determination of a PEFC equivalent circuit description. It also permits characterization of, in a very efficient way, both the static and dynamical behaviors of FC stacks.

The aim of this paper is to propose a new FC system durability diagnosis methodology by combining the advantages of behavioral modeling methodology and EIS methodology. Thus, in the first part of this paper, experimental results obtained on small power PEFC stacks under durability constraints are provided, and impedance spectrometry is performed on these stacks. Then, in the second part of this paper, the bases of fuzzy pattern recognition diagnosis are recalled. Finally, the last part

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TABLE I
TECHNICAL SPECIFICATIONS OF THE CONSIDERED PEFC STACKS

Number of cells	3
Nominal current	50A
Maximal power	100W
Cell area	100cm ²
Operating temperature	20°C to 65°C
Nominal operating temperature	55°C
Nominal air dew point	45°C
Operating pressure	Max 1.5 bar (abs)
Maximal differential pressure (anode/cathode)	0.6 bar
Media inlet	
Anode	Pure, dry hydrogen
Cathode	Humidified air
Coolant circuit	De-ionized water

of this paper is devoted to PEFC experimental diagnosis and identification of the causes of aging.

II. DURABILITY TESTS ON SMALL POWER PEFC STACKS

A. General Considerations

An FC stack could appear as an inherently reliable system due to the absence of any moving part. Nevertheless, the stack itself is prone to material degradation, and the Membrane Electrode Assemblies (MEAs) are subjected to mechanical constraints. In fact, two main causes can strongly affect the durability of a PEFC stack: the operating conditions (i.e., amount of reactant gas flows versus the load current demand, operating temperature, hygrometry levels of incoming gases, etc.) and some physical degradation causes (for instance, poisoning of catalyst sites, loss of proton conductivity in the membrane, sealing degradation, corrosion of plates, etc.) [10], [11].

Now, focusing on the aging mechanisms and the lifetime of an FC stack, what could be very interesting to know is the fact that the stack is at the beginning of its lifetime or at the end. Indeed, such information could be very useful for both engineers and the researchers during the research and development phase but in the future as well, considering market FC vehicles, in order to optimize the design and the control of the FC stack versus the aging process. Of course, the state of the FC stack should be deduced from as few measurements on the real system as possible and with as few sensors as possible. Moreover, in case of an ageing FC stack, it could also be very useful to diagnose what kind of life (i.e., operating conditions) this FC stack has had.

In our case, we choose to work only with current/voltage measurements in the static and dynamical modes. In particular, regularly time-spaced impedance spectrometry has been done to characterize the dynamical behavior of the FC stack. The tests are performed on identical small power PEFC stacks (three-cell stacks and an electrical power of about 100 W). The main technical specifications of these stacks are given in Table I. Of course, these low-power FC stacks are far away from the requirements of real experimental power FC devices dedicated to transportation applications. Nevertheless, a scaling factor has been used here to adapt the power solicitations coming from a real electrical vehicle onto the nominal power

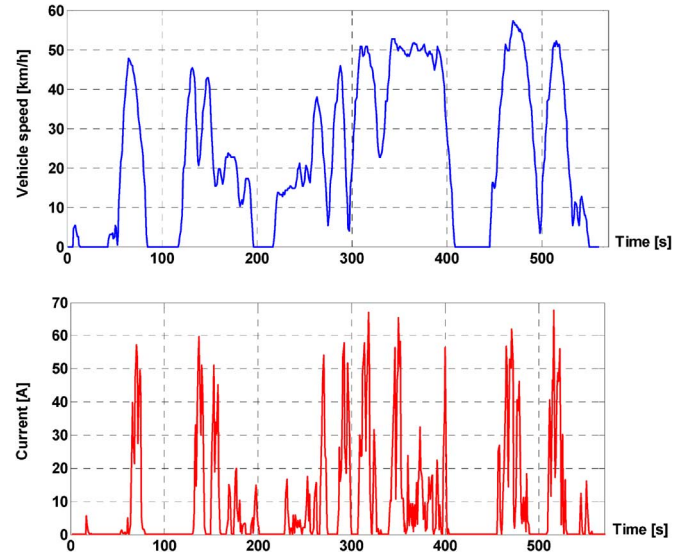


Fig. 1. Evolution of the current demand (derived from the vehicle real speed) on a cycle.

deliverable by these FC stacks. This allows a significant cost reduction of the experimental tests and will be discussed in detail in the next section.

