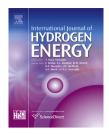


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PEMFC stack voltage singularity measurement and fault classification



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ABSTRACT

The study summarized in this paper deals with non-intrusive fault diagnosis of Polymer Electrolyte Membrane Fuel Cell (PEMFC) stack. In the proposed approach, the diagnosis operation is based on the stack voltage singularity measurement and classification. To this aim, wavelet transform-based multifractal formalism, named WTMM (Wavelet Transform Modulus Maxima), and pattern recognition methods are combined to realize the identification of the PEMFC faults. The proposed method takes advantage of the non-linearities associated with discontinuities introduced in the dynamic response data resulting from various failure modes. Indeed, the singularities signature of poor operating conditions (faults) of the PEMFC is revealed through the computing of multifractal spectra. The obtained good classification rates demonstrate that the multifractal spectrum based on WTMM is effective to extract the incipient fault features during the PEMFC operation. The proposed method leads to a promising non-intrusive and low cost diagnostic tool to achieve on-line characterizations of dynamical FC behaviors.

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Introduction

One of the main issues to be treated in Fuel Cell (FC) development remains the detection and identification of disturbances occurring during operation. Various internal and external factors are known to affect the useful life of PEMFCs. When any FC operational parameters (e.g. gas pressures and stoichiometry rates) are deflected from their nominal values,

the stack performance can be strongly impacted. Stack and/or cell voltage changes can reveal some damage phenomena and failure modes such as electrode flooding, membrane drying, gas leakage, loss of electroactive surface area. In this context, the proposal of diagnostic tools aims to provide potential solutions for improving stack durability, reliability, efficiency and for optimizing system development. Recent literatures have provided several diagnostic methods that are either based on physical models or data-driven approaches [1–5].

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Model-based diagnosis methods include for instance the so-called "equivalent circuits" models [6] operating with electrochemical characterization measures or voltage—current data, analytical models [7,8] which require an exhaustive knowledge on the multi-physical mechanisms (thermal, electrical, electrochemical and fluidic) and the numerous parameters that govern the process of FC operation. But, the non-linear nature of these phenomena, the reversibility or irreversibility nature of the damage, and the interactions between the various components of the FC system make really difficult the task of modeling.

Instead, the data-driven diagnosis approaches, without any sophisticated modeling process, have emerged in recent times and attracted the interest of researchers in the field of energy converters diagnostic. Thus, numerous publications have proposed signal processing based methods as diagnostic FC tools: Fourier transform [9], wavelets [10], wavelet transform based multifractal formalism [11], etc.

Many natural and artificial phenomena such as turbulence, diffusion limited aggregates, and electrical discharges exhibit complex structures. Since these phenomena are highly nonlinear and non-stationary, regular analyses such as Fourier decomposition cannot characterize them effectively. To encounter this limit, multifractal analysis was introduced in the last years [12] and showed its relevance in various fields such as diagnosis in medicine [13], in biology [14], and in finance [15]. So, multifractal analysis describes the scaling behavior of a distribution of measures in a geometrical and statistical mode. It consists in computing the set of punctual singularity strengths of a signal that can be represented in form of a concave curve (singularities spectrum). In mathematics, a singularity is a point at which the function (or signal) fails to be well-behaved (becomes degenerate) in some particular way, such as the non-differentiability and the discontinuity.

Our study is launched to develop some tools for FC dysfunction diagnostics, which require minimum of instrumentation (non-intrusive diagnostic tools). In fact, our method uses the information contained in the singularities of the stack voltage signal which reflect the state-of-health of the FC in a dimensionally reduced representation. That consists in quantifying the irregularities (or singularities) degree of the voltage according to the introduced operating faults. For this purpose, continuous wavelets and multifractal formalism are first used to compute the singularity spectrum of the voltage signal. Then, the spectrum is classified using pattern recognition methods, named Support Vectors Machine (SVM) and K-Nearest Neighborhoods (KNN) methods. More details about

the experimentation step, the applied methods and the obtained results are given in Sections materials and methods and results and discussion, respectively.

Materials and methods

Experimental process

The experimented FC is an 8 cell PEMFC stack manufactured by CEA LITEN in France. It has been designed for automotive applications. It is made of metallic gas distributor plates. The electrode active surface is 220 cm². The nominal current I is 110 A and the nominal current density is 0.5 A cm⁻².

The PEMFC stack was operated under a variety of conditions using the 'in-house' self-developed 1 kW test bench of the FC test platform in Belfort - France. More descriptions of the test bench are given in details in Ref. [16].

