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A method to increase the Fuel Cell internal humidity: application in low consumption vehicle prototype

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ABSTRACT: The water content within the polymeric membrane is essential to have good proton conduction and high efficiency of a fuel cell system. The internal relative humidity becomes more relevant if the fuel cell is used in not stationary applications such as automotive industries.

In this paper a new technique to increase the fuel cell efficiency acting on the internal humidity will be presented. In order to understand the potentialities and the limitations of such technique, the method was studied in a theoretical approach and then applied on a fuel cell (1 kW PEMFC) that supplies the energy for the traction of a prototype vehicle. Finally has been verified that the membrane water content is strictly related to the hydrogen consumption and for some applications in which not life-time but weight and performance need to be considered first (e.g. unmanned aerial vehicles or military single-use equipment), the filling method could be advantageous over humidified system or non-humidified systems.

To ensure the proper success of the procedure is also applied the differential method to fault detection as flooding or drying conditions.

1. Introduction

Fuel cells are comparatively efficient and reliable; they have no moving parts and operate without combustion process, producing high-quality of electric power. During the past years, fuel cells technology has received enormous attention from research institutions and companies as novel electrical energy conversion systems.

Today the fuel cells technology is performed in: automotive propulsion, distributed power generation and in small portable devices (battery replacement).

They use hydrogen as a fuel to drive an electrochemical process that produces electricity, with water and heat as the only by-products. This means that they are not subject to the Carnot Limits (theoretical limit on the efficiency of an engine based on the flow of heat between two reservoirs), and that they can effectively extract more energy from fuel than combustion-based methods.

Traditional internal combustion engines have efficiencies, typically, around 30 %, whereas fuel cells can achieve the 40-70 %. Although this technology was proven as feasible, there are some problems that must to be solved: the water management (from technical point of view), the design and the costs, first of all for automotive applications.

In this research a new humidification method is studied, implemented and applied on a Fuel Cell PEM (PEMFC) of 1 kW power (Tab. 1), in order to improve the proton conductivity of the polymeric membrane, increasing its humidity. Starting from the basic definitions and equations, the proposed technique and the differential method is applied on the stack. Then, the experimental results are presented and the advantages of the “Filling” method are discussed.

1 kW PEMFC features	
Nominal Power [W]	1000
Number of cells	40
Active area [cm ²]	61
Nominal Current [A]	35

Table 1

If all the energy from the hydrogen fuel, its “calorific value”, heating value, or enthalpy of formation, is transformed into electrical energy, then the EMF would be given by:

$$E = \frac{-\Delta h_f}{2F} \quad (1)$$

For higher heating value HHV $E=1.48$ V while for lower heating value LHV $E = 1.25$ V.

These are the voltages that would be obtained from a 100% efficient system, with reference to the HHV or LHV. [Ref.1]

The actual efficiency of the cell is then the actual voltage divided by these values, or:

$$\text{Cell efficiency} = \frac{V_c}{1.25} \cdot 100\% \text{ (with reference to LHV)} \quad (2)$$

However, in practice it is found that not all the fuel that is fed to a fuel cell can be used and some fuel usually has to pass through unreacted.

A *fuel utilization coefficient* can be defined as:

$$\mu_f = \frac{\text{mass of fuel reacted in cell}}{\text{mass of fuel input to cell}} \quad (3)$$

This is equivalent to the ratio of fuel cell current and the current that would be obtained if all the fuel were reacted. The *fuel cell efficiency* is therefore given by:

$$\eta = \mu_f \cdot \frac{V_c}{1.25} \cdot 100\% \quad (4)$$

It is also necessary to define the *stoichiometry coefficient* λ as:

$$\lambda = \frac{\text{air flow through the PEMFC}}{\text{air stoichiometry flow}} \quad (5)$$

The water content in the membrane influences the performance of the polymer electrolyte in a significant way.

In this paper a method to achieve an adequate membrane humidification is presented.

The technique was performed on a Fuel Cell that has been in non-operational state for more than 3 months. The hydrogen supply is in a dead-end mode with periodic manual purging to release impurities and the procedure is relatively simple. When Fuel cell has no load applied on it, the anode side is filled with demineralized water. After few seconds the same side must be purged. At this stage is necessary to ensure that the amount of water removed is equal to the water introduced inside the PEMFC. Then, the procedure is repeated more times (it depends on the membrane humidification to be obtained).

This method should be performed with caution to avoid the flooding problem because a relative humidity too high produces accumulation of liquid water on the electrodes, which can become flooded and block the pores, this makes difficult the gas diffusion. When an electrode is flooded, water forms a thin film that prevents the reactant gas from touching the catalyst and so to react. This phenomenon leads to a lower active area and thus a higher current density. Ref. [2, 3]

To show if the flooding occurred, the filling technique is combined with others methods which ensure the proper success of the filling such as the *differential method*. Ref. [4]

Any fault induces a voltage drop [Ref. 5]. Hence, cell voltage is a good failure indicator, easy to implement. Water forms at the cathode, that frequently flooding; if a stack keeps working in cathode flooding conditions, water starts accumulating and a fast diffusion from cathode to anode can lead to membrane perforation.

Otherwise in high-temperature operating conditions, the electrolyte membrane can dry out.

When the water content in a MEA reduces drastically, temperatures can rise rapidly, leading to serious damages. Just as flooding, drying effects on a single cell cannot be detected monitoring the entire stack voltage in operational conditions. This can be done with the differential method.

In particular Ref. [6, 7] show that flooding or drying only affects some cells or groups of cells. For example, the water produced by the electrochemical reaction can easily accumulate at the outlet of the stack. In Park's drying experiments on 8 cells stack, the most affected cells were 4th, 5th and 6th [Ref. 8].

Hence a flooding may occur in the inlet and outlet of the stack while a drying may occur in the center of the stack. This is the reason why, instead of monitoring all the cells, it could be useful to monitoring only a group of cells, for example, cells located at the inlet and outlet in order to detect a flooding, and cells located in the center of the stack for detecting a drying situation.

Thus three main areas of the stack are instrumented with voltage sensors: the inlet, the center and the outlet. The detection principle is based on the monitoring of the differential between the voltage in the center ΔV_{center} and the inlet/outlet voltages ΔV_{in} and ΔV_{out} respectively.

This principle allows to generate two new state-of-health indicators of the PEMFC stack:

$$\Delta V_{\text{diff,in}} = \Delta V_{\text{in}} - \Delta V_{\text{center}} \quad (6)$$

$$\Delta V_{\text{diff,out}} = \Delta V_{\text{out}} - \Delta V_{\text{center}} \quad (7)$$

In case of no fault, all voltages are constant or drop similarly due to load variation: the differential voltage is more or less equal zero.

