Design of Experiment

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Fuel Cell Project

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Introduction

The Proton Exchange Membrane (PEM) fuel cell requires a relatively stringent environment for operation. The conditions in the systems of the cell must be kept within a certain range for the cell to operate properly and utilize hydrogen fuel most efficiently.

For example: The membrane in a PEM fuel cell requires water to maintain its proton conductivity. If the membrane dries, the performance of the cell decreases. Alternately, if the water in the air stream becomes oversaturated, the excess water condenses, blocking the path of the air, cutting off the oxygen fuel, which reduces efficiency. The stack's temperature is another consideration. The cell's performance is below optimum before it reaches "operating" temperature (usually 75-100ºC), but must not reach excessive temperatures which may effect the humidity of the membrane and its conductivity. The mass flow of the hydrogen also needs to be enough for the reactions to occur so the cell generates power, but not excessive enough for the cell to be expelling unused fuel.

Purpose

The PEM fuel cell that was built last quarter does not yet have specifications for optimal operating conditions; the cell has been running using random operating conditions. The purpose of this project is to identify a combination of values, or a range of values, for variable factors that maximize the efficiency of the cell. In this case, efficiency is defined as being the amount of power created per amount of fuel consumed.

Initial Design

Originally, a range of the standard values were going to be used as a benchmark and a factorial design of experiments was going to be created using those values. Because the current operating conditions were chosen rather arbitrarily, published values seemed to be a valid starting point. Benchmarking the values, however, was difficult due to either an excessively wide of range of found values, or simply the inability to find standard values.

Due to the fact that published data was unavailable, benchmarking using the current operating conditions for the cell was the next best option. The cell was run using these conditions, which were established by last quarter's design team. The current pull in the cell was steadily increased until there was a significant drop-off in voltage. A polarization and power curve were created. From these graphs, the maximum power output of the cell at these specific operating and load conditions can be determined.

Variable Factor Selection

Although there are a countless number of factors that contribute to the efficiency of the cell, the factors chosen to vary in this experiment are the most operationally critical and are most easily monitored and adjusted. The chosen factors to vary and their current operating values are as follows:

Mass Flow of Hydrogen (centiliters/minute)	5 sL/min
Mass Flow of Air (centiliters/minute)	20 sL/min
Temperature of inflow air (degrees Celsius)	80ºC
Relative Humidity of Hydrogen	.8
Relative Humidity of Air	.6
Pressure Difference (psi)	<10 psi

Table 1: Current values for variable factors

A range for testing purposes was then chosen around those values and is as follows:

Mass Flow of Hydrogen (centiliters/minute)	5 g/min (fixed)
Mass Flow of Air (centiliters/minute)	10-30 g/min
Temperature of inflow air (degrees Celsius)	75-100ºC
Relative Humidity of Hydrogen	.7-.95
Relative Humidity of Air	.5-.75
Pressure Difference (psi)	<10 psi

Table 2: Chosen range of values for variable factors

Simplifications had to be made due to the inability to easily vary and monitor the effect of certain factors. The pressure difference between the air and hydrogen is currently monitored to stay below 10 psi. This constraint will be used for all testing runs and the value will not be varied in order to make sure the fuel cell is not damaged during testing. The temperatures of the inflow hydrogen and inflow air also, while being varied together, will not be varied relative to one another.

Factorial Design

A factorial design, specifically a three level (3^4) factorial design, was chosen for this project. In the first complete trial, there are four factors with three levels. Factorial designs investigate all possible combinations of the levels of factors in each complete trial. By approaching the trial systematically, the number of factor adjustments remains at a minimum.

The initial trial will give a good indication of the significance of each factor and that specific factor’s favorable level. The experiment can then be broken down further and more testing can be executed with more precise levels of factors being identified as favorable.

It was determined that performing a factorial design for all combinations of the chosen levels of these factors would be tedious and time consuming. Also, because fuel consumption is the dilemma that has created the need for alternate power sources such as fuel cells, it would be appropriate for the mass flow rate of hydrogen to be fixed and the other factors could be combined at different levels to yield the most power output of the cell for that given mass flow rate of fuel. As such, we would be identifying a combination of conditions that utilized a given amount of hydrogen fuel to its maximum energy potential.

Next, levels of the variables were chosen. The range of values must be divided into increments so that combinations of different quantities of these factors can be tested. The initial increment levels that were chosen would have yielded 1080 test runs. This necessitated the factor increments to be made significantly larger. Certain combinations of factor levels can then be identified as favorable for the cell’s operation. Once those factors have been identified, the scope of the factor levels can either be narrowed, or the initial range of the factors can be adjusted. For example, if the test runs that included the higher level of the relative humidity of hydrogen yielded significantly more power than the runs where the relative humidity of hydrogen lower, the other factors can be divided into smaller increments and combined with the greater humidified hydrogen for more thorough testing. Or, a selection of factor combinations can be run again and the relative humidity of the hydrogen can be increased outside of its original range.

