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Improved Round Trip Efficiency for Air Independent Regenerative Fuel Cell Systems

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Technical Report

RPT1736



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Executive Summary

The goal of this Office of Naval Research sponsored project is to develop high energy density energy storage systems for unmanned underwater vehicles (UUVs). ONR is interested in regenerative fuel cells for UUV applications due to the capability for high energy storage density vs. batteries. Improved fuel cell and electrolyzer efficiency are desired for higher energy density and faster refueling times. ONR funded the first phase of this effort as a collaborative project between Proton Energy Systems (d/b/a Proton OnSite) and W.L. Gore to make advancements in membrane technology, increasing the electrolyzer efficiency. Focus was on incorporation of reinforced membranes to enable lower ionic resistance through membrane thickness reduction.

Current commercial electrolysis stacks utilize 7-10 mil (225-250 μm) membranes, which can contribute up to 300 mV of overpotential at operating current densities. Past studies at Proton have indicated that 5-mil Nafion represents a practical limit for standard membrane thickness in electrolyzers, with failures seen under accelerated stress testing conditions, particularly at elevated temperatures. Past collaboration with Gore had shown robustness with full Gore MEAs even at 60-90 microns (~2-4 mil) but poor electrochemical performance. During the work reported here, a press technique was successfully developed to attach Proton electrodes to membranes from W.L. Gore without significant damage to the membrane.

Based on the studies performed to date, it was demonstrated that the resulting MEA was able to pass through an accelerated stress test and continue to operate for 1,000 hours, while showing improved electrochemical performance over N115. Membrane samples were pressure cycled 300 times and then allowed to operate at 80°C. Both the 60 and 90 micron samples passed this test. Both samples also passed the performance metric of achieving 1.9V at 2000 mA/cm², with the 90 micron sample at 80°C and the 60 micron sample at 65°C. The hydrogen permeation rate for these samples was similar to that of N115, but without any further processing, the crossover was found to be unacceptable at pressures over 200 psi or currents less than 1000 mA/cm². Using Proton's proprietary process for mitigating hydrogen crossover, the crossover rate was reduced to more typical levels, but performance was reduced somewhat.

The overall outcome of this project was successful electrode attachment to the Gore membranes without resulting in critical weakening of the membrane. This work is highly important in Proton's long term roadmap for increases in electrolysis efficiency because it demonstrates a pathway for significant reduction in operating voltage. A critical issue still to address is the hydrogen crossover. While the intrinsic permeability of the membrane is very consistent with existing commercial membranes for electrolysis, existing mitigation strategies for controlling this crossover in the cell have not yet been fully successfully applied to the W.L. Gore membranes. Proton has a National Science Foundation project which will assist in exploring the crossover issue. Additional ONR funding is expected in order to scale up the samples to Proton's 89 cm² MEA platform.

Introduction:

Proton is currently performing on a multi-phase program for ONR for air-independent energy storage applications. Regenerative fuel cells are an ideal candidate for this application due to the high energy density and cycle life vs. battery solutions. However, efficiency improvements are needed to enable higher current densities and more compact electrolysis modules. The first phase of this program was designed to investigate the potential advantages of using W.L. Gore's proton exchange membranes in Proton electrolyzers. Preliminary testing of Gore's MEAs conducted at Proton has shown that the thin, supported membranes can be extremely durable in the electrolysis environment and can offer a significant reduction in operating potential over Proton's state-of-the-art MEAs. In these tests it was observed that MEAs fabricated by Gore could run for several thousand hours but did so at very high overpotentials, typically running 200-300 mV higher than Proton MEAs of similar thickness. Conversely, MEAs that were fabricated via Proton's standard process with Gore's membranes showed a reduction in operating potential of several hundred millivolts but displayed such poor durability that they could not run for more than a few hundred hours before failing.

Therefore, the focus of this program was on adjusting Proton's electrode attachment conditions in order to realize the performance gains offered by the thinner material without sacrificing durability. Characterization of the physical properties of the membranes was also performed to ensure that the inherent hydrogen gas permeation for the Gore membranes would not be significantly higher than 5-mil Nafion. However, without any treatment, the crossover of both the Gore membranes and standard Nafion are unacceptable at low current or high pressure, and Proton has an internal process for treating Nafion membranes to mitigate this issue. Issues that still need to be addressed are 1) how to modify Proton's internal processes for the Gore membrane to reduce the hydrogen crossover to acceptable levels for normal operation and 2) scale up to larger MEA sizes and stack sizes.

Technical Approach:**Process/Data**

A key operational goal for this program was to fabricate an MEA that was able to run through an accelerated stress test (AST) procedure involving cycling the differential pressure in the cell between 0 and 400 psi 300 times, and then to continue to operate at steady state for 1,000 hours. The performance target for the new materials was to show an improvement over the performance of 5 mil Nafion membrane and to reach 1.9V at 2000 mA/cm². The initial test plan involved first collecting material property data for each thickness of the Gore material for comparison to N115 and N212. The major part of the effort involved processing studies to effectively attach the catalyst electrodes. Through recent development efforts, milder conditions have been identified for hydrocarbon membranes that were used as a starting point for optimization of key press parameters. Each new set of parameters was then evaluated for effectiveness in terms of electrode transfer and adhesion to the membrane, operating potential, and long-term durability. Operational testing was planned for the bench top 25 cm² test cell and for GC hardware. The size of the samples purchased from Gore prevented scaling up to larger cell sizes in this phase.