B. Considered Ageing Tests

Two types of durability tests have been done (and considered in this paper) on these small power FC stacks.

The first testing conditions are the simplest ones. The stack (noted as FC1 hereinafter) is operated under nominal conditions, stationary conditions, and constant load current (50 A) during 1000 h. The FC is fed with dry hydrogen and humidified air (dew-point temperature of 25 °C), and the set of anode/cathode stoichiometries is fixed to 2/4, respectively. Of course, due to the anode stoichiometry, the stack is operated in an open mode: Both anode and cathode flows are controlled by flow regulators placed upstream from the stack.

The second test is performed on the same type of FC stack (noted as FC2 hereinafter). The only difference resides in the fact that this second stack is operated under dynamical load current. The current solicitation has been defined as follows. First, the power needed by the drive train of a light electric car (Citroën car of A-segment) running on a real transportation mission profile (time versus speed of vehicle, urban cycle called Hyzurb HURBANC [12]) was computed with VEHLIB software developed by the French National Institute for Transport and Safety Research. This simulation software can be presented as a collection of tools, which are developed in the Matlab/Simulink environment and allow the modeling and evaluation of electric and hybrid powertrains [13], [14]. Then, in a second step, the calculated power reference, which is representative of the light car operated on the selected kinematics, had to be adapted to the power capabilities of the investigated FC, and it was finally brought back to a load current cycle with convenient amplitudes for the FC stack (Fig. 1). This last operation was done here by means of a simple constant coefficient. The conversion of the electrical power profile that is needed by the car drive train (estimated at its dc bus level)

into an FC current profile has been made using the following assumptions. The power consumption of the FC ancillaries and the losses generated by the dc/dc converter placed between the stack and the dc bus were neglected in the calculation of the FC power from the power level at the dc bus. Moreover, the use of a simple constant coefficient to translate the FC power profile into a stack current reference means that the FC voltage variation versus aging time (not known *a priori* for a specified load current) was not considered.

The current dynamical load cycle is continuously applied during 1000 h to the FC stack. During the strongest power peaks corresponding to the vehicle acceleration phases, the load current reaches the value of 70 A. The average current over the whole cycle is equal to 12.5 A. In the case of such high dynamical mission profiles, the reactive gas supply has to be properly controlled. The rates of hydrogen and air flows feeding the FC are initially defined, owing to a particular gas-supply strategy: Two profiles for hydrogen and air flow references are first computed from the load current profile, considering the nominal 2/4 anode/cathode stoichiometry set.

C. Electrochemical Impedance Spectrometry

EIS is a method that is commonly used by electrochemists in order to obtain a better understanding of electrochemical device behavior. Impedance spectroscopy is not only a powerful technique that allows characterizing the stack in dynamical conditions, but it also allows giving information about FC static behavior (e.g., determination of membrane resistance as a function of gas hydrations). The study of the dynamical FC behavior is carried out considering a static operating point (in our case 35 A) and a small sinusoidal alternating part around it (an amplitude of 1 A and a frequency range from 10 MHz to 30 kHz). The real and imaginary parts of the FC impedance are calculated from the measured current and voltage alternating components. The EIS is done two times a week on the FC stack. During the measurements, the stack remains at its nominal operating conditions (in terms of temperature and stoichiometry rates). Moreover, the dew point of incoming air is kept at 25 °C.

Figs. 2 and 3 show the two sets of Nyquist diagrams (impedance plots) that are measured during the aging tests on the two considered low power FC stacks. As shown in these figures, the Nyquist diagram is drastically evolving during the aging tests. Moreover, the impedance spectrum is composed of several parts.