If flooding occurs at the inlet/outlet, ΔV_{in} or ΔV_{out} drops and $\Delta V_{\text{diff,in}}$ or $\Delta V_{\text{diff,out}}$ becomes negative.

The feedback information is simple, fast and based on the real-time operating conditions without adding any external disturbances.

2. Experimental part

The National Instruments equipment was used for the acquisition of the voltages. The input signals are acquired, conditioned, buffered and then sampled by a single 12-bit ADC. The only restriction provided by the module is that the maximum voltage between ground and each single-ended voltage channel must be below 10 V.

The PEMFC of 1.0 kW has 40 cells and 41 cooling channels. Figure 1 shows, in particular, the disposition of the pins into the cooling channels in order to have 6 cells monitored in the inlet, 6 in the center and 6 in the outlet areas of the stack.

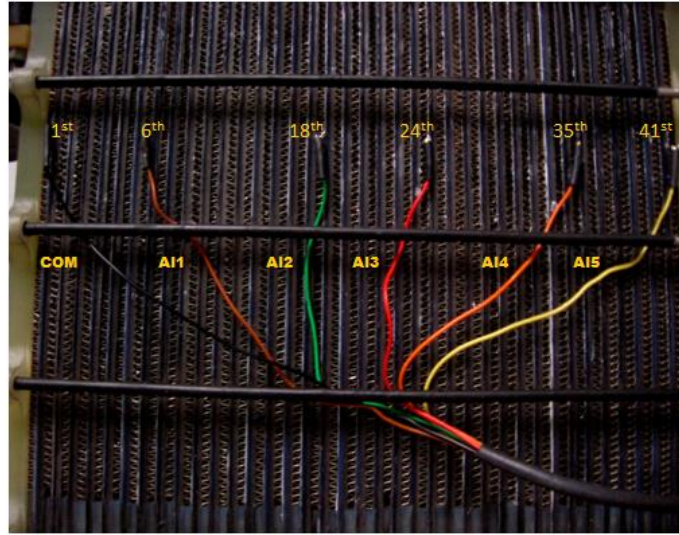


Figure 1 Pin connections from the PEM fuel cell and the corresponding number of the cooling channel

The OCV (Open Voltage Circuit) of the stack (almost 40 V) is well above the maximum input voltage for the module; then a voltage divider has been made (Fig. 2).

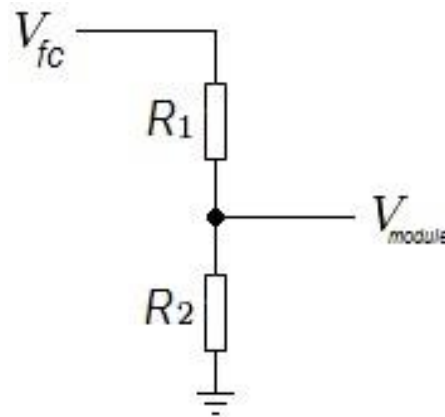


Figure 2 Voltage divider scheme for a single wire

First, the transfer functions were calculated, assuming the maximum voltage over the entire stack to be 40 V. Using the tension divider formula, the transfer functions can be expressed by:

$$H_n = \frac{V_{module,n}}{V_n} = \frac{R_{2,n}}{R_{1,n} + R_{2,n}} \quad \text{with } n = 1, 2, 3, 4, 5 \quad (8)$$

Where: $V_{module,n}$ is the generic voltage after the voltage divider (module input) and V_n is the generic actual voltage of a group of cells. Both voltages are referred to the common ground. Table 2 reports the values of the resistances used.

