Available online at www.sciencedirect.com

ScienceDirect

Journal of Power Sources 169 (2007) 85–91

JOURNAL OF POWER SOURCES

www.elsevier.com/locate/jpowsour

Description of gasket failure in a 7 cell PEMFC stack☆

Attila Husar[a,*], Maria Serra[a], Cristian Kunusch[b]

[a] Institut de Robòtica i Informàtica Industrial, Parc Tecnològic de Barcelona, Edifici U, C. Llorens i Artigas, 4-6, 2a Planta, 08028 Barcelona, Spain
[b] Laboratorio de Electrónica Industrial Control e Instrumentación, Facultad de Ingeniería, UNLP, Argentina

Abstract

This article presents the data and the description of a fuel cell stack that failed due to gasket degradation. The fuel cell under study is a 7 cell stack. The unexpected change in several variables such as temperature, pressure and voltage indicated the possible failure of the stack. The stack was monitored over a 6 h period in which data was collected and consequently analyzed to conclude that the fuel cell stack failed due to a crossover leak on the anode inlet port located on the cathode side gasket of cell #2. This stack failure analysis revealed a series of indicators that could be used by a super visional controller in order to initiate a shutdown procedure.

Keywords: Polymer electrolyte membrane fuel cells; Stack failure; Gasket; Indicators

1. Introduction

Polymer electrolyte membrane fuel cell stacks fail generally due to: failure of the gasket, failure in the membrane, and/or failure of the catalyst. This article is going to examine both analytically and physically the failure of a gasket in a 7 cell stack.

The stack is an ElectroChem® 7 cell stack (Part #EFC-50-03-7-ST) with Nafion 115 MEA's, with a catalyst loading of 1 mg PT cm^{-2} and Toray carbon fiber paper as the gas diffusion layers (GDL). The gasket is a precision grade silicone material specially cut for the bipolar plate design and it should be able to maintain its integrity up to 220 °C [1].

The stack was being tested under a variety of pressures (1–4.5 bar_a), temperatures (25–60 °C), relative humidity (0–100%), and current (0–5 A). Both air and oxygen were used on the cathode and only pure hydrogen was used on the anode. The cell failed after approximately 20 h of operation. The test station that was used in these experiments was an ElectroChem® MTS-150 in combination with an HSA gas humidifier.

The stack design is based on a bipolar configuration where the cathode of one cell is electrically connected to the anode of the adjacent cell while the gas is connected in parallel through the bi-polar plates' flow fields. Fig. 1 shows a cross-sectional view of a generic bipolar configuration with the correct gas flows.

The main components of the stack are the membrane electrode assemblies (MEA), the catalyst in the center on the anode and cathode side of the membrane, the gaskets on the perimeter of the MEA with gas diffusion layers, and the graphite bipolar plates. The gasket generally has two sealing functions: overboard leak and crossover leak. An overboard leak is when the gas from either the anode or the cathode side escapes to the environment. A crossover leak is when the gas from one side of the membrane crosses over to the other side of the membrane. A crossover leak can occur in two ways: one is a leak between the gasket and the membrane in the inlet or outlet ports and the other is by passing through the membrane itself. There is an expected crossover leak rate of hydrogen and oxygen through the membrane due to diffusion through the membrane which can be estimated for any given stack based on the number of cells in the stack, active area, and type and thickness of the membrane assuming that the seals are perfect. This flow rate due to diffusion is measured in $cm^3\ s^{-1}$ and is also related to the partial pressure and temperature of the gas. However, the loss in voltage due to the hydrogen diffusion should only be seen at open circuit voltage because, as the current increases, the hydrogen partial pressure decreases on the anode side which reduces the driving force of the diffusion [2]. The oxygen crossover, on the other hand, should not be perceived in the open circuit voltage.

☆ This paper is presented at the 2nd National Congress on Fuels Cells, CONAPPICE 2006.

* Corresponding author at: Institut de Robòtica i Informàtica Industrial, Parc Tecnològic de Barcelona, Edifici U, C. Llorens i Artigas, 4-6, 2a Planta, 08028 Barcelona, Spain.

E-mail address: ahusar@iri.upc.edu (A. Husar).

