

Description of Gasket Failure in a 7 Cell PEMFC Stack

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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 six hour 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 visual control 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/cm² 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 to 4.5 bar_a), temperatures (25 to 60°C), relative humidity (0 to 100%), and current (0 to 5 amps). Both air and oxygen were used on the cathode and only pure hydrogen was used on the anode. The cell failed after approximately 20 hours 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. Figure 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), gaskets on the perimeter of the MEA with a gas diffusion layer and catalyst in the center on the anode and cathode side of the membrane, and 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, and 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 $\text{cm}^3\text{-sec}$ 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.

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 figure 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 done 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 figure 3. It was done in three sections. The first section of the test was done with anode and cathode pressures 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. Figure 3 shows an increasing 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 done on this stack due to the lack of equipment. No combustion occurred during the leak test because nitrogen was being used on the anode.

After performing the pressure decay leak check it was decided to continue running the stack because the test did not show any overboard leaks and the crossover leak was not identified nor quantified yet.

The stack temperature was the indicator that led to the final shutdown of the stack. The stack was being operated with oxygen and hydrogen and no humidification. The stack temperature reached 90°C without current or resistance heaters enabled. The hot point in the stack was in the middle cells, which read a temperature of 91°C. The pressure in the stack could not be maintained. Both of these symptoms indicated a crossover leak and combustion. So the stack was purged with nitrogen, and declared inoperable.

The data for these final hours with respect to pressure and temperature is shown in figure 4, which shows spikes in temperature as the pressures decayed. The cell temperature measurements were taken at eight different points throughout the stack located in the bipolar plates in between each cell. For simplicity purposes they were called measurements 1 through 8, where measurement 1 is located between the end plate and cell #1, where cell #1 is the first cell the gasses encounter when they enter the stack.

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.

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

In figure 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 to 8 started to decline as was expected. However, the temperature measurements 1 to 3 continued to increase. This temperature rise showed signs of a breach possibly in cell #1 and/or #2.

Figure 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 figure 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 the oxygen due to combustion which, in turn, caused more oxygen to be forced 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 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 figure 9. Therefore, the oxygen breach should only affect the subsequent cells.

Cell #2 in Figure 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 indicates that there was 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. Figure 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 an off white color indicating degradation.

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.

Figure 12 illustrates what a good cell with no degradation of the gasket should look 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 Figure 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 Figure 14. 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.

Figures 15 & 16 show images of the silicon gasket material taken with the scanning electron microscope. In figure 15 the silicon gasket material was exposed to combustion from cell #2, and figure 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 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 though it may 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 cross over to the anode side causing the stack to heat up significantly.

4. Acknowledgments

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5. References

1. Silicone
http://www.stockwell.com/newstuff/newpdfs/500_solid_silicone/510_co_hr_9275_high_perf_clear_silicone.pdf
2. Barbir, F., *PEM Fuel Cells Theory and Practice* (Elsevier Academic Press USA, 2005).
3. ElectroChem Inc website:
<http://www.fuelcell.com/pdfs/FuelCellStack.pdf> (accessed May 2006)

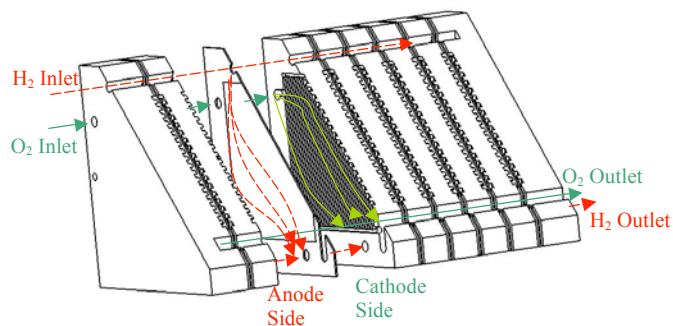


Fig. 1. A cross sectional view of a typical parallel flow of the reactant gases through a fuel cell stack