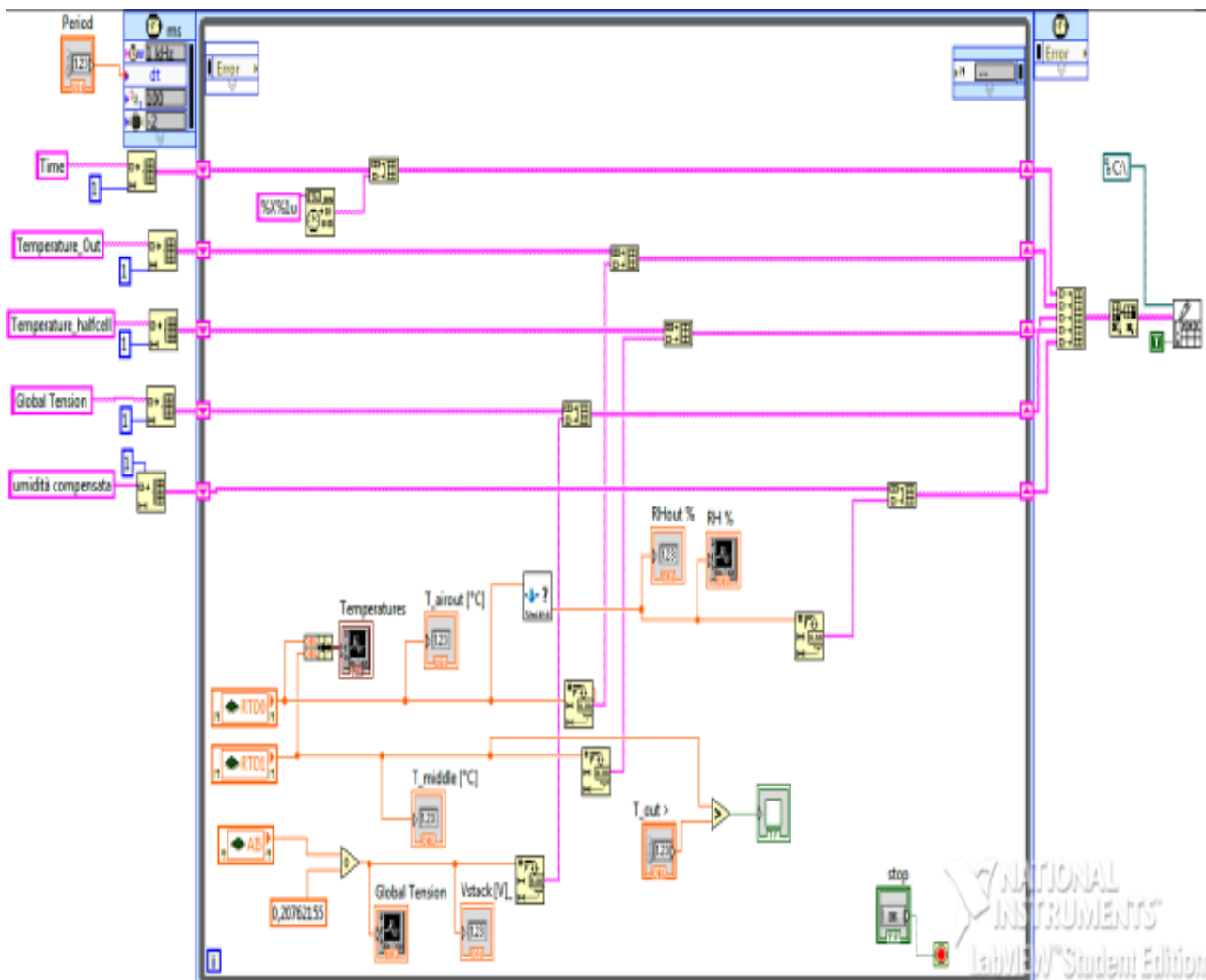


Figure 3 Virtual Instrument for the voltages acquisition

	R_1 [k Ω]	R_2 [k Ω]	H
A11	-	-	1
A12	52,2	52,3	0,500
A13	60,3	31,7	0,345
A14	52,2	15,8	0,232
A15	60,3	15,8	0,208

Table 2 Resistances used for the voltage divider and relative transfer coefficient

Regarding the software programming, the Virtual Instrument (Fig. 3) performs the following operations in a while loop:

- Acquires the module tensions (automatically referred to the common ground)
- Divides each of this tension by the corresponding transfer coefficient, obtaining the fuel cell real voltages:

$$V_n = \frac{V_{module,n}}{H_n} \quad (9)$$

- Computes V_{in} , V_{center} and V_{out} as follows:

$$\Delta V_{in} = V_{module,1} \quad (10)$$

$$\Delta V_{center} = V_{module,3} - V_{module,2} \quad (11)$$

$$\Delta V_{out} = V_{module,5} - V_{module,4} \quad (12)$$

- Calculates the differential voltages using Eq. (6) and (7).
- Builds up string vectors containing time: ΔV_{in} , ΔV_{center} , ΔV_{out} , $\Delta V_{diff,in}$ (V inlet - V center), $\Delta V_{diff,out}$ (V outlet - V center).

In order to check the capability of the differential method to recognize fault condition, first of all, it was applied on the fuel cell that has been in non-operational state for more than 3 months.

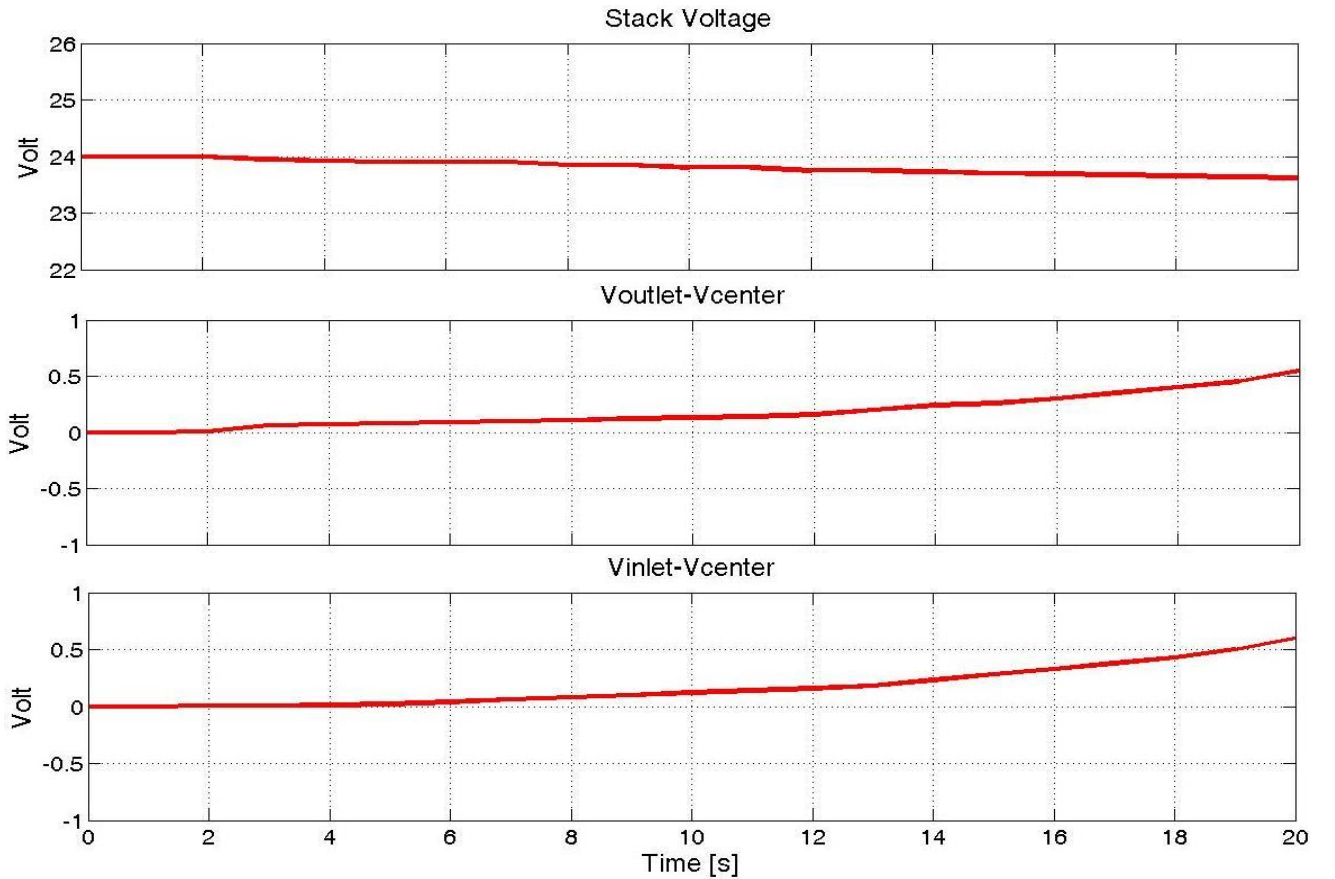


Figure 4 Stack with polymeric membrane dry

Figure 4 shows a drying case obtained by experimental tests carried out at constant current (7 A). In this situation, because the polymeric membranes of the center cells are drying, their resistances increase resulting in a voltages drop of the center cells only. The results are reported in figure 4 where it is possible to observe that the global stack voltage does not drop significantly, but thanks to the differential voltages, the fault can be detected and the stack is disconnected after about 20 seconds of operation.