0378-7753/$ – see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.jpowsour.2007.01.078

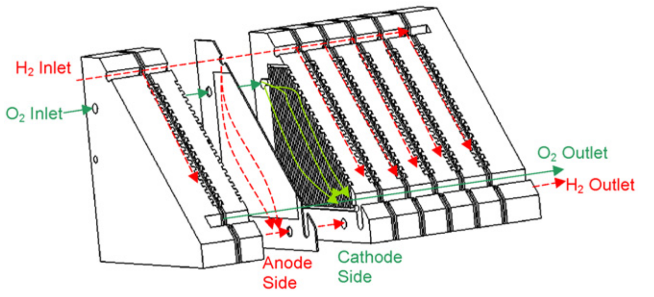


Fig. 1. A cross-sectional view of a typical parallel flow of the reactant gasses through a fuel cell stack.

The manufacturer recommended operating conditions for the stack are: H_2/O_2, 1.02 Stoichiometry for anode and cathode, 60 °C stack temperature, no humidification and a gas pressure of 4.50 bar_a (50 psi_g) for both the anode and cathode [3]. The data analyzed in this work were based on these conditions.

The stability of the stack over time with respect to individual cell voltage is shown in Fig. 2. As can be seen in this figure the stack was stable under these conditions. The variation in the cell voltage can be attributed to changes in the current due to impedance spectroscopy tests performed on the stack. Cell #7 of the stack had consistently showed a poor performance when compared to the other cells in the stack. However, this issue will not be analyzed in this work as it is unrelated to the ultimate failure of the stack.

As the testing went on, the cell voltages started to unexpectedly change and the pressure could not be maintained with the back pressure regulators. This marked the beginning of the end for this stack. The following section will describe the stack's final day of operation.

2. Gasket failure description

2.1. Analysis of data from the last day of operation

In the morning, an overboard leak check was performed on the stack using nitrogen on the anode and air on the cathode. A leak check was needed because on the previous run the pressure on the anode side could not be maintained. However, there were no other signs of failure. The procedure for the pressure decay leak check is as follows: a flow of nitrogen and air is applied to the stack, the system is pressurized to 4.5 bar_a, then the inlet and outlet to the stack are closed and the system left alone for a given amount of time. The pressure decay leak check is presented in Fig. 3. It was done in three sections. The first section of the test was done with anode and cathode pressures maintained at the same value (4.5 bar_a). The second part of the test was done with the cathode pressure left at 4.5 bar_a and the anode pressure reduced to 4.2 bar_a. The final part of the test was carried out in reverse order, the anode pressure was increased back up to 4.5 bar_a and the cathode pressure was reduced to 4.2 bar_a. The slope of each section of the test is indicated on the graph. Fig. 3 shows a positive slope of the lower pressure side of the membrane indicating that gas from the higher pressure side of the membrane is crossing over to the low pressure side. The exact rate is unknown because a true crossover leak check was not performed on this stack due to the lack of equipment. No com-

