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THE DYNAMIC MULTIFRACTALITY IN PEMFC STACK VOLTAGE SIGNAL AS A TOOL FOR THE AGING MONITORING

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Abstract

Fuel Cell aging monitoring and diagnosis are key-issues for scientists and industrials who intend to spread this technology. In the present work, we propose an efficient method that enables the extraction of valuable information which contains indicators on the aging of a studied PEM Fuel Cell (PEMFC). In this context, we investigate the possibilities offered by the Wavelet Leaders based Multifractal Analysis (WLMA), which is a suitable method to study the clustering variability of non-stationary fluctuating signals. A continuous aging test of 124 hours conducted under nominal operating conditions serves as data input for the development of our diagnosis method. Singularity spectra (SS) are calculated on the stack voltage signals at different periods of time. We observe that stack voltage over extended periods of operating time leads to different types of singularities, i.e., we find a narrow range of values of Hölder exponent h with non-zero fractal dimension $D(h)$ according to the operating time of the FC. We conclude that this dynamic multifractality of the stack voltage can be considered as a pertinent tool for the monitoring of the FC aging state.

Keywords: Fuel Cells; PEMFC; Aging monitoring; Singularity analysis.

1. Introduction

Fuel cells (FC) will play an important role in future sustainable cities. Because of their high efficiency rates, zero or low-emissions, silent operation, high power density and flexible operating range, low-temperature polymer electrolyte membrane fuel cells (PEMFC) are suitable for vehicle applications and for combined heat and power stationary energy systems [1]. A decisive factor for the widespread marketing of PEMFCs is the extension of lifetimes and reliability rates. Actually, researchers and industrials work together in order to reach the following FC lifetime targets: at least 5000 operating hours for transport applications, whereas the actual lifetime can be limited for vehicles operating under real-world conditions, and up to 40000 hours for stationary applications [2]. To achieve these objectives, the FC state-of-health monitoring constitutes an important challenge in this area. It is expected that the level of requirements for the safety of the FC will increase with a systematic automation control, fault diagnosis and monitoring process able to optimize the FC performance and durability.

Despite the high cost of both FC stack and continuous aging life tests, various research teams could successfully perform some experiments and analyses in order to better understand the degradation mechanisms related with the aging phenomena [3, 4]. Some of the works conducted in this area led to the development of numerical models, which enable to understand the interactions between different electrochemical mechanisms and the FC aging degradation phenomena [5-7].

The Electrochemical Impedance Spectrum (EIS) technique has been widely applied for the characterization and analysis of the FC degradation related to the aging phenomena [8]. This technique is efficient in the characterization of FC, to gain a deeper understanding of the FC aging and to develop the associated knowledge models. However, the usual EIS technique shows some limitations for real-time diagnosis applications. In particular, additional, complex and expensive equipment (i.e. electrochemical impedance spectrometer) is generally required.

In this context of diagnosis method dedicated to FC survey and control-command, we choose to consider only the “free” evolution of the FC voltage at the stack terminals (i.e. without introducing any additional load current perturbation or solicitation as it is the case for EIS measurement), and to base our diagnosis on the study of the morphology of this one and only signal (the individual cell voltages of the stack are not considered). In our previous works, we demonstrated the multifractality of the PEMFC voltage signal under various operating conditions and the effectiveness of this technique as a diagnosis tool was highlighted [9, 10]. This technique named singularity analysis is based on the use of wavelet leaders and multifractal formalism. The goal of this present work is to demonstrate the interest of studying the local regularity of the FC stack voltage signal in order to research and extract some relevant indicators of the FC aging.

2. Experimental and materials

2.1. Investigated PEMFC description

The experimented FC is an 8 cell stack. It was designed for automotive applications and manufactured by CEA (Alternative Energies and Atomic Energy Commission) in the framework of the French National Research Agency “ANR” DIAPASON2 Project. The FC is made of metallic gas distributor plates. The electrode active surface of a cell is equal to 220 cm². A summary of the FC nominal operating conditions is given in Table 1. During the project, this stack was used during a long test campaign including various fault modes (as starvation sequences, short-circuits, CO poisoning), and the final aging test described in the next subsection.