The following increment levels will be combined and chosen; the factorial combination yields 81 test runs:

Mass Flow of Hydrogen (centiliters/minute)	5 g/min (fixed)
Mass Flow of Air (centiliters/minute)	10, 20, 30 g/min
Temperature of inflow air (degrees Celsius)	75, 85, 95ºC
Relative Humidity of Hydrogen	.7, .8, .9
Relative Humidity of Air	.5, .6, .7
Pressure Difference (psi)	<10 psi

These tests will thus determine the optimal relative humidities and temperature for operating the fuel cell stack. It will also determine the minimum amount of air that can be used in proportion to the hydrogen flow in order to make sure the stack is not starved for oxygen.

Final Design

It was discovered that modifications to the initial design were necessary soon after the initial design experiments were begun. The limitations were primarily due to the constraints of the test stand.

First, the mass flow of hydrogen was reduced after it was discovered that 5 sL/min was grossly inefficient. After several runs, the data was analyzed to determine the fuel utilization according to the following equation:

$$U = \frac{IN}{2\dot{n}_{H2}F} \qquad (1)$$

For the initially chosen flow rate of 5 sL/min, this indicated that the fuel utilization of the fuel cell was around 8%, which is unacceptably low. This indicates that mass transfer limitations play a very important role in the operation of the fuel cell. This could be improved in later fuel cell design by decreasing the channel depth or increasing the channel width, thereby increasing the amount of contact the hydrogen has with the diffusion layer. Also, increasing the cell size or connecting multiple cells in series would allow for a more complete reaction of the input hydrogen, due to the longer period of time in which the hydrogen was allowed to react. Modifying the cell, however, was not in the scope of this project. Therefore, the hydrogen flow was set as low as possible in order to minimize the amount of wasted fuel. The lowest flow rate that we could achieve with the current test stand was 2 sL/min. This gave provided a maximum fuel utilization rate of about 20%.

Another limitation of the test stand was attributed to the mass flow rate controllers. As the flow rate of air exceeds 12 sL/min, the controller could not manage large oscillations in the flow rate. Because this could affect testing in an unknown manner, the maximum flow rate was adjusted to be 15 sL/min, which allows for a satisfactorily broad range of flow rates with minimal interference due to oscillation. Consequently, the other flow rates were changed to be 6 and 10 sL/min.

Taking these limitations into account we decided to test according to the following range of factors:

Mass Flow of Hydrogen (centiliters/minute)	2 sL/min (fixed)
Mass Flow of Air (centiliters/minute)	6, 10, 15 sL/min
Temperature of inflow air (degrees Celsius)	75, 85, 95ºC
Relative Humidity of Hydrogen	0.7, 0.8, 0.9
Relative Humidity of Air	0.5, 0.6, 0.7

Table 3: Final range of values for variable factors

The final issue that was encountered with the test stand was the achievable temperature range to the input gases. Since the gases are heated by water-filled heat exchangers, the maximum temperature that can be achieved is theoretically 100 degrees Celsius. According to the humidification calculations (see spreadsheet in Appendix 2) for the 95 degree runs, achieving a relative humidity of 0.9 would have required the gases to be heated up to 101 degrees. Therefore, this block of testing was not performed, reducing the total number of data runs to 72.

In addition to the constraints caused by the test stand, a major change was made to the order in which we were changing the variable levels. After running several tests, it was clear that the system took a long period of time to achieve steady-state conditions. However, steady-state occurred very quickly after changing mass flow rates. Therefore, the testing order spreadsheet was modified in order to minimize the number of times that the temperature of the inlet gasses needed to be changed.

Data Analysis

There are a number of ways to measure the performance of a fuel cell. For different cases, efficiency, fuel utilization, or pure power output can be the best indicators of how a fuel cell is performing. However, for this projects fuel cell, the best indicator was the maximum power output. Because the mass flow rate of Hydrogen (the limiting reactant) was held constant, the efficiency and fuel utilization are effectively equivalent, since both represent the amount of hydrogen that is being burned to produce energy. Also, since the fuel flow rate is held constant, the maximum efficiency and fuel utilization will occur when the output power from the fuel cell is the highest. Thus, all options simplify to using the maximum power output as an indicator of cell effectiveness.

To analyze the data, the peak power output of the cell was determined for each set of conditions. There are two ways this can be determined: first, one can simply look at the data and pick the point with the highest output power. However, this strategy can give strange results since it does not do a good job of distinguishing between statistically significant data and outlying points. The second option, is to find a way to average the data points near the peak power, using all of the points to statistically find the average peak power for the run. This method, however, has one major drawback: the shape of the power curve is not necessarily standard. Therefore, a model needs to be developed that best exemplifies the shape of the power curve. If the model is not a good fit to the data, the errors in the model can propagate through to the result.