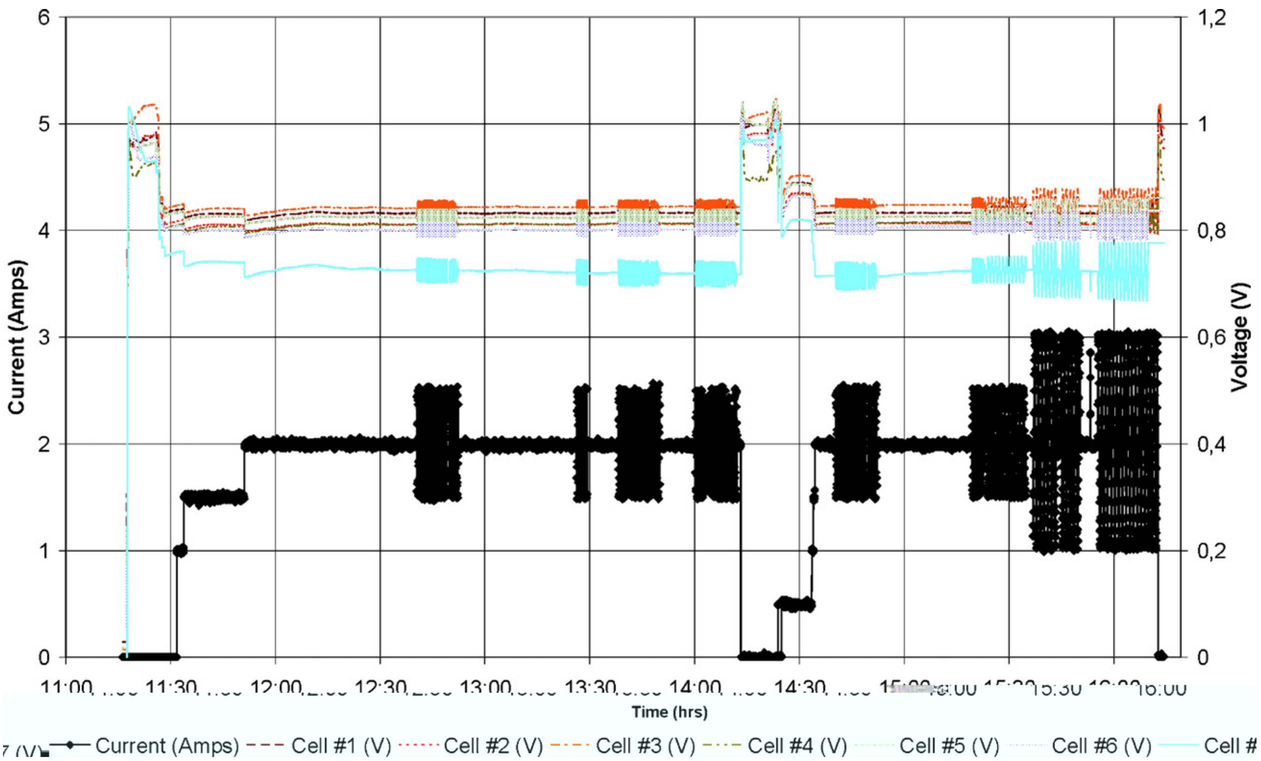
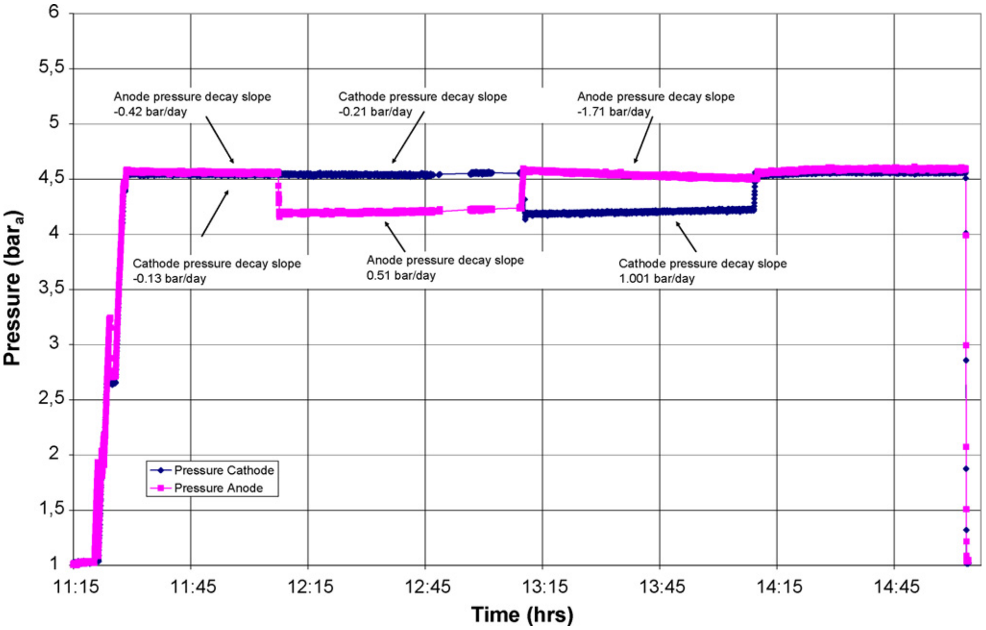


Fig. 2. Normal voltage current response of the 7 cell stack a few days before failing, H_2/O_2, 1.02 Stoichiometry for anode and cathode, 60 °C stack temperature, no humidification and a gas pressure of 4.50 bar_a (50 psi_g) for both the anode and cathode.


6
5,5
5
4,5
4
3,5
3
2,5
2
1,5
1
Pressure (bar$_a$)
11:15
11:45
12:15
12:45
13:15
13:45
14:15
14:45
Time (hrs)
Anode pressure decay slope -0.42 bar/day
Cathode pressure decay slope -0.21 bar/day
Anode pressure decay slope -1.71 bar/day
Cathode pressure decay slope -0.13 bar/day
Anode pressure decay slope 0.51 bar/day
Cathode pressure decay slope 1.001 bar/day
Pressure Cathode
Pressure Anode