- 1) An inductive part, which is present at high frequencies (between 4 and 30 kHz). That plot can be associated with the pseudoinductance part due to all the metallic components of the complete tested FC stack.
- 2) A first capacitive arc (frequency range from 130 Hz to 4 kHz) that corresponds mainly to charge transfer phenomena (electrons and protons). A larger arc suggests a difficult transfer of the electrical charges in the double-layer capacities.
- 3) A second capacitive arc (for frequencies between 0.2 and 130 Hz) that is related to mass transport (ions in the gaseous phase). A larger arc diameter (estimated on the real axis) means that the diffusion of the species,

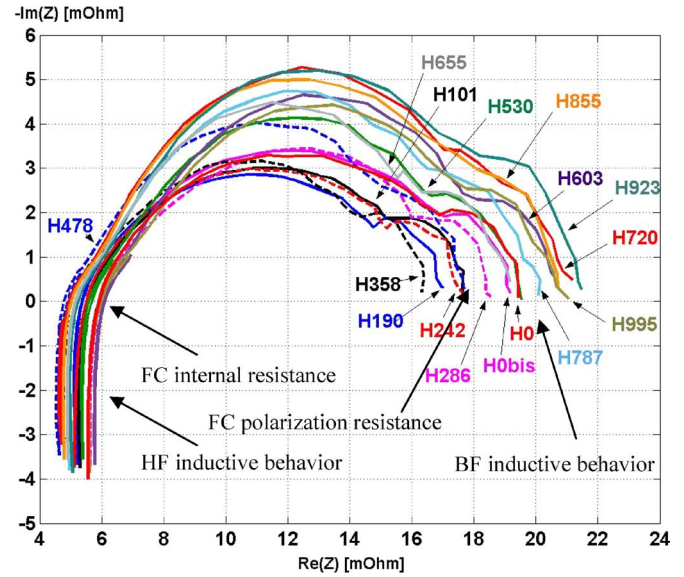


Fig. 2. Evolution versus time (in hours) of the impedance spectrum for the stack FC1.

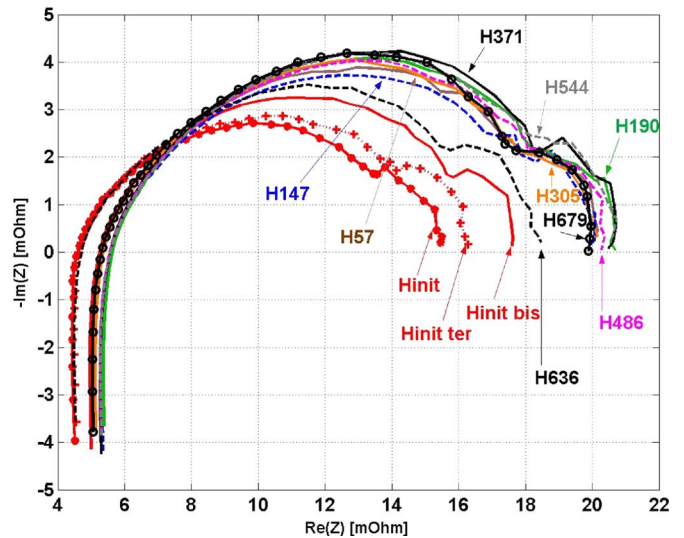


Fig. 3. Evolution versus time (in hours) of the impedance spectrum for the stack FC2.

from the FC plate channels to the electrodes, occurs with some problems. It can be due, for instance, to the insufficient reactive gas supply or to the accumulation of a too large water quantity in the gas diffusion layers (flooding phenomenon).

- 4) Another inductive part (for frequencies below 0.2 Hz) that is difficult to explain. Some authors related this low frequency inductive part to the relaxation of intermediate species in an electrochemical–chemical–electrochemical oxydoreduction reaction mechanism [15].

