Diagnosis of a Fuel Cell Stack Using Electrochemical Impedance Spectroscopy and Bayesian Networks

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***Abstract*—In the first part of the paper, a novel architecture of impedance spectrometer is presented. The instrument is dedicated to the characterization and diagnostic of large fuel cell stacks operated in galvanostatic mode. It allows impedance measurements on cells located in the middle of the stack, where common mode potentials are usually too high for commercial devices. In the second part of the article, probabilistic methods (Bayesian Networks) are used to provide efficient diagnostic of a proton exchange membrane fuel cell stack. Experiments are performed on a twenty cell assembly using electrochemical impedance spectroscopy and from the test results, a Bayesian Network is proposed to ensure the diagnosis of the investigated fuel cell (study of drying and flooding phenomena). A maximum rate of good classification equal to 91% has been reached.**

***Index Terms*— Electrochemical Impedance Spectroscopy, Diagnosis, Probabilistic method, Bayesian Network, Fuel Cell.**

I. Introduction

IMPORTANT research programs are launched worldwide to increase the durability and reliability of batteries, supercapacitors and Fuel Cell (FC) generators [1], [2], [3]. To reach this aim, a considerable research effort is made in order to understand the physical and chemical phenomena involved in these new electrochemical components through the development of diagnostic methodologies [4], [5]. In this context, Electrochemical Impedance Spectroscopy (EIS) is a powerful experimental method to characterize and diagnose electrochemical systems. EIS technique allows measuring the impedance of the investigated system over a wide frequency band. The real and imaginary parts of the system impedance are calculated from the measured current and voltage alternating components. Each impedance spectrum is a series of single impedance measurements at discrete frequency points. EIS can be performed in two modes: potentiostatic (potential control mode) and galvanostatic (current control mode). Data obtained by EIS are often expressed graphically using a Nyquist plot. Numerous papers deal with EIS as a diagnostic tool for FC generators [6], [7]. EIS reveals valuable information about the reaction mechanism of an electrochemical process: different reaction steps will dominate at specific frequencies, and the frequency response shown by EIS can help to identify the rate limiting steps. In the case of a FC impedance study, the main EIS benefit is the resolution of dynamic processes occurring at different timescales in the system like charge (electrons, protons) transfer and mass (reactant molecules) transport. However, the use of EIS technology is costly and often strongly limited by the electrical specifications of the available spectrometers. Generally, commercial spectrometers have a compliance voltage of about 12 V and a maximal current range of 50 A. Moreover, the common mode potential of commercial impedance spectrometers might be limited to a few Volt units only. These limitations are usually not compatible with the characterization of large battery banks or FC stacks needed for transport and stationary applications [8]. Besides, the use of EIS for the characterization of electrochemical devices leads to different constraints. Firstly, the stationary state of the element under test is required. Secondly, the record of a cell impedance spectrum is time consuming (generally between 2 and 45 min, due to the low-frequency investigated range).

FC converts directly chemical energy into electricity, heat and water [9]. FC appears to be a promising technology by the possibility of obtaining higher system efficiencies [10]. However, FC suffers from a lack of maturity in its technological development. EIS is thus used as an in-situ and non intrusive technique to understand the underlying physical mechanisms in FC Membrane Electrode Assembly (MEA), to acquire specific knowledge about the FC component behaviors [11], especially in terms of durability [12], and to facilitate the controls of FC systems [13]. In particular, EIS has been widely employed by FC experimenters to study and improve diffusion phenomena at cathode side [14]. EIS can also be used to study the ionic conductivity of membranes [15], pollutant effects [16] and catalyst loading [17]. So far, EIS has been commonly applied to the study of single cells. Only a limited number of works has been done on FC stacks [7], [18], [19] and only few research has been devoted to the characterization and diagnostic of individual cells in FC stacks through EIS methods [20], [21]. In many theoretical studies, FC stacks are modeled as a single "mean" cell assuming that all the individual cells have an identical behavior [22], [23]. This hypothesis can be done for short stacks and for safe stacks operated near nominal conditions. However, when dealing with the issues of diagnostic (i.e. cell failures, operations in severe conditions) and large stacks dedicated to

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embedded generators, the “mean” cell models are not sufficient for diagnostic purposes. For instance, in the case of a multi-cell generator, these models are not useful to explain some large distributions in the individual cell voltages possibly caused by the gas supply design (e.g. configuration of the channels in the FC bipolar plates) and the cooling system configuration.