The first filling cycle was performed using a pump to inject water inside the anode compartment as shown in Fig. 5. The initial temperature of the stack was 30°C. The 3-way valve needs only for switching the operative fluid (water) or gas (hydrogen).

Defining '*residential time*' (*RT*) as the time in which the water remains inside the stack, if the *RT* is too long the flooding condition may occurs, therefore for the considered fuel cell the *RT* is about 15 seconds. Thus after this time the anode side is purged with hydrogen (max hydrogen pressure for our Fuel cell system is 0.5 bar), paying attention to remove completely the water inside the channels for the hydrogen supplying. To avoid hydrogen waste the nitrogen for the purging phase has been used. The water injected, completely fills the anode side and must be the same quantity of the water recovered with the purging phase.

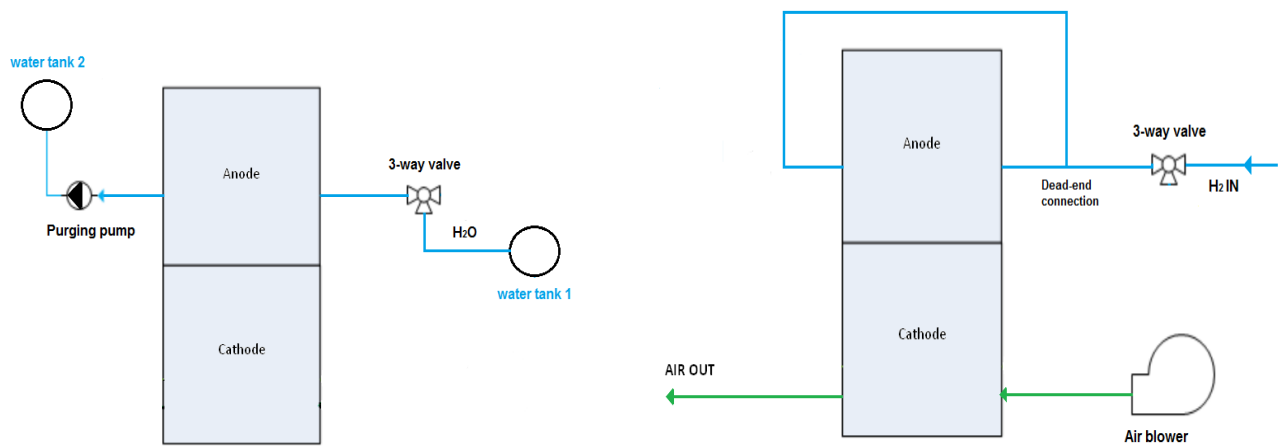


Figure 5 Filling scheme (left side) and operative scheme (right side)

The *residential time* (15 seconds) is not a mandatory time but it is the result of many tests conducted on the same stack in order to understand the optimal *RT* and avoid flooding conditions. A *residential time* greater than 30-35 seconds leads often an inconvenient excess of water in the anode compartment. Probably the *RT* value depends on the membrane thickness and the channels size and it could be different for another PEM Fuel cell. Further tests may clarify the relation between the residence time and the Fuel Cells features.

Directly after the 1st water filling cycle, the differential method is applied on the stack to verify its state of health during a transient. Figure 6 shows that there are no cells perturbed more than others and thus their voltage drops are similar, generating a constant differential voltage which means a filling technique correctly operated.

The filling procedure was performed 3 times and each time was used the differential method to see if the flooding occurred.