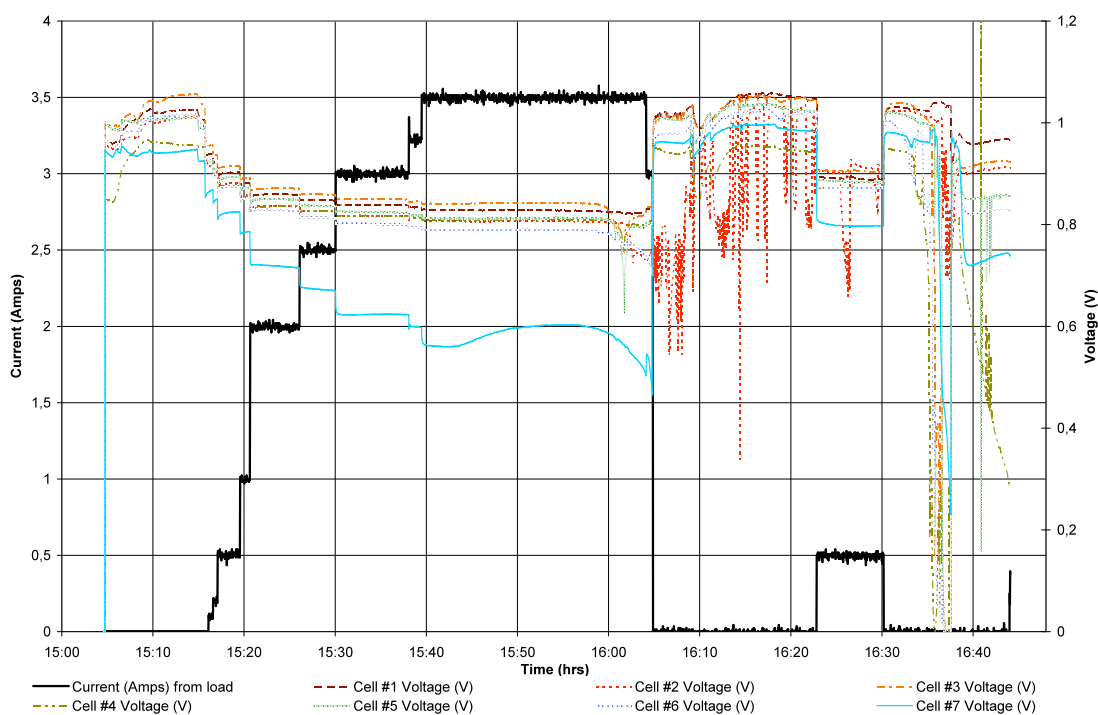


Fig. 2. Normal voltage current response of the 7 cell stack a few days before failing, H₂/O₂, 1.02 Stoichiometry for anode and cathode, 60°C stack temperature, no humidification and a gas pressure of 4.50 bar (a) (50 psig) for both the anode and cathode