Several variables were identified at the outset of this effort that were most likely contributing to the loss of durability observed in previous testing with Proton electrodes on Gore's membranes. First, it was theorized that the standard press conditions used for Nafion were causing some degree of thermal breakdown in the Gore material. In Proton's standard process, the press box is

held under load and heated to high temperature in order to allow the binder in each electrode to flow and adhere to the membrane surface. Gore has indicated that its membranes have a different internal structure than Nafion, which is designed to provide additional mechanical stability to such thin material. It was anticipated that using milder conditions could help to improve membrane durability, but changing conditions by too much can lead to poor electrode transfer and negatively impact the cell's operating potential. Adding additional dwell time at the milder conditions was a strategy identified to counteract the slower decal transfer and produce high quality electrodes.

In addition, the idea of developing a completely alternate process was identified if flexing these variables was unsuccessful. This process would represent a significant deviation from Proton's standard technique, but it would allow for a drastic reduction of the press cycle time. A press cycle typically lasts several minutes to press the MEA. It was believed that the membrane may be able to withstand harsher conditions for a short period of time without being damaged significantly.

Membrane physical characterization:

Membrane samples were received from Gore with nominal thicknesses of 90 microns, 60 microns, and 45 microns. All membranes were tested for water uptake, ion exchange capacity/equivalent weight, and nitrogen diffusion rate for relation to hydrogen diffusion, with N115 as the baseline. Dimensional measurements were collected as well. This characterization data is included in Table 1 below. The Gore material has similar intrinsic properties to N115 in terms of water uptake and equivalent weight, but shows a significantly lower gas diffusion rate when normalized by thickness.

Membrane Sample	Average Measured Thickness (mil)		Water Uptake (%)	Equivalent Weight (g/mol)	Normalized Nitrogen Diffusion Rate, 200 psi (mL*mil/minute)				Normalized Nitrogen Diffusion Rate, 1000 psi (mL*mil/minute)			
	Dry	Hydrated			25°C	40°C	60°C	80°C	25°C	40°C	60°C	80°C
N115	4.96	5.98	40.48	1025	.825	1.20	1.85	2.70	3.55	5.03	8.00	12.0
Gore 90 µm	4.22	6.20	40.74	1027	.620	.779	1.15	2.02	2.44	3.30	4.96	7.55
Gore 60 µm	3.01	4.40	42.41	1074	.461	.685	1.04	1.38	2.31	2.79	4.37	6.50
Gore 45 µm	1.88	2.57	39.35	1080	.784	.912	1.27	1.58	2.88	3.83	5.91	8.17

Table 1: Summary of membrane characterization data.

Processing Studies: Traditional Method:

After collecting data on the material properties of all samples, the focus shifted to evaluating the electrode attachment method. An experiment was designed to illustrate the impact of lowering press temperature on cell performance using the 25 cm² bench top cell hardware. MEAs were pressed with standard Proton electrodes and Gore's 90 micron membrane at varying temperatures and polarization curve data was collected for each. The desired output of this experiment was to determine whether the performance would decline consistently with the reduction in press temperature or if the temperature could be reduced to some threshold value before any impact on performance was observed. The results of this testing can be seen in Figure 1 below.

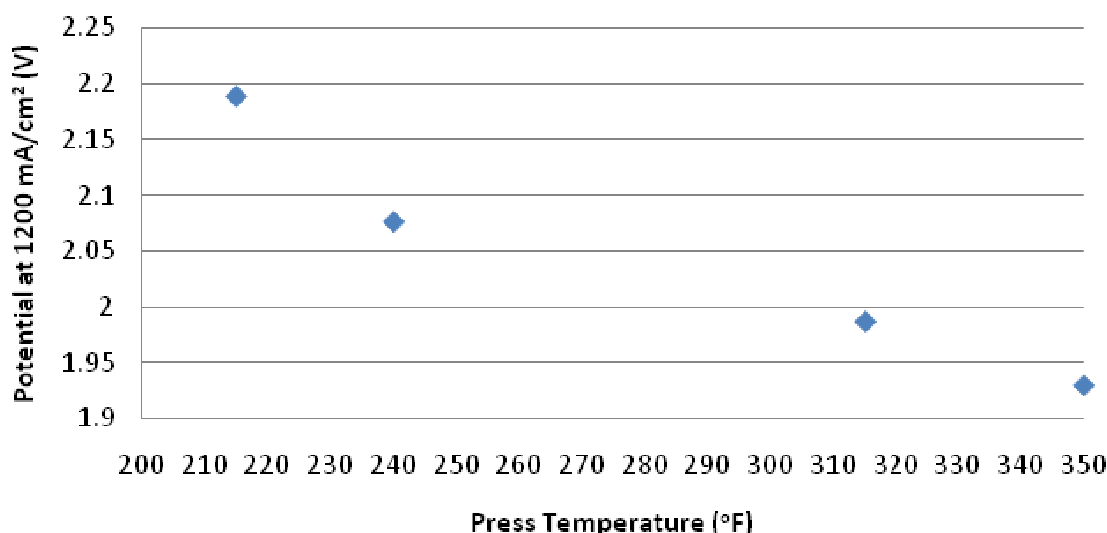


Figure 1: Impact of press temperature on MEA performance in 25 cm² hardware.