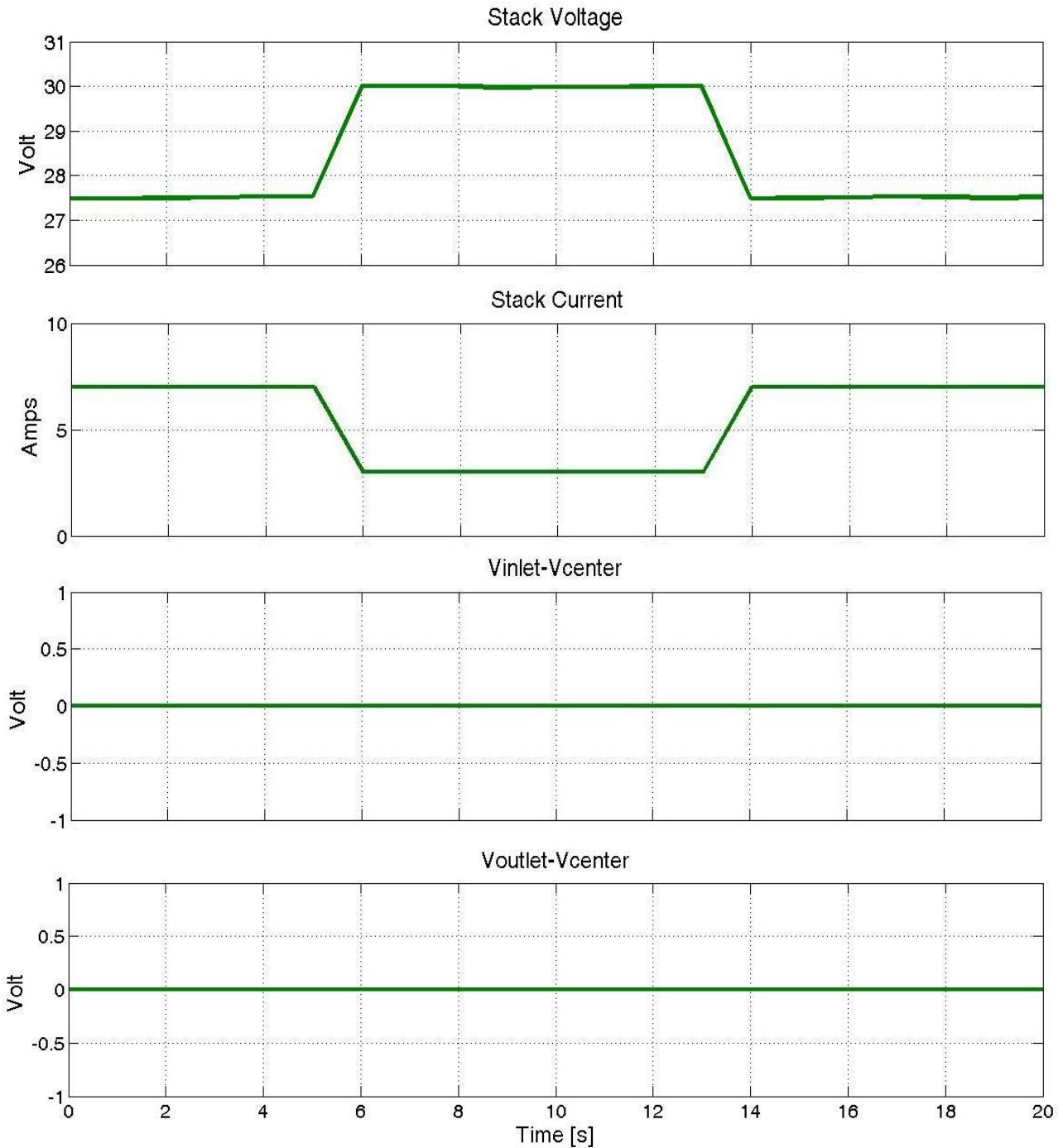


Figure 6 Healthy stack with current variation after 1st Filling cycle

Sometimes, after the purging phase, there may still be water inside the channels for the hydrogen supplying. This problematic issue can be detecting using the differential method and fix it operating once the pump. As shown in Fig. 7, the global stack voltage does not drop significantly. On the contrary monitoring the three groups of cells makes it possible to detect the early fault.

In Ref. [9], is explained also, that a common method for removing excess water inside the PEMFC is using the reaction air flowing through it and the correct variation of the air stoichiometry λ would maintain the RH_{out} (outlet relative humidity) proximal to saturated conditions. In our tests we don't take into account the possibility to control the blower fan speed as a humidity function so there is no control between humidity and air flow.

The stoichiometry coefficient influences both the availability of oxygen as well as the humidity of the membrane. A low λ limits the availability of oxygen because the air is depleted of oxygen when it reaches the end of the airflow channels.

In Ref. [10] the performance of a PEMFC with different level of fuel and air stoichiometry utilization has been tested. However the maximum efficiency, defined as above, occurs at about 80% of fuel utilization (H_2) and 25% of air utilization.

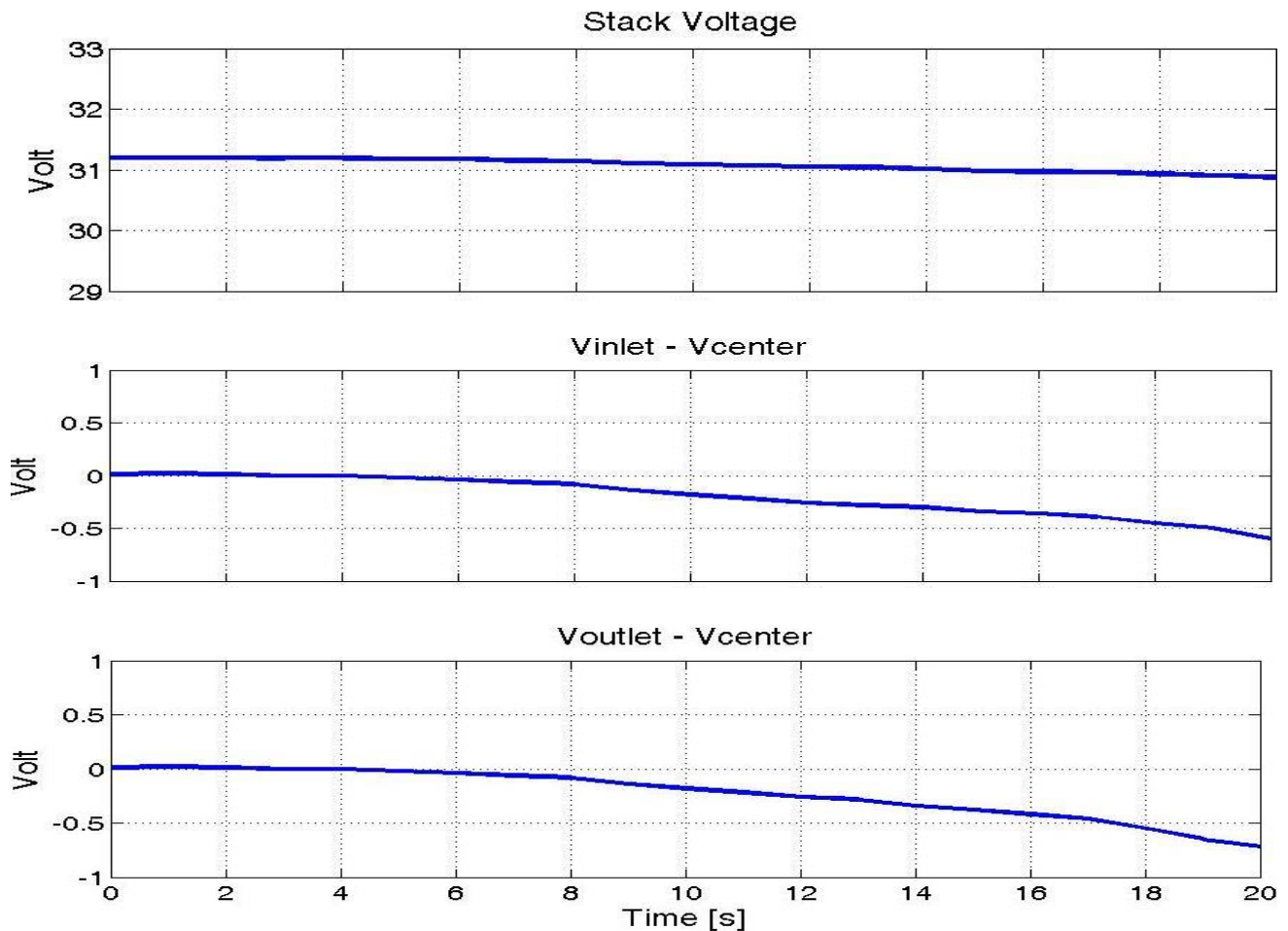


Figure 7 Flooding case

In the Fig. 8 are shown the different polarization curves measured at the start (when no filling is performed and $T_{\text{initial stack}} = 20\text{ }^{\circ}\text{C}$) and after each filling procedure. The voltage increasing, is due to a better humidification of the polymeric membrane thanks to the water filling technique.