In addition, these impedance spectra enable the measurement of the FC stack internal resistances (for high frequency zero imaginary part of the Nyquist diagram) and the FC stack polarization resistances (for low frequency zero imaginary part of the Nyquist diagram). A higher internal resistance, due to the insufficient humidification of the membrane (drying

phenomenon), brings obviously worse FC performance. As the polarization resistance corresponds to the global sum of the various resistances (internal resistance, transfer resistance, etc.) that are linked with the different FC physical internal phenomena, a higher polarization resistance leads to a lower stack voltage.

The aim of this paper now is to define if some interesting information can be extracted from these impedance spectra to identify stack-performance degradation and, in this case, the cause of this degradation. The next part of this paper will, thus, focus on the recall of some rudiments of knowledge about system diagnosis and, particularly, fuzzy pattern recognition-based diagnosis methodology.

III. FUZZY PATTERN RECOGNITION

A. General Considerations on Fault Detection and Isolation

In any type of system, the basic concept of fault diagnosis consists of the following tasks [16]: fault detection and isolation (has a fault occurred within the system, and where has the fault occurred?) and fault analysis (what is the type of the fault?). In order to perform these tasks, the following steps are carried out: residual generation, i.e., the generation of signals which reflect the faults and the residual evaluation, i.e., the decision making as to type and time of occurrence of the fault. Depending on the method of residual generation, the methods of fault detection fall into one of three categories: signal-based, analytical model-based, and knowledge-based.

The signal-based fault detection is the most commonly used form. It consists of the setting of thresholds on signals or symptoms extracted from the system. Such a method has the advantage of simplicity. Nevertheless, the main drawbacks of this method are that many sensors are often needed and that a modification of the operating conditions may cause false alarms or even mask faults.

The analytical model-based methods can overcome some of the drawbacks of the signal-based methods, but they need an accurate and complete model of the system under consideration. However, such a model does not often exist, particularly in industrial applications.

In the case of modeling uncertainty, or the presence of vague or incomplete knowledge about the system, an alternative is required, which is not based upon the existence of an exact mathematical model of the system. Within this framework, fuzzy logic can propose an interesting alternative. In fact, no analytical model of the system is required; moreover, fuzzy logic offers the chance to combine heuristic knowledge with any model knowledge which may be available.

Fuzzy logic can be used, in the fault detection and isolation process, as a residual generator [17], [18] or as a residual-analysis tool [19]. An alternative to these two approaches is to have a one-step fault detection scheme based upon a fuzzy system; this last method has already been used to perform steady-state diagnosis on an FC system [7]. One of the most interesting and powerful residual-analysis tools is fuzzy clustering. The idea behind fuzzy clustering is basically the same as that of pattern recognition. Online, the degree (fuzziness) to which the

current data belongs to each of a set or predefined clusters is determined, and this results in a degree-of-membership [7] to each of the predetermined faults.

B. Residual Analysis via Fuzzy Clustering

Clustering is the allocation of data points to a certain number of classes. Each class is represented by a cluster center, which can be considered as the point that best represents the data points in the cluster. The idea behind fuzzy clustering is that each data point belongs to all classes with a certain degree of membership; this degree is dependent upon the distance to all cluster centers. For fault diagnosis, each class could correspond to a particular fault. Overviews of fuzzy clustering [20], [21] and applications in system diagnosis process have been presented in many papers [22], [23].

One fuzzy clustering algorithm is the fuzzy k -means algorithm [24]. The idea here is to divide the n different data (i.e., measures on the considered system) in K classes $\{C_1, \dots, C_K\}$. Each datum \mathbf{p}_j belongs to a class C_i with a membership degree $u_{i,j} \in [0, \dots, 1]$. Thus, a partition \mathbf{U} can be created

$$\mathbf{U} = \begin{bmatrix} u_{1,1} & u_{1,2} & \cdots & u_{1,n} \\ u_{2,1} & u_{2,2} & \cdots & u_{2,n} \\ \vdots & \vdots & \ddots & \vdots \\ u_{K,1} & u_{K,2} & \cdots & u_{K,n} \end{bmatrix} \quad (1)$$

which is subject to

$$\sum_{i=1}^K u_{i,j} = 1 \quad \forall j \in \{1, \dots, n\} \quad (2)$$

and to

$$\sum_{j=1}^n u_{i,j} > 0 \quad \forall i \in \{1, \dots, K\}. \quad (3)$$

Relation (2) underlines the fact that the sum of membership degrees to the K classes for one data must be equal to 1. Relation (3) implies that each class must have at least one element.

