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Unique Fuel Cell Test Fixture Allowing Independent Control of Gas Sealing and Electrical Contact Pressure

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This paper describes the development of a unique single cell proton exchange membrane (PEM) test fixture to independently investigate the effects of gas sealing and assembly pressure. The test fixture is shown to provide useful insights into the effects of pressure on contact resistance. Two versions of gas diffusion layers (GDL) were tested to demonstrate that contact pressure significantly affects cell performance. In general, performance tends to increase with clamping force until a plateau is reached, after which performance drops rapidly. The rapid drop at high GDL pressure is due to puncture of the membrane or GDL mechanical failure. The performance improvements with pressure may be due to reductions in electrical or ionic resistance.

Introduction

PEM fuel cells are typically comprised of membrane electrode assemblies (MEA), hot pressed to carbon gas diffusion layers, and sandwiched between two electrically conductive plates containing gas flow channels. The gases are contained within each compartment (anode and cathode) of the cell by seals that are compressed between the oversized membrane and smooth outer surfaces of the flow plates. The correct compression of the seals is crucial to confine the gases, maintain high open circuit voltages, and minimize fuel crossover. Likewise, the amount of compression between the flow channel plates and each GDL is critical to provide good electrical conductivity, prevent the fragile GDL from compressive failure, and to limit GDL intrusion into the flow channels, which can restrict bulk gas flow.

It is difficult to create seals that are perfectly sized to allow good sealing and optimum electrical contact. Ideally, seal thickness will be slightly larger than GDL thickness to allow for some compression before electrical contact is made. Unfortunately, the dimensions involved are so small (common GDL thickness ~0.18 mm) that the tolerances needed to ensure proper fit are difficult to maintain. Additionally, swelling of the membrane when humidified, hand built MEAs constructed in research laboratories, and varying MEA designs from one test to the next only compound the sealing problem.

Experimental Approach

A unique single cell test fixture has been built to independently apply pressure for gas sealing and for GDL-MEA-bipolar plate contact as shown in Figure 1. The fixture consists of a polycarbonate plate (1) fitted with a typical (albeit small, 1cm²) graphite

flow channel (2). A circular groove (2) is machined in the graphite to secure a small o-ring. A mating polycarbonate plate (3) has a mating o-ring groove and o-ring, but instead of a conductive flow channel, it has a hole (4) through which a conductive (solid aluminum) cylinder (5) is inserted. This cylinder has its own o-rings (6) that create a gas tight chamber when it is inserted into the conjoined polycarbonate plates. The cylinder has a gas flow channel machined into one end (7) with manifold holes and hose fittings (8) that can supply gas from the other end. This configuration allows a typical MEA to be clamped securely between the o-rings of the two polycarbonate plates while the position of the cylinder and graphite flow block are axially varied to provide precise control of the GDL contact pressure. In this way, the pressure loaded onto the GDL can be varied without changing the gas sealing.

Figure 1. Exploded view of test apparatus and photo of actual rig.

Two linear springs were added to the test fixture to apply measurable pressure to the MEA where the force applied is directly proportional to the length of the springs as shown on the right in Figure 1. Testing was performed on small, 1 cm^2 fuel cells manufactured in-house using two different GDL constructions. The “standard” GDL had a single microporous layer (MPL) composed of PTFE and Vulcan XC-72 carbon black ink painted onto the electrode side of a Toray carbon paper (0.19 mm thick) GDL with a loading of 2 mg/cm^2 carbon and a 40% PTFE to carbon paper ratio, by weight. The second GDL type was similar to the first but with an additional PTFE coating (40% PTFE to carbon paper, by weight) applied to the current collector side of the GDL. Figure 2 shows a cross-sectional view of each GDL.

Figure 2. Schematic cross-sectional view of GDL compositions.

The catalyst ink for anode and cathode electrodes for both cells was made using a colloidal method after Uchida ⁽¹⁾. A 60:1 ratio of butyl acetate to Nafion solution was formed, and then carbon supported platinum catalyst (30% Pt by mass) was added in a ratio of 40% Nafion to catalyst by weight. This ink was then ultrasonically stirred for several hours and applied by airbrush to the MPL side of the GDL. Several layers were applied, with heated, forced-air drying between applications, until a catalyst loading of 0.4 mg Pt per cm² was achieved. The catalyst coated GDLs were then hot-pressed onto Nafion 115 membranes with 800 N/cm² pressure for 2 minutes at 135°C.

