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Signal Processing for the Impedance Measurement on an Electrochemical Generator

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Abstract: Improving the life time of batteries or fuel cells requires the optimization of components such as membranes and electrodes and enhancement of the flow of gases [1], [2]. These goals could be reached by using a real time measurement on loaded generator. The impedance spectroscopy is a new way that was recently investigated. In this paper, we present an electronic measurement instrumentation developed in our laboratory to measure and plot the impedance of a loaded electrochemical generator like batteries and fuel cells. Impedance measures were done according to variations of the frequency in a larger band than what is usually used.

The electronic instrumentation is controlled by Hpvee® software which allows us to plot the Nyquist graph of the electrochemical generator impedance. The theoretical results obtained in simulation under Pspice® confirm the choice of the method and its advantage. For safety reasons, the experimental preliminary tests were done on a 12 V vehicle battery, having an input current of 330 A and a capacity of 40 Ah and are now extended to a fuel cell.

The results were plotted at various nominal voltages of the battery (12.7 V, 10 V, 8 V and 5 V) and with two imposed currents (0.6 A and 4 A).The Nyquist diagram resulting from the experimental data enable us to show an influence of the load of the battery on its internal impedance. The similitude in the graph form and in order of magnitude of the values obtained (both theoretical and practical) enables us to validate our electronic measurement instrumentation. Different sensors (temperature, pressure) were placed around the device under test (DUT). These influence parameters were permanently recorded. Results presented here concern a classic loaded 12 V vehicle battery.

The Nyquist diagram resulting from the experimental data confirms the influence of the load of the DUT on its internal impedance. *Copyright © 2008 IFSA.*

Keywords: Fuel cell on load, Impedance Spectroscopy method, Impedance measurement, Nyquist graph.

1. Introduction

Energy and climate perturbations are now well established challenges. The use of hydrogen in fuel cell is an essential vector, and has been the focus of intensive study in recent years as promising alternative energy sources. Thus, various studies have been carried out in the electronic-physic fields of these kinds of generator [1] [2]. The improvement of the effectiveness and the life time of fuel cells require to optimize components such as membranes and electrodes as well as to enhance the fluid exchanges process. Impedance measurement is a possible metrological tool for this kind of characterizations. It's a powerful technique, which can provide useful information on the electro-chemical systems in a real and very short time [3]. This technique can be considered as a good tool to determine the status of charge of batteries or fuel cells.

Impedance measurement on a battery or fuel cell must be done on a load. This is necessary in order to evaluate the performances in real conditions. Fuel cell components that can affect the impedance include current collectors, porous electrodes, the catalytic layer and the membrane [4]. We are interested in developing a real time diagnostic based on impedance measurement of a battery or fuel cell on load. The method is based on the impedance spectroscopy (EIS) of battery or fuel cell from which the Nyquist diagram is plotted.

The analysis and the shape of the diagram can provide information to follow the generator performances on a load. This approach may help in identifying many characteristics of the system as well as the kinetic resistances, the ohmic resistances, the electrolytic, contact and porous layer resistances or the transport limitations of the reactant [3].

The environmental measurement is essential; it allows us to know at what state the impedance measurement was made. To achieve this, several sensors, like temperature, pressure, current, flow of gases, and more sensors recover data and measurements of the DUT.

2. Theoretical Study

2.1. Method

A simulation test under Pspice® was done in order to validate the choice of the method and its ability to provide some accurate and exploitable results. The electrochemical generator is represented by an electronic model given in Fig. 1.

The variable load is represented by a MOSFET, and the internal impedance of the DUT is represented by Randle's circuit (R_{ohm} = 10 mΩ, R_{act} = 90 mΩ, and C_{dc} = 300 µF). These values correspond to what one can expect for a PEM fuel cell. These estimated values are based on the work carried out by Noponen [5], Wagner [6], Brunetto and al. [7]. They have done measurement on PEMFC with the same dimensions as our fuel cell on which the experimental measurements are expected to be done. They found that the real part is ranging from 3 mΩ and 200 mΩ and the imaginary part is close to 15 mΩ.