As seen below in Figure 1 (a representative polarization and power curve), the polarization curve for the fuel cell was linear throughout the data range. Since the power curve is simply the integral of the polarization curve, it is clear that a parabola can effectively model the power curve. For all but one data set, the parabola model has a correlation coefficient of greater than 0.99, indicating that the parabolic model satisfactorily fits the data.

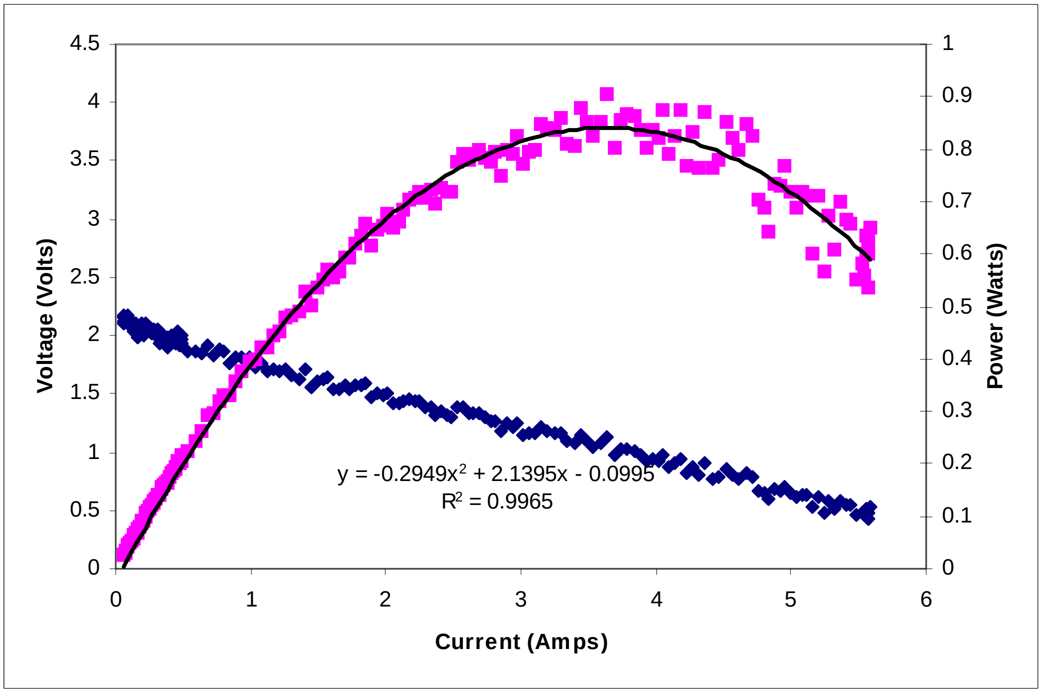

Figure 1: This is a representative polarization/power curve that was collected. Note that since the polarization curve is roughly linear, a parabolic model is a good description of power curve data.

The collected data was fit to the quadratic model, shown below:

$$P = C_2 I^2 + C_1 I + C_0 \tag{1}$$

As such, the current at the maximum power could be found by setting the derivative of the model equal to zero:

$$0 = 2C_2 I + C_1 \tag{2}$$

Finally, the current related to that maximum power value could be plugged back into the original model to determine the maximum power output of the cell, $P_{max,calc}$, according to the model. The results of this analysis, plus the peak power determined by the test stand, P_{max}, can be found in Appendix 1.

Results and Discussion

It is clear from the data that the humidification of the input gases is not an important variable for the operation of this cell. Graphs of the maximum power output versus the relative humidity of the fuel cell for each run can be seen below in Figures 2 and 3, for air and hydrogen, respectively. In both cases, the maximum power for each test run appears to have no dependence upon the humidity and is indicated by the fact that the regression lines are horizontal