The cluster centers $\mathbf{W} = [\mathbf{w}_1, \dots, \mathbf{w}_K]^t$ are determined so that they minimize the following convergence criterion $J_m(\mathbf{U}, \mathbf{W})$:

$$J_m(\mathbf{U}, \mathbf{W}) = \sum_{j=1}^n \sum_{i=1}^K (u_{i,j})^m \cdot \|\mathbf{p}_j - \mathbf{w}_i\|^2 \quad (4)$$

where $m > 1$ is an exponent that fixes the degree of fuzziness of the algorithm (typically $m = 2$).

The whole algorithm can be written as follows.

- 1) Initialize the cluster-center matrix $\mathbf{W}(0)$ by randomly choosing K data among the n available data.
- 2) Fix the parameter m value and the value of a threshold ε . The algorithm will be stopped when the maximum

number of iterations (iter_{\max}) is reached or when the maximal modification (between two iterations) on an element in matrix \mathbf{W} remains below ε value.

3) Evaluate the initial fuzzy partition $\mathbf{U}(0)$ owing to

$$u_{i,j}(0) = \frac{\left(\frac{1}{\|\mathbf{p}_j - \mathbf{w}_i\|}\right)^{\frac{2}{m-1}}}{\sum_{k=1}^K \left(\frac{1}{\|\mathbf{p}_j - \mathbf{w}_k\|}\right)^{\frac{2}{m-1}}},$$

$$i \in \{1, \dots, K\} \quad j \in \{1, \dots, n\}. \quad (5)$$

4) $\text{iter} = 1$.

5) Repeat.

5.1 Evaluate the new cluster-center matrix $\mathbf{W}(\text{iter})$ owing to

$$\mathbf{w}_i = \frac{\sum_{j=1}^n (u_{i,j})^m \cdot \mathbf{p}_j}{\sum_{j=1}^n (u_{i,j})^m}, \quad i \in \{1, \dots, K\}. \quad (6)$$

5.2 Evaluate the new fuzzy partition $\mathbf{U}(\text{iter})$ according to (5).

5.3 $\text{iter} = \text{iter} + 1$.

6) Until $\max_{i,j} |u_{i,j}(\text{iter}) - u_{i,j}(\text{iter} - 1)| > \varepsilon$, and $\text{iter} \leq \text{iter}_{\max}$.

In the next part of this paper, this fuzzy clustering algorithm will be implemented in the Matlab environment. It will then be used to detect both the occurrence of a behavioral degradation (i.e., a drop of the nominal polarization curve) on the FC stack and the cause of this degradation.

IV. EXPERIMENTAL RESULTS

A. Relevant Parameters on the Impedance Spectra

Considering Figs. 2 and 3, the first step is to define, on these impedance spectra, the most relevant parameters characterizing the aging of the FC stacks. In our case, only two parameters have been considered.

- 1) First, the difference between polarization resistance and internal resistance; what is, in fact, the width of the impedance spectrum considering the real part of the impedance? This parameter is subject to important variations in case of degradation resulting from a loss of mass transfer rate of reactants inside the FC stack [25].
- 2) Second, the maximal absolute phase value of the Nyquist plot. This value is very simple to measure in an experimental way, yielding to the maximal phase displacement between the FC current and voltage. This value is also subject to important variations in case of degradation of the electrolyte membrane. Indeed, this maximal absolute phase value is generally measured on the first capacitive arc, relating to charge transfer (electrons and protons) at reaction interfaces and across the membrane.