While this experiment was being conducted, some initial performance testing was also being carried out on 28 cm² MEAs. The first two attempts utilized MEAs pressed on 90 micron membrane, using press conditions identified during complementary work on a 2009-10 Phase 1 STTR on hydrocarbon membranes. 90 micron membranes were used in the first round of testing because, as the thickest samples available, it was believed that these would have the highest intrinsic durability. The membranes used were hydrated by boiling in water. Each MEA exhibited fair transfer and adhesion, but more catalyst was left on the substrates than is normally seen in the manufacturing process. Initial acceptance test protocols showed that the in-cell resistances were significantly higher than normal. When testing was started, each cell was planned to operate at steady-state for 100 hours to allow the potential to stabilize before beginning the AST. Allowing the potential to stabilize before beginning the AST would allow for comparison to the operating potential after cycling to verify that the test did not have a negative effect on the MEA performance. However, the first two MEAs showed operating potentials of about 2.2 V at 1.86 A/cm² and failed for high %LFL before beginning the AST.

It was decided to re-confirm the durability performance of the Gore-fabricated MEAs, to make sure that the early failures with the revised processing conditions were still due to the Proton process parameters rather than a change in the Gore material. Gore provided a set of half-MEAs with platinum catalyst on 45 micron membranes which were used for this purpose. Two of these membranes were positioned back-to-back to form an equivalent 90 micron MEA (RND1033601). The dry half-MEAs were die cut and built into a cell and then hydrated in cell with a single six hour heat soak at 65°C. The potential was expected to be high due to the use of two platinum electrodes, since platinum is not an optimal catalyst for oxygen evolution. This cell was not able to reach the desired 1.86 A/cm² current density due to high potential, and instead was operated at 0.7 A/cm² with a potential of about 2.35 V. As anticipated, this cell was able to withstand the AST and did not show any negative effects in the post-AST operation phase. A post-operational ATP showed about a 40% increase in in-cell resistance compared to the initial ATP but no change in gas crossover rate.

Advanced Processing Studies

After seeing no durability improvement with temperature it was decided to attempt to press an MEA utilizing a process closer to typical fuel cell MEA fabrication. Based on input from Will Johnson at Gore, the load was more of a concern than the standard temperatures used in Proton's production fabrication process. Therefore, the first try was done with a 90 micron membrane and a set of 25 cm² production catalyst decals. The press load was reduced by half, and the temperature was increased back to the original production level. There was no attempt to keep the membrane hydrated during pressing, which enabled thinner platens and shorter dwell times. When the cycle completed and the MEA was removed it was observed that the substrates were difficult to separate. The membrane had fused to the substrate in the areas around the catalyst decals and was damaged on removal from the substrate. Despite this poor result, electrode transfer and adhesion appeared to be very good. The membrane and substrate are pictured in Figure 2.

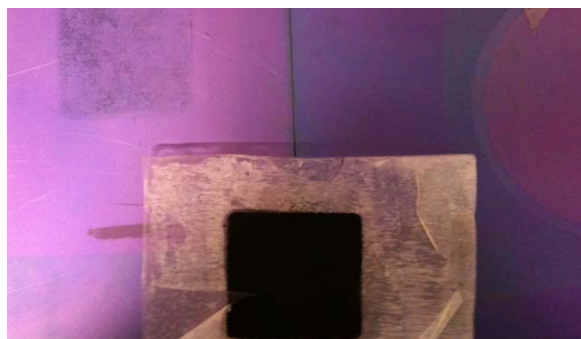


Figure 2: Result of first attempt to press an MEA with a dry membrane. Despite the obvious membrane damage, very little catalyst was left on the substrates.

Membrane Thermal Evaluation:

Several additional trials were run in an attempt to determine what factor was causing the damage and to find a way to alleviate it. The test was repeated with no electrodes to isolate the membrane impact. The temperature was reduced significantly and all other conditions were kept the same. The membrane responded in the same way as in the first trial, fusing to the substrates and tearing when the substrates were separated.

In each case the membrane seemed to hold its shape, suggesting that the material was fusing to the substrate chemically rather than simply melting or dissolving because of the elevated temperatures. The membrane material was reported by Gore to be stable to temperatures of about 320°F, again suggesting that the milder press cycle should not cause such severe damage. In response, the previous test was repeated with an inert Teflon sheet on top of each of the substrates. After this cycle, no damage to the membrane was observed. This result suggests that there was some interaction between the membrane and the decal substrates that caused the observed damage.