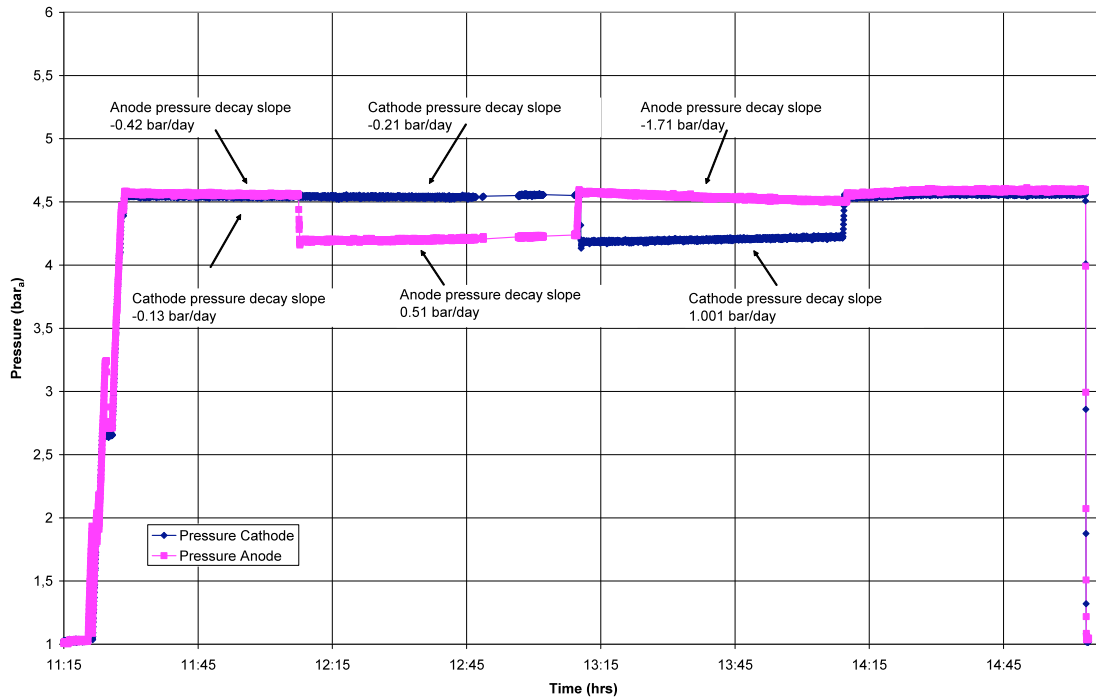


Fig. 3. Pressure decay leak check of stack at ambient temperature with air on the cathode and nitrogen on the anode

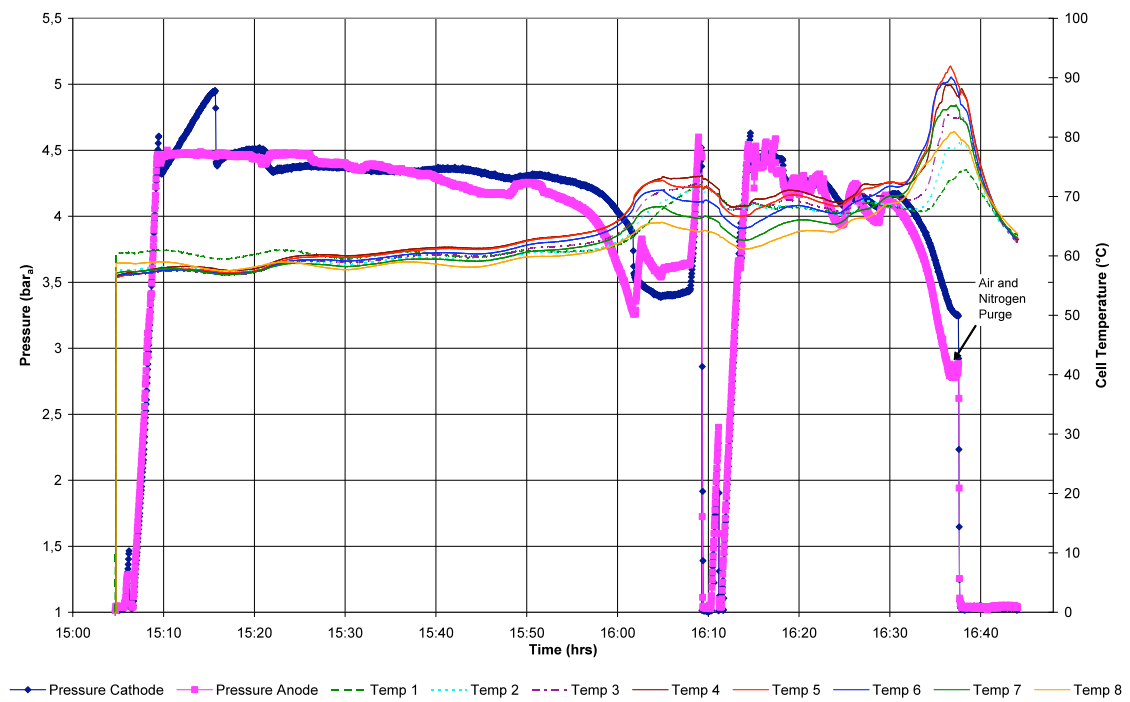


Fig. 4. Pressure of the reactant gasses vs. cell temperature during the last hours of operation. *H₂/O₂, no humidification??*

