IMPLEMENTATION OF A LABORATORY INFRASTRUCTURE FOR THE DEVELOPMENT OF RELIABILITY AND DURABILITY STUDIES OF PROTON EXCHANGE MEMBRANE FUEL CELLS AT IPEN-CNEN/SP

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ABSTRACT

In this paper the implementation process of a laboratory infrastructure at IPEN - CNEN/SP for the development of reliability and durability studies of fuel cells is described. The outfit for this infrastructure includes fully automated test stands for proton exchange membrane fuel cells operation, safety systems for the laboratory and electrolysers for hydrogen and oxygen supplies in a large scale. In addition, the test protocols used in long term and accelerated life experiments of proton exchange membrane fuel cells are briefly discussed. Furthermore, some preliminary results of the tests that have been conducted in this laboratory are presented.

KEY WORDS

PEM fuel cells; Long term tests; Accelerated Life Tests; Durability; Reliability.

1. INTRODUCTION

One of the big challenges of the 21st century is the search for new energy sources that may substitute the ones currently ruling the global economy, ensuring energy sustainability in the future, and at the same time being clean, efficient and economically feasible. In this context, one of the most promising technologies is the fuel cell. There are many types of fuel cells, which are classified according to the electrolyte used and the operating temperature. The Proton Exchange Membrane Fuel Cell (PEMFC) is one type of technology that has been shown good results for both mobile and stationary applications. They provide high power density, are robust and easy to operate, besides having inherent advantages, such as high efficiency and very low pollutants emission [1-3].

Despite the great progress made in the last few years, cost, reliability and durability are still the major obstacles to be overcome before having a widespread acceptance of this technology in the market. According to the United States Department of Energy (US DOE), only when the cost of the PEMFCs is lower than $50 kW^{-1} this technology may compete with other energy sources. On the other hand, the use of PEMFCs to power automobiles will require the fuel cells to be as durable as the internal combustion engines used in current vehicles. This means a lifetime of at least 5,000 hours of operation, considering all the operating conditions. For stationary applications, the lifetime of the PEMFCs must surpass 40,000 hours to compete with the current stationary systems of power generation. However, the lifetimes of the state-of-the-art PEMFCs used in mobile and stationary applications are shown to be of the order of 1,700 and 10,000 hours, respectively [4].

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In this context, reliability and durability studies are fundamental to the improvement of fuel cell technology, since they cover the identification of degradation and failure mechanisms, which may provide feedback for the developing projects. The operating conditions of the fuel cells are very important to these studies, since they can affect the overall performance of the devices. In general, the required targets for the performance decay rates for the majority of fuel cell applications are, by the end of their life cycle, around 20% of the initial efficiency of the system, that is, in the range of 2–10 $\mu V.h^{-1}$. Long term durability tests are ideal for analyzing the degradation mechanisms of various components of the PEMFCs. However, to operate these fuel cells for some thousand hours, besides not being practical, is a very costly procedure. In this sense, Accelerated Life Tests (ALTs) can be used to provide a fast elucidation of the main degradation mechanisms and durability estimations within a period of time more appropriate to the development of the projects. Therefore, the protocols of ALTs must be developed considering that the results obtained may be compared to the ones generated in long term tests. It must be assured that the conditions in which these tests are performed do not insert new failure mechanisms, that is, failures which are not associated with the actual operating conditions of the devices [4].

Since 2000, the Nuclear and Energy Research Institute (IPEN), which is a State of São Paulo autarchy, associated to the University of São Paulo (USP) for educational purposes, supported and operated technically and administratively by the National Nuclear Energy Commission (CNEN), has been carrying out a research area in efficient energy sources with low environmental impact, including the analysis and development of systems associated with fuel cells. The Fuel Cell and Hydrogen Centre at IPEN-CNEN/SP has developed, among other technologies, a sieve printing technique for the production of Membrane-Electrode Assemblies (MEAs) for PEMFCs of geometrical dimensions similar to the commercial ones. The production has been successful in terms of efficiency of the MEAs as long as they have been presenting high electrical current density [5]. During these years, the researches have been focusing on the optimization of the efficiency of the MEAs, by increasing the geometric area of the electrodes and the production scale. In 2010, IPEN delivered one hundred MEAs (250 cm^2 of geometric area) to a Brazilian company named Electrocell. These MEAs were used in the assembly of the first module of 5 kW of nominal power made with PEMFCs entirely manufactured with national technology.