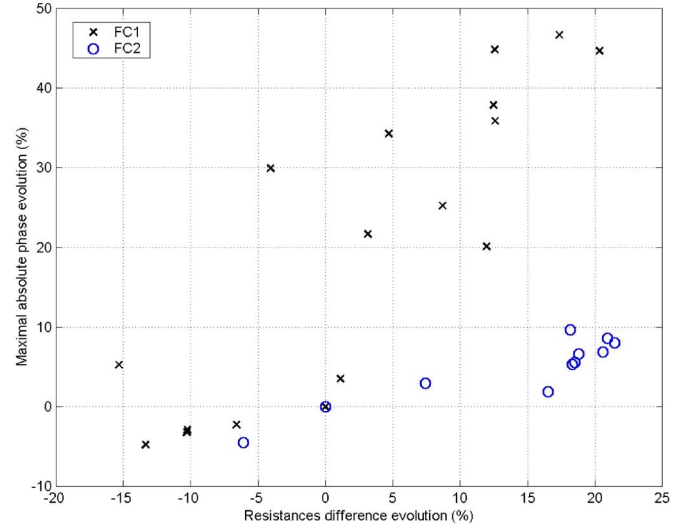


Fig. 4. Time evolutions of the two considered parameters (difference between polarization and internal resistances and maximal absolute phase value of Nyquist plot) for the two FC stacks.

B. Application of the Fuzzy Clustering Algorithm to Diagnose FC Stack State

Time evolutions of the two parameters used for the diagnosis process and already presented in the last paragraph are considered in terms of percentages versus their initial values. Thus, Fig. 4 shows the experimental evolutions of the two considered parameters (difference between polarization and internal resistances, maximal absolute phase value) obtained for the two FC stacks FC1 and FC2. As it is shown in this figure, the evolution percentages of the two considered parameters are very important during the 1000 h of experimental tests that have been carried out. This point underlines the fact that the choice of these two parameters is obvious in characterizing the evolutions of the two FC stacks. Moreover, under constant current solicitation, the maximal absolute phase of the Nyquist diagram varies with time in a much larger area (about 40% to 50%) than in the case of a @dynamical current solicitation (only about 10% variation). Considering the time evolution of the difference between polarization and internal resistances, this evolution is important and of the same order (from -15% to $+20\%$) for the two FC stacks.

The fuzzy clustering algorithm is now applied on the obtained experimental measured points to produce three different clusters: The first one should be dedicated to the characterization of the “rated” behavior of the considered FC stack (FC1 or FC2), the second one should be dedicated to the characterization of the typical degradation of the first stack (under static current solicitations), and the third one should be dedicated to the characterization of the typical degradation of the second FC stack (under dynamical current solicitations). The maximum number of iterations here is $\text{iter}_{\max} = 300$, the value of the fuzziness parameter m is fixed to 2, and the threshold ε is fixed to 10^{-6} . Fig. 5 shows the obtained results.

In Fig. 5, three different clusters (the center of each cluster is marked by a square) are shown; each one is related to a specific behavior of the FC stack (rated behavior, static current solicitation evolution, and @dynamic current solicitation evolution).

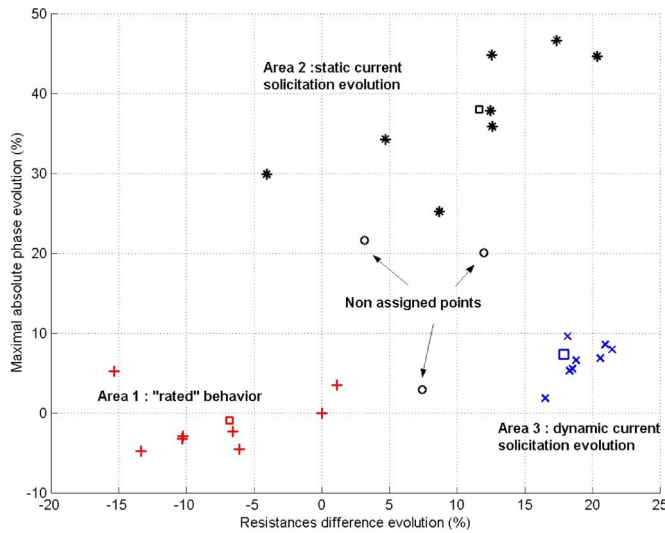


Fig. 5. Fuzzy clustering algorithm applied on the experimental data.

