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► **To cite this version:**

Elodie Pahon, Daniel Hissel, Samir Jemei, Nadia Yousfi Steiner. Signal-based diagnostic approach to enhance fuel cell durability. *Journal of Power Sources*, 2021, 23, pp.230223 (8). hal-03359954

HAL Id: hal-03359954

<https://hal.science/hal-03359954>

Submitted on 2 Aug 2023

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Signal-based diagnostic approach to enhance fuel cell durability

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Abstract: To face the dependency to fossil fuels and limit the carbon emissions, fuel cells are a very promising technology and appear to be a key candidate to face the increase of the energy demand and promote the energy transition and energy mix. To meet future needs for both automotive and stationary applications, fuel cells will require improved durability. In this paper, a new concept to enhance fuel cell performance and lifetime is provided, by introducing a disruptive and high-efficiency diagnostic approach based on signal processing. The results show that the failures on the fuel cell system can be detected and isolated efficiently thanks to fault indicators. The innovative concept proposed here, can guarantee improvements of the durability and of the reliability of the fuel cell system by detecting abnormal operating conditions rapidly and efficiently.

Keywords: Proton exchange membrane fuel cell; diagnostic; signal processing; wavelet transform; entropy.

1. Introduction

In the framework of energy transition, new technologies such as hydrogen energy and fuel cell systems must be investigated. This is particularly relevant for the mobility [1][2]. Hydrogen has a high energy efficiency which is three times more important than gasoline (120MJ/kg for hydrogen vs. 43 MJ/kg for gasoline). Another strong asset of hydrogen is its abundance on Earth (more than 90% in atoms number) which makes it an almost inexhaustible resource. Hydrogen-energy can be used as a

way to store electricity (i.e. coming from renewable energies that are intermittent) and as a supplying gas to generate electricity through a fuel cell. The electrolysis of water is an appropriated solution to produce hydrogen without greenhouse gas emissions, by using electricity supplied by renewable energies, as wind and solar, to split water molecules into hydrogen and oxygen. The recent sharp decline in the cost of renewable energy suggests that the production of hydrogen from renewable energies through a power-to-gas process might become more profitable[3].

Based on the Technical Roadmap of the US Department of Energy (DoE) proposed in 2017[4], two main bottlenecks are still hindering fuel cell vehicles from being as competitive as gasoline vehicles. The system cost and durability targets to achieve by 2020 are 40 \$/kWe (for a projected market of 50k units per year) and 5,000 hours respectively, and even more in 2025 (35\$/kWe – 8,000h) compared to 45 \$/kWe and 4,130 hours obtained today. Therefore, the key challenge, in research and development, is to decrease the cost while simultaneously improving the durability; that is completely contradicted. A solution to increase the durability of fuel cell systems, without increasing their cost, is to work at the system level and consider fault detection and isolation (FDI) methods. These solutions allow avoiding performance losses in a fuel cell system while preserving durability. Literature reported various approaches to fault diagnostic dedicated to fuel cell systems[5]–[8]. A disruptive approach is proposed, based on signal processing, for improving the performance and the durability of fuel cell systems, without being intrusive and without any additional costs. The state-of-health diagnostic approach focuses on the measurements that are always available on a commercial fuel cell system. Among them, the fuel cell stack voltage is obviously always accessible. Thus, the implementation of state-of-health (SoH) estimation techniques, by using only the voltage signal, is of highest interest, from a scientific, technical and also economical point of view. In this paper, some relevant results validated on experimental tests are obtained, allowing to distinguish efficiently between different states-of-health (or faults) of the fuel cell system. It is then a strong asset for extending the fuel cell lifetime by acting on the different actuators of the system.

The layout of the paper is as follows. First, the degradation mechanisms occurring on a fuel cell system and the diagnostic principle are introduced. Secondly, the signal-based approach is detailed with a focus on the entropy feature that is considered as a state-of-health indicator. Then, the experimental tests and signal generation is presented. This step allows performing the diagnostic tool and building an entropy range for each operating condition.

2. Health management framework

2.1. Degradation mechanisms.

A fuel cell system is complex, multi-physics and multi-scale with strong non-linearities. Moreover, a fuel cell system is composed of several ancillaries that are related to the different physic domains. The cooling system refers to the thermal management of the device. The production of water and the gas supplying are directly related to the electrochemical and fluidic phenomena. Finally, the electrical power converter and the control are the power management parts. Figure 1 presents the global architecture scheme of the test bench used in order to generate data (ageing data or diagnostic data).