2. Objectives

This paper aims to describe the implementation process of a laboratory infrastructure for the conduction of reliability and durability studies of fuel cells at IPEN-CNEN/SP, as well as to present some preliminary results of the tests that have been performed in this laboratory. In addition, the test protocols used in long term and accelerated life experiments of proton exchange membrane fuel cells are discussed.

3. Methodology

3.1. Implementation of a laboratory infrastructure at Ipen-Cnen/SP for the development of reliability and durability studies of fuel cells

The lack of information on the reliability and durability of fuel cells being developed at IPEN-CNEN/SP justified the need for the installation of a laboratory infrastructure dedicated to studies of this nature. Thus, in mid-2006, a research area entitled "Reliability and durability studies of proton exchange membrane fuel cells" started to be developed. Financial support was granted to two projects proposed in this area: FAPESP (Project No. 06/51278-5) and FINEP (Project No. 01.08.0339.00). In general, the projects that are part of this research area aim at the study of the PEMFCs and components that are being produced at IPEN-CNEN/SP in a laboratory scale, using national technology for their manufacture. The ultimate objective is to obtain data that qualify these PEMFCs for a pre-commercial production.

To achieve the goals of the project linked to FAPESP, which lasted three years (from April/2006 to April/2009), financial resources were required for the construction and assembly of a fuel cell operation panel manufactured by a Brazilian company named Electrocell and for the acquisition of an electrolyser that provides hydrogen (H_2) and oxygen (O_2) with a capacity of 10 m^3h^{-1} and 5 $m^3\,h^{-1}$, respectively. Moreover, with an additional amount granted by FAPESP in March 2008, it was possible to purchase the components for the assembly of five PEMFCs of 25 cm^2 and a microcomputer, and to contract a service to develop a system for data acquisition and remote monitoring of the fuel cells.

Regarding the project supported by FINEP the financial resources were used for the purchase of an additional electrolyser, the installation of an exhaust and air cooling system in the laboratory, and for the acquisition of two fuel cell test stands, fully automated, manufactured by a German company named FuelCon.

3.2. DESCRIPTION OF THE TEST STAND MANUFACTURED BY ELECTROCELL

A control panel containing five test stations was developed and assembled by the company named Electrocell in the laboratory at IPEN-CNEN/SP (**Figure 3.2-1**). This panel was used for long term tests as well as accelerated life tests that were conducted during the development of the FAPESP Project No. 06/51278-5. The key features and components of this panel are the following:

- 5 independent test stations;
- Gas temperature and cell temperature automatic controllers;
- Pressure gauges and rotameters to control inlet gases pressure and flow;
- Manual back pressure system to control gas pressure in the cell at each station;
- Purge system with N_2;
- Electronic load with operating range from 500 mA to 50 A to control the electric current generated by the fuel cell;
- Microcomputer and touch screen video monitor in each test station;
- Connection hose with quick disconnect device;
- Sensors for monitoring H_2 leakage to the laboratory environment.

Figure 3.2-1 Side view of the fuel cell operation panel manufactured by Electrocell

Furthermore, to make the man-machine interface with the electronic load, Electrocell provided a software called "scDinamica" that performs the following functions: monitoring of fuel cell voltage and electric current during operation; adjustment of the electric current that simulates a system load (communication with the electronic load) and measurement of the fuel cell voltage corresponding to the current value previously set; data acquisition during the operation of the fuel cell, record of fuel cell voltage and current density over the time; elaboration of graphs (voltage x current density, power x current density, voltage x time and electric current x time) storing the records in electronic files in the format *.csv or *.xls.