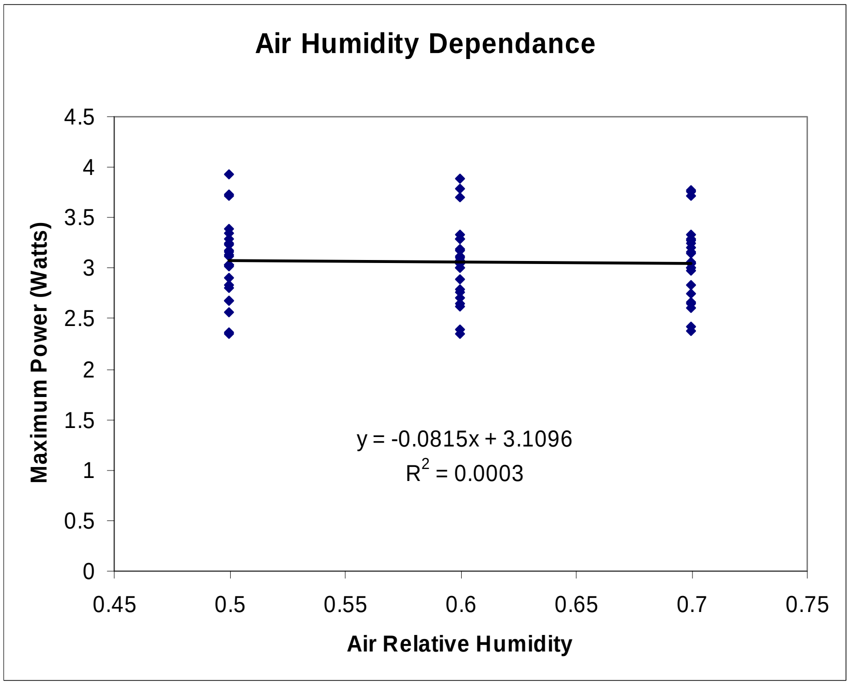

Figure 2: This graph shows the maximum power versus humidity of the inlet air. Since the linear regression yields a horizontal line, it is clear that the maximum power has little or no dependence upon the humidity of the air.

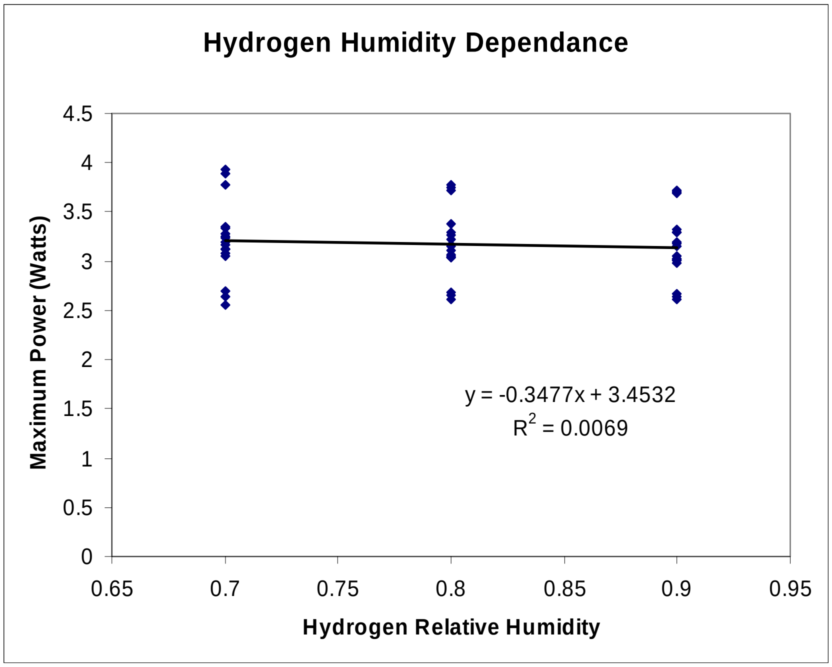

Figure 3: This graph shows the maximum power versus humidity of the inlet hydrogen gas. Since the linear regression yields a roughly horizontal line, it is clear that the maximum power has little or no dependence upon the humidity of the hydrogen gas.

However, unlike the humidity, the temperature of the inlet gases to the fuel cell stack has a dramatic effect upon the power output of the stack. From our testing, it appears that the fuel cell performs best with lower inlet gas temperatures. In Figure 4 (below) it can be seen that both the current and, consequently, the maximum power decreased as the inlet gas temperature increased.