Table 1. The PEMFC nominal operating conditions.

Coolant flow: deionized water	2 l/min
Anode stoichiometry rate (H ₂)	1.5
Cathode stoichiometry rate (air)	2
Absolute pressure for H ₂ and air inlets	150 kPa
Maximum differential pressure between anode and cathode	30 kPa
Temperature of the cooling circuit	80°C
Anode and cathode relative humidity rates	50 %
Nominal Current	110 A

2.2. Continuous aging life test

The PEMFC stack was operated under its nominal operating conditions during 124 hours in continuous mode, using the 1 kW test bench developed on the FC test platform in Belfort - France. The monitoring and controls of the FC test bench parameters are done through National Instruments Materials and a specific software associated with a friendly human machine interface developed with Labview. More detailed descriptions of the test bench are available in [11].

The plot of the stack voltage evolution during 124h is given in Figure 1. Some EIS measurements were conducted during the aging test at three different times after the initial aging test start: 3h later, 47h later, and 120h later (Figure 2). The EIS measurements were performed in galvanostatic mode at the nominal current by imposing additional AC sinusoidal solicitations (at various frequencies ranging from 0.1 Hz to 10 kHz, and with maximal peak amplitudes of 10A).

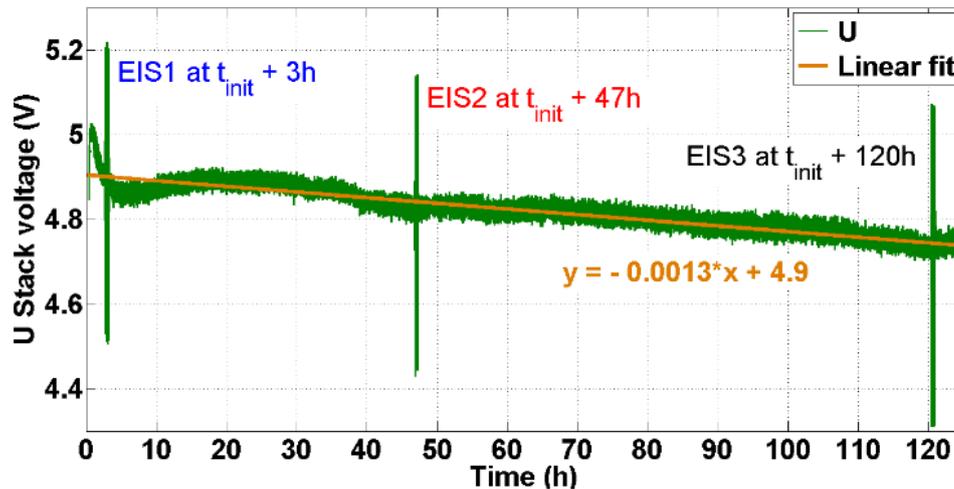


Fig. 1. Evolution of the stack voltage during 124 continuous operating hours with three EIS characterizations (3h after the beginning of the aging test, 47h later, and 120h later).