3.3. DESCRIPTION OF THE TEST STAND MANUFACTURED BY FUELCON

The two test stands manufactured by the German company Fuelcon (**Figure 3.3-1**) were acquired with funds granted by FINEP and are equipping the laboratory at IPEN-CNEN/SP. The FuelCon test stands have the following features and components:

- Multi-range load with U, I, P or R control;
- Fully automated control of pressure, temperature, flow and humidification of gases (O_2 and H_2);
- Automatic control for cell heating and cooling during operation;
- Electronic back pressure control, exhaust drain and dry gas by-pass;
- Safety control systems, including emergency manual button, automatic shutdown of the electronic load, automatic closing of valves, automatic cooling and purging of the cells in case of emergencies, etc.;
- Automatic humidification system including flexible hose trace heating;
- Software "FuelWork" used to control the operation of the fuel cells in the test stands, allowing the edition of the procedures for the automatic operation of the fuel cells, including automatic data collection for the elaboration of graphics, and automatic shutdown and cooling of the system at the end of the test. Furthermore, the synoptic diagram of the system presented on the computer screen allows the user to control all the operating parameters of the test;
- Data acquisition system;
- Fully automated for unattended operation (remote monitoring and operation);
- Embedded high frequency impedance analyzer (1 test stand).

Figure 3.3-1 Test stands manufactured by FuelCon

3.4. PROCEDURES FOR CONDUCTING DURABILITY TESTS OF FUEL CELLS AT IPEN-CNEN/SP

The recommended steps to be followed during the performance of the durability tests are:

1. System heating: the initial operating conditions of the fuel cell and the system are set according to **Table 3.4-1**;
2. Fuel cell cycling: in this step the fuel cell current is increased up to the value corresponding to a single cell voltage of about 300 mV and, immediately afterwards, the fuel cell current is decreased to zero in order to reach the open circuit voltage (OCV). This process is repeated for at least 15 cycles;

3. Fuel cell conditioning / Stabilization of the operating conditions of the system: in this step the fuel cell current is adjusted in the electronic load to the value corresponding to a single cell voltage of about 600 mV. The fuel cell voltage stabilization is reached when the variations in the cell voltage are lower than +/- 5 mV during the last hour before ending the conditioning step. The most important factor in this step is that the cell voltage is stable before the actual measurement step starts;
4. Elaboration of the initial polarization curve: the initial curve, which shows the fuel cell voltage as a function of the current density, will be compared with the polarization curve drawn by the end of the test. This analysis may be useful for making the diagnosis of a possible loss of fuel cell performance;
5. Specific test (long term or accelerated test) to measure the variables of interest: in both cases cited above, i.e., in the long term test and accelerated life test, some test protocols are followed. These protocols have been developed by the European Commission under the project entitled "Fuel Cell Systems Testing, Safety & Quality Assurance". More specifically, the long term tests are based on the *Test Module PEFC SC 5-1* (Testing the voltage-power as function of time - Steady test for a PEFC single cell) [6] and *Test Module PEFC SC 5-6* (Testing the voltage and the power as a function of time at a fixed current density - Long term durability steady test for a single PEFC) [7]. The accelerated tests are based on the *Test Module PEFC SC 5-4* (Testing the voltage and the power as a function of the current density following an on/off profile versus time - Accelerated ageing on/off cycling test for a PEFC single cell) [8]. These protocols aim to form a pre-normative and pre-regulation base to be established in the European Union, with the support of the International Partnership for the Hydrogen Economy (IPHE). For this reason, they are recommended by INMETRO (National Institute of Metrology, Standardization and Industrial Quality), which is the Brazilian representative organization in this partnership.
6. Elaboration of the final polarization curve: as mentioned in Step 4, the initial and final polarization curves can be compared in order to diagnose a possible loss of fuel cell performance;
7. System shutdown and cooling: before concluding the system shutdown and cooling phase, it is recommended to perform the purge of the system piping with N_2.