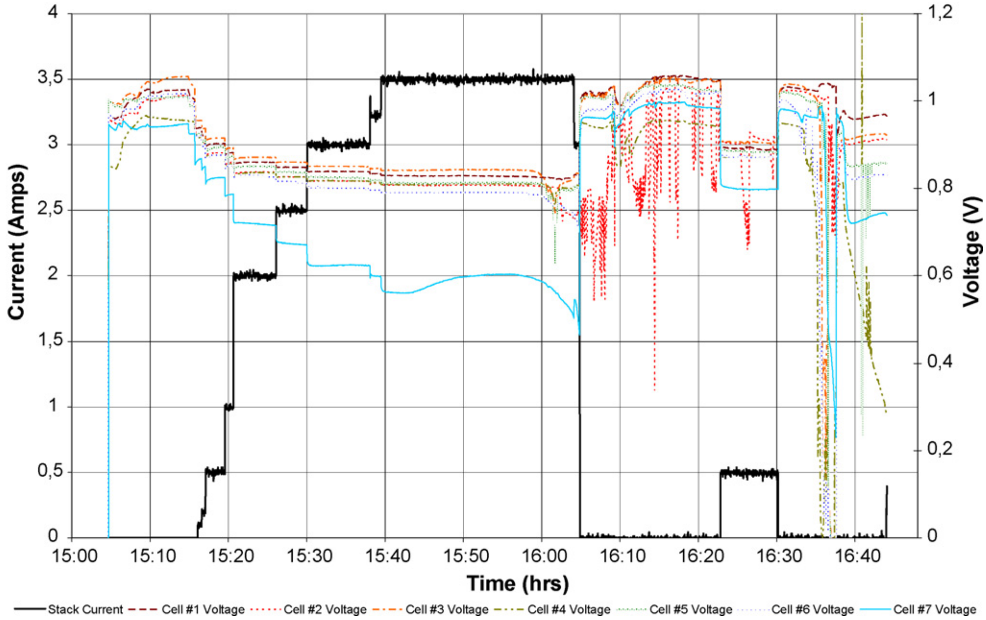


Fig. 5. Stack current vs. individual cell voltage during the last hours of operation, H_2/O_2, no humidification and a gas pressure.

In an attempt to stabilize the pressure the anode and cathode gas flows were increased. However, this attempt did not work and the pressures continued to fall.

Fig. 5 depicts the stack current versus the individual cell voltages. It shows how, after the current was reduced to 0 at around 16:05, the open circuit voltages became very unstable especially that of cell #2.

In Fig. 6 the time between 15:50 and 16:10 is examined with respect to pressure and cell temperature. When the load was removed from the stack at 16:05 the temperature measurements 4–8 started to decline, as was expected. However, the temperature measurements 1–3 continued to increase. This temperature rise showed signs of a breach possibly in cell #1 and/or #2.

Fig. 7 explains how a breach in one cell affects the flow of the reactant gases when the pressure of the anode is higher than the cathode. Hydrogen enters directly into the breached cell and combusts in the catalyst layer, therefore increasing the temperature for just that one cell.

At 16:09 there was a temporary shut down of the stack where the pressure was reduced to 1 bar_a.

In Fig. 8 the time between 16:30 and 16:45 is examined with respect to anode and cathode pressure and temperature. Initially both pressures were equalized. However, the anode pressure started to decline more rapidly than the cathode because hydrogen was being consumed at twice the rate of oxygen due to combustion which, in turn, caused more oxygen to be forced