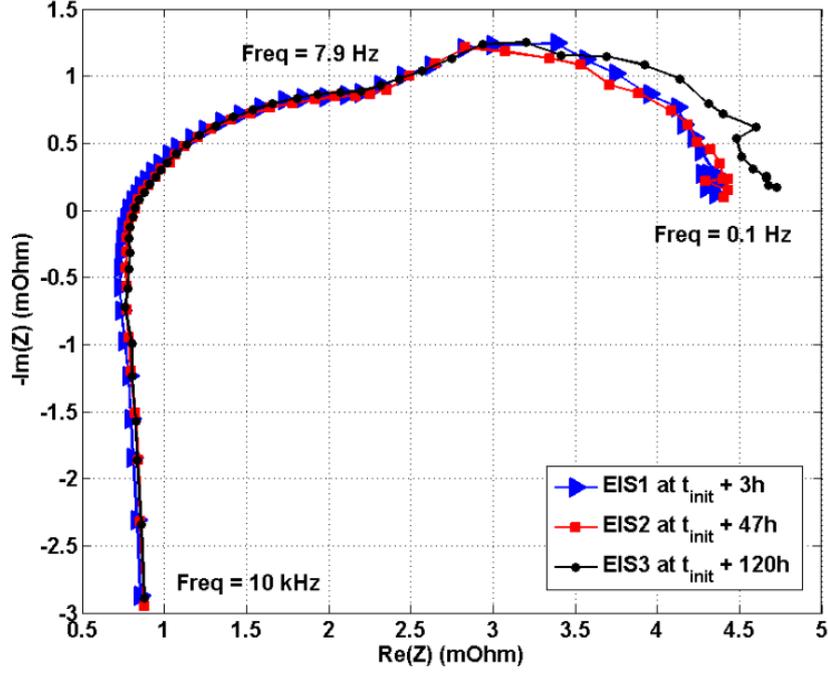


Fig. 2. EIS characterizations of the stack at different times during the aging test.

3. Regularity analysis of voltage signal

The method we have chosen is based on the local regularity analysis of the FC stack voltage, which is a typical non-linear signal. Actually, it is reasonable to assume that the operation of the stack during long term affects the morphology of the produced voltage. The analysis of its local fluctuations can therefore provide direct information on the dynamics of the device and on its state-of-health as well. To perform a right characterization of these complex signals, some appropriate and robust techniques are obviously necessary. In our study, we adopt the multifractal analysis of voltage signals based on wavelet leaders [12] in order to extract some latent information hidden in the singularity features and then to identify the signature of singularities for different operating time periods. Indeed, the multifractal features can fully display the distribution of signal singularities, while the geometric characteristics and the local scaling behaviors are described more precisely. The principle of the multifractal formalism consists to calculate two sets of coefficients associated to the signal:

- the Hölder exponent (h), that quantifies the local regularity strength of the signal,
- the fractal dimension ($D(h)$) that associates each cluster of points with the same regularity strength h to a non-Euclidean dimension [13].

It is well known that joint transformations in time and scale (or frequency) provide information on how the regularity of the signal varies over time.

Recently, a new formalism called *wavelets leaders* was built from the discrete wavelet transform. Its mathematical basis can be described as follow. Let a function $\psi(t)$ with a compact time support, called *mother-wavelet* [8] which satisfies the condition: $\int_{\mathbb{R}} t^i \psi(t) dt \neq 0$, where $i = 0, 1, \dots, N_\psi - 1$ and the vanishing moment $N_\psi \geq 1$. So, a family of wavelets $\psi^i(t)$ can be generated; it is sometimes called *daughter-wavelets*.

The templates of $\psi(t)$ dilated to scales 2^j and translated to time positions $2^j k$ where j is the multiresolution parameter, with an orthonormal basis of $L^2(\mathbb{R})$, can be formulated as follow [12]:

$$\psi_{j,k}(t) = \frac{1}{2^j} \psi\left(\frac{t-2^j k}{2^j}\right), j \in \mathbb{Z} \text{ and } k \in \mathbb{Z} \quad (1)$$

Then, the discrete wavelet transform of a signal $X(t)$ is defined by the following formula [14, 15]:

$$d_X(j, k) = \int_{\mathbb{R}} X(t) 2^{-j} \psi(2^{-j} t - k) dt \quad (2)$$

Assuming the dyadic interval λ and the dyadic cube Γ [12, 15, 16]:

$$\lambda = [k \cdot 2^j, (k + 1) \cdot 2^j] \quad (3)$$

$$\Gamma = 3\lambda = \lambda_{j,k-1} \cup \lambda_{j,k} \cup \lambda_{j,k+1} \quad (4)$$