Table 3.4 -1: Input variables of the test

Input	Value / range	Control accuracy
Fuel gas composition	H_2 (5.0 analytical / 5.0 ECD)	-
Oxidant gas composition	O_2 (4.0 analytical)	-
Fuel cell temperature	75^oC	± 3^oC
Fuel gas temperature	90^oC	± 3^oC
Oxidant gas temperature	80^oC	± 3^oC
Inlet fuel gas pressure	1 bar	± 2%
Outlet fuel gas pressure	-	-
Inlet oxidant gas pressure	1 bar	± 2%
Outlet oxidant gas pressure	-	-
Fuel gas flow rate / stoichiometry	50% to 100% in excess of the	-
Oxidant gas flow rate / stoichiometry	gas stoichiometry	

4. RESULTS

4.1. LONG TERM LIFE TESTS

Some preliminary long term tests have been performed in the laboratory at IPEN-CNEN/SP using the stands manufactured by Electrocell and FuelCon. The single PEMFCs tested and referred in this work incorporate the technology developed at IPEN-CNEN/SP and have a geometric area of 25 cm^2. The electrodes were manufactured with commercial Pt/C catalyst from BASF with average loads of 0.4 and 0.6 mg cm^{-2} at the anode and cathode, respectively. Input variables of the test are shown in **Table 3.4-1**. For these tests, the current was kept constant at a value that stabilizes the cell potential of about 600 mV. Polarization curves were taken in the beginning and in the end of the test in order to analyze the loss of cell performance as a function of operating time. In the graph of **Figure 4.1-1** the curve *cell voltage x time* obtained from the long term test performed on the station manufactured by Electrocell is shown. This test lasted 500 hours and data related to the performance decay rate and some other parameters are summarized in **Table 4.1-1**. In **Figure 4.1-2** it can be observed that, for the polarization curves obtained before and after the 500-hour test, the variation in cell performance was very slight.

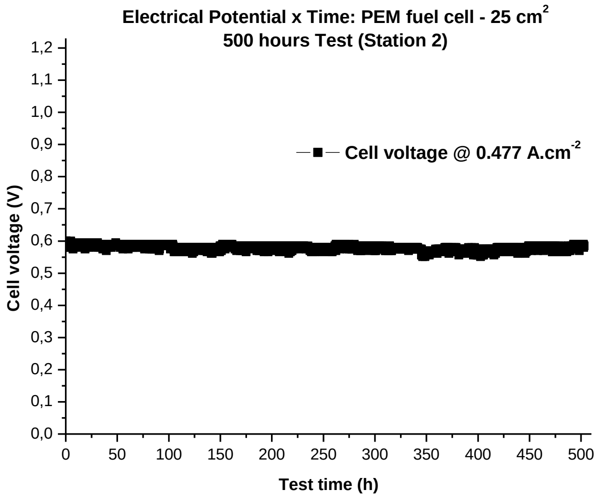

Figure 4.1-1 Graph of cell voltage *vs.* time obtained from the 500-hour test conducted in the fuel cell test stand manufactured by Electrocell

Table 4.1-1 Summary of the results obtained from the long-term test conducted in the fuel cell test stand manufactured by Electrocell

Total test time	503.00 hours
Number of cell voltage values observed	3018
Mean of cell current density during test	0.526 A cm^{-2}
Mean of cell voltage during test	0.581 V
Loss of cell voltage by the end of test	2.500 %
Cell voltage decay rate during test	0.030 mV h^{-1}
Mean of cell power density during test	0.306 W cm^{-2}
Loss of cell power density by the end of test	2.129 %
Cell power density decay rate during test	0.013 mW h^{-1}

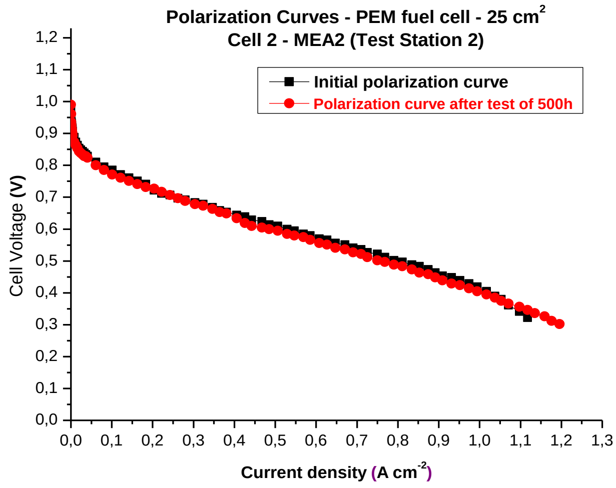

Figure 4.1-2 Polarization curves (initial and final) obtained from the 500-hour test conducted in the fuel cell test stand manufactured by Electrocell