It is well known that FCs are complex systems with many different properties and processes influencing their operation. Therefore the identification of an error in operation proves to be quite complex. To avoid this difficulty, we have proposed an experimental procedure that makes the faults recognition easy. Indeed, we introduced some faults associated with different parameters (cathode stoichiometry rate 'FSC', anode stoichiometry rate 'FSA', gas pressure 'P', cooling circuit temperature 'T', and carbon monoxide 'CO' poisoning at the anode side) with changes from the nominal operating values. During each PEMFC fault, the singularity spectrum was computed on the produced voltage stack. More details on the set of faults scenarios proposed in this work are summarized in Table 1. Note also that a reconditioning procedure of the FC was used between each fault test: the FC was operated in nominal operating conditions ('Ref'state) during about 15 min with the aim to recover the nominal performances.

The fault scenarios considered in this work have been selected in the framework of the DIAPASON 2 project supported by the French National Research Agency (ANR). The scenarios duplicate various failures which are representative of the application environment (electric vehicle or stationary generator). The faults considered correspond to potential ineffective operations of the FC system ancillaries/actuators (e.g. failure of the air supply subsystem in the FC generator). The faulty mode limits have been specified with the help of the PEMFC manufacturer (CEA LITEN of Grenoble — France). The trade-offs concerning the parameter ranges explored, the selected parameter levels have been found by considering the capabilities of the FC in terms of performance (a total FC collapse or severe FC

Parameters value	Nominal conditions (Ref)	Cathode flow fault (DFSC)	Anode flow fault (DFSA)	Inlet gas pressures fault (DP)	Cooling circuit temperature fault (DT)	CO Poisoning (DCO)
FSC	2	1.3	2	2	2	2
FSA	1.5	1.5	1.3	1.5	1.5	1.5
P (bars)	1.5	1.5	1.5	1.3	1.5	1.5
T (°C)	80	80	80	80	75	80
Presence of CO (ppm)	0	0	0	0	0	10

degradation had to be avoided), by taking into account the technological limitations of the FC test bench, and the possible duration of the experimental campaign as well.

Singularity spectrum calculation

The singularity spectrum is computed from the FC voltage signal [11]. This one is associated with the Hölder exponent 'h' (parameter used to quantify a singularity degree of a point) and the Hausdorff dimension 'D(h)' (dimension of set of points, named 'IsoHölder' that exhibits the same value of h) [17,11].

In our application, the singularity spectrum D(h) gives us some information about the behavior of the stack through the degeneration of the FC voltage signal as a function of different operating conditions. Each FC fault (e.g. the FSC fault) imposes its own stamp on the morphology of the stack voltage.

One of the powerful properties of the Wavelet Transform (WT) is that the strength of the singularities points in the signal can be detected and measured at multiple scales through the local maxima coefficients (WTMM) [18,19]. The other fundamental advantage of using the WT is that the skeleton defined by the WTMM provides an adaptive space-scale partitioning from which the D(h) singular spectrum can be extracted. It is well adapted to reveal the hierarchy that governs the spatial distribution of singularities of multifractal measures.

This specification of the WTMM is related to the partition function defined by the statistical moments of $q \in \mathbb{R}$ order given by (1):

$$Z(q,s) = \sum_{l \in I(s)} \left(\sup_{(u,s') \in I, s' \le s} \left| W_{\psi}(X)(u,s') \right|^q \right) \tag{1}$$

where:

- $W_{\psi}(X)$ is the continuous wavelet transform of the signal X, using the mother wavelet of type ψ at a location u and a scale s:

$$\forall u \in \mathbb{R}, \forall s > 0, \ W_{\psi}(X)(u,s) = \frac{1}{\sqrt{s}} \int_{\mathbb{R}} X\psi\left(\frac{x-u}{s}\right) dx \tag{2}$$

- l is a maxima line obtained after linking different wavelet transform maxima points detected at different scales for position u.
- L(s) is the set of maxima lines that exists at the scale s.

The applied wavelet must satisfy the regularity criterion which is related to the number of vanishing moments defined by (3).

$$\forall k = 0, \ 1 \cdots N - 1: \quad \int\limits_R x^k \psi(x) dx = 0 \tag{3}$$

where:

 ψ is a wavelet with N vanishing moments. The best wavelet examples are the derivatives of the Gaussian. In our study, the Mexican hat wavelet (2nd derivative of the Gaussian) is applied and $0 \le h \le N$.