Moreover, three experimental data have not been assigned to a specific cluster; in fact, their membership degrees never exceed to 0.6 (whatever cluster is considered); this numerical value has been chosen here as a threshold to define the belonging of a point to a specific cluster. Clearly, these nonassigned points correspond to transient behavior of the considered FC stack versus durability. The state of the stack is going from a cluster (i.e., a kind of behavior) to a new cluster (i.e., a new kind of behavior). Of course, the fourth cluster could also be added, referring to this transient behavior. Nevertheless, the authors suggest limiting the number of clusters to three; each of these three clusters relates to one of the “steady states” of the considered FC stacks (rated behavior at the beginning of the experiments, state after 1000 h of static current solicitation, and state after 1000 h of dynamic current solicitation). Moreover, the name of the clusters could also have been changed into “young” (for the cluster relating to the rated behavior), “middle aged” for the transient behavior (nonassigned points), and “old” (for the two last clusters obtained after 1000 h of ageing test) if the “age” of the stack is wanted as an output of the diagnosis process.

C. Results and Discussion

As presented in the last paragraph, the proposed methodology is able to detect, on a PEFC stack, a durability performance deviation from “rated” performances and to detect what “kind of life” the FC stack has had before this degradation occurs. Moreover, the fuzzy nature of the clustering algorithm enables two main important things. First, minor variations around the rated behavior are not considered as real degradations; these deviations can occur due to small variations in experimental conditions (flows, temperature, load, etc.). Second, the algorithm also shows that some experimental points cannot be assigned to a particular pattern. Clearly, those experimental points are reflecting the fact that the FC stack behavior is under evolution, going from a first pattern (i.e., a first state) to another one (i.e., another state).

The proposed diagnosis methodology is, thus, able to detect a deviation of the operating point from the rated performances and to propose explanations about the cause of this performance degradation. Then, the obtained results could be exploited in two ways. First, it is possible to perform better understanding of degradation mechanisms of the FC stacks and, thus, increase their performances while acting on their design. Second, it is also possible to use these results in a real-time control (as no computational time-consuming FC model is required) of the FC powertrain, modifying the current requested from the FC stack in order to increase its durability.

V. CONCLUSION

FCs are nowadays considered as interesting alternatives for embedded power generation on next-generation vehicles. Nevertheless, before seeing those vehicles in the market, many technological bolts are still to be overcome. Among these bolts, durability and reliability of FC power generators are addressing a particular interest around the world. Moreover, when talking about durability and reliability, efficient real-time diagnosis of the FC stack and system appears to be a major issue in order to be able to propose a control law that is intended to prevent permanent deterioration of the embedded FC power generator.

This paper presents experimental durability results on the small power low-temperature FC stacks. Two types of aging constraints for the FC stack have been explored in this paper: under steady-state current and under a real transportation load current cycle. The behaviors of the two stacks have been studied for a 1000-h duration time, and degradations have been evaluated, owing to the EIS on the Nyquist evolution plots. Two values have then been extracted from these Nyquist plots: the difference between polarization resistance and internal resistance of the considered FC stack and the maximal absolute phase value of the Nyquist plot for the considered FC stack. Evolution over time of these two values in a 2-D space is then considered for diagnostic purposes. Finally, fuzzy-clustering methodology is applied to identify clusters in this 2-D space; each of them relates to a specific behavior or aging process of the FC stack. This methodology appears very promising for the diagnosis of FC stacks, which is mainly due to the fact that efficient models of FC stacks, including durability considerations, are, at the present time, still not available.

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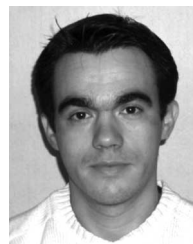
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