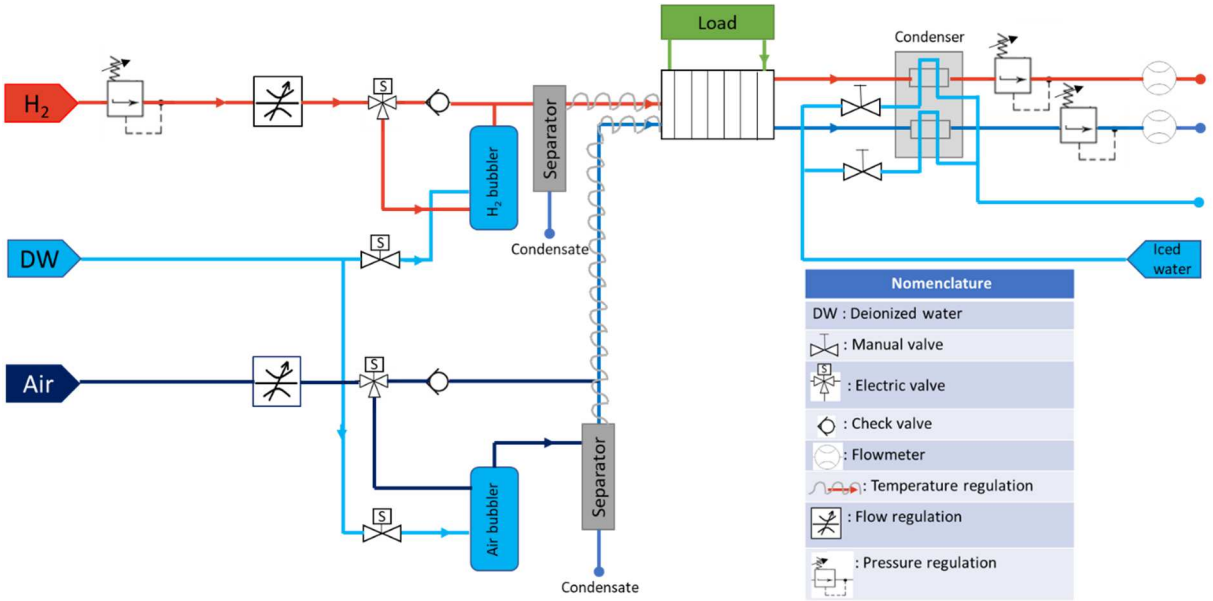


Figure 1. Overview of a fuel cell system

Each component of the fuel cell system can fail. The more complex the system, the higher the number of possible failures. In the literature[9], different kind of failures are mentioned:

- Sensors faults: the measurement given by a sensor is wrong, therefore the information delivered to the user is also wrong. However, the system properties are unchanged.
- Actuators faults: the controllers and regulation actions on the system are modified or stopped. The system properties are affected.
- Subsystem faults: some malfunctioning provide changes on the system properties. In the case of a multi-components system (i.e. fuel cell system), each component has to ensure a specific function. So, the whole system is able to provide service if all components are working well. Therefore, some faults on a component impact the performance of the whole system and their effects can propagate and have negative effects on other components. The phenomenon is called *fault propagation*.

The sensors and actuators faults are often neglected based on assumptions that considering them as perfect components. Generally, when talking about faults (or failures) it is referred to subsystem faults. In the proposed case, it suggests faults occurring on the fuel cell system that means fuel cell stack and ancillaries (especially compressor, pump, filters and converter). Failures are due to three main causes:

- Manufacturing process[10]: nanostructure, materials, assembly, design
- Operation[11][12]: extreme operating conditions, severe faults, fault propagation
- Environment[13]–[17]: pollutants, impurities, weather, vibrations, negative temperature

The faults linked to the manufacturing process are usually not considered during the diagnostic because they lead to a sharp and rapid degradation of the fuel cell system even to an inability to run it. The only solution to correct the fault is to rebuild the device. For instance, a too high tightening torque during the fuel cell stack assembly makes the subcomponent crushed and unusable[18].

Another example could be a bad design of the channels in the bipolar plates that impact the gas and coolant flows and exhaust process.

Conversely, for the environmental and operating conditions, it is possible to find solutions without considering a new component. Some additional devices can be added to solve some non-optimal conditions, as filters for impurities or heaters for negative temperatures.