The major implementation steps of the multifractal spectrum computation based on the WTMM method can be formulated as follows [11]:

- Calculate the Wavelet Transform (WT) of the signal at multiple scales.
- ii. Find the local maxima of WTs in each scale.
- iii. Chain the wavelet maxima across scales.
- iv. Compute the partition functions Z(q, s) using the Formula (1).
- v. Compute $\tau(q)$ with a linear regression of $\log_2 Z(q,s)$ as a function of $\log_2 s$:

$$\log_2 Z(q,s) \approx \tau(q) \log_2 s + K(q)$$
(4)

and the Hölder exponent :
$$h = \frac{d\tau(q)}{dq}$$
 (5)

vi. The singularity spectrum D(h) is estimated using the Legendre transform [11]:

$$D(h) = \inf_{q \in \mathbb{R}} (qh - \tau(q) + c) \tag{6}$$

where c is a constant.

The multifractal measures given by (6) describe the behavior of the objects not only at different scales s, but also for different statistical moments q. The parameter q serves as a "microscope" for exploring different regions of the singular measurement:

- For values q > 0, the strongly singular measures are enhanced.
- For values q < 0, the less singular areas are emphasized.

More explanations and details about the multifractal algorithm and an illustration of the different steps for calculating the singularity spectrum based on continuous wavelets transform are given in Ref. [11].

Feature selection and classification

In the case of the proposed diagnostic strategy, the singularity spectrum of the stack voltages is simply constituted as a vector whose dimension equals the number of 'h' and 'D(h)' values.

To avoid the pattern recognition problems called curse of dimensionality [20] which can be caused by too large dimension data, it is necessary to select only the pertinent features to be classified. The feature selection operation allows not only the reducing of the computational complexity, but also improves the performance of the classification method.

Various techniques have been developed for selecting the pertinent features to ensure a good discrimination of data for various application areas. Actually, there are two general approaches: filters and wrappers [21,22]. In the case of the filter type methods, features are selected based on the intrinsic characteristics which determine their relevance or discriminant powers with regard to the target classes. On the other hand, in wrapper type methods, feature selection is

"wrapped" around a learning method: the usefulness of a feature is directly judged by the estimated accuracy of the learning method. The latter requires expensive computation to search the best feature.

Minimum Redundancy — Maximum Relevance (MRMR) feature selection technique was recently introduced for gene selection application [23]. This method selects the features according to their relevance to the concerned target and the redundancy among the features themselves. Both the relevance and redundancy are quantified by estimating the following Mutual Information (MI):

$$I(x,y) = \sum_{i,j} p(x_i, y_j) \log \frac{p(x_i, y_j)}{p(x_i) p(y_j)}$$
(7)

where x and y are two variables, p(x, y) their joint probabilistic distribution and p(x), p(y) their respective marginal probabilities.

MI is defined to estimate how one vector is related to another and to measure the level of "similarity" between variables or features.

The Minimum Redundancy condition consists in selecting the features that are mutually maximally dissimilar. It can be calculated as follow:

min
$$A_{I}$$
, $A_{I} = \frac{1}{N^{2}} \sum_{i,j \in S} I(x_{i}, y_{j})$ (8)

where N is the number of features in S.

The Maximum Relevancy condition allows measuring the level of discriminant powers of features that are attributed for different target classes $C = \{C_1, C_2, ..., C_k\}$. It can be expressed as follow:

max
$$B_I$$
, $B_I = \frac{1}{N} \sum_{i=S} I(C, x_i)$ (9)

Thus, the MRMR feature set is obtained by optimizing the conditions given by Eqs. (8) and (9). The different possibilities to make the single criterion function can be formulated as follow:

$$\max(B_I - A_I) \tag{10}$$

$$\max(B_I/A_I) \tag{11}$$

Formula (10) expresses the so-called Mutual Information Difference "MID" and Formula (11) defines the Mutual Information Quotient "MIQ".

The MRMR selected singularity features are then classified using the pattern recognition methods named Support Vector Machines 'SVM' and K-Nearest Neighbors 'KNN'.

SVM method has shown its effectiveness in the fault diagnosis domain in recent years [24]. This method embodies many important characteristics, such as: good generalization performance, the absence of local minima and the sparse representation of solution. It is based on kernel function for projecting the original input space into highly dimensional one, making possible the linear separation of training samples. The data discrimination is achieved by drawing an optimal hyperplane that defines a boundary that maximizes the margin between data samples in two classes. So, standard

SVM is a binary classifier (2 classes). For multi-class situations, a multi-class classifier can be constructed using binary classifiers, such as one-against-others or one-against-all [25], and other approaches where the multi-class classifier is constructed directly [26]. In this paper, we used SVM and Kernel Methods Matlab Toolbox [27].