The next step was to repeat this trial with a temperature of 350°F to determine whether or not the membrane could handle the high temperature with the alternative substrate. Under these conditions the membrane did not appear to be altered dimensionally, but several small brown spots appeared across the sample. This phenomenon is pictured in Figure 3. Since the temperature was the only variable that had been changed since the previous test it was assumed that these spots represented a thermal breakdown within the material. Several additional tests

were conducted in the same manner with a series of temperatures, and it was determined that 270°F was the upper limit for pressing this material.



Figure 3: Membrane samples pressed between Teflon at 350°F (left) and 270°F (right). The brown spots that appeared are likely an indication of thermal breakdown within the material.

Membrane Processing: Refined Parameters

This new information fed into the design of the next membrane durability test. One theory for the failure of the MEAs pressed at low temperature was that the poor catalyst distribution resulted in hot spots in the MEA, causing mechanical failure during operation. A new GC MEA was therefore pressed with a hydrated 90 micron membrane at 270°F (RND1034402). It was anticipated that increasing the press temperature to 270°F would improve electrode quality without causing damage to the membrane. The MEA was pressed with a reduced press load and the dwell time was kept at 5 minutes. Electrode transfer was much better in this case than in the 215°F test, with cell resistances on the initial ATP within the normal range. Still, the cell exhibited high operating potential. This cell again exhibited a hydrogen crossover failure well before successful completion of the test procedure, after about 200 hours.

After seeing failures in previous testing with seemingly good quality electrodes, a series of tests were run to explore the possibility of reducing the process cycle time significantly through the use of a dry ink process. In the first attempt, 60 micron membrane was used with an existing 0.1 ft² cathode ink decal. The electrode, printed on Teflon, was placed on top of a production substrate, the membrane was placed on top of the decal, a bare Teflon sheet was placed on top of the membrane, and a production substrate was placed on top of the sheet. This set of conditions successfully produced a half-MEA with excellent electrode transfer and adhesion and much shorter press cycle. The membrane also showed no signs of damage. After several iterations an ink decal was successfully attached to a Gore membrane sample at these new conditions with excellent catalyst transfer and adhesion.

While the alternate pressing technique shows promise, it would require significant changes to other processes in order to be feasible for production. Ideally, a hydrated version of the shorter press conditions can be found. Additional trials were therefore done with variations of the new press conditions to try to find a combination conducive to working with hydrated membranes. Hydrated membrane samples were pressed on production substrates with two-minute and one-minute cycle times to find out if the shorter exposure time would keep the membranes from being damaged. However, the membrane still showed the same level of damage as in previous attempts, fusing to the substrate.

Electrochemical Characterization

Having demonstrated success with the alternate process and the need for further refinement with the traditional process, it was decided to restart operational testing with MEAs fabricated through the alternate process. As anticipated, all electrodes exhibited good transfer and adhesion to the membrane. 90 micron membrane samples were evaluated first in this series because it was anticipated that the thickest membrane would have the best intrinsic durability. One MEA had two Pt electrodes, for comparison with the 2 45 micron Gore $\frac{1}{2}$ MEAs. The two-cell stack (RND1035701) successfully operated for 100 hours at 50°C, 200 psi, and 1.86 A/cm², completed 300 pressure cycles, and continued to operate at steady state before temporarily suspending the test at 333 hours of continuous operation. The high hydrogen back-diffusion rate through the thin membrane resulted in a consistent 19% LFL reading. Figure 4 below shows a comparison of the operating potential trends for these two cells before and after the pressure cycle period. The data shows no increase in potential after completion of the AST procedure and relatively stable performance before and after, suggesting that the AST did not cause any damage to the membrane.

The next step in the process was to evaluate the 60 micron material. All pressing conditions were kept the same and the ink decals were printed from the same batches. The 60 micron MEA (RND1101401) was also able to successfully operate for a 100 hour break-in period, 300 pressure cycles, and a 100+ hour post-cycle period. The test was temporarily suspended after 447 hours with a final potential reading of 1.907 V. As expected, the CG sensor reading for this cell was higher than for the 90 micron stack, hovering around 30% at steady state conditions. Proton normally uses a proprietary process to mitigate hydrogen crossover in Nafion, which was not used in these early tests in order to assess the inherent performance of the Gore materials. As discussed later in this report, early work was performed late in the program to determine the viability of Proton's process for these membranes. Figure 5 shows a comparison of the pre- and post-cycle voltage trends for the 60 micron MEAs. Again, the operating potential did not increase in response to the AST, suggesting that the membrane was not damaged during this process.