Some examples of works can be mentioned. X. Yuan *et al.* have tested a six-cell PEMFC assembly [7], [20]. The experimental set-up was made of a commercial impedancemeter (Solartron 1260 gain / phase analyzer), a TDI electronic loadbank to generate polarization currents (DC currents) and a home-made rotary cell switch unit to measure the impedance spectra of the cells. To study the causes of Nafion® membrane dehydratation, M. Ciureanu has tested a PEMFC stack made of 5 cells using a similar set-up [24].

Various methodological solutions have been proposed in the literature to ensure the diagnostic of PEMFC applications. Some examples of works using stochastic (statistical) solutions can be mentioned [25], [26]. In their study, Hernandez *et al.* have analyzed cell voltage probability density functions as clustering parameter [25]. The method proposed was applied successfully for the diagnosis of flooding electrodes. Nevertheless, in this paper, little information only is given about the accuracy in fault detection. Roy et al have proposed a similar approach but they used EIS at low frequencies (0.1-100Hz) to discriminate fault conditions [26]. But precise information about the accuracy of the fault detection module is also missing. Other solutions based on probabilistic approach have been proposed [27]. In their work, Riascos *et al.* have used the Bayesian theory to diagnose multiple fault conditions (faults in the air reaction fan, in the refrigeration system, in membrane permeability to hydrogen and fault in hydrogen pressure) [27]. Bayesian Networks (BN) provide a natural tool for dealing with three problems that occur throughout applied mathematics and engineering, and which are: uncertainty, decision and reasoning. BN can be expressed under the form of probabilistic graphical models in which nodes represent random variables, and the (lack of) arcs represent conditional independence assumptions. The fault diagnosis procedure of Riascos *et al.* is based on the online monitoring of variables easy to measure in the system such as voltage, electric current, and temperature. We can notice that, in this work, the fault diagnosis stage is not realized from experimental data but with data given by an empirical static FC model in fault conditions [27]. The problems of data quality related with the sensor accuracies, the possible drift of the FC system performances during data acquisition and other natural imperfections of FC generators are not taken into account in the FC model, which can impact the effectiveness of the diagnostic module. In [27], the authors mention that several tests have been conducted to verify the effectiveness of the diagnosis tool, and that in all the tests performed, the diagnosis process has always indicated the true cause as the most probable one. If the Bayesian theory applied in the work of Riascos *et al.* [27] seems to be particularly promising and adapted for FC diagnostic “in simulation”, the BN approach has to be validated in practice.

In this article, we present a new architecture of impedance spectrometer, which permits diagnosis investigations on large electrochemical devices like FC stacks, and we aim to highlight the capabilities of BN for the diagnosis of real FC stacks. Our impedance spectrometer is described in Section II. Some EIS test results obtained on a twenty-cell PEMFC are presented and analyzed by means of a BN in Section III.

II. Impedance Measurement System Development

A. *Architecture of the spectrometer*

The new impedance spectrometer has been designed using National Instruments materials and Labview® software [28] (Fig. 1). A 1000B PXI chassis is used to implement the different measurement cards. Two types of trigger lines are included in the chassis backplane, which leads to a high precision for the synchronization between the different cards. The synchronization of all digital multimeters (DMM) cards PXI-4071 is guaranteed by a 28.8 MHz internal reference clock. The trigger signal of PXI-4071 cards is produced by a function generator (PXI-5406). An arbitrary function generator (PXI-5406) is connected to the control circuit of the electronic loadbank. The waveform generator can generate five different types of signals (sinus, square, ramp, triangle, and arbitrary) in the voltage range 0-10 V. The electronic load transforms the actuating signal into a regulated load current with DC and AC component parts (galvanostatic operation). To measure and control the current, a high accuracy LEM current sensor ITB-300S is used. The current measured by the sensor is changed into voltage by a high precision resistance (5 Ω, 0.2%). The voltage converted by the resistance is measured by a DMM. The stack voltage is measured directly at the “boundaries” of the assembly. At the same time, the voltages of the cells are acquired indirectly using a multiplexer card (PXI-2527). In the current system configuration, the multiplexer card can switch up to 31 modules / channels (individual cells or groups of cells). Adding two PXI-2527 cards would permit to perform impedance tests on 93 modules with 500 V maximum compliance voltage and 700 V common mode potential.