Among these post-manufacturing faults, it can be considered:

- Water management faults: the membrane needs to be well-humidified in every operating condition to avoid flooding (an excess of water inside the stack) or drying (a lack of humidity of the electrolyte)
- Gas starvation faults: fuel and/or oxidant are not supplied in sufficient quantity to enable an optimal electrochemical reaction
- Short-circuit faults: the electrical power converter has been short-circuited
- Poisoning faults: pollutants are existing inside the fuel and/or oxidant
- Cooling faults: the control of the temperature of the fuel cell system is no more efficient

In all cases, these faults impact the fuel cell performance and lead to a voltage decrease. As mentioned, a degradation or a malfunctioning of the balance of plant (BoP) component will cause an unavoidable operating condition on the fuel cell stack that implies a faulty state-of-health. For example, an oxidant starvation could be due to a failure of the compressor. An increase of stack temperature may come from a fault on the cooling system (i.e. pump or heat exchanger). In addition, the different phenomena (electrical, electrochemical, fluidic, thermal) present many different characteristic time responses[19]. In brief, the fastest concerns the electrical phenomena. Then comes the time constant of the electrochemical reactions (oxidation and reduction), the gas diffusion, the liquid water transport, the poisoning, the temperature effects and the degradation and ageing of the fuel cell. Dijoux et al.[20] suggest a fault classification according to time responses, effects, reversibility and involved parameters.

2.2. Fault diagnostic approach.

Diagnostic is a crucial point for fuel cell systems as it is a way to increase their lifetime and their reliability. As the durability of fuel cell systems needs to be improved, it is necessary to be able detecting as soon as possible a fault or an unavoidable condition that could degrade the system faster, deeply and permanently.

The diagnostic approach is usually defined as a two easy-to-follow steps that consist in detection and identification of a fault. The principle of the fault detection is given in Figure 2 with the performance indicators that are based on the ratio of false alarms, non-detection or detection delay. An overview of the fault identification is presented on Figure 3. The performance indicators are directly linked to misclassifications results.

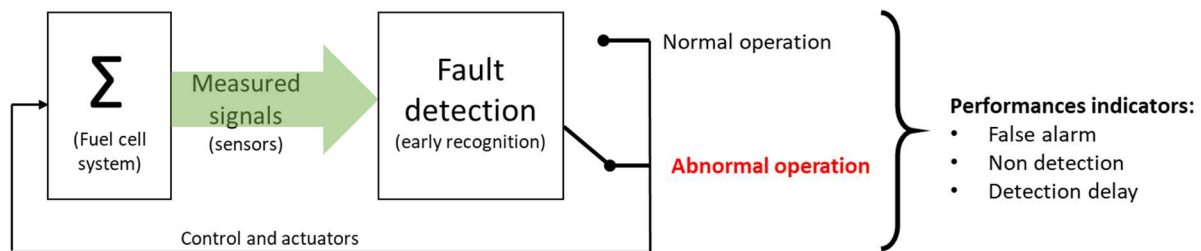


Figure 2. Fault detection principle and performance indicators

Several strategies for developing fault diagnostic exist, that may be divided in three main groups: model-based[21], data-based[22] and signal-based[23], [24] approaches. For the model-based approaches, inlets and outlets are linked by a mathematical relation. The method analyzes the residuals between the sensor measurements and the expected values, derived from diagnostic-oriented model of the considered system. For a fuel cell, this approach is often inadequate because of the lack of knowledge on the physical phenomena occurring inside the fuel cell and/or on the values of the internal parameters involved in the behavior of a fuel cell stack. The data-based approaches depend on heuristic knowledge. Moreover, a large database is needed, in order to train the algorithm and easily make the fault detection and identification. With a signal-based method,

signal processing is used. It is an efficient tool for extracting valuable features that can reflect the occurrence of certain types of faults.