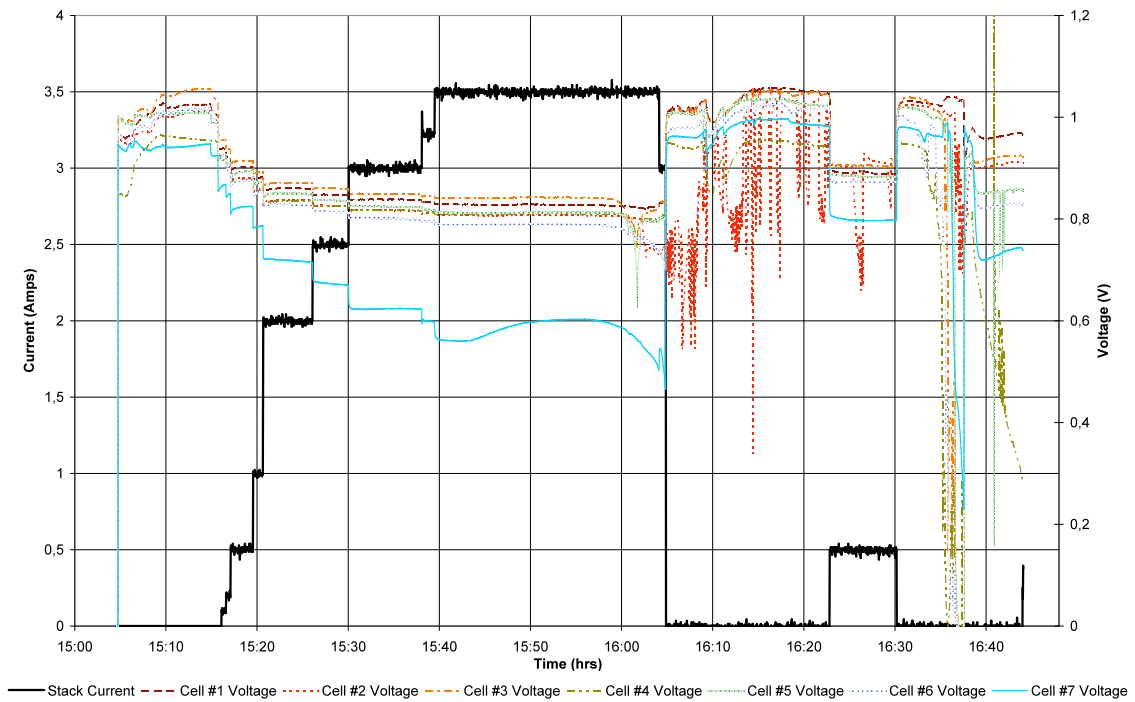


Fig. 5. Stack current vs. individual cell voltage during the last hours of operation, H₂/O₂, no humidification and a gas pressure??