Γ denotes the union of the interval λ with its two adjacent dyadic intervals [12]. Therefore, the *wavelet leader* is the local supremum of the wavelet coefficients located in the dyadic cube over all finer scales [14]:

$$L_X(j, k) = \sup_{\lambda' \subset \Gamma} |d_{X,\lambda'}| \quad (5)$$

The formula (5) indicates that to compute $L_X(j, k)$, which consists of the largest wavelet coefficient $d_X(j', k')$, we consider the indexes $(k - 1)2^j \leq 2^{j'} k' < (k + 2)2^j$ at all finer scales $2^{j'} \leq 2^j$. A possible scheme illustrating this definition is given in Figure 3.

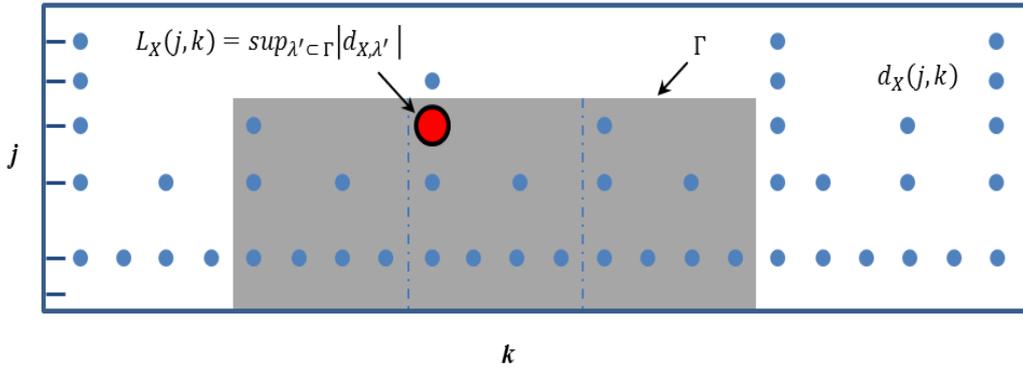


Fig 3. Illustration of the principle used in tracking the wavelet leaders L_X (black and red circle). They are calculated from the discrete wavelet coefficients $d_X(j, k)$ (blue dots) by taking the supremum in the time neighborhood $\Gamma = 3\lambda$ over all finer scales $2^{j'} < 2^j$ (area in grey).

The principle of the multifractal analysis based on wavelet leaders (WLMA) consists in analyzing the behavior of structure functions of the voltage signal, based on its discrete wavelets transform, in the limit of small scales. Let us define the structure functions $S_L(q, j)$, which quantify the spatial average of wavelet coefficients at a given scale 2^j :

$$S_L(q, j) = \frac{1}{n_j} \sum_{k=1}^{n_j} |L_X(j, k)|^q \quad (6)$$

Where q is the order of the statistical moment, $n_j \approx n / 2^j$ is the number of leaders available at each scale, and n the length of the sample.

Then the scaling exponent $\xi_L(q)$ is calculated by evaluating the behavior of the logarithm of structure functions in the limit of fine scales:

$$\xi_L(q) = \liminf_{j \rightarrow 0} \left(\frac{\log_2 S_L(q,j)}{j} \right) \quad (7)$$

In other terms, the structure functions $S_L(q,j)$ exhibit a power law behavior with respect to the scale analysis, in the limit of fine scales:

$$S_L(q,j) \cong C_q 2^{j\xi_L(q)} \quad (8)$$

where C_q is a constant.

The SS representing the function $h \rightarrow D(h)$ is obtained by the Legendre transform of the scaling exponents:

$$D_q(h) = \inf_{q \neq 0} (1 + qh - \xi_L(q)) \quad (9)$$

As for multifractal analysis, the vanishing moment should be larger than the largest singularity exponent; moreover, the overlarge vanishing moment will introduce border effects. Therefore, the reasonable analyzing wavelet is selected as Daubechies 3 ('Db3') function (with 3 vanishing moments, see Figure 4).