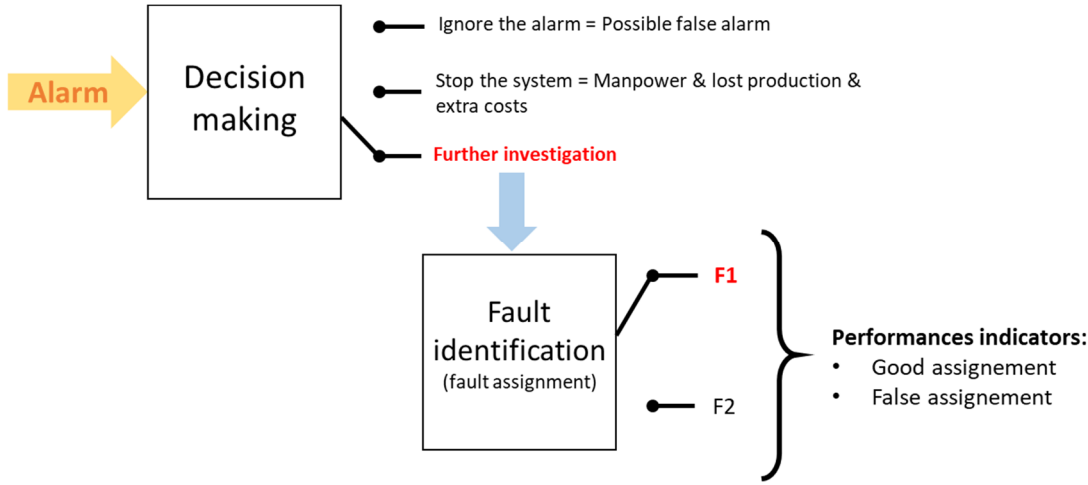


Figure 3. Fault identification principle and performance indicators

3. Signal-based approach

The diagnostic is done in the time-frequency domain. The aim is to characterize non-stationary signals, in the frequency domain. The non-stationary signals vary along time; this is the case for many signals that can be measured on a fuel cell system. The wavelet mathematical transform is an efficient tool for analyzing signals and extract both temporal and frequency changes in a considered signal compared with other approaches as Fourier transform. In this work, the given signal-based approach developed to diagnose a fuel cell system during its lifetime uses the wavelet transform to detect abnormal operating conditions combined with a specific degradation indicator called entropy to identify the fault and its intensity.

Entropy is a measure of complexity[25]. Thus, this measure can be used to evaluate the spectral richness of a signal as well as transients changes or behaviors. Although entropy is a very suitable tool for signal processing of electrochemical systems, the literature is poor, except for diagnostic of batteries[26]–[28]. Regarding fuel cells, the proposed approach is nevertheless a disruptive one.

The given signal-based approach dedicated to fuel cell diagnostic is composed of five steps:

- Data acquisition: the signal is acquired in different operating conditions (nominal, variation of different parameters one by one to an upper or lower limit values). In this work, only the stack voltage is used as it is an easy-to-measure value that is available in all fuel cell systems without any extra cost.
- Data preprocessing: outliers recorded are removed as well as unavoidable shut-downs of the fuel cell. The signal is then decomposed in several time series and labelled with the corresponding state-of-health, according to the operating conditions and expert knowledge.
- Wavelet transform: the signal is analyzed in a time-frequency framework. The high and low frequencies are decomposed in order to allow displaying the global signal at different scales and getting information about degradation along frequency range. The wavelet coefficients are computed for next step.
- Feature extraction: the entropy degradation feature is then computed (see section 3.3).
- Performance evaluation: the diagnostic result is obtained, based on the entropy value and on their value and trend.

The two first steps are directly linked to the experimental part with: *i)* the acquisition of voltage measurement at a given acquisition frequency and for various operating conditions and *ii)* the post experimental tests data treatment and labeling with the actual states-of-health of the fuel cell, defined by a human expert.

The wavelet transform is a mathematical tool that allows a time-frequency analysis unlike the traditional Fourier transform. When using the short-time Fourier transform technique, a “time-localized” Fourier transform is obtained with a fixed window size for all frequencies. The wavelet transform enables variable window sizes to analyze different frequency components in a given signal[29]. Two parameters have to be defined before using the wavelet analysis: the type of considered wavelet and the number of decompositions needed.

3.1. Wavelet family choice.

The “mother wavelet” selection plays an important role for detecting and localizing different types of transients[30]. In this paper, the orthogonal and biorthogonal wavelets families are considered. There are needed to ensure the inversibility of the decomposition. Moreover, the orthogonality is a valuable property that conserves the energy of the signal which is helpful in the reconstruction of a signal from the coefficients[31]. A method for reaching the best wavelet depending on the available signal, is to consider the error between the signal $s(t)$ and the signal $s'(t)$ after the reconstruction. The wavelet corresponding to the lowest error is chosen. The error is given by the equation 1.

$$\varepsilon = \frac{\sum_{i=1}^N |x_j - x'_j|}{N} \quad (1)$$

Where:

x_j : sample j from $s(t)$.

x'_j : sample j from $s'(t)$.

N : total number of samples.

The study shows that the Daubechies wavelet allows to minimize the error ε and more precisely the Daubechies order 4 (db4). This result is in accordance with the literature as *db4* is widely used for diagnosis task for energy sources[32].