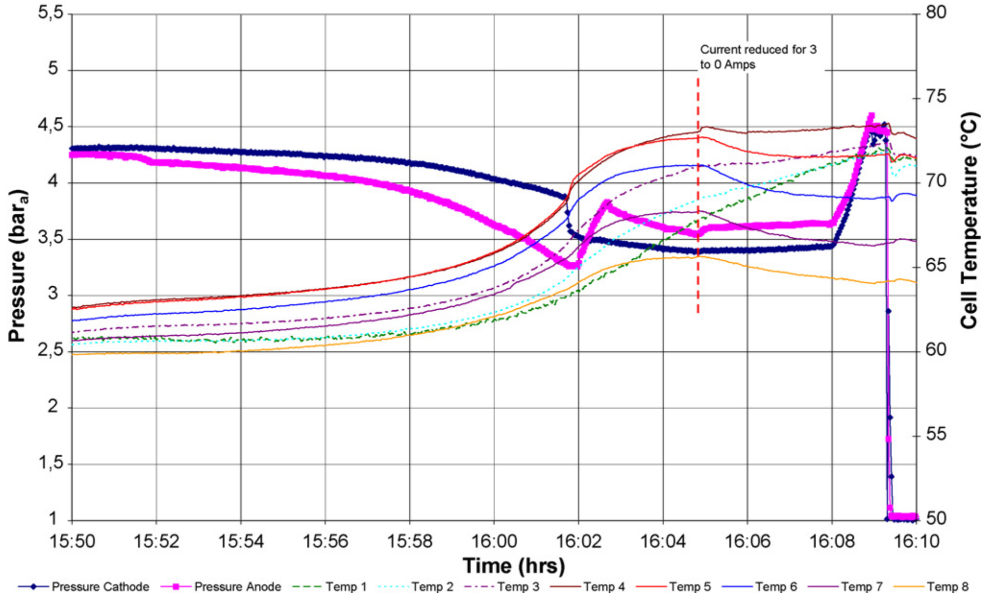


Fig. 6. Zoom of Fig. 4 between 15:50 and 16:10. Increase in temperature in cells #1 and #2 with no electric load or external heaters on the stack and an anode reactant pressure higher than the cathode pressure when the load was removed.

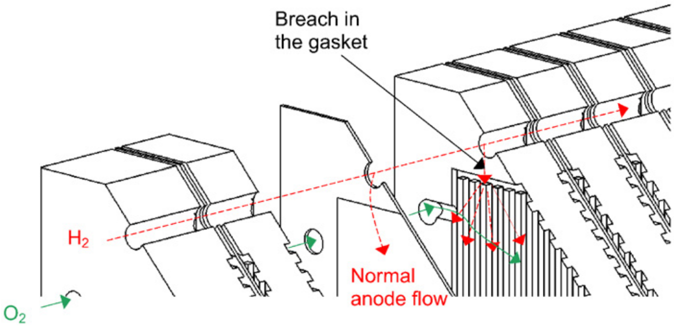


Fig. 7. Flow of the reactant gases when there is a breach in the seal at the anode inlet port that goes into the cathode flow field. Anode pressure is higher than the cathode's.

into the anode inlet port thus, increasing the rate of combustion in the cells located after the breach. Once the nitrogen purge was initiated on the anode and the air purge on the cathode the pressures began to stable out and the cell temperatures decreased. Shortly after, the pressure was reduced to atmospheric and the stack was declared inoperable.

Since the breach is reversible it should permit the oxygen to enter into the anode inlet port if the cathode pressure was higher than that of the anode as shown in Fig. 9. Therefore, the oxygen breach should only affect the subsequent cells.

Cell #2 in Fig. 10 showed a unique behavior when the anode pressure was higher than the cathode pressure. The cell lost about 20 mV and became unstable. Then, when the anode pressure was decreased to be lower than the cathode's the cell stabilized. This indicated the presence of a hydrogen crossover in cell #2.

Since the stack was declared inoperable it was disassembled to be physically examined.

Upon opening the stack it was clear that it was cell #2's cathode side gasket that had failed. Fig. 11 shows the cathode gasket and GDL for cell #2 while still attached to the bipolar plate. The interface between the gasket and the GDL is a white color indicating degradation.

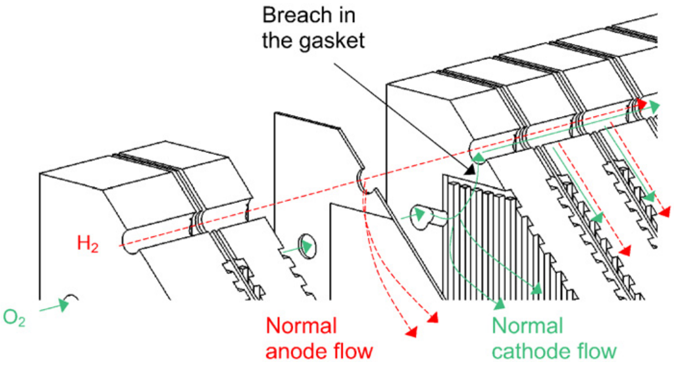


Fig. 9. Flow of the reactant gases when there is a breach in the seal at the anode inlet port. Oxygen is entering into the anode inlet port. Cathode pressure is higher than the anode's.

Interestingly, this was the only cell that had water droplets on the gas diffusion layer and a few of the droplets were brown in color which is a sign of contamination possibly from combustion on the cathode side.

Fig. 12 illustrates what a good cell with no degradation of the gasket looked like.

When the gasket and MEA were peeled off of the anode side of the bi-polar plate it revealed that there was severe degradation of the gasket material around the anode inlet port towards the GDL as shown in Fig. 13. There was also some degradation around the cathode inlet. This degradation occurred on the cathode gasket.