The KNN method is based on the metric measure between the query sample and the training samples. There are various metrics for measuring the nearness between two samples, such as: Euclidean distance metric, Hamming distance and Mahalanobis one, correlation distance metric, etc. [28]. According to the KNN rule (K is the number of chosen neighbors), a query sample should be assigned to the same class of the sample in the training set that is nearest. This method can be costly in terms of time and memory when the training set contains many high dimensional samples.

Results and discussion

Diagnosis results

In this work, we adopt the multifractal analysis of voltage signal in order to extract and identify the singularity signature ("average" singularity spectrum) of defaults related to the imposed poor operating conditions. The computing of the "average" singularity spectrum is based on 10 voltage profiles acquired for each operating scenario. Each voltage profile covers 3000 points acquired at a frequency close to 11 Hz (using the monitoring data system of the FC experimental test bench). More details on the computing of the "average" singularity spectrum can be found in Ref. [11].

The diagram of Fig. 1 gives an overview of the different steps applied in the proposed diagnosis methodology.

Some examples of stack voltage recorded for different operating conditions and their multifractal spectra are plotted in Figs. 2 and 3. As we can see at first sight, each operating default gives its own stamp on the morphology of the stack voltage. The singularity signature of each fault is revealed by the distinguished singularity spectra. Therefore, the singularity parameters can be used as discriminant parameters to develop a PEMFC non-intrusive diagnostic tool.

After running the MRMR programs [29], we obtain the optimal features that facilitate the detection of the fault operating conditions of the PEMFC. Both MID and MIQ selecting techniques have been used and compared to rank the importance of the 100 singularities strengths distributed in the voltage signal associated to 100 Hausdorff dimensions which quantify these singularities. The optimal features set selected by both MID and MIQ techniques contains 70% of Hölder exponent 'h' features and 30% of Hausdorff dimension ones. Besides, 90% of these selected features are extracted from the left side of the singularity spectrum (Fig. 4). Therefore, we can conclude that the punctual singularity strengths of the stack voltage reveal very useful informations on the physical and electrochemical phenomena inside the FC.

On the one hand, the combination of the SVM classifier with MID feature selection technique offers 84.29% of good classification rate with 15 selecting features. 86.96% of good classification rate was obtained using the same classifier coupled with

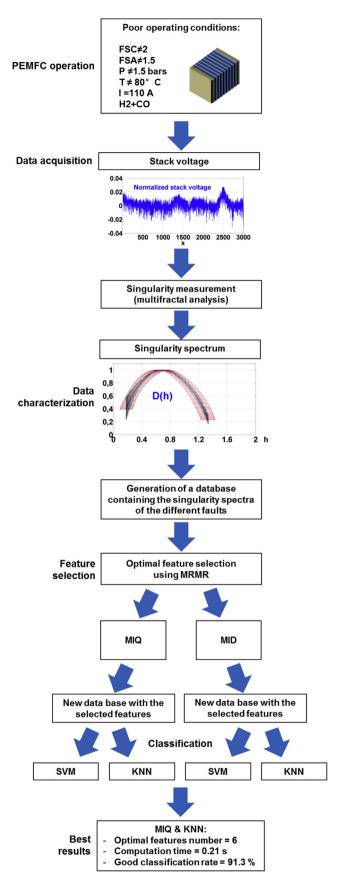


Fig. 1 – Synoptic diagram showing the different steps of the proposed PEMFC diagnosis strategy.

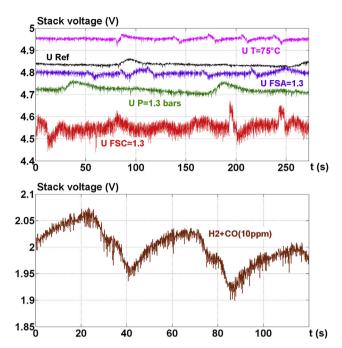


Fig. 2 — Stack voltages vs. time for various faults, i.e. operating conditions (Ref = nominal operating conditions). Note that the stack voltage evolution displayed for the CO fault was recorded 30 min after the beginning of the fuel poisoning.

MIQ technique and 6 optimal features. On the other hand, the KNN classifier associated to MIQ technique gives 91.3% of good classification rate against 86.96% when coupled with MID technique, and with 6 selected features. Therefore, the optimal features selected by using MIQ technique offer the best discrimination between the 6 classes described previously.

The results were analyzed according to the global classification error (in %) and using the confusion matrices between the true and the estimated classes for all the observations in the database. The results obtained with the MIQ selecting technique and the adopted supervised classification methods are reported in the confusion matrices displayed in Tables 2 and 3.