3.2. Decomposition level threshold.

The decomposition level choice is linked to the acquisition frequency and the frequency range as given by Equation 2, coming from Ibrahim et al. work[32].

$$i = \left\lceil \frac{\log\left(\frac{f_{acq}}{f_{FC}}\right)}{\log(2)} - 1 \right\rceil \quad (2)$$

where:

f_{acq} is the acquisition frequency of the signal $s(t)$ (1Hz).

f_{FC} is the frequency domain of the fuel cell (between 100mHz to 1kHz)[21]

A mean frequency $f_{FC} = 10Hz$ is considered and the acquisition frequency of the stack voltage is 1 Hz. Thus, five decomposition levels are needed.

The dedicated wavelet decomposition process is presented in Figure 4. The detail signal D_i contains the high frequency information linked to the initial signal $s(t)$ namely frequencies comprised within the $\left[\frac{1}{2^{i+1}} f_s, \frac{1}{2^i} f_s\right]$ Hz interval (when considering a dyadic decomposition[33]). The approximation signal A_i contains low frequencies of the initial signal $s(t)$ which are in the range of $\left[0, \frac{1}{2^{i+1}} f_s\right]$ Hz (when considering a dyadic decomposition[33]).

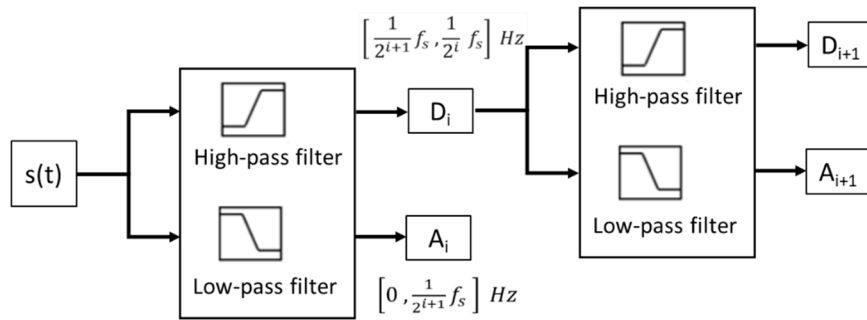


Figure 4. Global wavelet decomposition process

3.3. Entropy feature.

This study involved the signal processing method using wavelet transform and an entropy feature as a degradation indicator. The entropy gives information on the order/disorder level according to a response of a multi-frequency signal [28] and gives the complexity range of a signal $s(t)$. It measures the homogeneity of the spectral distribution of a signal from a discrete wavelet transform (DWT). An orthogonal wavelet is needed to ensure the invertibility of the decomposition. The expression of the energy is simplified. For a signal s and $C_i(k)$ the coefficients coming from the DWT with the decomposition level $i=1, \dots, N$ at time k , the energy at the level i is given by:

$$E_i = \sum_k |C_i(k)|^2 \quad (3)$$

The total energy is:

$$E_{tot} = \sum_{i=1}^N \sum_k |C_i(k)|^2 = \sum_{i=1}^N E_i \quad (4)$$

The relative energy at level i is:

$$p_i = \frac{E_i}{E_{tot}} \quad (5)$$

p_i defines a probability distribution of energy. The entropy of this distribution is given by:

$$Entropy(s) = - \sum_{i=1}^N p_i \cdot \ln p_i \quad (6)$$

With $\sum_{i=1}^N p_i = 1$, and $p_i \cdot \log_2 p_i = 0$ if $p_i = 0$

Equations (3) and (6) indicate that the entropy of the wavelet coefficients is bounded by:

$$0 \leq Entropy(s) \leq \ln N \quad (7)$$

in which $Entropy(s)$ will be equal to:

- **zero**, if all wavelet coefficients are equal to zero except one;
- **$\ln N$** , if the probability of energy distribution for all the wavelet coefficients is the same (i.e., $1/N$).

This leads to the conclusion that the lower the entropy value is, the higher the energy concentration will be[34].

Then, in order to measure the distance between probability distributions of data sequences X and Y , the relative entropy RWP is defined as:

$$RWP = D(X||Y) = \sum_{x \in X} p(x) \log \frac{p(x)}{p(y)} \quad (8)$$

with $p(x) \log \frac{p(x)}{p(y)} = 0$ if $p(x) = 0$, and $p(x) \log \frac{p(x)}{p(y)} = \infty$ if $p(y) = 0$.

The smaller the relative entropy is, the more similar the distributions of the two data sequences (X and Y) will be. Thus, a small relative entropy value between the signal (i.e., data sequence X) and its corresponding wavelet coefficients (i.e., data sequence Y) is expected [35].