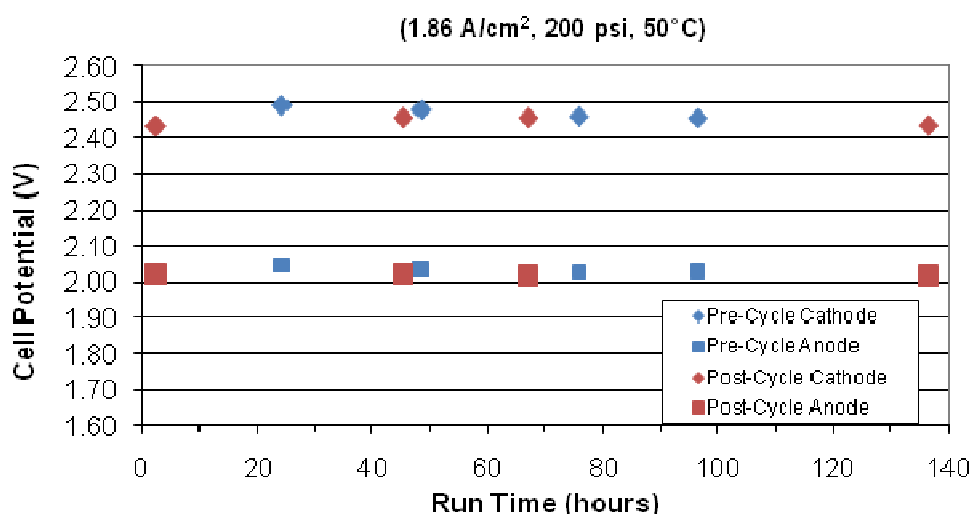


Figure 4: Comparison of voltage trends before and after pressure cycling: 90 micron membrane

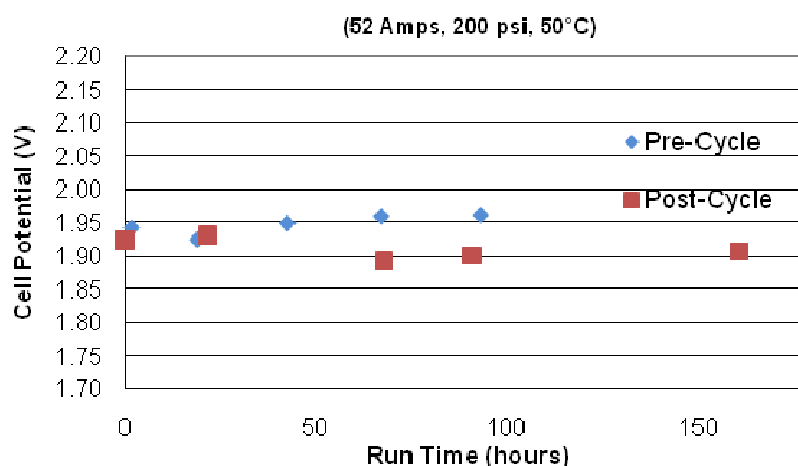


Figure 5: Comparison of voltage trend before and after pressure cycling: 60 micron membrane

45 micron membranes were evaluated in the same way, with all MEA fabrication conditions kept the same. The stack built with this cell (RND1103501) showed signs of failure and shut down for high gas crossover after 56 hours of operation at steady state. Review of the operating data showed a sudden spike in %LFL and drop in voltage at the time of shutdown. Post-operation testing showed a slight decrease in cell resistance and just a slight increase in nitrogen diffusion rate, but indicated an electrical short across the membrane. Disassembly of the stack showed an unusually high amount of cathode catalyst transfer to the cathode GDL which may have contributed to the failure. Removal of a large amount of catalyst could potentially result in highly-resistive “hot spots” where a thermal breakdown of the membrane could occur. Before the failure occurred, the cell had been running at 1.995 V and showed a CG reading of about 35% LFL.

The operating temperature was increased briefly on the 90 and 60 micron stacks to collect data on how the overpotential changes with temperature. Similar data was collected for 5 mil Nafion. This data is shown in the plot in Figure 6.

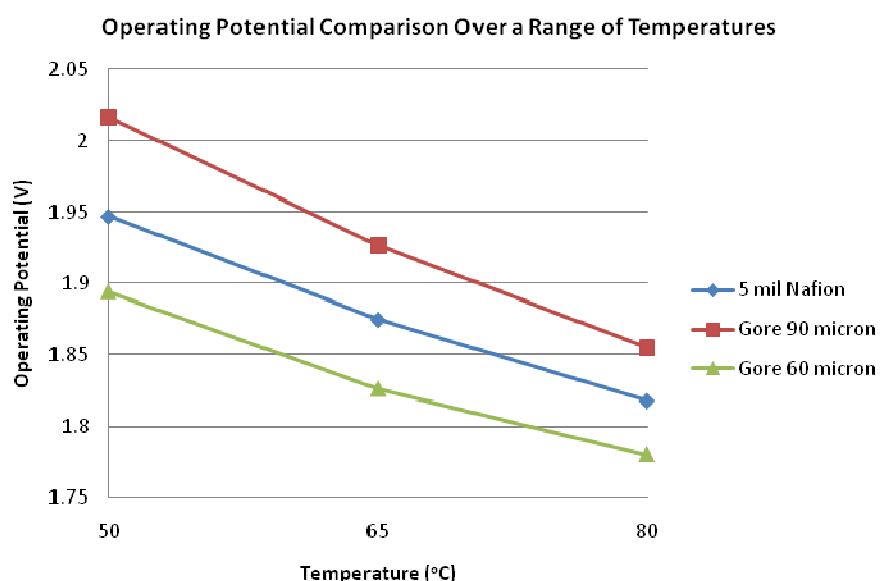


Figure 6: Comparison of Gore membrane to Nafion for several temperatures.