Finally, when the gasket was totally detached from the membrane it was discovered that a piece of the gasket material was fused to the membrane at the anode inlet port as seen in Fig. 14.

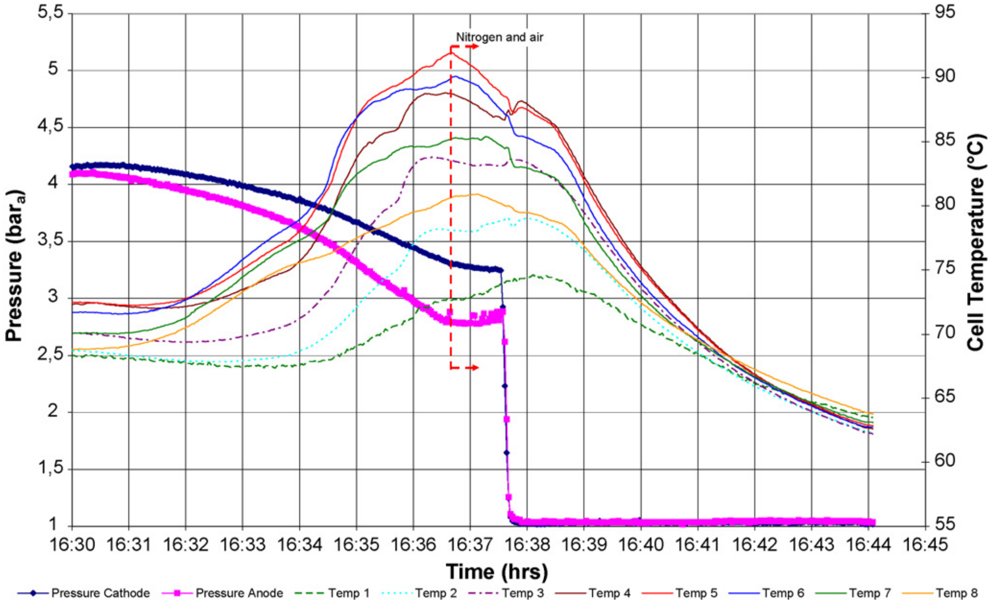


Fig. 8. Zoom of Fig. 4 between 16:30 and 16:45. Cathode pressure is higher than the anode's and the hot spots in the stack are from cell #3 to #6 indicating that the oxygen was entering into the hydrogen stream through the inlet port.

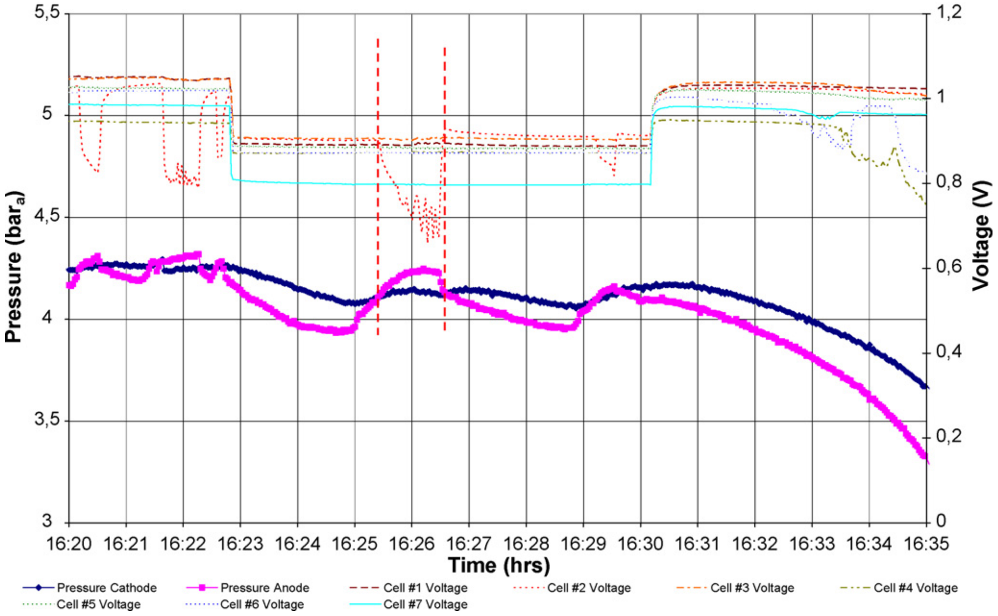


Fig. 10. Temporary instability of cell #2 when the anode pressure is higher than the cathode's. Hydrogen enters directly into the cathode flow field of the cell.