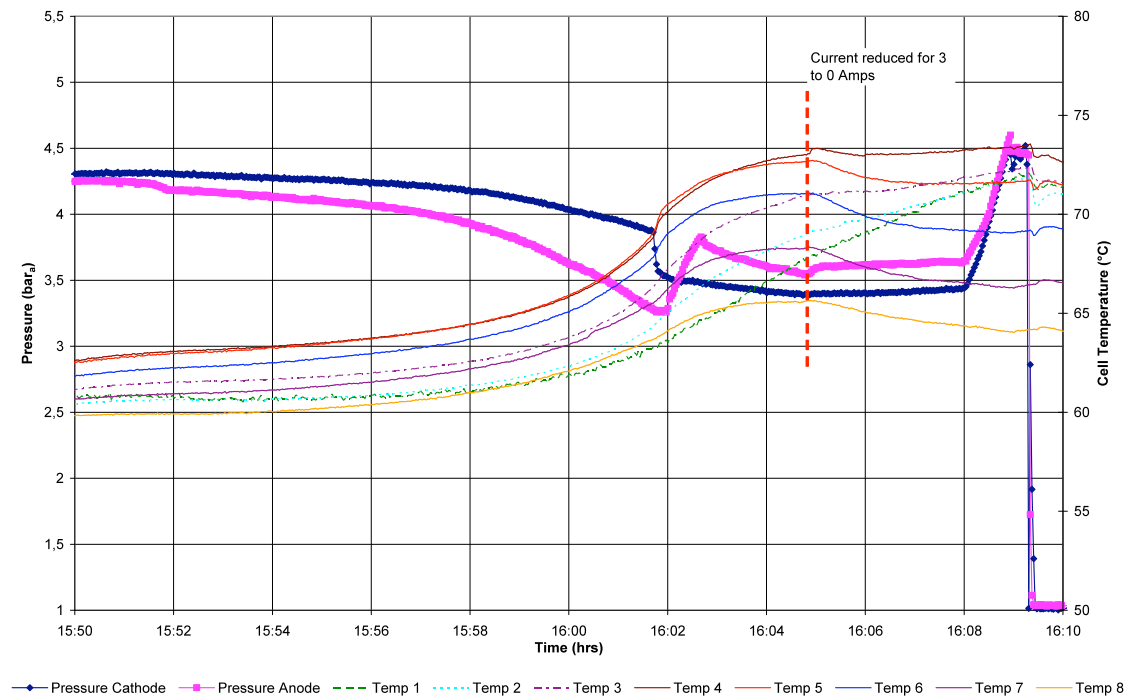


Fig. 6. Zoom of fig 4 between 15:50 and 16:10. Increase in temperature in cells #1 and #2 with no electric load or external heaters on the stack and an anode reactant pressure higher than the cathode pressure when the load was removed

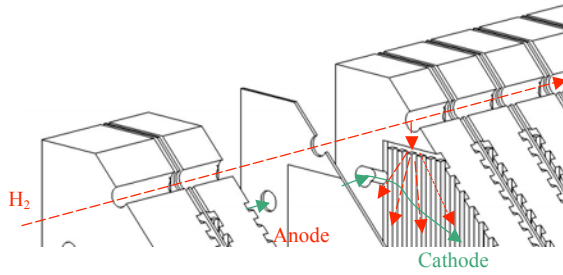


Fig. 7. Flow of the reactant gases when there is a breach in the seal at the anode inlet port that goes into the cathode flow field. Anode pressure is higher than the cathode's