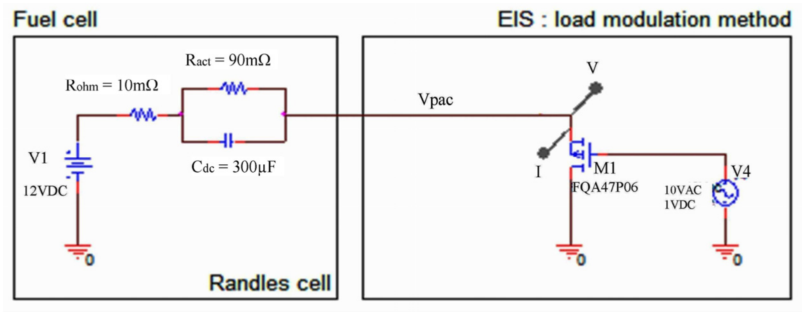


Fig. 1. Electric representation of the load modulation method (simulation).

2.2. Results

The frequency range used for this simulation is the same for the experimental measurement. It is ranging from some mHz to 10 kHz. Fig. 2 shows the result of this simulation in the Nyquist graph, where the imaginary part (Z'') versus the real part (Z') of the complex impedance is plotted. A perfect semi-circle is obtained.

As it can be seen, the negative sign before the imaginary part (Z''). The results of this simulation allowed us to make a comparison with experimental results in order to validate this method and to examine its feasibility.

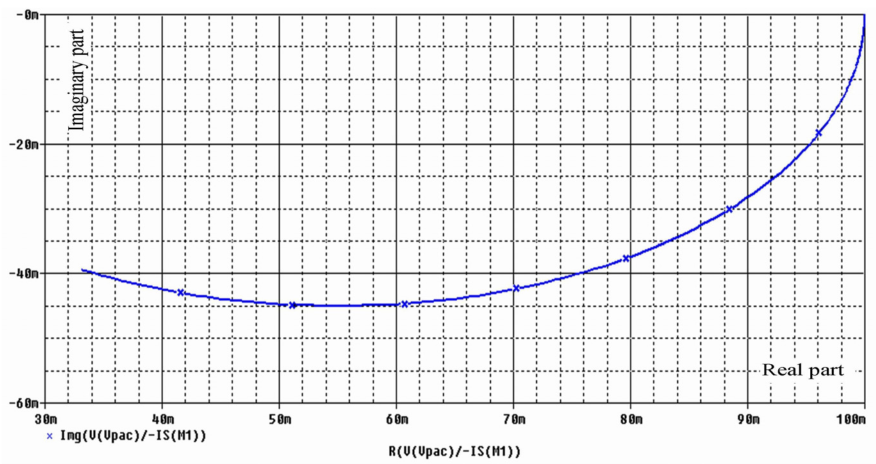


Fig. 2. Impedance Nyquist Graph of the simulated method.

3. Experimental Study

3.1. Methodology

The principle of the impedance spectroscopy is very simple and is shown on Fig. 3. It consists on measuring the electric potential according to the variations of the frequency according to the applied current.

The Impedance Spectroscopy (IS) has the advantage, compared to other methods, to have a less influence on battery or fuel cell during the tests. It can provide more information on the status of the charge. Measurements are generally carried out without load. It is useful to cover a large frequency range in order to obtain more information from the impedance spectrum generated. For a Proton Exchange Membrane Fuel Cell (PEMFC), the impedance spectrum was generated in a frequency ranging from 1 Hz to 10 kHz [8].

However, Walkiewicz et al [9] did studies between 1 mHz and 65 KHz. The number of points collected by decade varies between 8 and 10 points. The principle of measurement is to add a signal, at constant frequency, to the output of the voltage of the battery when it is delivering the desired current. The superimposed signal can be obtained by three methods: potentiostatic, galvanostatic or load modulation method.

Among these three methods, we have selected the load modulation method. It consists in varying the resistance of the load according to the signal that we would like to superimpose. Thus, the impedance of the generator under test can be obtained by the ratio of the voltage of the battery and the current coming from the battery. Fig. 3 shows an electric diagram of this method.