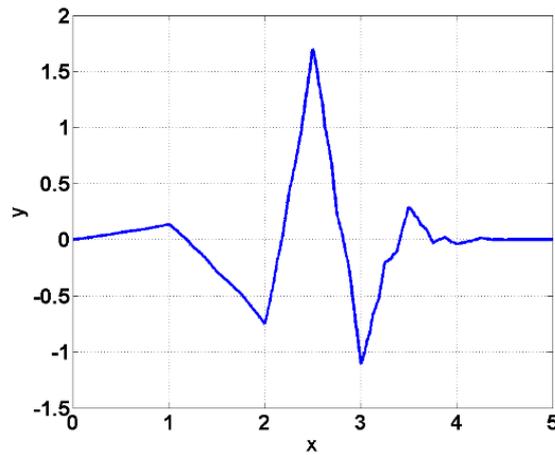


Fig 4. Typical Daubechies wavelet's, with 3 vanishing moments ('Db3').

The concept of Hölder exponents is that positive exponents imply that the function is continuous and has a given number of derivatives, while negative exponents imply that the function encloses transitions, jumps and eventually divergences to infinity. As the last characteristics are not observed in the studied voltage signals, we must avoid all negative values of h and $D(h)$. To obtain the entire curve $D(h)$ by the Legendre transform, both positive and negative values of q -order are needed, with $q \in [q^-, q^+]$. Notice that $q = q^-$ and $q = q^+$ amplify the small and large fluctuations of the signal. In this study, both q^- and q^+ are selected avoiding the negative values of Hölder exponent h and Hausdorff dimension $D(h)$. So, for our study and according to the observed FC voltage morphology, $q \in [-4, +5]$.

The SS computed using the WLMA analysis for the stack voltage signal acquired under nominal operating conditions is reported in Figure 5 (at the bottom). The singularities of the studied voltage signal provide $\xi_L(q)$

values with a curved q -dependency and the SS is in the form of a concave arc. So, we conclude that the studied voltage signal exhibits a multifractal organization.