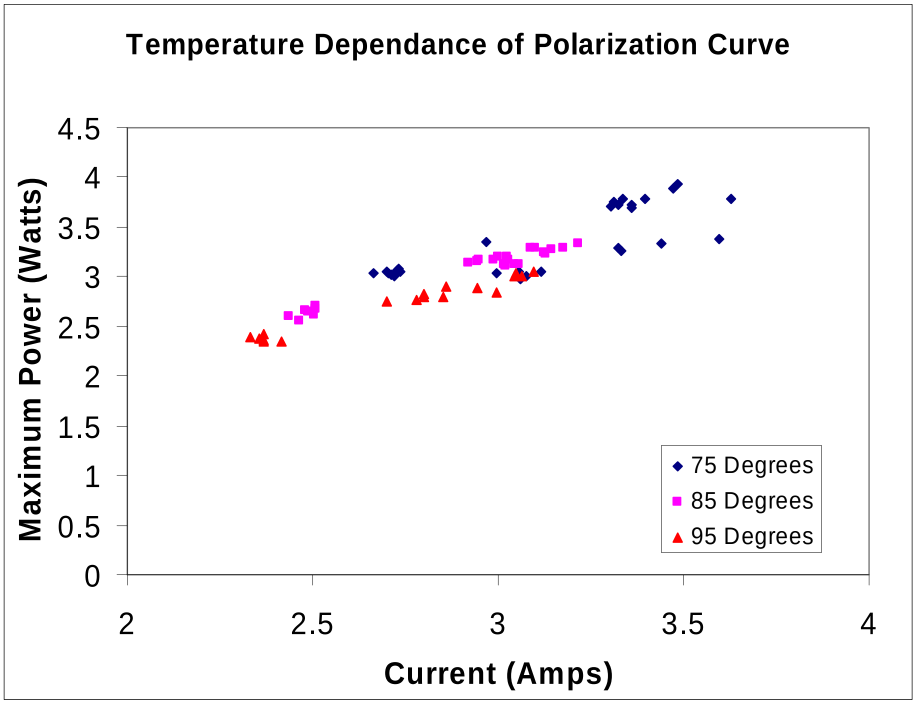

Figure 4: This graph shows the resultant current and maximum power output of the cell for different temperatures of the inlet gases. Note how the data point groups have lower currents and power outputs for higher temperatures. This indicates that the fuel cell was performing best when the inlet gases were cooler.

Figure 5, the maximum power versus temperature, below, shows the same trend. However, it may be easier to interpret since the trend is indicated by the slope of the line. Since the line has a negative slope, this indicates that the fuel cell performed better with lower inlet gas temperatures. Yet, it is clear that there is a large range of maximum power outputs for each temperature. From this graph, it is not clear why there is such a large range of values; it however became clear after the dependence upon the air flow rate was analyzed.

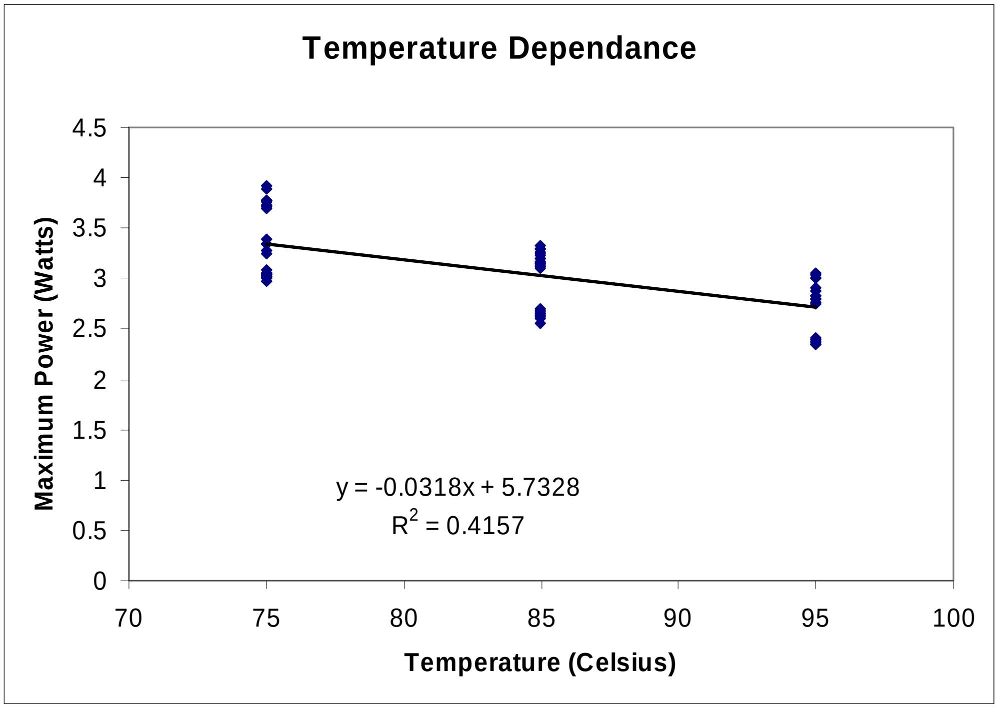

Figure 5: This graph plots maximum power versus the temperature of the inlet gases. It is another way to see the trend that the fuel cell was delivering more power at lower temperatures.

Similar to the temperature of the inlet gases, the data also shows that the flow rate of air was important for how well the fuel cell performed. In Figure 6, below, the fuel cell delivered a higher maximum power output for a lower flow rate of air. Similar to the temperature dependence, there is a wide range of maximum power outputs for each air flow rate.