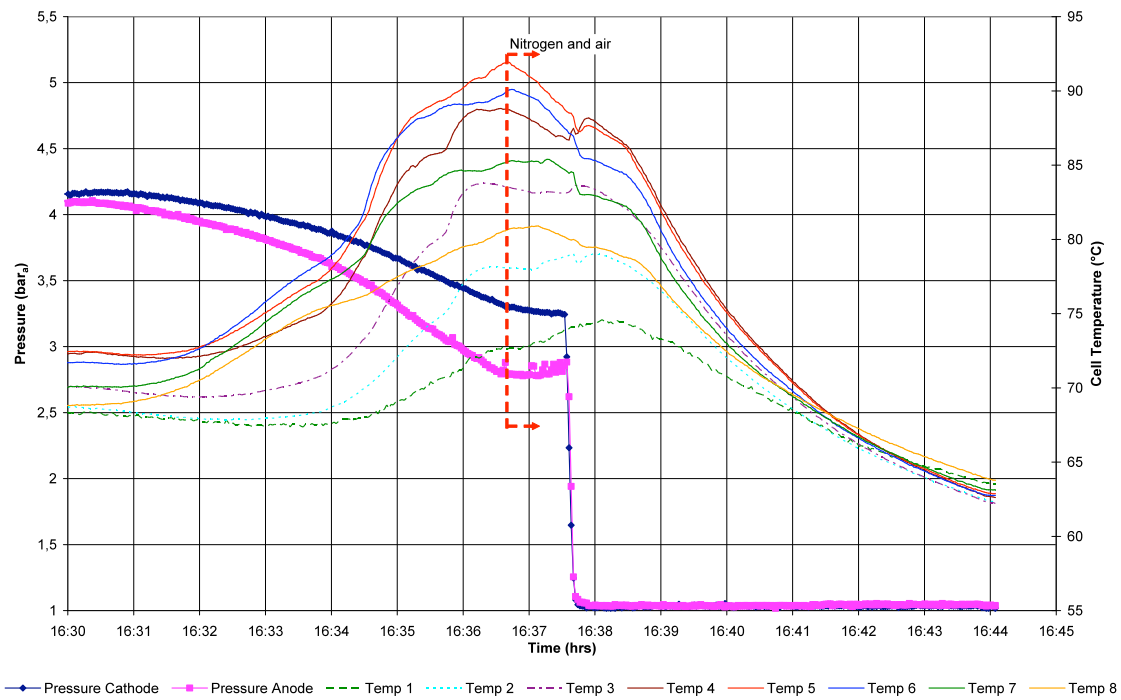


Fig. 8. Zoom of fig. 4 between 16:30 and 16:45. Cathode pressure is higher than the anode's and the hot spots in the stack are from cell #3 to #6 indicating that the oxygen was entering into the hydrogen stream through the inlet port

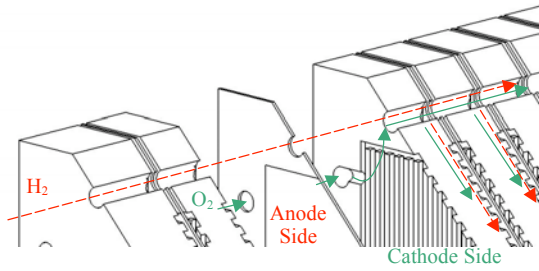


Fig. 9. Flow of the reactant gases when there is a breach in the seal at the anode inlet port. Oxygen is entering into the anode inlet port. Cathode pressure is higher than the anode's

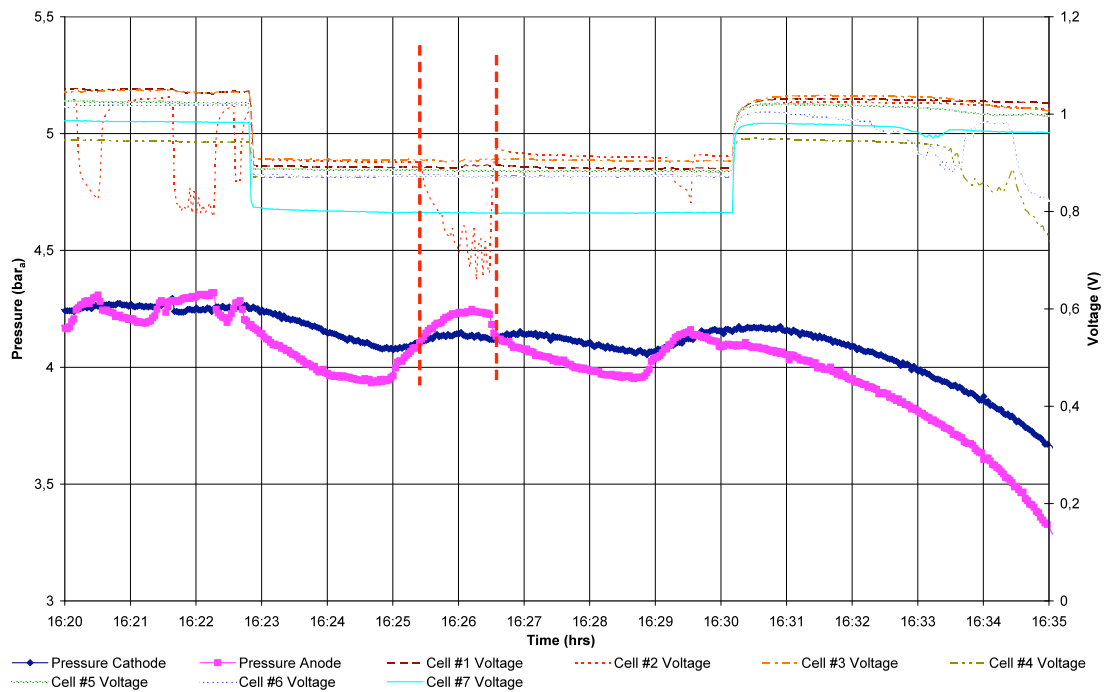


Fig. 10. Temporary instability of cell #2 when the anode pressure is higher than the cathode's. Hydrogen enters directly into the cathode flow field of the cell