The 90 micron Gore material shows higher operating potentials than N115 for each temperature and the 60 micron membrane shows consistently lower potentials. The 90 micron membrane shows a larger percent improvement than the other two going from 50°C to 80°C.

Both the 90 and 60 micron cells were placed back on test in order to achieve the final operational target for this program: durability to 1,000 hours of operation. For the remainder of the testing, the operational temperature was increased to 80°C as opposed to 50°C to demonstrate durability at even harsher conditions. The voltage trend for the 90 micron stack is shown in Figure 7. At the end of the test the Pt-Pt cell was running at 2.298V and the standard cell was running at 1.883V at 80°C and 400 psi differential pressure. The stack was showing a 25% LFL CG reading under these conditions. Before removing the test, polarization curve data was collected for each cell.

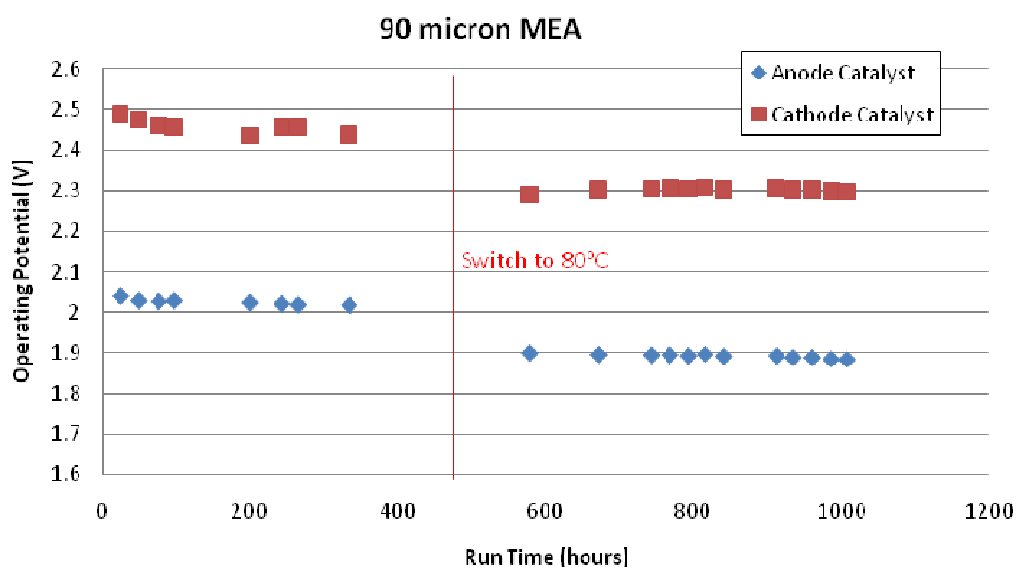


Figure 7: Voltage trend for 90 micron cell stack.

During this test, the current density applied to the stack was increased well beyond the typical operating level for a Proton electrolyzer to demonstrate the soundness of the cell design and its ability to run in a higher capacity system. This stack was operated for a very short duration at current densities as high as 5.38 A/cm², where it showed an operating potential of 2.56V (Figure 8). The polarization curve was also linear up to these current densities, indicating negligible mass transfer differences.