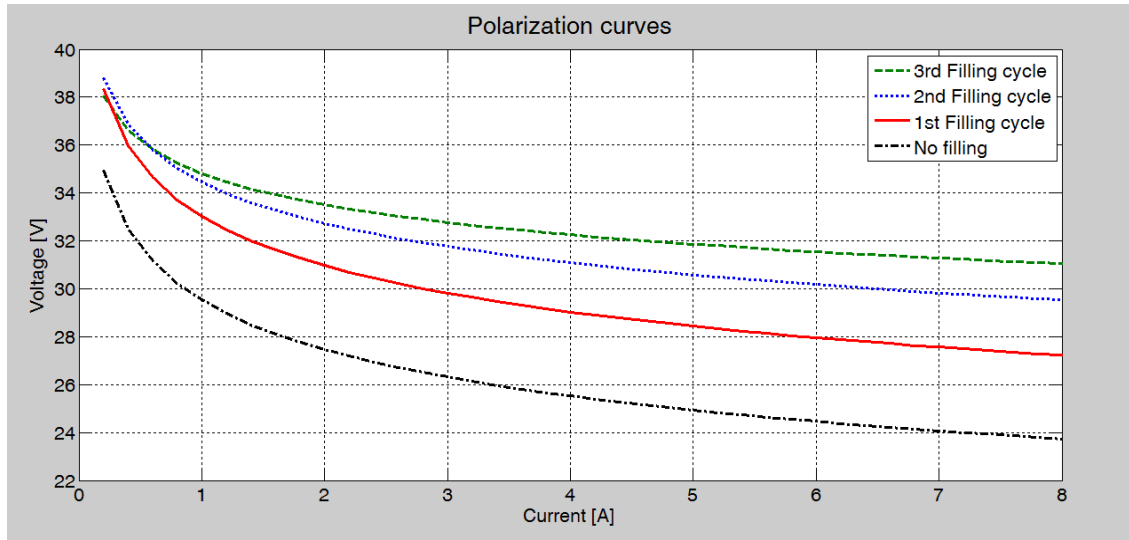


Figure 8 Polarization curves at different Filling cycles

3. Results and discussion

The time needed for both one filling cycle and flooding check (using the differential method) is about 5 minutes. There's no rule to decide precisely how many filling procedures can be execute (3 filling cycles in our case). Of course, higher is the number of filling cycles higher is the possibility to get flooding conditions. Every time that the filling technique is performed there is a total voltage increasing as shown in figure 8.

In order to have an indicator of the maximum filling cycles possible, we define a new parameter " ζ ":

$$\zeta_j = \frac{V_j(I) - V_{j-1}(I)}{V_1(I)} \cdot 100 \quad \text{for } j=2, \dots, n \quad (13)$$

Where $V_j(I)$ is the j -th polarization curve (function of the current), n is the number of the polarization curves (4), ζ is the *filling increasing rate* (FIR) end $V_1(I)$ is the polarization curve with no filling (black curve in Fig. 8). In order to make a simple FIR calculation, the current is fixed at 7 A, thus the $V_j(I)$ functions become a voltage values $V_j(7A)$.

Then is possible to extrapolate from Fig. 8, 4 different voltage values $V_j(7A)$ (one for each polarization curves) and calculate the *filling increasing rate* (Table 3).

	$V_j(7A)$ [Volt]	$V_j(7A) - V_{j-1}(7A)$	ζ
j=1 - No filling curve	24	-	-
j=2 - 1 st filling curve	27.6	3.6	15 %
j=3 - 2 nd filling curve	29.8	2	8.3 %
j=4 - 3 th filling curve	31.3	1.5	6.3 %

Table 3. Filling increasing rate calculation

The FIR value and the outlet humidity plot (Fig. 9) are good indicators of the membrane state humidification. The maximum number of filling cycles is reached when the FIR value is minor than 8 %. Also in this case the threshold FIR value is not a mandatory and it depends on the PEM Fuel Cell.

Applying Eq. (4), and considering $\mu_f = 0.95$, it is possible to calculate the efficiencies related to the 7 A of current, as following explicated:

$$\eta_{\text{No Fill}} = \mu_f \cdot \frac{(V_1(7A))/40 \text{ cells}}{1.25} \cdot 100 \% \approx 46 \% \quad (17)$$

$$\eta_{3\text{rd Filling cycle}} = \mu_f \cdot \frac{(V_4 (7A))/40 \text{ cells}}{1.25} \cdot 100 \% \approx 60 \% \quad (18)$$

Concerning the Eq. 17 and 18, the numerator is a voltage average and there is an efficiency increase of 14%.