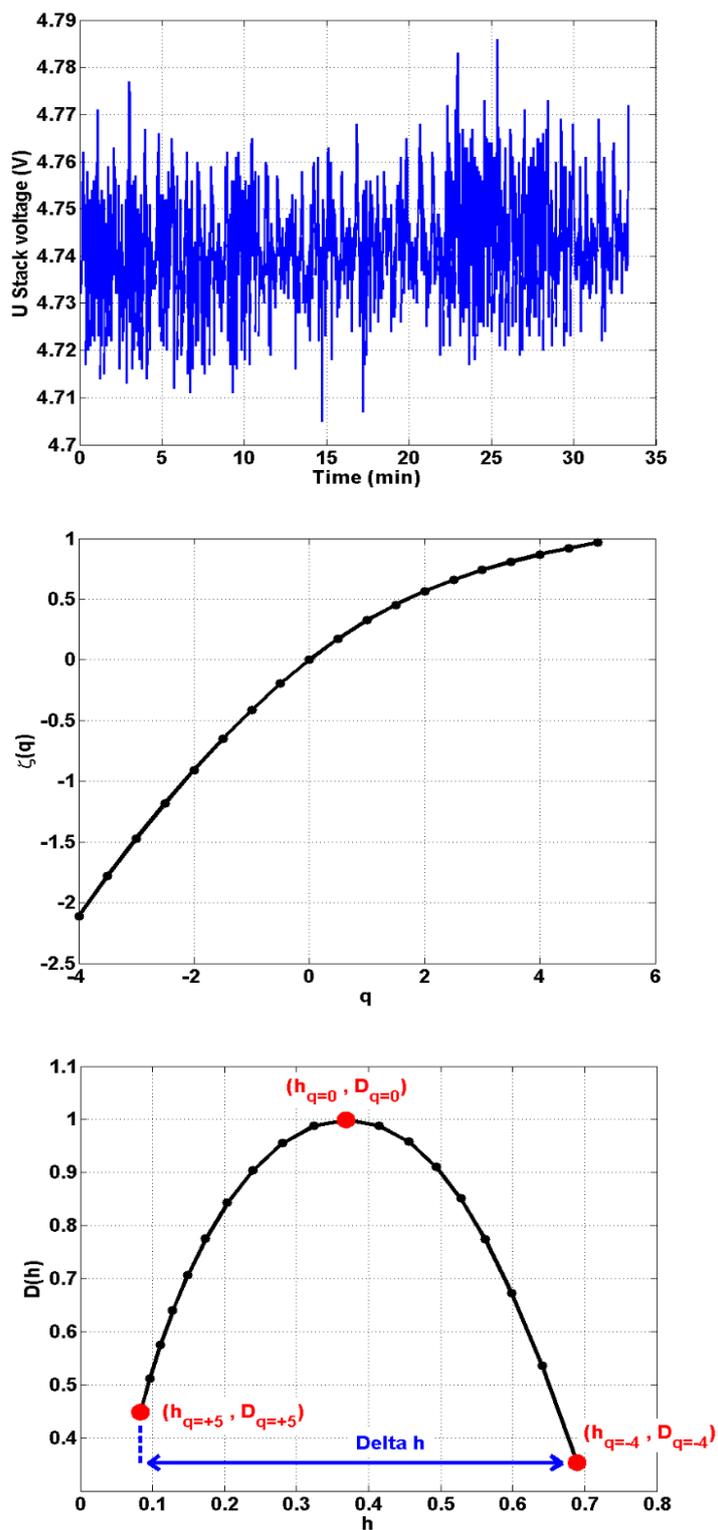


Fig 5. Typical curves of scaling exponent $\xi_L(q)$ (in the middle) and singularity spectrum (SS) (at the bottom) obtained by applying multifractal analysis on the voltage signal drawn in (at the top).

4. What about the dynamic multifractality in the stack voltage?

We have analyzed the acquired voltage signal at different time periods of the aging test: 2.5 hours after the initial aging test start, 50 hours later, and 122 hours later. These time periods are chosen according to the EIS acquisition times listed above, and in order to make possible some comparisons between the two methods (EIS and singularity analysis). Indeed, we cannot apply both methods exactly at the same time due to the disturbances added in the voltage signal by the EIS characterization sequences. The obtained SS are reported in Figure 6.

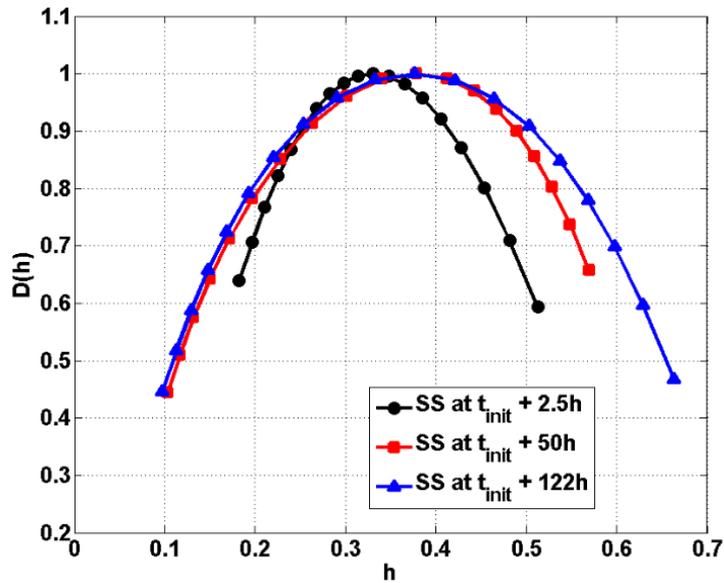


Fig 6. Singularity spectra obtained at three time periods of the aging test.

The SS results indicate that the degree of voltage signal multifractality exhibits strong variations with time. After 2.5 operating hours, the stack produces a voltage with weakest multifractality. Then, 50 hours later, the SS became larger because it encloses more singularity clusters. At 122 hours of operation, the SS becomes even wider, indicating a stronger multifractality.