All testing was performed using room temperature hydrogen and air, both of which were humidified to 100% relative humidity by bubbling through columns of room temperature water. The gases were supplied to the cell at ambient pressure (exhausted to atmosphere), each with a flow rate of approximately 100 ml/min. The fuel cell was also held at room temperature for all testing. For each MEA, the test fixture was not opened and the MEA was not removed between any of the pressure increases.

Results and Discussion

Five different load cases were examined for each test performed. Figure 3 and Figure 4 show the resulting polarization curves for the normal GDL and the GDL with extra PTFE operating at room temperature on humidified hydrogen and air. These figures clearly show a performance increase in both cases for a contact pressure increase from 1.6 to 3.1 MPa. The performance improvements with increasing pressure after this level are not as significant, but the trend does seem to persist. These trends contradict similar experiments reported in the literature performed on cells operating at elevated temperatures. Lee et al. ⁽²⁾ found optimum cell compression by varying bolt torques for more pliable GDL materials, but found the lowest torque setting (equivalent to 1.6 MPa) to be optimal when using Toray carbon paper at 80°C operating temperature. Escribano et al. ⁽³⁾ studied compression ranging from 0.5 MPa to 10 MPa and found that the best performance was achieved at their lowest compression setting for a cell operating at 80°C with 1.5 atmosphere air. Ge et al. ⁽⁴⁾ observed performance similar to Lee et al. where compression optimums were found for cloth GDLs, but carbon paper GDLs exhibited the best performance with the lowest compression setting, at 65°C operating temperature.

These literature results indicate that increases in GDL compression reduce GDL pore sizes, ultimately leading to reduced mass transport and excessive concentration losses. The different operating characteristics observed in the current study may be due to room temperature operation where activation and ohmic losses play a more significant role than mass transport losses. This can be seen qualitatively in Figures 3 and 4 by the lack of the familiar drop-off in voltage at high current density, simply because the current density achieved here at room temperature is not high enough to enter the concentration polarization region. The case for this conclusion can be bolstered by work from Kitahara

et al.⁽⁵⁾. Like the data reported here, they found performance improvements with assembly pressure, including a plateau region above 460 kPa in testing at 80°C, which they attributed primarily to reductions in ohmic polarization. It seems that Kitahara et al. never experienced the reduced pore size and associated concentration losses observed by others⁽²⁾⁻⁽⁴⁾ at elevated temperature, simply because their maximum assembly pressure (0.77 MPa) was sufficiently low.

Figure 3. Polarization curves for a fuel cell under increasing GDL contact pressure for a normal GDL.

Figure 4. Polarization curves for a fuel cells under increasing GDL contact pressure for a GDL coated with an extra layer on PTFE.

Figure 5 and Figure 6 show impedance spectroscopy results for the fuel cells utilizing each type of GDL. The trends observed for the GDL containing extra PTFE show a correlation between assembly pressure and resistance. The left intercept, or bulk resistance, decreases and the right intercept, or total resistance, also decreases with increasing contact pressure. The curves shift to the left but essentially retain their size and shape throughout the contact pressure variation. However, the corresponding Nyquist plot for the normal GDL (Figure 5) shows unique behavior. The bulk resistance decreases with pressure as expected, but the total resistance increases with increasing

pressure, resulting in loop sizes that “grow” larger with increasing contact pressure. This behavior is not currently understood, but may be due to breakdown of the original microstructure of the catalyst layer.

Figure 5. Nyquist plots for a fuel cell under increasing GDL contact pressure for a normal GDL .

Figure 6. Nyquist plots for a fuel cell under increasing GDL contact pressure for a GDL coated with an extra layer of PTFE.

Figure 7. Current interrupt test plots of voltage recovery following a short circuit for a normal GDL.

Figure 8. Current interrupt test plots of voltage recovery following a short circuit for a GDL with an extra PTFE coating.

Figure 7 and Figure 8 show current interrupt tests performed on both GDL configurations at each pressure level. It should be noted that the two plots each contain compilations of 5 independent experiments, with each of the 5 results presented as staggered in time by 250 milliseconds (to clarify the presentation of each data set). For this test, a resistor (0.5 ohms) was placed across the fuel cell electrodes until the voltage stabilized, after which the resistance was instantaneously removed. The instantaneous voltage recovery shown as vertical dashed lines represents the portion of overpotential resulting from resistance losses. The measurements for the normal GDL clearly show decreasing ohmic resistance as contact pressure is increased. The results for the GDL coated with extra PTFE show a similar trend with one outlier (3.1 MPa).