Some preliminary long term tests have also been performed using the test stands manufactured by the German company FuelCon. These tests were performed for 500 hours continuously, differently from the tests conducted at the Electrocell stand. In this latter, the tests had to be periodically interrupted due to the humidifiers which have to be filled up with water manually. In the FuelCon stands, the humidifiers are automatically supplied with de-ionized water, so that the tests do not need to be interrupted. In the graph of **Figure 4.1-3** the curve *cell voltage x time* obtained from the long term test performed on the station manufactured by FuelCon is shown. After about 300 hours of test, the cell voltage decreased substantially. One possible explanation might be the water accumulated in the flow channels, observed when the cell was opened at the end of the experiment. The polarization curves obtained before and after the 500-hour experiment performed in the test stand of FuelCon are shown in **Figure 4.1-4**.

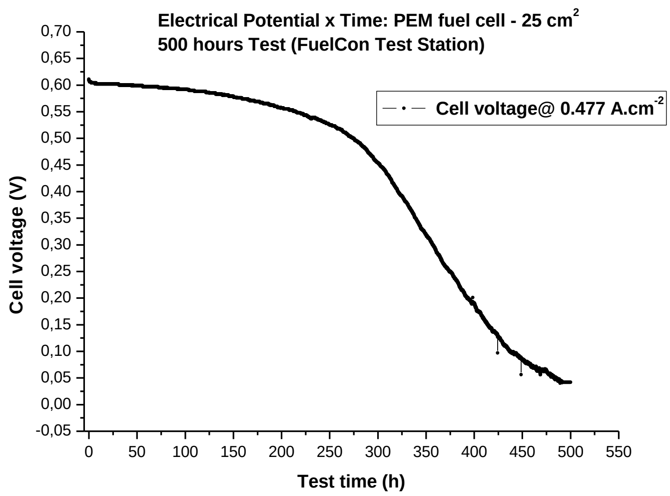

Figure 4.1-3 Graph of cell voltage *vs.* time obtained from the 500-hour test conducted in the fuel cell test stand manufactured by FuelCon

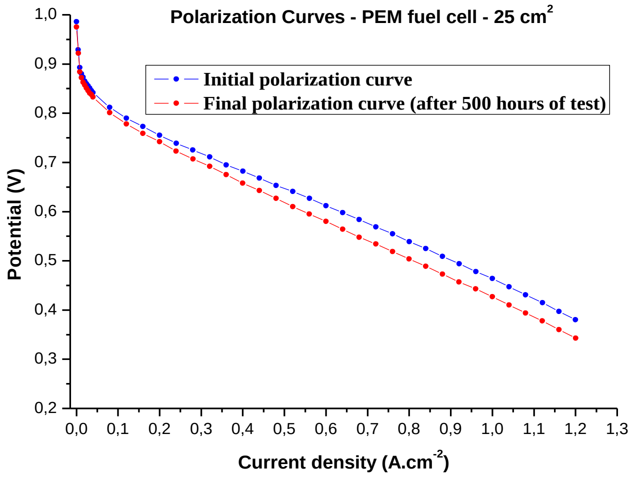

Figure 4.1-4 Polarization curves (initial and final) obtained from the 500-hour test conducted in the fuel cell test stand manufactured by FuelCon

4.2. Accelerated Life Tests

The scope of the accelerated test performed in the laboratory at IPEN-CNEN/SP was to determine the evolution of the voltage of a PEMFC operating in specified conditions during a durability test performed in cycling conditions of the electrical current. The main objective was to determine the evolution of both open circuit voltage (OCV) and voltage on load in terms of Vh^{-1} when submitting the cell to a specific load profile including “on” and “off” phases of 15 minutes. After the initial polarization curve, the current density was fixed again to a value corresponding to a load current for a first “on” phase of 15 minutes, e.g. 16 A. Then the cycling consists on the following profile:

- Phase “*off*” = 15 min at i=0 A cm^{-2}
- 10 sec at I=4 A or i=0.16 A cm^{-2}
- 10 sec at I=8 A or i=0.32 A cm^{-2}
- 10 sec at I=12 A or i=0.48 A cm^{-2}
- Phase “*on*” = 15 min at **I**=16 A or i=0.64 A cm^{-2}