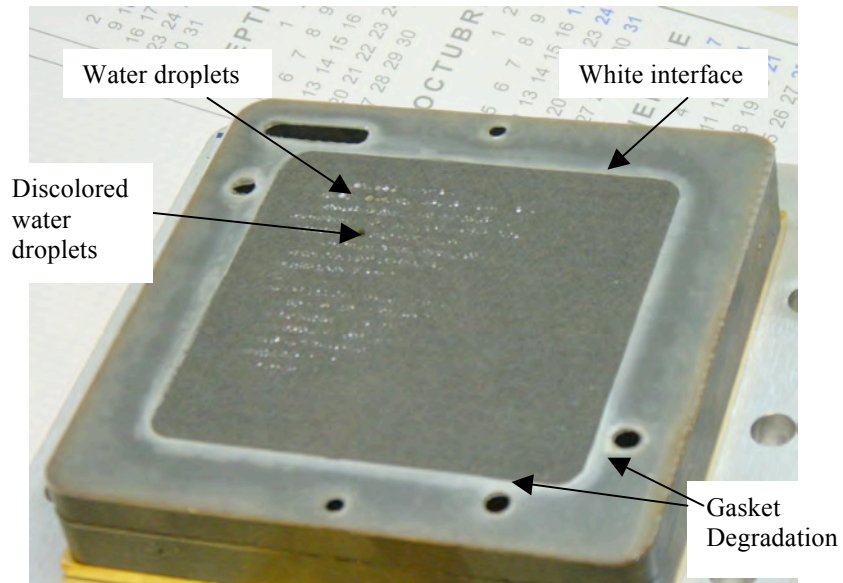


Figure 11. Cathode GDL and gasket of cell #2 before it was removed from the bi-polar plate