The voltage signals associated to aging phenomena contain some sharp and impulsive variations with relatively close magnitudes.

To illustrate the obtained results, multifractal features estimated from the SS such as those displayed in Figure 5 ($h_{q=-4}$, $h_{q=+5}$, $h_{q=0}$, Δh , $D_{q=-4}$ and $D_{q=+5}$) are reported in Table 2. As we can observe, the SS width increases over time.

These results highlight a dynamic multifractality in the voltage signal produced by the stack at different times of the durability test. The indicators selected (SS pattern features) can therefore be used as tools for the FC aging monitoring.

Table 2. Multifractal features obtained in the three time periods of the FC aging test.

	$h_{q=-4}$	$h_{q=0}$	$h_{q=5}$	Δh	$D_{q=-4}$	$D_{q=+5}$
SS (~2.5h)	0.513	0.330	0.182	0.331	0.593	0.640
SS (~50h)	0.570	0.379	0.103	0.467	0.658	0.445
SS (~122h)	0.664	0.377	0.097	0.567	0.467	0.446

5. Additional discussion

The stack voltage evolution curve vs. aging time given in Figure 1 shows a voltage decrease of about 0.2V for the 124 hours of FC operation. The fitted line of Figure 1 shows that the mean degradation rate was about 1.3 mV/h (for a “mean” cell). This high degradation rate can be attributed to the fact that, before the aging experiment, the FC stack was first operated under quite severe conditions during a previous test campaign, which could partially damage the FC stack and affect its performances.

The results of the EIS characterizations of the stack displayed in Figure 2 show that the high frequency resistance (or internal resistance) related with the FC membrane resistance remains less sensitive to any aging phenomena. A slight increase of about $57\mu\Omega$ is noticed according to the FC operating time (124h). No obvious performance degradation was found on the membranes during this study.

In contrast, the low frequency resistance increases of about 0.4 m Ω . The larger low-frequency arc obtained on the EIS measurement at $t=120h$ suggests some diffusion problems in the FC.

Now, if we analyze the SS results (Figure 6), we observe an important widening of the SS as a function of aging time (Table 2). This behavior of the spectrum indicates the enrichment of the stack voltage by singularity clusters (low and strong frequency oscillations).

The SS computed at 2.5 operating hours exhibits a weak width ($\Delta h = 0.331$) with a dominant singularity strength $h_0 = 0.33$. Then, 50 hours later, the FC produces a voltage signal with more fluctuating clusters ($\Delta h = 0.467$). And finally, at 122 operation hours, this behavior amplifies and the SS becomes even wider ($\Delta h = 0.567$).

Intuitively, the widening of the SS versus aging time can be related with the increase of the low frequency resistance measured through the EIS characterization, and possibly with gradual problems of gas diffusion in the FC.

6. Conclusion

The multifractal analysis based on wavelet leaders (WLMA) described in this paper gives us important insights about the time dynamics of the FC stack voltage during the considered aging test periods.

The PEMFC stack voltage was analyzed using a mathematical microscope (discrete wavelets) at small scales. Its structure and clustering properties were then quantified using the multifractal formalism. This approach enables the access to hidden information enclosed in the FC voltage signal. The computed multifractal features at three time periods of the aging test show a dynamical behavior in the clusters (or iso-Hölder subsets) formation in the voltage signal according to the aging time.

We conclude that this dynamic multifractality of the stack voltage can be considered as a pertinent non-intrusive tool for the monitoring of the FC aging state. Moreover, this tool presents an interesting computational performance for an implementation in real time. In fact, the computing time is estimated at about 0.7 seconds for a voltage profile covering 6000 time points under the computer system characteristics of: Intel® Core™ i7-2960 XM, CPU @ 2.7 GHz, RAM:16 GO.

In future works, we intend to apply the proposed technique during longer FC aging tests and with other FC devices in order to validate the proposed approach.

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