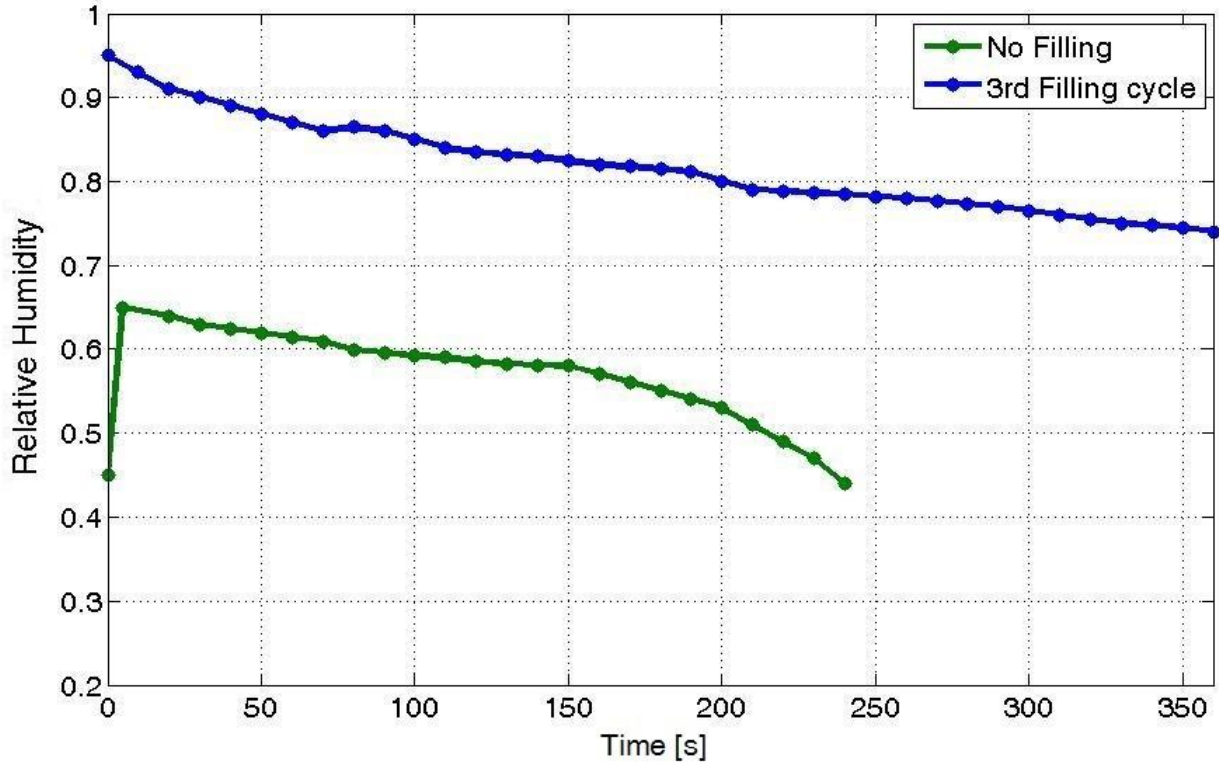


Fig. 9 Relative humidity differences between filled and unfilled PEMFC

Figure 9 illustrates the outlet humidity when the Filling technique is operated 3 times and when it's not (black curves in the Fig. 8). Both tests were executed at constant current of 7 Ampere.

Considering the curve with no Filling (green curve in Fig. 9), the fast humidity decreasing after 150 seconds is due to a high temperature generated by the Fuel Cell (initial conditions: Time= 0 s, $T_{amb} = 22\text{ }^{\circ}\text{C}$ $RH_{amb} = 50\%$, constant during the tests; final conditions: Time= 243 s, $T_{stack} = 58\text{ }^{\circ}\text{C}$ $RH_{outlet} = 43\%$). The humidity behavior depends on the variation of the membrane resistance which increases the temperature, sometimes above the recommended limit.

Also in Ref. [11, 12] the resistance variation was associated with fault detection of flooding and drying. Completely different is the humidity trend of the output air when the 3rd water filling is performed. The humidity decreases slowly as consequence of a hydrated membrane and it is maintained between 95 % and 75 % for more than 5 minutes (initial conditions: Time= 0 s, $T_{amb} = 21\text{ }^{\circ}\text{C}$ $RH_{amb} = 50\%$, constant during the tests; final conditions: Time= 360 s $T_{stack} = 51\text{ }^{\circ}\text{C}$ $RH_{outlet} = 74\%$).

Concerning the results shown in Fig. 9, there's no air flow adjustment during the test and the λ value is kept constant to 8.

One goal of the technique proposed is to demonstrate the filling technique capability to increase the membrane humidity, thus the current request during the tests is quite low and in consequence there is a low water production by the reaction. A second reason for such low current is because 7 Ampere is the typical current provided to drive the prototype.