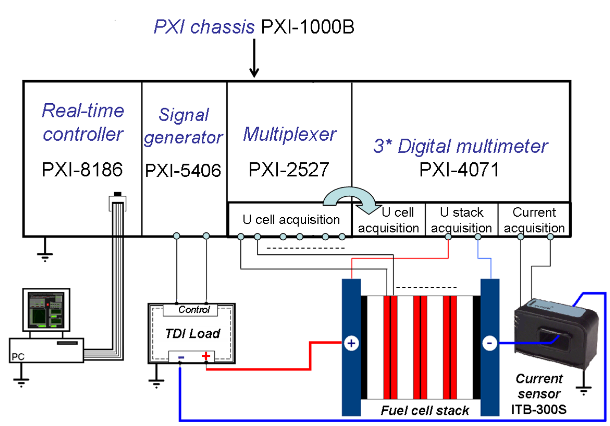

Fig. 1 Scheme of the experimental EIS measurement system.

B. Hardware specifications

As summarized in Table 1, the new high voltage impedance spectrometer exhibits strong capabilities for the characterization and diagnostic of multi-cell PEMFC stacks. With regard to its high compliance voltage and common mode potential, the impedancemeter allows investigating the behavior of FC stacks including more than 100 elementary cells [28]. However, the frequency bandwidth is limited in comparison with other commercial solutions (Zahner IM6 and Solartron Modulab). Nevertheless, the frequency bandwidth exhibited by our system corresponds well with the classical frequency bandwidth of PEMFC generators. Obviously, dimensions, weight and cost were also taken in consideration during the design phase of the instrument.

C. Software development

A specific Human – Machine Interface (HMI) for the control and acquisition of impedance spectra has been developed under Labview Real-Time (Fig. 2). The choice of developing the software on a Real-Time target was done with the goal to optimize the synchronization between the acquisition cards (to reduce error on complex impedance) but also to control acquisition time (deterministic task). Some subroutines have been developed to automatically perform impedance measurements on the cells to be tested. Figure 2 shows a FC test bench and the impedancemeter during the characterization of a PEMFC stack.

Table 1 General specifications of impedancemeters.

Parameters		***Our system***
Current range (A)	AC component (i~)	$7{\cdot}10^{-3} < i\sim \leq 450$
		$I + i\sim \leq 450$
	DC component (I)	$i\sim/2 < I \leq 450$
		$I + i\sim \leq 450$
Voltage range (V_{rms})	Compliance voltage	±700
	Common mode potential	±500
Theoretical frequency bandwidth (Hz)		$6.7{\cdot}10^{-3}$ - $36{\cdot}10^{+3}$
Voltage acquisition way		31
Dimensions [height, width, depth] (mm)		178 × 270 × 379
Weight (kg)		15
Cost (k€)		19.5

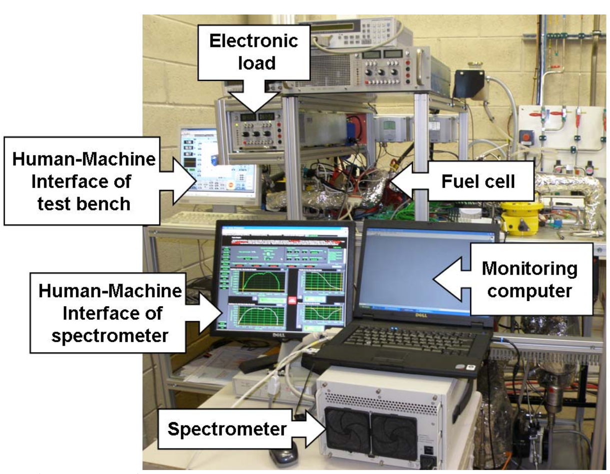

Fig. 2 Impedance spectroscopy measurement on a PEMFC stack

III. Experimental Diagnosis on Twenty-Cell Stack

Bayesian Networks have been extensively applied to fault diagnosis [27]. BN are a probabilistic reasoning formalism introduced by [29]. A BN is a structure that graphically models relationships of probabilistic dependence within a group of variables. The Bayesian approach is particularly adapted where uncertainty, decision and reasoning are problematic. The clear readability of causal links between some variables is a major advantage for the comprehension of physical mechanisms which occur in a system, compared to other formalisms. At the opposite of other formalisms such as neural networks, an integral causality between variables exists. Moreover, BN can be employed with incomplete data bases (due to aberrant measures, crash sensor...) as well as mixed data, that is to say, including the knowledge of human and experimental data. Finally, BN offer the possibility to adapt this network and the probability distribution by a gradual adaptation with new training data. Thus, the network previously reached can be perfectly adapted to changing circumstances over time (as it is the case for FC ageing). Nevertheless, BN is not well suited to applications where the set of data to be analyzed (e.g. experimental data) is limited. In addition, BN use only discretized variables, which is a drawback for continuous values.