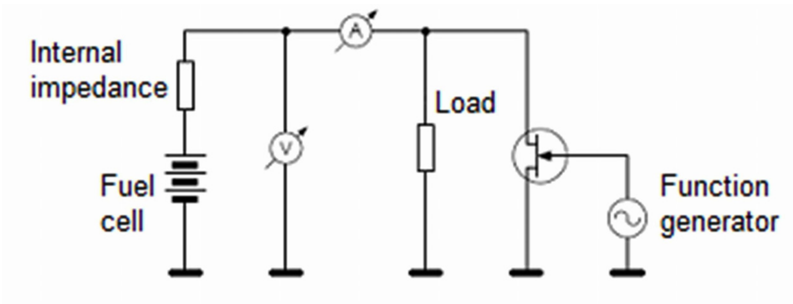


Fig. 3. Electric representation of the load modulation method [10].

3.2. Principle of the Electronic Measurement Instrumentation

The principle of the electronic instrumentation is presented in Fig. 4. The current is controlled by an analogical current regulation. This choice allows us to have a more linear, fast and reliable regulation. The instrumentation uses a VXI system stand, which controls different electronic cards. Software, under Hpvee®, was developed for automatic impedance measurements of the DUT. Two synchronous detections were used to filter the noise and reject the 50Hz. They filter a very narrow band around the processed voltage and current signals. Thus, it is possible to filter all the noise and to detect the amplitude of the useful signal at frequency fixed. These two synchronous detectors are controlled by four square signals for which the differences in phase are 90°. The real and imaginary parts of the DUT impedance are deduced from their outputs by simply using Ohm's law. These two parameters (real and imaginary part of the complex impedance) are then plotted in the Nyquist diagram.

The instrumentation is controlled by dedicated Hpvee® software. This software controls a VXI system stand containing several measuring devices in the form of plug-in circuits: a low frequency generator (HPE1340A), a multimeter (HPE1326B), a 4-Channel D/A Converter (HPE1328A), a 16 ways multiplexer (HPE1351A) and an input/output circuit (HPE1330B).

The instrumentation is composed of six modules. Each module provides signals to the other for an automatic measurement. This modular approach is for the prototype development and will be integrated in one specific card at the end. The card can then be integrated within a vehicle as an embedded system.

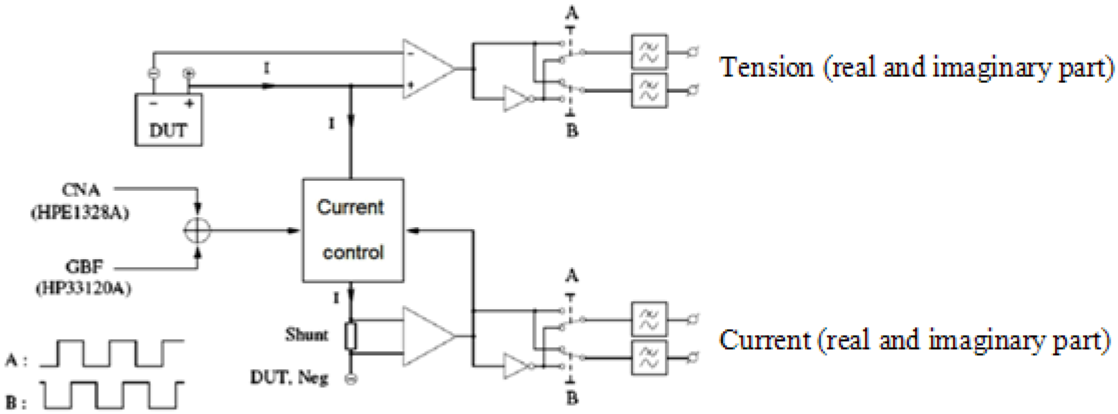


Fig. 4. Principle of the electronic measurement instrumentation.

The "Power supply" module provides the supply to the various electronic circuits, it delivers a tension of +/-13 V and +/-7.5 V. The "Signals generator" module provides the control square signals for the tension and current synchronous detections, as well as the current imposed signal. These signals are generated from the sinusoidal signal delivered by a GBF. The frequency scanning is controlled by the Hpvee® program, which changes the frequency value step by step. The "Current control" module drives the load by an imposed current while running. This current is a square signal, generated by the "Signals generator" module; it is composed of a DC part which represent the imposed current and an AC part which represents the frequency on which this current is imposed. The "Amplification" module amplifies the imposed current signal, measured at the Shunt resistance terminals. This amplified signal, which is disturbed, is transmitted to the synchronous detection (current). The "Synchronous detection" module allows the amplification of the signal coming from the DUT, and the recovery of the real and imaginary part of this signal by the synchronous detection.