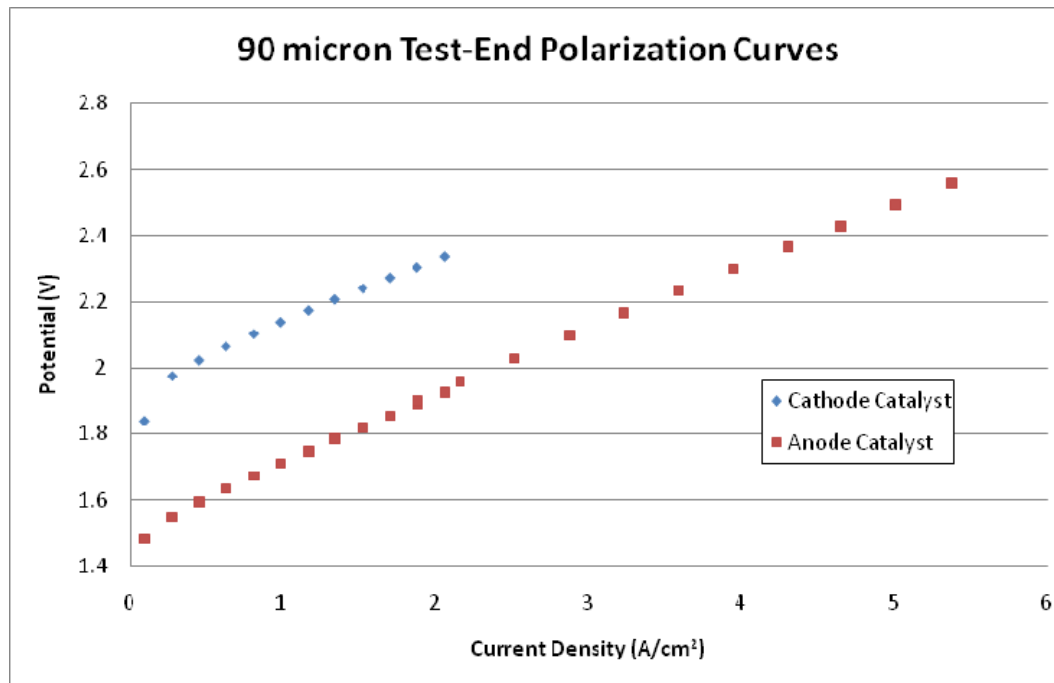


Figure 8: High current density polarization curve data collected for 90 micron stack after 1,000 hours of total operation.

The 60 micron cell was also able to complete its 1,000 hours of operation without failure. Due to high gas crossover rate at the elevated temperature, however, this cell was operated at only 200 psi. At the completion of the test this cell was running with a potential of 1.78 V at 80°C. The voltage trend for the life of this cell is shown in Figure 9. Under these conditions, the CG sensor displayed a reading of 33% LFL. After completing the 1,000 hour durability test, high current density polarization curve data was collected for this cell as with the 90 micron cells. The data is summarized in Figure 10. This MEA showed successfully met the final performance target for the program by displaying an operating potential of less than 1.90V at about 2,500 mA/cm².

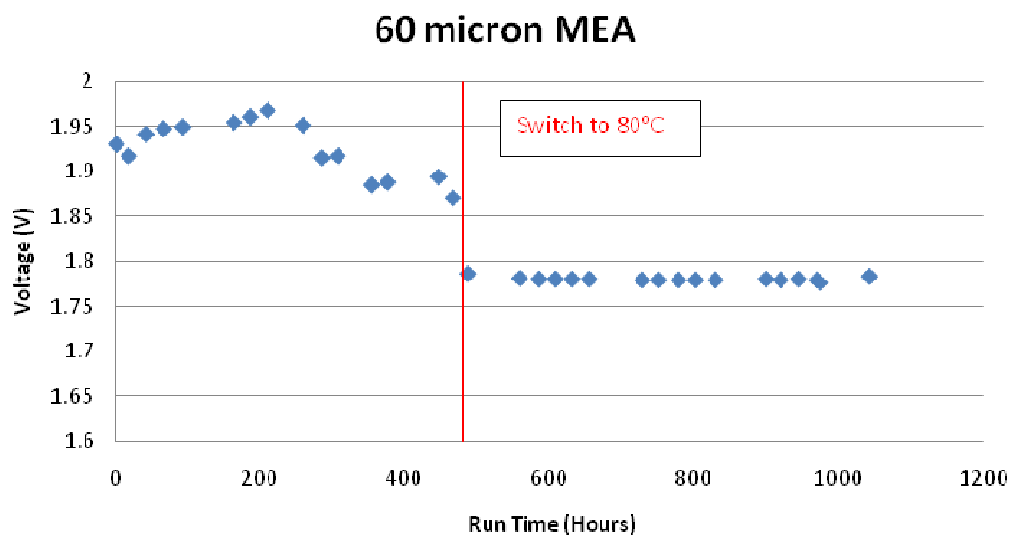


Figure 9: Voltage trend for 60 micron cell.