To go further into the definition of a BN [30]: $\mathcal{B} = (\mathcal{G}, \theta)$ is a BN if $\mathcal{G} = (X, E)$ is a Directed Acyclic Graph (DAG) whose vertices represent a set of random variables, and if $\theta_i = [\mathbb{P}(Xi/X_{Pa(Xi)})]$ is the matrix of conditional probabilities of node i knowing the state of its parents $Pa(X_i)$ in $\mathcal{G}$. One hypothesis imposed by the Bayesian theory is the following one: for each variable X_i, all variables $Pa(X_i)$ must be such that X_i is conditionally independent from X_j knowing $Pa(X_i)$. A BN $\mathcal{B}$ is a probability distribution on X, for which joint distribution can be simplified as follows:

$$\mathbb{P}(X_1, X_2, \ldots, X_n) = \prod_{i=1}^{n} \mathbb{P}(Xi/X_{Pa(Xi)}) \tag{1}$$

This decomposition of the joint distribution allows obtaining powerful inference algorithms, making of BN modeling and reasoning tools, when uncertainty or incomplete data are present [30]. They are also useful for classification problems where the interactions between different variables can be modeled by conditional probability relations. The construction of a graph to describe a diagnostic process can be executed in two ways:

- Given by an expert. The expert(s) need(s) to know all relationships among variables (the relationship between son and parent nodes).
- Based on probabilistic methods (K2, MWST, SopLEQ… [27, 30]) using databases of records.

The construction of a Bayesian structure $\mathcal{G}$ based on knowledge can be relatively simple; but its performance depends completely on the human expert knowledge about that domain [27]. When dealing with diagnostic applications and classification tasks, some BN structures must be proposed. One of them, the naive Bayes classifier is based on the socalled Bayesian theorem and it is particularly well suited when the dimensionality of the inputs (measurements) is high. Despite its simplicity, naive Bayes can often outperform more sophisticated classification methods [30]. The naive Bayes classifiers make the assumptions that each variable $X_{i,\dots,}\ X_n$ are conditionally independent from the default variables (X_C) (Fig.3). The Bayes naive classifier selects the most likely classification $X_C(n)$ given the values $X_i(n)_{,\dots,}\ X_n\ (n)$.

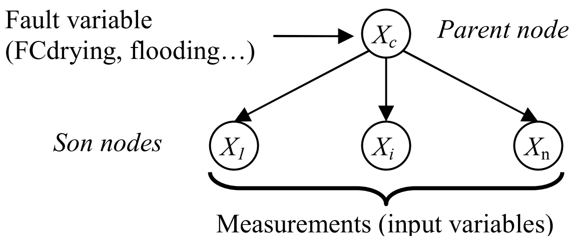

Fig. 3 The example of the naive Bayes classifier.

A. Database generation

In our study, the investigated twenty cell stack is assembled with commercial perfluorosulfonic MEAs, graphite bipolar plates and electrodes with 100 cm² area. The experimental data have been recorded according to a fractional experimental design at 2 levels (Table. 2) [12]. This experimental design deals with the impact of five factors: anodic Relative Humidity (RHa) [35, 75%], cathodic Relative Humidity (RHc) [35, 75%], anodic Stoichiometry Factor (FSa) [1.8, 3], cathodic Stoichiometry Factor (FSc) [2, 3], and FC Temperature (T) [60, 80°C] for a current density of 0.5 A/cm². The aim of this test matrix is to induce different operating states into the FC generator: from drying to flooding state. Some impedance experiments (tests Nr. 1, 2, 4, 10, 12) have not been recorded due to the critical fault conditions induced by these tests on the FC performance. The experiments and EIS measurements have been performed using a 2 kW testbench developed in-lab [31].

Table 2 Fractional experimental design at 2 levels and 5 factor.