4. Experimental tests and signals generation

An in-lab test bench developed for diagnostic and characterization tasks is used. Three PEMFC have been tested on the same test bench under several and extreme operating conditions (please refer to Table 1). The fuel cells have an active area of 220 cm² for both of them and 100 cm² for the other one, the nominal operating temperature is 80°C and the nominal current density is 0.5 A/cm². They are composed respectively of 8, 40 and 20 cells. The manufacturer also recommends a range of anode and cathode stoichiometries. The requested values evolve between 1.1 and 2.5 for anode side and between 1.1 and 5 for the cathode side.

To collect voltage signals in various operating conditions, a first experiment under nominal operating conditions is performed. Then, a one-by-one parameter is changed, as air stoichiometry (λ_{air}), cooling flowrate $Q_{cooling}$ or hydrogen purity Q_{fuel} , in order to cause a faulty condition on the fuel cell stack. The values of the parameter are progressively increased or decreased until a given threshold, defined by the physical limits of the test bench. A last test is then another time performed in nominal operating conditions.

For example:

- For the faulty condition F1 (low air stoichiometry), $\lambda_{air} = 2$ (nominal value); $\lambda_{air} = 1.6$; $\lambda_{air} = 1.4$; $\lambda_{air} = 2$.
- For the faulty condition F2 (high air stoichiometry), $\lambda_{air} = 2$ (nominal value); $\lambda_{air} = 3$; $\lambda_{air} = 3.5$; $\lambda_{air} = 4$; $\lambda_{air} = 5$; $\lambda_{air} = 2$.
- For the faulty condition F3 (cooling failure), $Q_{cooling} = 9.8$ l/min (nominal value); $Q_{cooling} = 4.9$ l/min; $Q_{cooling} = 0$ l/min; $Q_{cooling} = 9.8$ l/min.
- For the faulty condition F4 (fuel purity), $Q_{fuel} = \text{pure H}_2$ (nominal condition); $Q_{fuel} = \text{H}_2 + 10$ ppm CO; $Q_{fuel} = \text{pure H}_2$; $Q_{fuel} = \text{H}_2 + 10$ ppm CO; $Q_{fuel} = \text{pure H}_2$.

The same tests are performed several times on the three fuel cell stacks. A constant acquisition frequency of 1Hz is always considered.

Some characterization as polarization curves (voltage vs. current) and electrochemical impedance spectra (EIS) are also performed during the different tests. In this study, the EIS measurements are only performed in order to see the degradation on the fuel cell. They provide more information about the degradation state of the fuel cell system and can be used to validate the efficiency of the proposed diagnosis method. Indeed, both methods (EIS and entropy) could be coupled to increase the robustness of the results, but the strategy to obtain an EIS on-board is less obvious to implement than the signal-based method.

Table 1. Technical specifications of the investigated fuel cells

State-of-health	Air stoichiometry	Temperature (°C)	Pressure (bar abs)	Cooling flow rate (l/min)	Carbon monoxide content in fuel (ppm)
Healthy (H)	2	80	1.5	9.8	0
Faulty (F) – Low air stoichiometry (F1)	1.6	80	1.5	9.8	0
	1.4	80	1.5	9.8	0
Faulty (F) – High air stoichiometry (F2)	3.5	80	1.5	9.8	0
	4	80	1.5	9.8	0
	5	80	1.5	9.8	0
Faulty (F) – Heating (F3)	2	80	1.5	4.9	0
	2	80	1.5	0	0
Faulty (F) – Fuel poisoning (F4)	2	80	1.5	9.8	10

As already said, only the voltage signal is here considered and analyzed for the fault diagnostic. This choice is made in order to limit the implementing cost of the diagnostic approach on an embedded system as no additional sensor is needed to perform diagnostic. **As an example, Figure 5 presents 3 evolutions of the fuel cell voltage during an experimental test with a) low air stoichiometry λ_{Air} (F1), b) Heating (F3) and c) fuel poisoning (F4).**

5. Diagnostic performance

This section presents the diagnostic results obtained thanks to the disruptive approach based on the wavelet transform combined with the entropy feature. The wavelet tool is used to detect some changes and/or transients in the fuel cell voltage signal. The time-frequency analysis gives some alerts based on specific frequencies that are directly linked to a specific fault (depending on the time constants of each physical phenomena[20]). Then, the extraction of entropy feature is performed in order to identify the fault and its intensity/severity. To reach this aim, the relative entropy (RWP) is observed for each operating condition and compared to the healthy condition that corresponds to the nominal operating conditions. The results for each type of fault are presented on Figure 6.

The first remark is that the developed algorithm is able to determine whether there is a fault or not. Secondly, the observed entropy evolves gradually and positively when the effect of the fault is increasing. For instance, when the water flow becomes non-optimal (F3), the RWP increases until reaching high RWP value when the cooling system is completely out of order. Thirdly, for all cases considered in this study, the computational speed of the algorithm is less than one second. This threshold is set according to the faults considered, which are mainly related to fluidic and thermal parameters whose time constants are greater than one second.