A confusion matrix shows how the predictions are made by the classification model or mtehod [30,31]. The rows

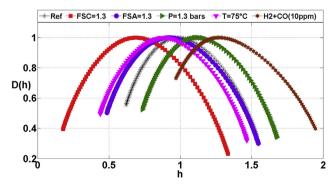


Fig. 3 – Average singularity spectra related with the different faults considered in Fig. 2.

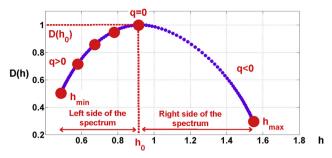


Fig. 4 — Examples of optimal features (red circles) selected using the MIQ technique. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

correspond to the known class of the data, i.e. the labels in the data (true classes). The columns correspond to the predictions made by the model (predicted classes). The value of each of element in the matrix is the number of predictions made with the class corresponding to the column for examples with the correct value as represented by the row. Thus, the diagonal elements show the number of correct classifications made for each class, and the off-diagonal elements show the errors made. The accuracy is the overall correctness of the model and it is calculated as the sum of the correct classifications divided by the total number of classifications.

So, as it is shown by the good classification rates results displayed in Tables 2 and 3, four failures of cathode flow, gas pressures, cooling circuit and CO poisoning are successfully identified (classification rate = 100%) by both KNN and SVM classifiers. However, 25% of the anode flow failure situations are confused with the optimal operation of the FC, and vice versa. This can be explained by the fact that the gap value

Table 2 — Confusion matrix of the obtained good classification rates using MIQ feature selection technique and KNN classifier.

Class	Ref	DFSC	DFSA	DΡ	DT	DCO
Ref	75	0	25	0	0	0
DFSC	0	100	0	0	0	0
DFSA	25	0	75	0	0	0
DP	0	0	0	100	0	0
DT	0	0	0	0	100	0
DCO	0	0	0	0	0	100

Table 3 — Confusion matrix of the obtained good classification rates using MIQ feature selection technique and SVM classifier.

Class	Ref	DFSC	DFSA	ΩP	DT	DCO
Ref	50	0	50	0	0	0
DFSC	0	100	0	0	0	0
DFSA	25	0	75	0	0	0
DP	0	0	0	100	0	0
DT	0	0	0	0	100	0
DCO	0	0	0	0	0	100

between the nominal value of the anode stoichiometry rate (FSA = 1.5) and the considered fault situation with FSA = 1.3 is small.

Computation costs

The proposed non-intrusive diagnostic method offers an interesting computational performance for implementation in real-time. In fact, the computing time of a singularity spectrum is estimated at about 7 s for a voltage profile covering 3000 time points under computer system characteristics of: Intel® Core™ i7-2960 XM, CPU @ 2.7 GHz, RAM:16 GB. The feature selection stage offers the possibility to reduce the computation cost of the characterization step to 0.21 s because only 6 optimal singularity features are considered (instead of 200 features for a complete singularity spectrum). The additional computing time for the classification step with KNN method is about 4 ms only.

Conclusion

The work uses the relevant information describing the regularity of the stack voltage to develop a new online and nonintrusive diagnostic tool for PEMFC without additional instrumentation. The proposed approach consists in combining MRMR feature selection technique (MID and MIQ) and pattern recognition methods (SVM and KNN). The results of singularity spectra based on continuous wavelets transform show that each fault imposes its own singularity signature on the produced voltage. The best good classification rate of the faults (obtained with MRMR MIQ and KNN) is close to 91% and the computation time makes possible the on-line implementations of the diagnostic tool in embedded FC generators. These results encourage us to further develop this study by considering some others faults or a combination of faults (by considering two or more failures simultaneously during the FC operation).

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Nomenclature

$\psi(x)$	wavelet function
$W_{\psi}(X)$	wavelet transform of the signal X
$\tau(q)$	scaling function
A_I	minimum redundancy condition
B_{I}	maximum relevance condition
С	constant
C_i	i th class
CO	carbone monoxide
D(h)	singularity (or multifractal) spectrum
FSA	anode stoichiometry factor
FSC	cathode stoichiometry factor
100	cathode stolemonietry factor

h Hölder exponent h(x) Hölder function

 h_0 Hölder exponent for q=0 h_{\max} Hölder exponent for q=+2 h_{\min} Hölder exponent for q=+2I (x,y) mutual information (MI) K number of nearest neighbors

l wavelet transform maxima chained to form a line

L(s) set of maxima lines that exists at scale s

MID mutual information difference MIQ mutual information quotient

MRMR minimum redundancy - maximum relevance

N number of features
P inlet gas pressures, bars

p(x), p(y) marginal probabilities of x and y

p(x,y) joint probabilistic distribution of two variables x and y

q order of statistical moments

s scale

T cooling circuit temperature

u position

Z(q,s) partition function

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