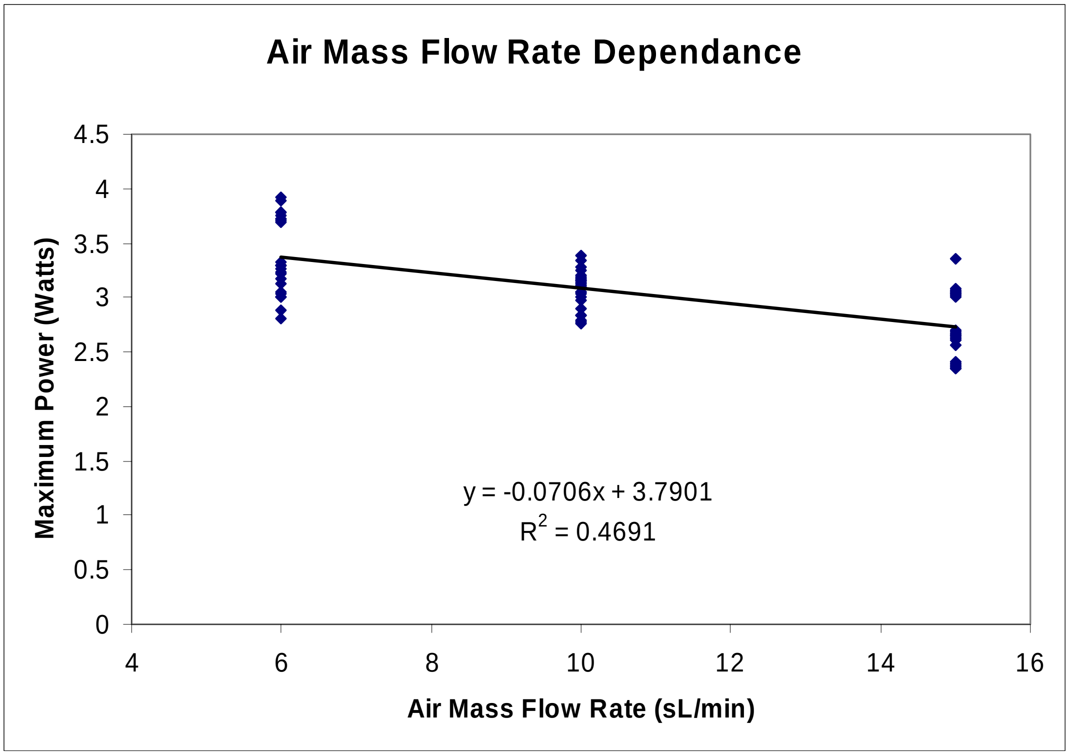

Figure 6: This graph shows the maximum power output of the fuel cell versus the air flow rate. As evidenced by the negative slope of the regression line, it is clear that the fuel cell performed better with lower air flow rates.

Taking all of these results into account, it appears that only the temperature and air flow rate were variables that were accounted for in the scope of this project that had a significant impact on the maximum power output. To determine if there was some other factor affecting the maximum power output, the results of Figures 5 and 6 were combined to see if each factor is the cause of the wide range of maximum power outputs for each independent variable. These graphs, Figures 7 and 8, can be seen below.

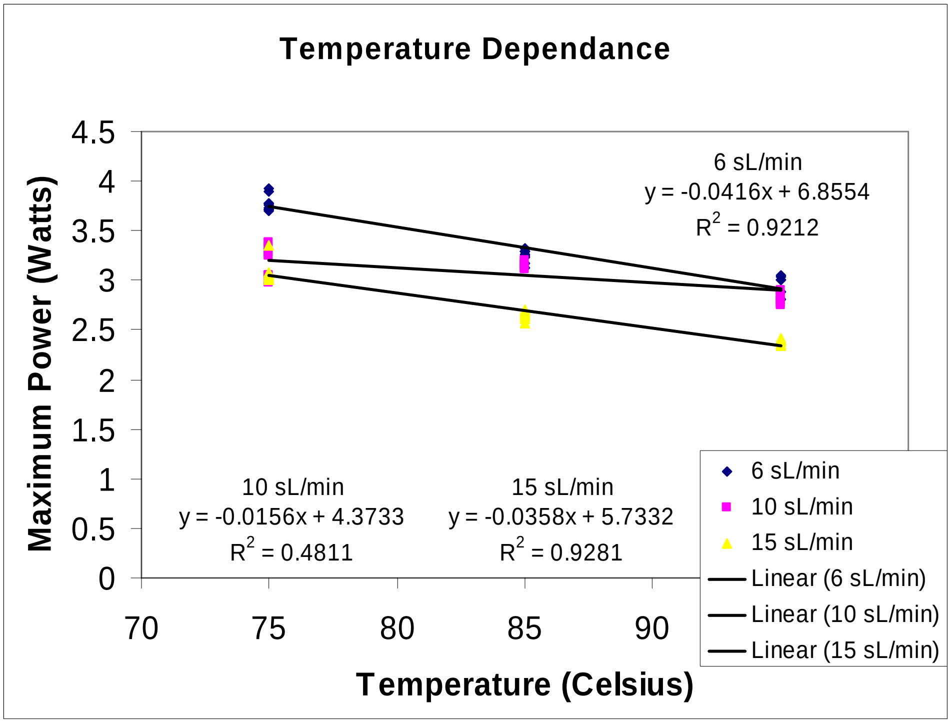

Figure 7: This graph shows that the range of power output values for each temperature was due to the different air flow rates. The lower the air flow rate, the higher the power for a given temperature.