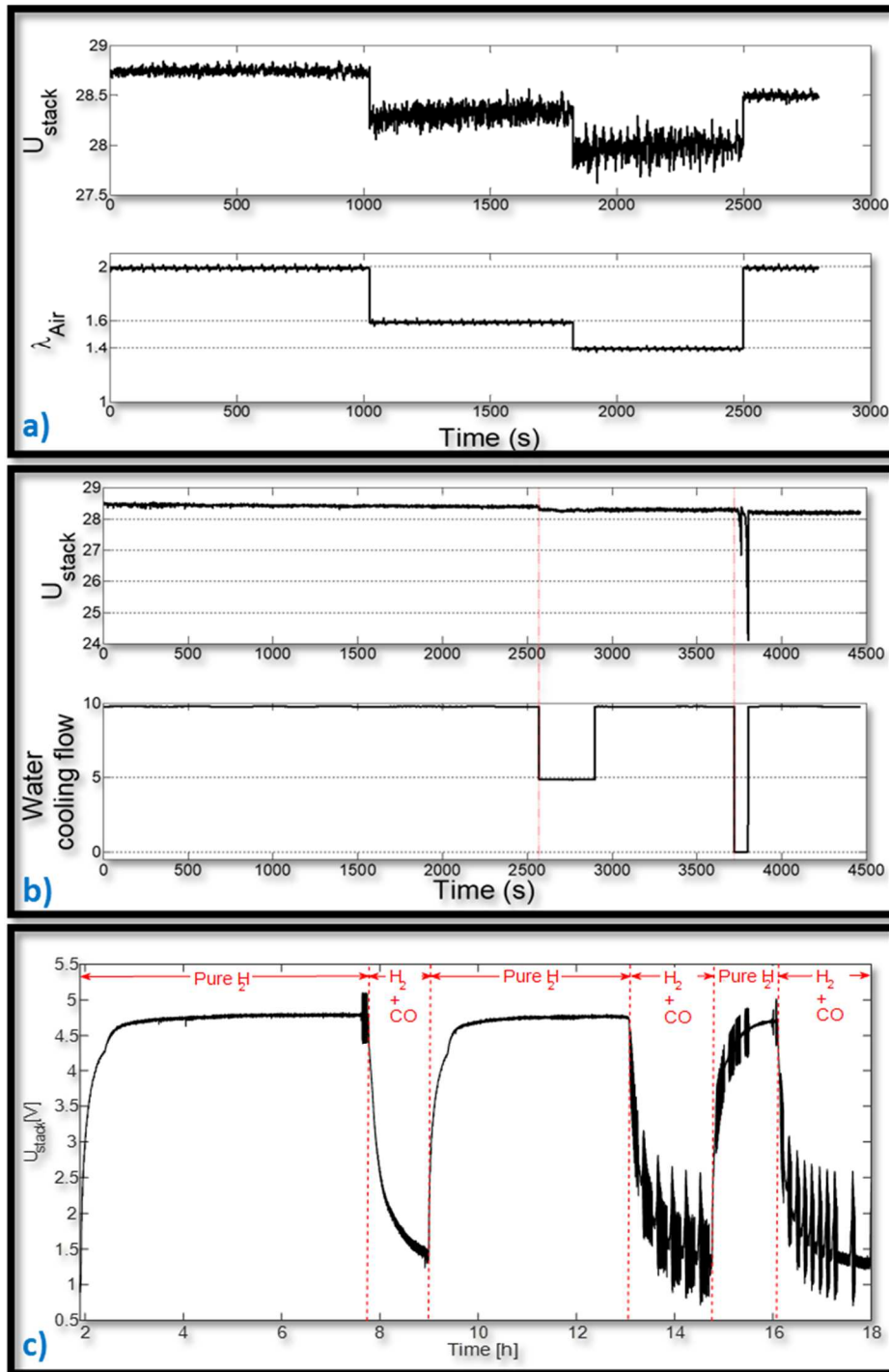


Figure 5. Impact of a) the air supply, b) the cooling flow and c) the CO presence in fuel (10 ppm) on the stack voltage – Oscillations on figure 5.c) represent the EIS measurement periods

As a conclusion, the entropy tool gives a clear information about the fault occurrence and the strength of the fault, by comparing a signal sample to another signal (defined as the reference signal). The transient states are also visible, which is very helpful in order to prevent any major

damage on a fuel cell system. Thanks to the RWP, the fault detection and the fault magnitude can be defined as well as the fault identification by observing the RWP ranges. Table 2 gives the resulting RWP ranges to be considered for the fault identification. Table 2 is built based on all the experiments performed on the three fuel cell systems to validate the efficiency and the robustness of the method. The closer the RWP is to the upper limit, the greater the fault intensity.