Tests Nr.	*RHa (%)*	*RHc (%)*	*FSa*	*FSc*	*T (°C)*	*EIS*
1	35	35	1.8	2	80	*
2	35	35	1.8	3	60	*
3	35	35	3	2	60	X
4	35	35	3	3	80	*
5	35	75	1.8	2	60	X
6	35	75	1.8	3	80	X
7	35	75	3	2	80	X
8	35	75	3	3	60	X
9	75	35	1.8	2	60	X
10	75	35	1.8	3	80	**
11	75	35	3	2	80	X
12	75	35	3	3	60	**
13	75	75	1.8	2	80	X
14	75	75	1.8	3	60	X
15	75	75	3	2	60	X
16	75	75	3	3	80	X

** : Cell(s) voltage(s) were too unstable for EIS .*

*** : The voltage of cell 20 was unstable and below 250 mV.*

In total, 231 impedance spectra were recorded at the boundaries of the FC stack, which represents 21 spectra per experience (stack impedance is measured one time, and then in parallel with each spectrum recorded on the 20 individual cells). Only one spectrum per individual cell and per experience was recorded (220 impedance spectra in total). Due to the fact that BN needs a lot of experimental data to obtain an efficient classifier, we propose to diagnose the FC from the stack spectra only. Some examples of stack impedance spectra recorded on the twenty-cell generator are shown in Fig. 4.

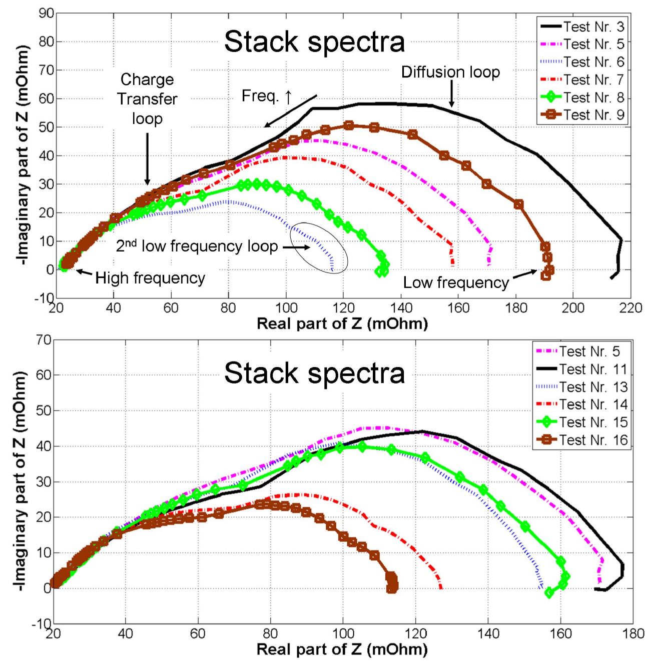

Fig. 4 Examples of impedance spectra recorded on the twenty cell stack.

As shown in Fig. 4, different shapes of stack impedance spectra are recorded following the experiment number and the operational parameters investigated. A linear region can be observed at high frequencies for all the experiments. In this linear region, Faradic processes are not significant, and the effects of both double-layer charging and proton-transfer through the Catalyst Layer (CL) dominate the overall response, which is typical of a porous electrode [32]. The value of 45° originates from the assumption that the proton-transfer resistance is uniformly distributed across the CL [33]. The stack impedances of Fig. 4 are dominated by oxygen diffusion. The decrease of the global FC performances (linked to the evolution of the polarization resistance) can mainly be attributed to the slow increase of the diffusion arc sizes, which is caused by insufficient air flow rates and a gradual accumulation of water inside the FC (especially in the electrodes and GDL at cathode side). Some stack impedance spectra, recorded for tests Nr. 3, 6, 7, 8, 9, 14, 16, exhibit three loops. The high frequency section (~ 10 kHz to 5 Hz) is due to the charge transfer associated with the Oxygen Reduction Reaction (ORR). When air is used as the oxidant gas, there is another low frequency loop (~ 5 Hz to 1 Hz) which is due to oxygen diffusion through the backing layer. The second low frequency loop (~ 1 Hz to 50 mHz) is rarely observed and described in the literature. Freire *et al.* [34] suggest that flooding in the cathode and membrane hydration are both contributing factors.

Taking into account the analysis made previously from the experimental results (Fig. 4) and considering the operational parameters of Table 2, a classification of failure modes can be proposed for the experimental domain explored. Five failure modes are defined: minor drying (tests Nr. 5, 8, 11), moderate drying (tests Nr. 3, 9), slight flooding (test Nr. 16), minor flooding (test Nr. 7, 14), moderate flooding (tests Nr. 13, 15), and a normal mode for test Nr. 6 (Fig. 5).