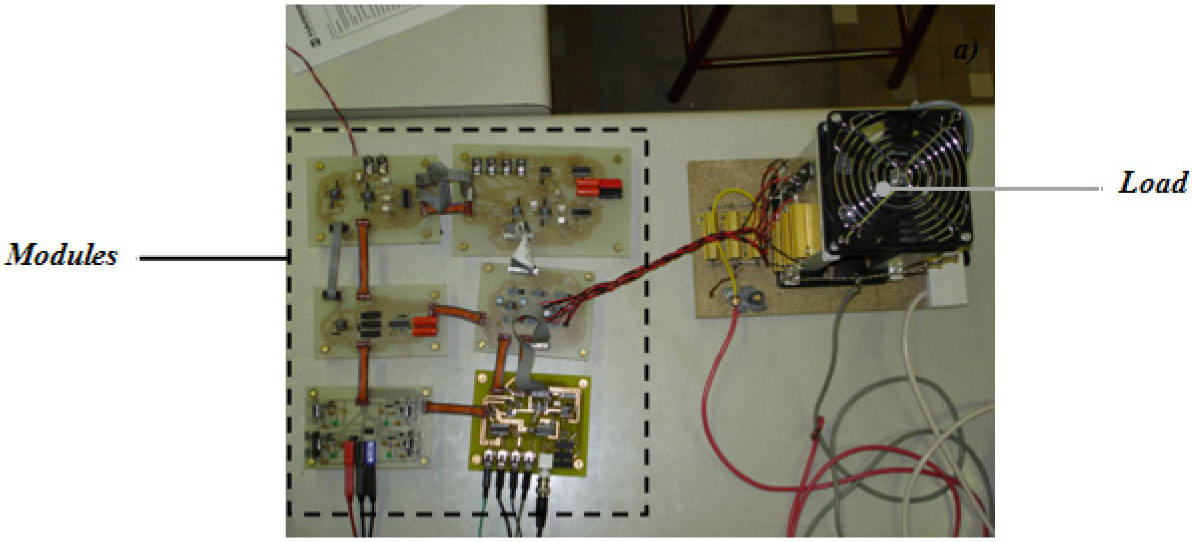
To test the system, we have developed a load which can support current up to 50 A (Fig. 5). All the systems surrounding, as GBF, the computer, the oscilloscope or multimeters, will be replaced in a more evolved version of the instrumentation by components able to provide the same functionalities.

4. Results and Discussion

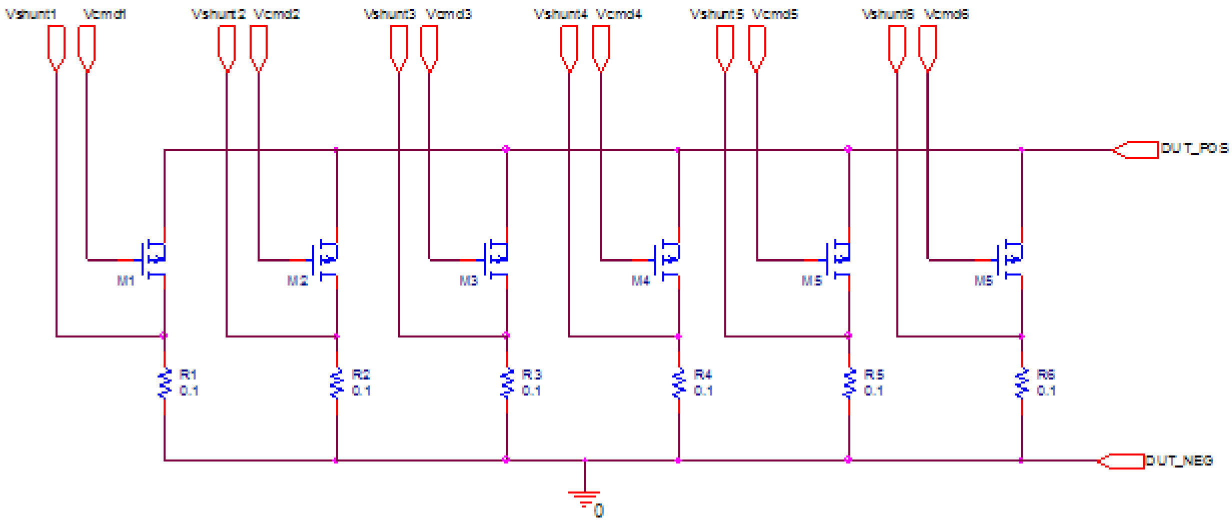
In order to avoid any risk for the fuel cell, preliminary results have been carried out on a classical (and cheaper) 12 V vehicle battery delivering a starting current of 330 A and having a capacity of 40 Ah. The impedance spectrum of a fuel cell and a vehicle battery are very close because the electrochemical processes are almost identical [11] [12]. However, spectrums are very depending on the values of components used in the Randles model. [13].