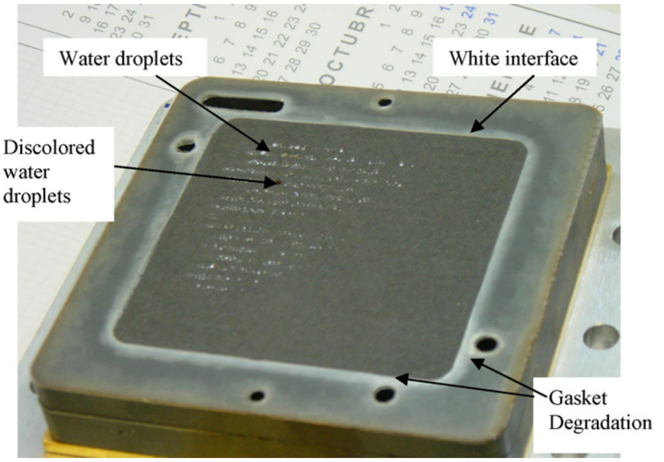


Fig. 11. Cathode GDL and gasket of cell #2 before it was removed from the bi-polar plate.

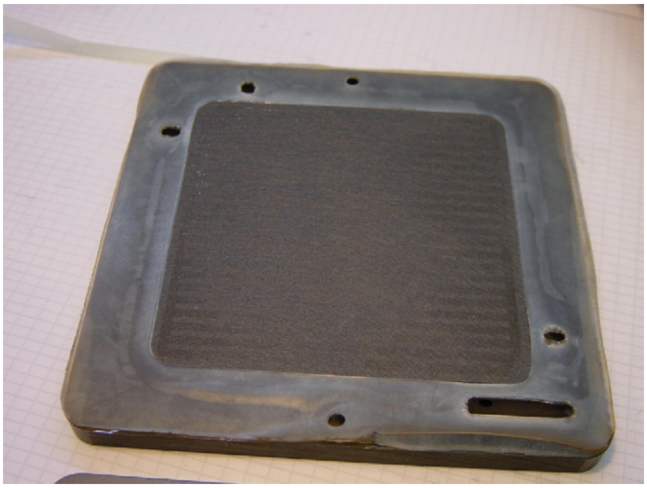


Fig. 12. Cell #5 gasket and diffusion layer which showed no degradation.

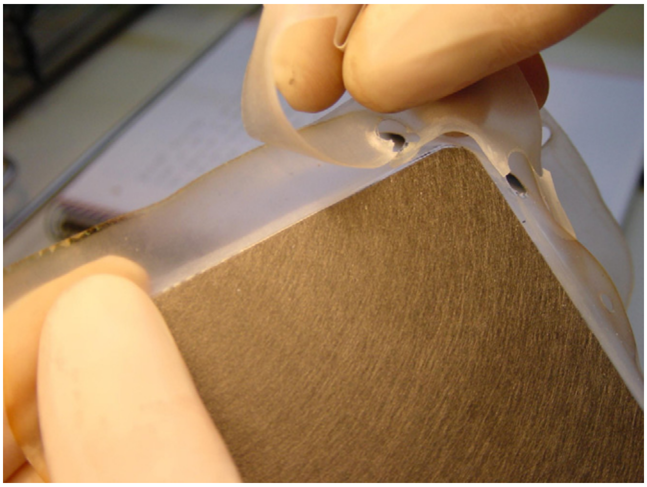


Fig. 13. Cell #2 cathode gasket degradation.

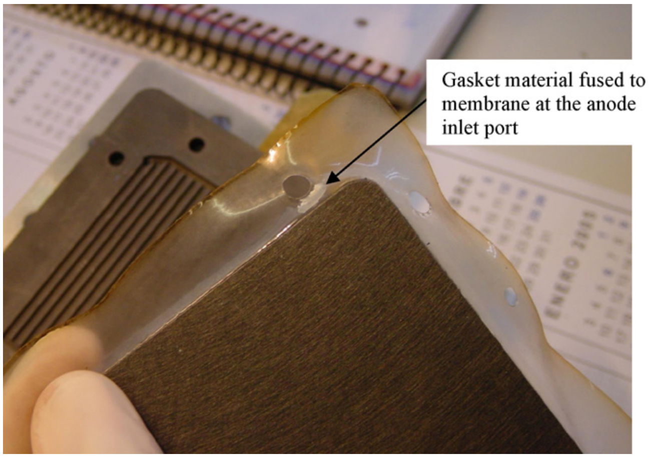


Fig. 14. Gasket material left on the membrane at the anode inlet port and on the cathode side of the membrane.