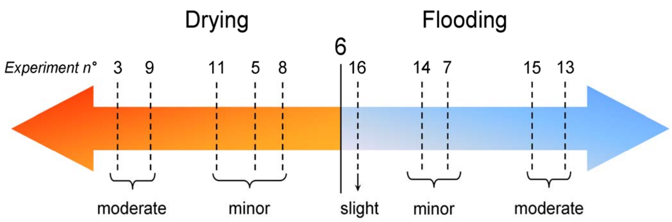

Fig. 5 Classification of failure modes.

B. Inputs of Naive Bayes Classifier

The naive Bayes classifier such as other Bayesian classifiers needs a vector inputs (measurements) to differentiate failure and normal modes. Similarly to the work of Latham [35], a set of multiple frequencies on the stack impedance spectra has been chosen as input variables (son nodes of Fig. 3). The selected frequencies are distributed on the entire impedance spectrum to avoid any duplication of information for frequencies which are very close. In this work, we propose to distribute evenly the frequencies used as input variables across the 42 measured frequencies in the range 5 kHz to 50 mHz. In order to limit the dimensionality and complexity of the BN, only one frequency per decade has been used (Fig. 6), which leads to the following values: 5 kHz, 500 Hz, 50 Hz, 5 Hz, 775 mHz (the frequency at 500 mHz was not measured) and 50 mHz. By considering the complex coordinates (real, imaginary), 12 variables (inputs) are obtained. The fault variable (parent node) includes all FC failure modality (e.g. moderate drying).

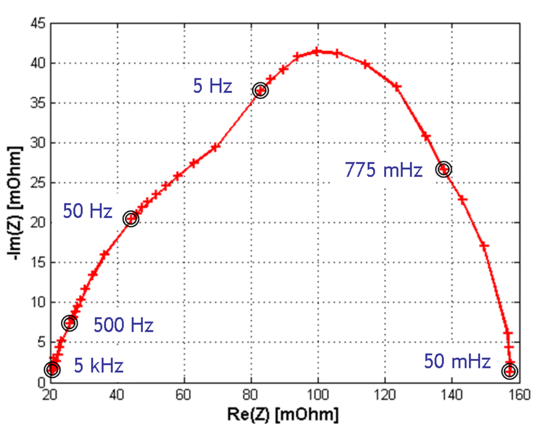

Fig. 6 BN input selection on stack impedance spectrum.

C. Results

Various factors such as Learning Database (LD) size and different methods for discretized input variables, and the effects of input variable number on the rate of good classification (identification of operating mode) have been studied.

Database size variable has an important impact on the right or wrong fault detection. When the LD size is less than 25% of the global database, our results show that the rate of false classification increases rapidly with LD size (e.g. rate of 58% for a LD of 5%). At the opposite, when LD represents more than 75% of the global database, our results indicate that the rate of good classification increases slowly with LD size (e.g. rate of 93.8% for LD equal to 90% and 94.3% for a LD of 95%). However, the same rate of good classification has been reached for a LD between 25% and 75% of the global database.

Different methods to discretize the input variables exist in the literature. In our study, the segmented tree method has demonstrated the best capability for the diagnostic of PEMFC stack. As a result, a maximum rate of good classification equal to 91.2% has been reached, while a manual discretization method and the K-Means algorithm permit to reach rates of good classification equal to 89.7% and 88.2% respectively. The high rate of good classification has been obtained for a learning database representing 75% of the experimental database and for input variables discretized by segmented tree [36].

IV. Conclusion and Perspectives

Our new impedance spectrometer is dedicated to the diagnosis of large electrochemical devices (like FC stacks) composed of a number of elementary cells. In its current configuration, the impedancemeter equipment costs approximately half as much as a commercial spectrometer. It enables the diagnostic of electrochemical sources composed of 31 modules (cells or groups of cells).

To complete the FC diagnosis procedure, the naive Bayesian classifier has been chosen in a first approach. Twelve input variables allow differentiating between 6 operating modes (5 in fault conditions). After a study of parameters that can influence the rate of good classification, an "optimized" classifier has been proposed. It is able to detect 91% of samples included in a validation database. Topics such as the study of fault effects in FC, the construction and improvement of BN structures for fault diagnosis in FC, and their association to fault treatment processes are still under study.

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