It is recommended to increase the current density step by step in order to avoid important voltage drop [8]. In the graph shown in **Figure 4.2-1,** the variation of cell voltage with time during the "on/off" current cycling test held in the measured period is presented. As can be seen in the polarization curves shown in **Figure 4.2-2**, the cell performance decreased slightly after 250 hours of "on/off" cycles. According to Lin *et al* [9], dynamic driving cycle has many effects on performance degradation and micro-structure of the PEM fuel cell, once it causes structural and electro-chemical changes within the catalyst layer and the membrane.

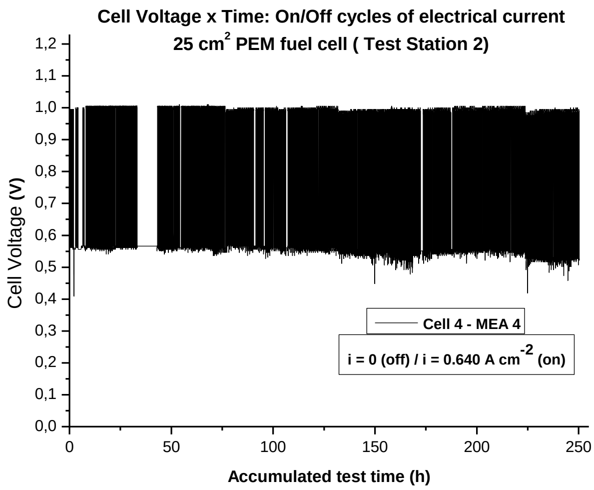

Figure 4.2-1 Graph of cell voltage *vs.* time obtained from the accelerated ageing on/off cycling test conducted in the test stand manufactured by Electrocell

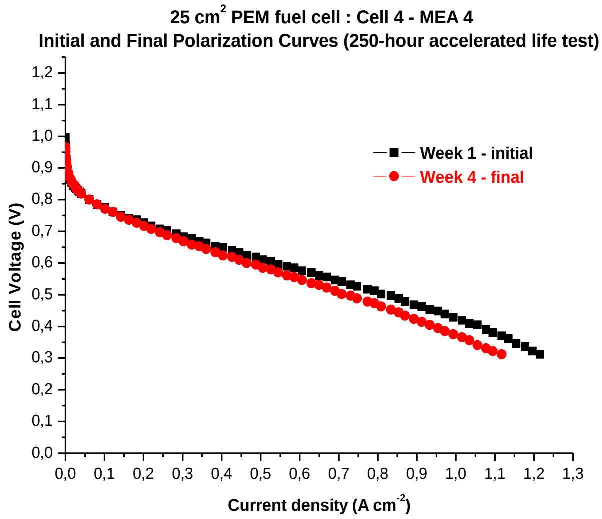

Figure 4.2-2 Polarization curves (initial and final) obtained from the accelerated ageing on/off cycling test conducted in the test stand manufactured by Electrocell

5. CONCLUSIONS

At present, durability, reliability and cost are the major barriers for the commercialization of PEMFC technology. Therefore, the implementation of a laboratory infrastructure specialized in reliability and durability studies at IPEN-CNEN/SP is an advance towards the development of this type of fuel cell technology in Brazil. Furthermore, the identification of major failure modes of fuel cells and the associated degradation mechanisms of their components generates a fundamental feedback for the projects currently being carried out in Brazil. The test stands that outfit the laboratory at IPEN-CNEN/SP are appropriate to perform long term tests and accelerated life tests with PEMFCs, which shall provide knowledge about the degradation of membranes, electrocatalyst layers, bipolar plates, crossover rates, etc. As a result, it will be possible to propose the necessary mitigation strategies to preserve the membranes as well as improvements in the design of the fuel cells may be implemented.

Although the preliminary results presented in this work are not elucidative enough to solve the PEMFCs degradation issues, they have been meaningful to the elaboration of a set of tests protocols that better evaluate the performance of the PEMFCs.

6. ACKNOWLEDGEMENTS

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