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

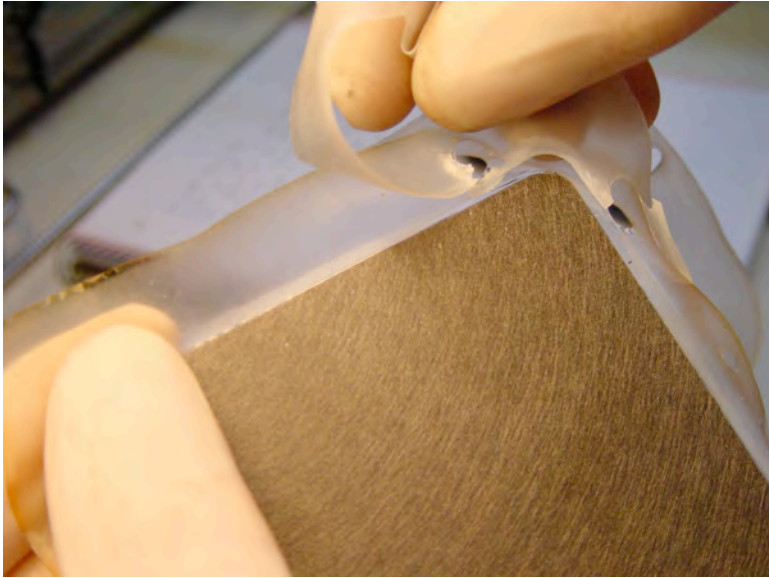


Figure 13. Cell #2 cathode gasket degradation

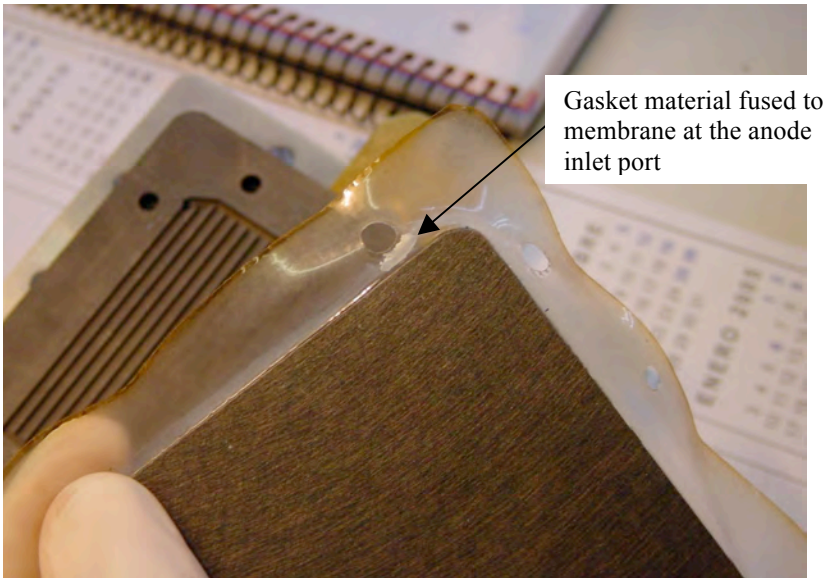


Figure 14. Gasket material left on the membrane at the anode inlet port and on the cathode side of the membrane

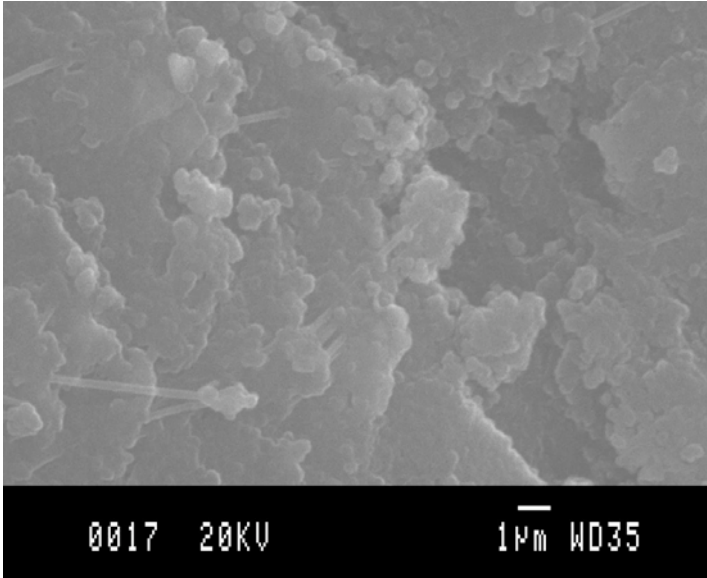


Fig. 15. Silicon gasket material that was exposed to the combustion from cell #2

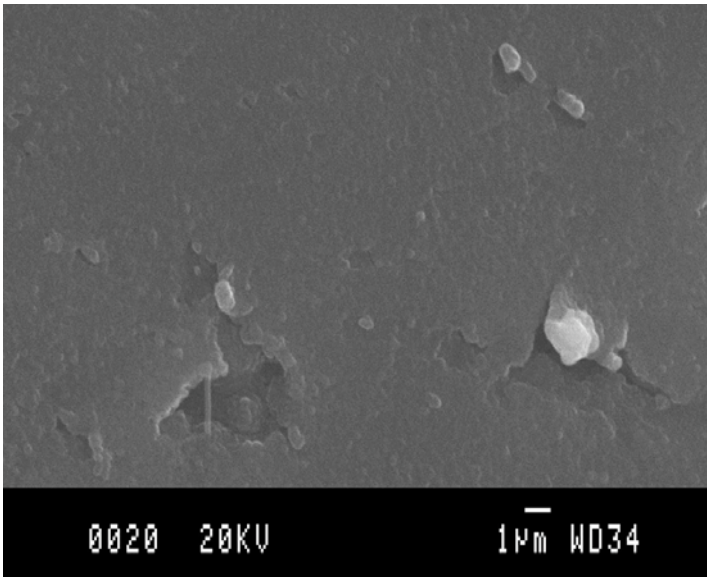


Fig. 16. Silicon gasket material that was not exposed to the combustion from cell #4