Figure 9 shows electrical resistance values for varying contact pressure. For this test, resistance was measured across the test rig electrodes with either no MEA installed, one non-teflonated GDL installed, one GDL with a microporous layer (normal GDL), or one GDL with MPL and extra PTFE layer. As expected, the resistance associated with the test rig only is low and constant regardless of assembly pressure. Likewise, the resistance

for a non-teflonated GDL is low and constant across all pressure levels. However, it is clear that adding PTFE to the GDL increases resistance and creates a pressure dependence. The resistance for the normal GDL is relatively large and decreases with increasing contact pressure, while the resistance for the GDL with extra PTFE is even larger and likewise shows a decreasing trend with pressure.

Figure 9. Dependence of electrical resistance on contact pressure for test rig and 3 different GDL configurations.

Another contradictory and intriguing aspect of the assembly compression and performance challenge can be found in a paper by Zhou and Liu⁽⁶⁾. Their modeling work shows that GDL electrical resistance never significantly affects cell performance, even if the resistance is increased by an order of magnitude. But the current results clearly show that ohmic losses decrease with increasing contact pressure. The increases in this work may be due to reductions in ionic ohmic losses, as well as electrical ohmic losses. Bazylak et al.⁽⁷⁾ found that GDL compression actually leads to better liquid water transport as a result of the creation of preferential water flow channels as the GDL (Toray) fibers are broken. The crumbling GDL and PTFE create hydrophilic flow channels in the otherwise hydrophobic GDL. Such channels could improve membrane and catalyst layer hydration, which could explain the ohmic performance improvements observed in the current results. Improved hydration results in better ionic conduction⁽⁸⁾.

The maximum value of 8.5 MPa was chosen for each GDL, not by design, but by necessity. Both MEAs failed as the pressure was increased beyond 8.5 MPa. For the normal GDL, the graphite fibers of the GDL were crushed so badly beyond this pressure that they entered, and completely filled, the gas flow channels, blocking gas flow to the cell. In the case of the GDL coated with extra PTFE, the cell failed because the more robust GDL punctured the membrane causing excessive crossover. The observation of the first failure mode in previous work led to the idea of building a more robust GDL by using an extra PTFE layer. The resulting second failure mode perhaps indicates that the properties of current GDLs and membranes are well matched, mechanically.

An additional facet of compression pressure that is not considered here is the durability of a fuel cell under higher pressure and stress conditions. The current testing showed that immediate failure occurred above 8.5 MPa, but the duration of testing was not long enough to give any indication of the long term performance at the other compression levels. It is reasonable to assume that high compression pressure may lead

to mechanical stresses that will eventually cause premature failure even for initially well performing cells. Similar concerns are noted by Escribano et al.⁽³⁾.

The results herein show that assembly pressure may be a critical design consideration for small PEM fuel cells operating at room temperature, such as those commonly proposed for future portable power devices like cell phones or lap top computers. The surface area-to-volume ratio of such power devices may be too small to allow self heating, and the miniature, simple nature of the device may exclude the use of external heating. Because these applications will need to be optimized for both size and weight, the physical structure necessary to provide 4-8 MPa of clamping pressure will not be trivial.

Conclusions

The design of a unique test fixture that allows independent sealing and GDL contact pressure has been described. Results from the test fixture show significant performance variations based solely on GDL contact pressure. The performance increases with clamping force until a plateau is reached, after which performance drops rapidly as the membrane is punctured or the GDL experiences mechanical failure. The development of this unique test fixture has allowed GDL compression to be easily studied because it provides an opportunity to vary the GDL contact pressure with no impact on gas sealing. Several observations are noteworthy:

- For room temperature operation, concentration losses were never a factor for any compression level.
- Ohmic losses were clearly reduced with increasing compression before failure.
- The reduced ohmic losses may be due to electrical *or* ionic conductivity increases that resulted from compression.
- Traditional GDL and membrane materials are mechanically well matched.
- The durability of PEM stacks that use high compression levels is not determined.
- The clamp load required in a small PEM cell to achieve high compression is not trivial.

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