Measurements were carried out at different nominal voltages (12.7 V, 10 V, 8 V and 5 V) with two imposed currents (0.6 A and 4 A). The choice of these limits current is arbitrary. Fig. 7 and 8 show the complex plane impedance plots (Nyquist diagram) carried out with the electronic instrumentation. Fig. 9 show the complex plane impedance plots carried out with the N3301A Agilent load [8] at a nominal voltage of 12.7 V.

The results obtained enable us to show the influence of the load on its impedance. Nyquist Graphs showed below were obtained by using the Hpvee® software developed for the opportunity, it is then transposed under Microsoft® Excel in order to plot the curves. Nyquist graphs are generally presented in the literature have a positive imaginary axis. Actually, values on the axis of the imaginary part are negative (capacitor effect), but by convention, when the graph is plotted, they are multiplied by -1.



(a)



(b)

Fig. 5. The six modules of the electronic measurement instrumentation with the load (a); the electronic representation of the 50 A load (b).

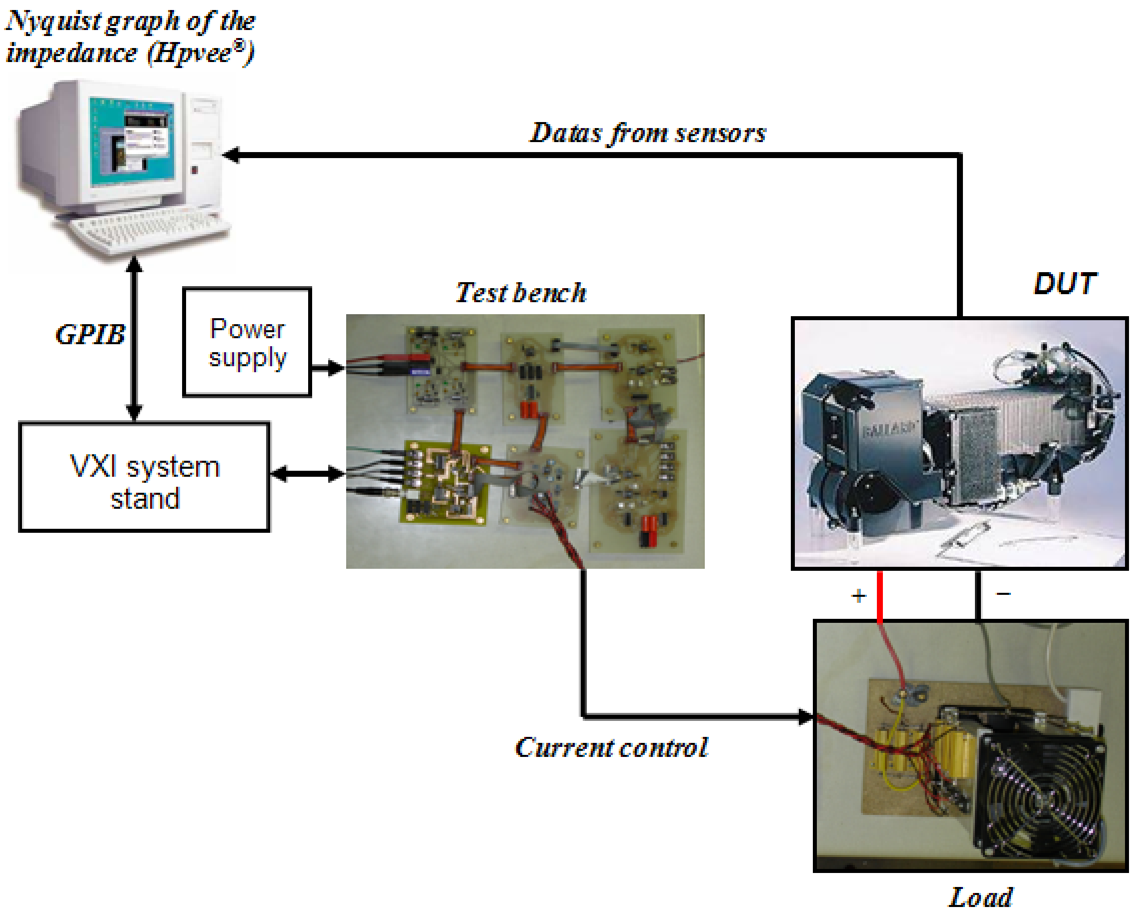


Fig. 6. An overall view of the electronic measurement instrumentation.

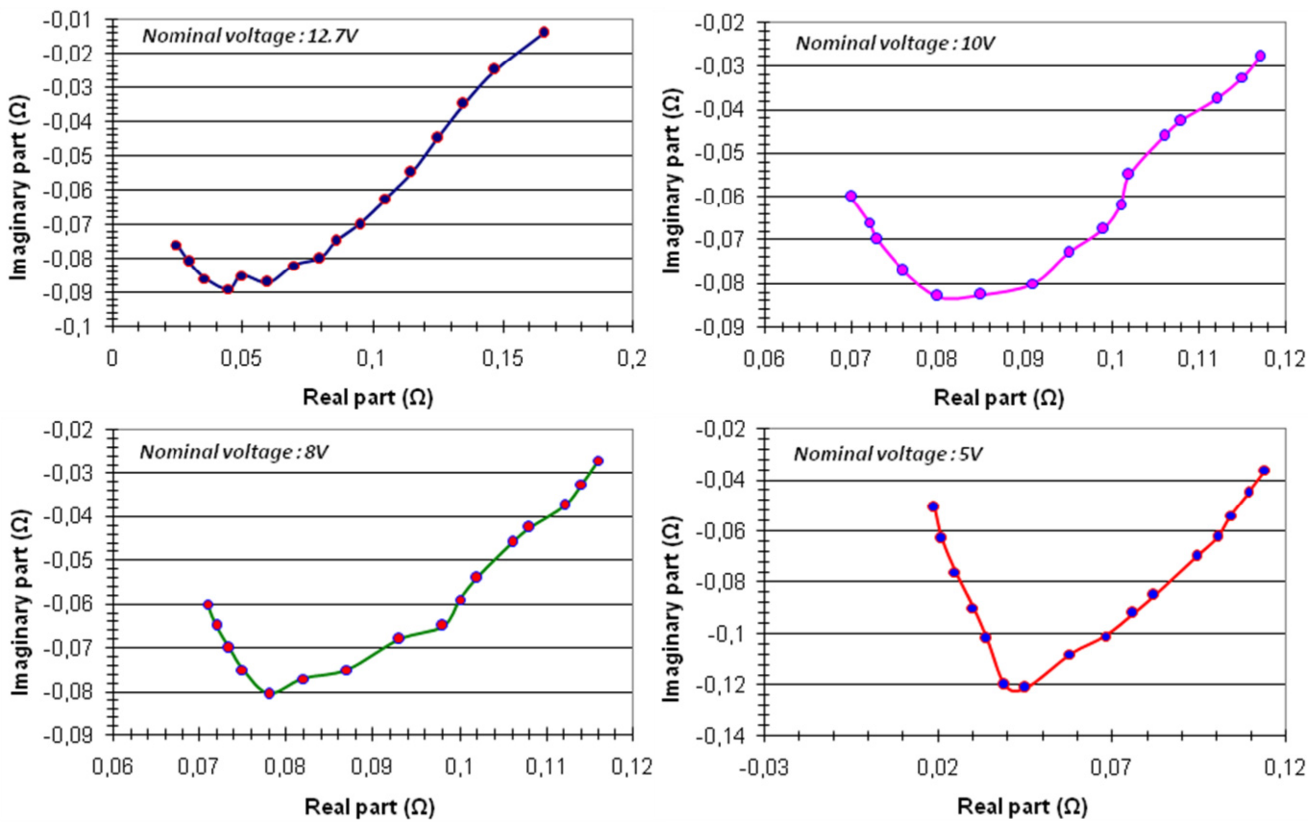


Fig. 7. Impedance of the battery at an imposed current of 0.6 A and at different nominal voltages
The impedance is carried out with the electronic measurement instrumentation.