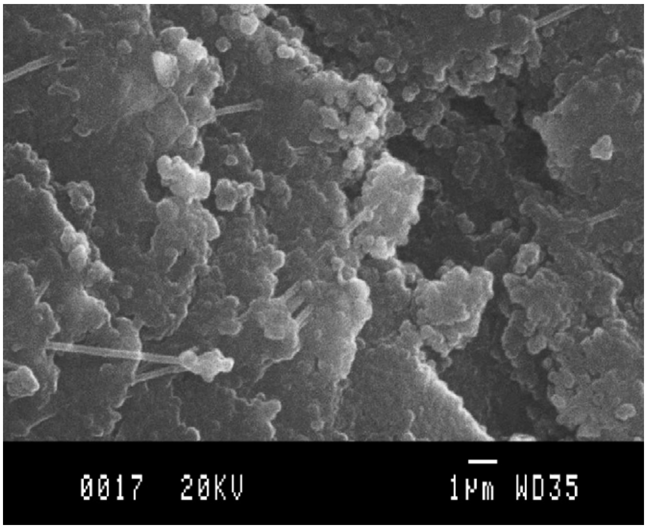


Fig. 15. Silicon gasket material that was exposed to the combustion from cell #2.

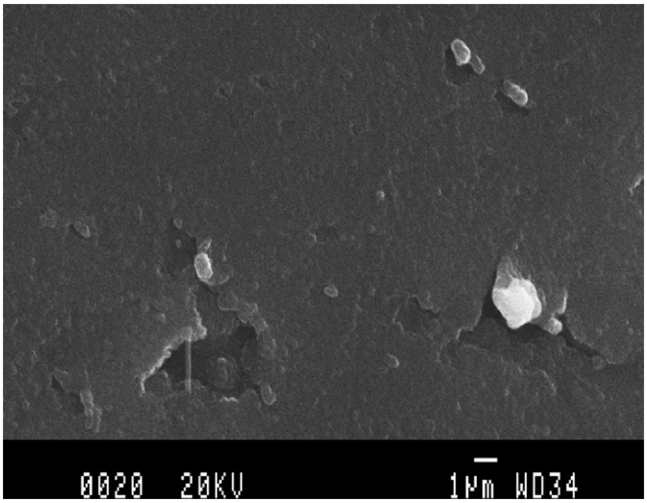


Fig. 16. Silicon gasket material that was not exposed to the combustion from cell #4.

This could be explained by the high temperature reached due to combustion that caused the gasket material to disintegrate and fuse to the membrane.

Cell #1 gasket also showed signs of degradation but not as severe as cell #2. The degradation can be noted by the discoloration of the gasket material around the edges of the GDL.

Figs. 15 and 16 show images of the silicon gasket material taken with the scanning electron microscope. In Fig. 15 shows the silicon gasket material from cell #2 that was exposed to combustion and Fig. 16 shows what the same material looks like under regular conditions.

The MEAs from cells #2 and #4 were also examined. An X-ray Energy Dispersing Spectroscopy was performed on both MEA samples which indicated the presence of both platinum and silicon. The presence of platinum was expected because it is used as a catalyst. However, the silicon indicated a possible contamination of the MEA.

3. Conclusion

The 7 cell stack failed due to a crossover leak on the cathode side gasket of cell #2. The main indicators of the failure were: an unexplainable rise in the stack temperature, pressures that could not be maintained, and the change in cell #2 voltage relative to anode and cathode pressure. The gasket failure occurred gradually over time which might have been started by loosening of the stack due to thermal cycling. Once started, the leak degraded the gasket further by exposing the gasket to the high temperature of combustion of hydrogen and pure oxygen. The combustion was initiated by the platinum catalyst on the membrane. Finally, the leak was so large that enough oxygen was able to crossover to the anode side causing the stack to heat up significantly.

Acknowledgment

This work has been partially funded by project CICYT DPI2004-06871-C02-01, from the Spanish MEC.

References

[1] Silicone. http://www.stockwell.com/newstuff/newpdfs/500_solid_silicone/510_cohr_9275_high_perf_clear_silicone.pdf.
[2] F. Barbir, PEM Fuel Cells Theory and Practice, Elsevier Academic Press, USA, 2005.
[3] Electro Chem Inc. website: http://www.fuelcell.com/pdfs/FuelCellStack.pdf (accessed May 2006).