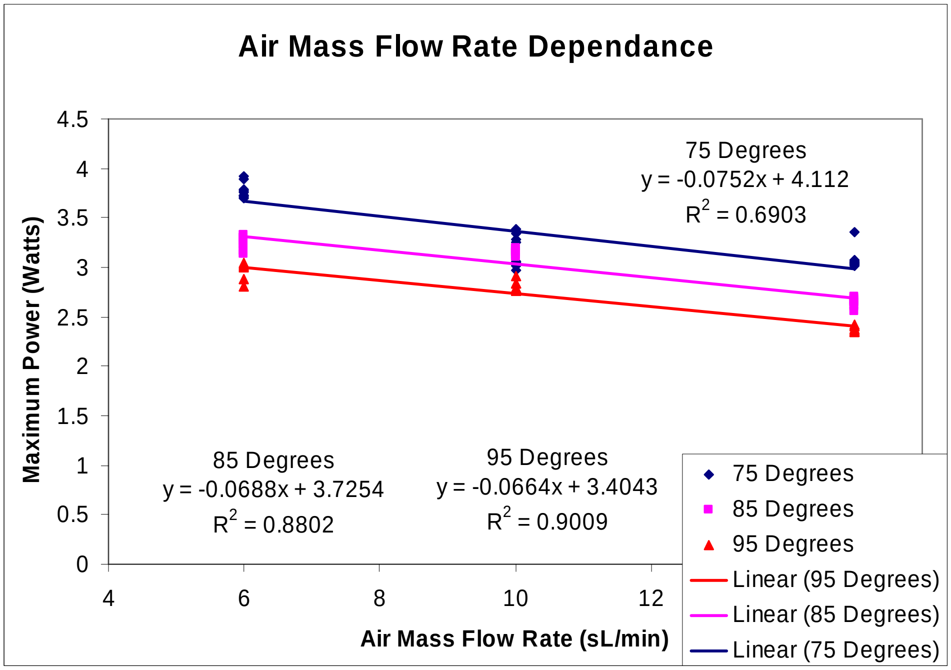

Figure 8: This graph shows that the range of power output values for each temperature was due to the different air flow rates. The lower the air flow rate, the higher the power for a given temperature.

From these graphs, it appears that taking into account the temperature and air flow rate can fully explain the maximum power output ranges. For the temperature graph, Figure 7, it appears that each group of data points for a given temperature have different values corresponding to different air flow rates; the data seems to fall into three separate groups corresponding to the three separate air flow rates. There appears to be some error in the data collected for 75 degrees and 10 sL/min since that data group does not follow the trends exhibited by the other data. Since that data group was collected on the same day, the first day of experimentation, it is likely that something was procedurally different that day, resulting in a lower than expected maximum power output.

Similarly, for the air flow rate graph, the range of power output data for each flow rate is explained by the different inlet temperatures. For this description of the data, all three lines appear parallel, separated by a vertical offset. This offset appears to correspond to the average power output difference between the temperatures. Finally, the correlation coefficients for each regression line are sufficiently large to indicate a good match. There is some error, likely due to the one day of testing, mentioned before, but, overall, it follows a linear model.

Conclusions

Overall, the humidification of the air and hydrogen streams appeared to have no effect on the performance of the fuel cell. However, the temperature and air flow rate both greatly influenced the performance of the cell. In addition, when both variables are taken into account, simultaneously, as in Figures 7 and 8, the data shows a very high correlation to a linear model with respect to temperature and air flow rate. This indicates that any other variables that might have been different during testing had little effect upon the performance of the fuel cell.

The independence of the performance of the fuel cell on humidification levels was not surprising. As past fuel cell testing on this project has indicated, a PEM fuel cell is either humidified or it is not. That is to say, either the fuel cell with function normally (when humidified), or it will fail if the membrane dries out (when it is not humidified). Since the humidification appeared to have no effect on the cells performance, it appears that, for the reasonable range of humidifications that were taken into account, the test stand satisfactorily humidifies the gas streams.

The fact that a lower temperature of the inlet gases resulted in better performance seems counterintuitive. Generally, it would be expected that a fuel cell will perform better at higher temperatures since the kinetics of the reaction are more favorable as the gases get hotter. However, at some point the performance of a fuel cell drops off as higher temperatures will cause the membrane to dry out. This can occur at any point in the cell, resulting in a drop of performance. In addition, one must remember that as the gases move through the fuel cell, they are reacting exothermically, which increases the temperature of the gas stream. Therefore, it is likely that the best inlet temperature for a fuel cell that lacks an external cooling system will be lower than the optimal operating temperature of the fuel cell. Thus, for hotter inlet gases, the temperature of the gases would eventually reach a point where the membrane was drying out, decreasing the performance.