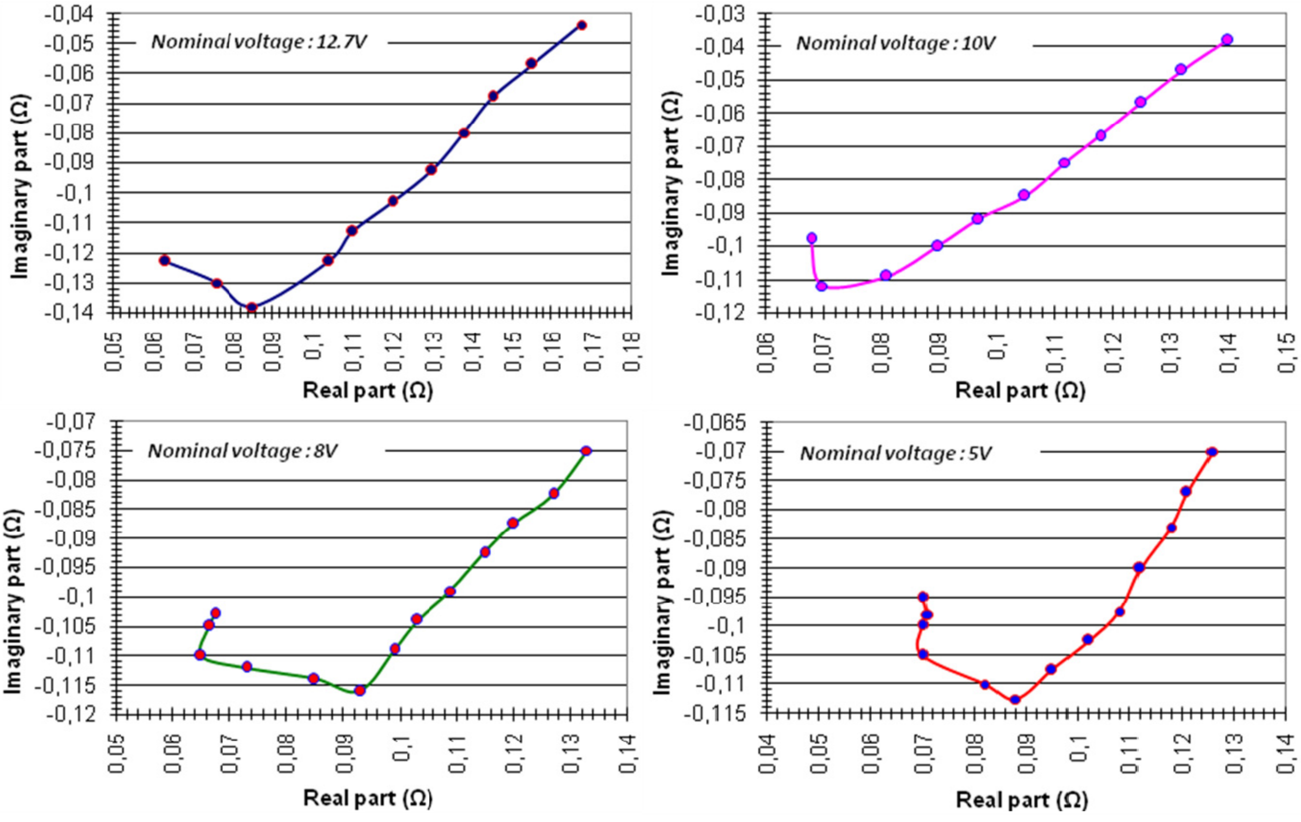


Fig. 8. Impedance of the battery at an imposed current of 4 A and at different nominal voltages
The impedance is carried out with the electronic measurement instrumentation.