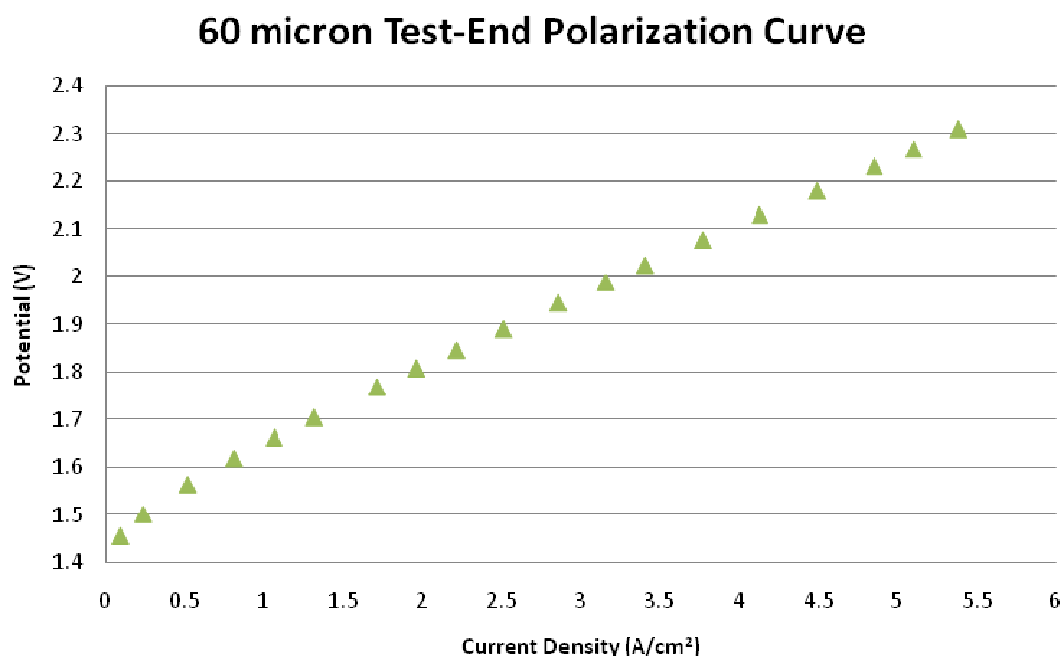


Figure 10: High current density polarization curve data collected for 60 micron stack after 1,000 hours of operation.

Hydrogen Crossover

In all of the operational testing done on this program, the thin membranes exhibited high hydrogen back-diffusion rates due to the lack of processing. In order to show the potential for correcting this, a 60 micron membrane was processed through Proton's proprietary process and was pressed with electrodes and tested. Gore's membranes proved to be challenging because of the use of a dry process on this program and because of how the material responded to the crossover mitigation process. Attempts to take the membrane through the process resulted in warping. One sample of the 60 micron membrane did platinize successfully, although it is unclear what was different in this sample. The anode ink electrode was printed from the same batch as in the 45 micron membrane evaluation, but the prints showed slightly lower catalyst loading in this case. Due to the reinforcement in the Gore samples, the x-y dimensional stability is very good, such that drying the membrane for the pressing process and then rehydrating it should not cause significant distortion of the electrode decals.

Discussion

The first phase of this program was largely successful, as a set of parameters was developed specifically for processing Gore's membranes that resulted in an MEA that could withstand the AST procedure and run for at least 1,000 total hours with a lower operating potential than N115. The durability targets were met by both the 90 and the 60 micron samples, with the 45 micron sample showing a failure before the initial 100 hour break-in period completed. The testing done on this program also showed that these membranes are stable at 80°C and that they can be treated to reduce the hydrogen back-diffusion rate to a level that is on par with MEAs with Nafion membranes. Based on prior work with Gore's materials, it is reasonable to assume that further reductions in operating potential can be achieved with a more optimized press process electrodes.

The bench top temperature evaluation suggests that, in the wet process, cell operating potential increased immediately and consistently when reducing the press temperature. The data appears to follow an approximately linear trend, though the sample size was relatively small. This effect is most likely a result of decreasing quality of electrode attachment to the membrane. All MEAs tested were pressed with the same dwell time, so it is still possible that a slight reduction in temperature could be offset with an increased exposure time. The information gathered from this experiment points out one of the limitations of the current pressing process, in attempting to use other membrane types than Nafion which may not be as thermally stable. As additional work, it would be useful to measure the temperature near the center of the press box to determine how hot the electrodes actually get during a typical cycle and how long they remain at that temperature.

Although it was unsuccessful on this program, the possibility of using a traditional process to fabricate MEAs with Gore's membranes still exists. The dry process works best of all the permutations tried in this study, but the work done so far is not enough to rule out other variations. The observation of thermal breakdown in the membranes above 270°F was an important piece of information to come out of this study. The spots that appeared in the material at sufficiently high temperatures provided a definitive measure of whether or not the material was responding negatively to the press conditions. What is still unclear is how this damage would manifest itself. If the ionomer was being damaged at these temperatures, performance would suffer. If the support structure was damaged, the durability could suffer. Although 270°F appeared to be the upper limit temperature based on the visual measurement, it is possible that the performance would not be affected by lowering the press temperature even further.

The dry pressing process developed under this program carries with it some significant benefits. The short cycle time could allow for a more efficient manufacturing process and is more conducive to automation than the current process. There are also some significant obstacles that would need to be overcome to use a dry process. One obstacle that has already been discussed in this report is the ability to incorporate hydrogen crossover mitigation processing into a dry press process. In general, this is an area that requires further development.

Conclusions

The work done on this program has shown the feasibility of using the thin, supported membranes from W.L. Gore to increase electrolyzer operating efficiency. A set of pressing conditions was developed to create an MEA with Gore's 60 micron membrane material and Proton's electrodes that could withstand an accelerated stress test procedure and operate for at least 1,000 hours without failure, while running at lower overpotentials than N115. Prior testing with Gore's membranes suggests that optimization of the fabrication conditions could result in further reduction of operating potential. Processing of the membrane has shown considerable success in reducing the rate of hydrogen back-diffusion rate, but additional optimization is needed.

Recommendations

- ◆ Further optimize dry press technique over temperature, press load, and dwell time
- ◆ Develop technique to mitigate hydrogen crossover in the Gore material without damaging it.

- ◆ Improve anode ink formulation.
- ◆ Investigate pressing hydrated Gore membranes with ink electrodes.