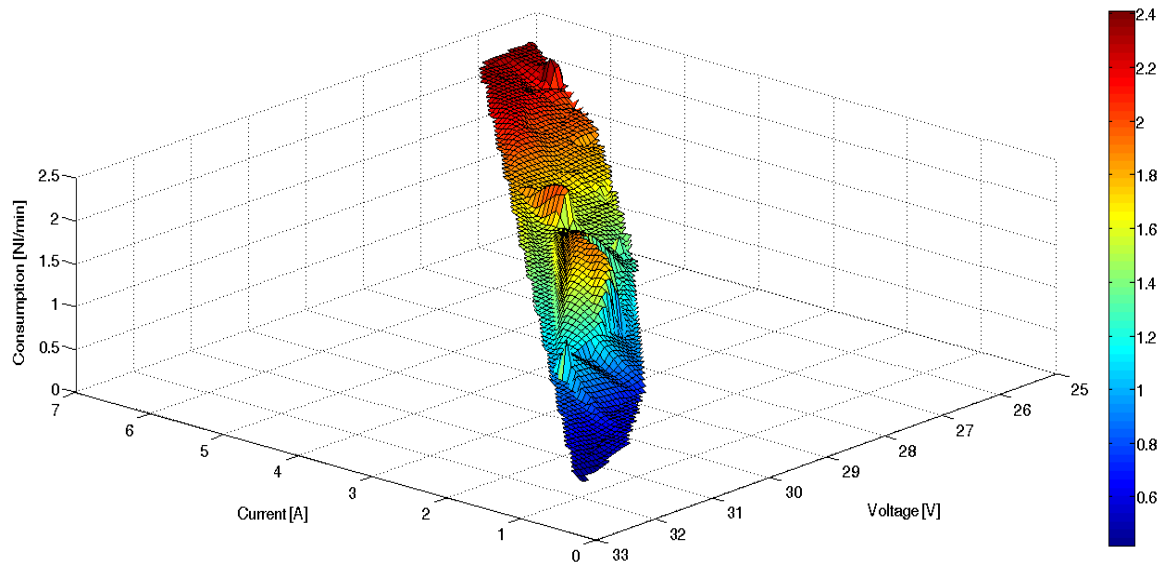


Figure 10 Current, voltage and hydrogen consumption before the filling technique

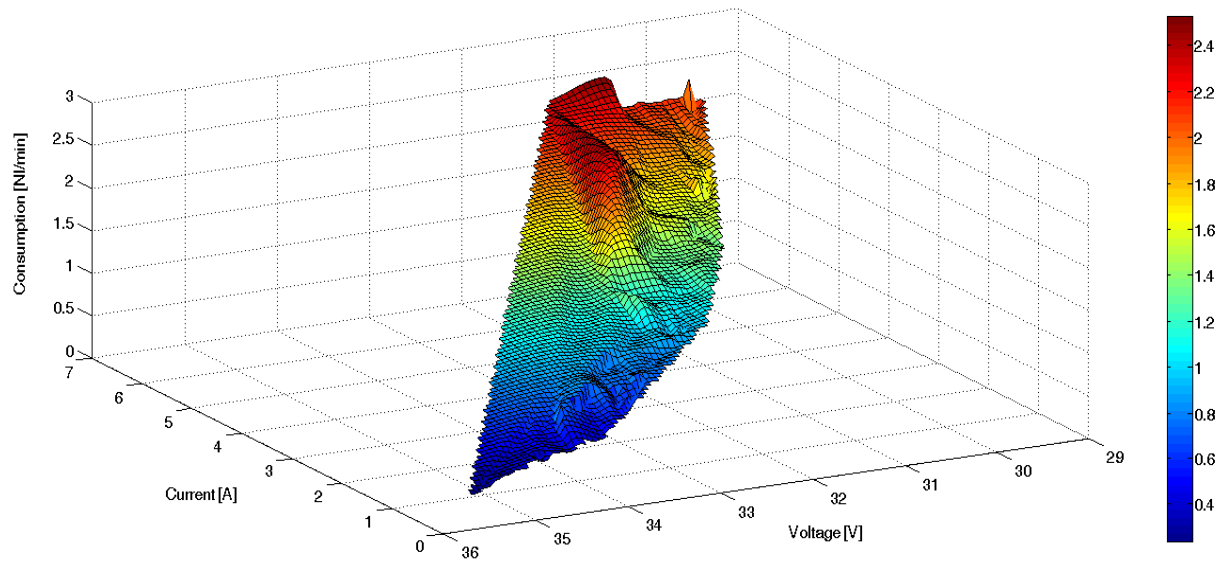


Figure 11 Current, voltage and hydrogen consumption after the 1st filling cycles

Figures 10 and 11 illustrate a 3D polarization curves having the consumption as 3th dimension. The effects of the filling technique, in terms of hydrogen consumption, before the 1st water filling (black curve in the Fig. 8) and after respectively are shown. There are evident differences; in fact Fig. 10 shows a profile with more consumption peak. The conclusion is that the peaks can indicate a low polymeric membrane humidification (the relative humidity of the outlet air was between 50% and 60%) but further experimental analysis will be made in order to understand what is the meaning of these consumption peaks.

The data shown in Fig. 10, 11 was obtained using the MATLAB software and the following instructions:

```

- current = [...];
- voltage = [...];
- consumption = [...];
- xlin1=linspace (min (current), max (current), 30);
- ylin1=linspace (min (voltage), max (voltage), 30);
- [C1, V1] =meshgrid (xlin1, ylin1);
- F1= griddata (current, voltage, consumption, C1, V1,'cubic');
- surf (C1, V1, F1);

```

The first 3 instructions are the experimental data of the current, voltage and consumption respectively. Considering the same hydrogen consumption it is possible to make a comparison for different current condition, and the results are shown in the Tab. 4.

NO FILLING TECHNIQUE			
Current [A]	Voltage [V]	Consumption [L/min]	Power [W]
4	29	1,4	116
5	27	1,8	135
6	26	2,2	156
WITH FILLING TECHNIQUE PERFORMED (1 st filling cycle)			
Current [A]	Voltage [V]	Consumption [L/min]	Power [W]
4	32	1,4	128
5	31	1,8	155
6	31	2,2	186

Table 4 Filled and unfilled PEMFC data

Analyzing the results it's clear that a high membrane humidification is essential to reduce the hydrogen consumption because there is a voltage increasing at fixed current. The currents considerate are the most used for the PEM considered in this study and for the type of automotive application.

3.1 IDRA*pegasus* powertrain system

IDRA*pegasus* is a carbon fiber monocoque (Fig. 12) optimized to reach low consumption results in extreme competition as the Shell Eco-Marathon. The shape was chosen in according to aerodynamic, mechanical packaging and powertrain layout constrains. In order to test the filling technique potentiality and limitations it was performed before the race, conditioning the 1 kW fuel cell in a properly way. An Actuator Control Board (ACB) manages the blower air flow and the current request considering as fundamental parameter the outlet relative humidity. The ACB decides also the best stoichiometric coefficient taking into account both cooling and drying effect. [13, 14]

Figure 13 shows the application of the fuel cell inside the vehicle. The current generated supply an electric motor (200 W) that gives the motion at the rear wheel.



Figure 12 IDRA*pegasus* prototype

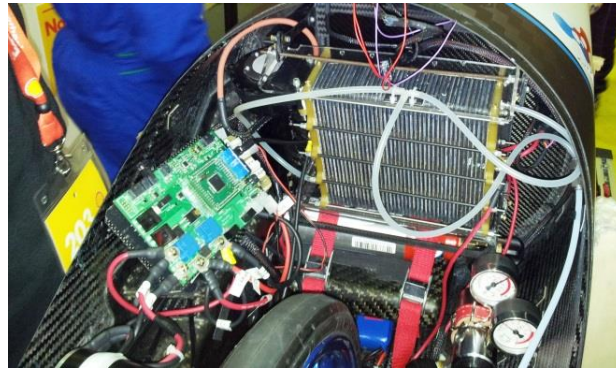


Figure 13 A detail of PEMFC inside the IDRA*pegasus* prototype

Conclusions

The relative humidity is a crucial variable to improve the PEMFC performance. A technique, which increases the water content of the membrane, was performed. This method is particularly effective when there is need to have a high humidity rate inside the PEMFC in the startup conditions. Another advantage is given by the fast water recovery of the dry membrane without any waste of hydrogen. In addition, the technique could be easily implemented in PEMFC equipment.

In order to ensure the success of the Filling method was implemented also the differential method in order to show if flooding conditions occurred. Experimental tests were conducted to verify the reliability of the proposed technique.

The water filling technique consumes less power for the auxiliary systems if compared with the traditional humidifier system because there is only one blower which is the reaction blower.

The Filling was operated in the last Shell Eco-Marathon competition (Rotterdam may 2014), and it's allowed to reach a consumption of 319 km/kWh, improving the results of about 30 km/kWh if compared to the previous year.

In our experience, the method has some limitations because it gives much stress to the fuel cell components so the fuel cell life-time is reduced. However it is still interesting for some applications in which not life-time but weight and performance need to be considered first, e.g. UAV applications or military single-use equipment.

Nevertheless, further tests will be made in order to investigate the long-terms effects of this method, especially on the platinum loading. For example, the Electrochemical Impedance Spectroscopy (EIS) provides more complete information about the FC state of health thanks to fuel cell impedance measure. Ref [15-17]

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Nomenclature

EMF = electromotive force.

HHV = higher heating value.

LHV = lower heating value.

V_c = single cell voltage

μ_f = fuel utilization coefficient.

η = fuel cell efficiency.

λ = stoichiometry coefficient.

ΔV_{in} = inlet cells voltage

ΔV_{center} = center cells voltage

ΔV_{out} = outlet cells voltage

$\Delta V_{diff,in}$ = inlet differential voltage

$\Delta V_{diff,out}$ = outlet differential voltage

H_n = transfer function

$V_{module,n}$ = generic module input

V_n = generic actual voltage

RT = residential time

RH_{out} = outlet relative humidity

ζ = Filling increasing rate

$V_j(I)$ = j-th polarization curves

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