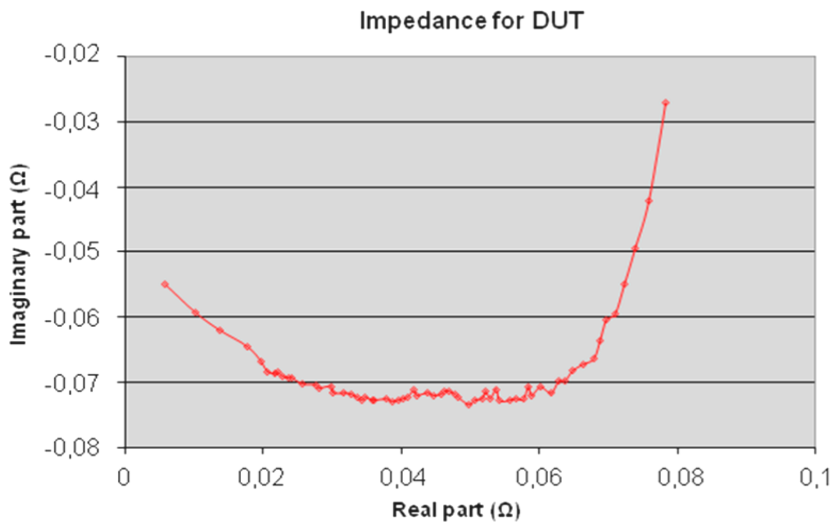


Fig. 9. Impedance of the battery at an imposed current of 4 A and at nominal voltage of 11 V
The impedance is carried out with the N3301A Agilent load.

In order to visualize the different phenomena and the effects occurring inside the battery, basically capacitor effect, we prefer to keep the negative imaginary axis.

As it can be seen, the shape of the curves shown in Fig. 7 demonstrates the ability of our system to measure the impedance of a DUT on load.

The shape of the curve obtained at a nominal voltage of 10 V and at current imposed of 0.6 A is similar to the shape of the curve shown in Fig. 9 using the Agilent load. As it can be seen, the curve become more linear when the nominal voltage of the battery decreases, which means a discharge of this latter? This phenomenon can be seen for a nominal voltage of 5 V (Fig. 7). A pseudo semi-circle obtained if we do not take into account of the right stiffness. The shape of this curve could be due to the weak nominal voltage at which this measure has been made. Below a nominal voltage of 4 V, our system of measure is not more capable to make some correct and exploitable measurement. This could be due to the level of tension drain/source of the Mosfets that must be important enough for measurement.

The experimental curves show the predicted behavior by the theory at low frequencies. Resistive effect is generated by a positive value at the level of the real axis, while capacitor effect by negative value at the level of the imaginary axis. We can also observe a variation of component values, basically resistances of diffusion, with the discharge of the battery.

The second set of measurement at imposed current of 4 A (Fig. 8) show that the curves have the same shape to those obtained with at imposed current of 0.6 A (Fig. 7), however, values of real and imaginary axes are different.

The environment of the device under test (DUT) is controlled by sensors that collect in real time data including temperature, gas flow and pressure.

The control of the environment of the DUT is essential to evaluate their influence on the correlation between the impedance measurement and the performances of the generator. Our next step is to establish this correlation between the impedance of the generator on the load and the humidity. A convenient humidity sensor will be placed in the stack heart.

This correlation will serve to establish a theoretical model that permit to predict the behavior of the loaded generator in real time. If the humidity decreases to much it should be possible to predict this and to stop the process before the deterioration of the membranes and thus the fuel cell destruction.

5. Conclusion

Impedance measurement is a powerful technique, which can provide useful information on the electrochemical systems in a real time very quickly. This technique can be considered as a good tool for batteries or fuel cells diagnosis in real time. The first aim of these tests is to validate our method. This paper presents the principle of measurement and the description of our test bench, as well as, the different electronic cards. Experimental results and simulation are in a good agreement but must be performed now on a loaded fuel cell.

The different Nyquist graphs show that a relationship could exist therefore between the status of the load and the internal impedance of the generator. In the case of the battery, as the one used in this study, the variation of the impedance is generally weak (in the order of milli-ohms) in the operating frequency range. The correlation between the curves obtained with our test bench and those obtained with the commercial Agilent load confirms that our test bench is good spectroscopic impedance instrumentation. It permits to measure and to plot the impedance of a battery or fuel cell versus frequency. The method should be a good tool for fuel cell diagnosis by controlling in real time the behavior of its membranes.

Acknowledgements

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- Smart sensors and systems;
- Sensor instrumentation;
- Virtual instruments;
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