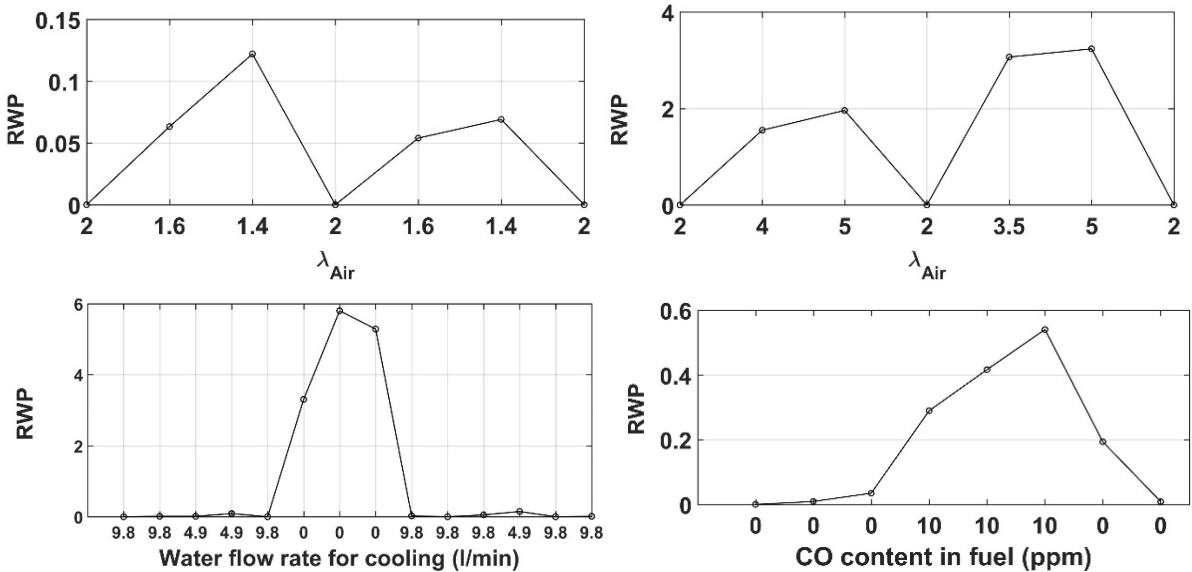


Figure 6. Fault identification results based on the entropy feature

Table 2. Resulting entropy values corresponding to different faults

Fault #	SoH	Entropy range
F1	Low air stoichiometry	[0 – 0.2]
F2	High air stoichiometry	[1.5 – 3.5]
F3	Cooling fault	> 3.5
F4	Fuel supply poisoning	[0.4 – 0.6]

6. Conclusion

The proposed disruptive diagnostic method based on a signal-based approach allows detecting a SoH variation on a PEM fuel cell stack. In order to reproduce key failures occurring on actual fuel cell systems, several experiments are here performed on a test bench. The fault diagnostic approach

combines the wavelet transform and an entropy feature that allow respectively detecting and identifying a fault and its intensity on the fuel cell system. The entropy is a reliable indicator of the SoH, by giving information on the fault detection and fault severity according to a reference signal, which is acquired in nominal/healthy conditions, and independently of the ageing time. The main contribution of this work is to design and validate a low-cost disruptive methodology for on-line diagnosis of fuel cell systems, based on signal processing.

This diagnostic tool is especially interesting for meeting the requirements of automotive large-scale applications and is a promising approach to improve the durability and the reliability of fuel cell systems. Finally, based on the diagnostic results, this method seems robust enough for real life scenarios, as the failures that are considered in this manuscript take into account the different ancillaries of the fuel cell system (air supply, fuel supply, cooling system) and their possible failures. These faults are the most likely and most recurrent on a fuel cell system and in a real application (e.g. automotive application). Finally, this approach is novel and inexpensive in terms of computing time, which could encourage the implementation of this online method for fast fault detection in future work.

7. Acknowledgment

The authors would like to thank the French ANR project DIAPASON2, for financially supporting this work and offering the possibility to use their data. This work has been supported by the EIPHI Graduate School (contract ANR-17-EURE-0002) and the Region Bourgogne Franche-Comté and the French National H2 & Fuel Cell Network (FRH2 CNRS).

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