A likely reason for the decreased performance with increased air flow is the pressure gradient that is created with higher air flow rates. Since the cell was run with a low hydrogen flow rate, a pressure drop was induced across the hydrogen channels in the fuel cell. The air flow rates were then higher than the hydrogen flow rate. Thus, the pressure drop across the air channels of the fuel cell would be much higher than the pressure drop across the hydrogen side. This results in a pressure gradient that is opposed to the transfer of hydrogen across the diffusion layer. This could possibly have a negative impact on performance and would explain the trends found for the air flow rate. However, fuel cells are very complicated, and there could also be other explanations for this result.

Experimental Design Conclusions

In retrospect, designing this experiment as a full factorial design might not have been the most time-effective way to go about running the tests. Since four factors were being varied, a fractional factorial experiment would have possibly been as thorough as necessary, but would have taken less time, allowing for a re-evaluation of testing parameters and a possible second round of testing. Fractional factorial design splits up the test combinations, choosing combinations dispersed throughout the testing sequence, that could show testing results with fewer test runs. For this, we would have analyzed the average change between the selected values for a factor rather than employing a one-factor-at-a-time method to graphing as we did in Figures 2, 3, 5, and 6. In addition, analyzing the data according to section 1.1 of Montgomery's Design and Analysis of Experiments, would have given us a direction in which to carry out further optimization as shown in section 1.2.

Recommendations

From this testing, we found there are a number of things that should be taken into account for future work on the fuel cell project.

Future Cell Design

The fuel cell that we tested was getting very low fuel utilization. This is likely due to the small size of the fuel cell compared to the flow rate of hydrogen that we were using. Therefore, it might be a good consideration for future design groups to look at either creating cells with shallower channels (which would increase the ratio of surface area of the diffusion layer to gas channel volume), creating larger cells (which would provide for a longer residence time, burning off more of the hydrogen), or designing the cells to work in series (which effective simulates a larger cell) to get a higher fuel utilization.

Test Stand Modifications

Overall, the test stand was helpful for the work that we did. In particular, the humidification system and the user interface worked well. Once the slew command was added to the UI, it became much easier to collect polarization data. Also, the system did a good job of controlling temperatures. While this did place a limitation on the maximum temperature available for testing, it was extremely effective in the range that it did allow. However, there were also a number of limitations that the test stand placed on the experiment. These include the following:

- Modify/Replace the Mass Flow Controllers – The mass flow controllers did not perform well. On start-up, they were particularly bad, allowing for several seconds of very high flow to occur before turning down the flow rate to what we set it at. Even after the test stand group wrote new start-up procedures to try and counter this, we found that the controllers were very finicky and often did not start up correctly. In addition, given the small size of the cells that the test stand will conceivably be testing, the lower limit of 2 sL/min is a bit high. To burn that much hydrogen, it would be necessary to build larger fuel cell stacks or string them up in series to burn off the hydrogen. Conversely, if a mass flow controller can be found that provides for good control on lower flow rates, that would be ideal for the hydrogen side of the system.

- Find better insulation – At one point, the temperature controls failed and one of the hot-pots began to boil and the insulation started falling away from the side of the hot-pot. It would be a good idea to get an adhesive that works over a wider range of temperatures for the insulation.
- Determine how close the humidifiers get to equilibrium – In order to put a number to the relative humidity, we used the equation, $\Phi = C \frac{P_{vap,pre-humidifier}}{P_{vap,post-heat}}$, which relates the vapor pressure of water at the pre-humidifier and post-humidifier temperatures along with a factor, C, which determines how close to equilibrium the gases get in the humidifiers. We assumed that the humidifiers would get to 80% of equilibrium since this is similar to what Duksu Hyun and Junbom Kim found in their study. However, this is not necessarily representative of our own test stand system. Therefore, performing tests to determine this coefficient, would be helpful. This test could be as simple as running air through the humidifier and measuring the mass flow of the air in and out of the humidifier. The difference in

mass flow between the input and output would represent the rate at which water was being absorbed by the air and could, thus, be used to determine how close to equilibrium the gases get.

Literature Sources

1. Montgomery, Douglas C. Design and Analysis of Experiments. 5th ed. John Wiley & Sons, Inc. 2001. New York.

2. Hyun, Duksu and Junbom Kim. "Study of External Humidification Method in Proton Exchange Membrane Fuel Cell." Journal of Power Sources. Issue 126. 2004.

3. NIST: Chemistry Webbook. http://webbook.nist.gov/chemistry/ May 4, 2005.

Appendix 1

“Copy of Fuel Cell Humidification” – In Attachment

Appendix 2

“Overall Data” – In Attachment

Appendix 3

http://students.washington.edu/akrinke/fuelcell/ - in May 19, May 20, May 27, June 2.