Effect of Temperature on the In-Plane Permeability in the Gas Diffusion Layer of a PEM Fuel Cell

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The in-plane permeability is an important property of the gas diffusion layer (GDL) in a Proton Exchange Membrane Fuel Cell. It governs the transport of reactants and water vapor from the channel to the GDL regions under the land of the bipolar plate, and the bulk mass transport in the case of interdigitated flow fields. The current focus in literature has been on studying the effect of porosity on this property. These studies have traditionally been conducted at room temperature. However, typical PEMFCs operate most efficiently between 65 and 90°C. In this study, an ex-situ test setup has been developed to investigate the effect of temperature on the in-plane permeability of the GDL. The in-plane permeability of humid air is measured for system temperatures of 25, 40, 60 and 80°C for samples from Toray Inc., SGL Corp. and Mitsubishi Rayon Corp.

Introduction

The transport of humid air through a GDL in a Proton Exchange Membrane Fuel Cell (PEMFC) plays an important role in designing the water management strategy. Permeability (1)(2)(3) and diffusion (4) have been identified to have a direct impact on the mechanism of water transport through the GDL.

Water transport through a GDL occurs in the through-plane and the in-plane directions. It has been shown that in the through-plane direction, diffusive transport takes precedence (4) while for the in-plane direction; permeability does play an important role. Mass transfer in the interdigitated flow field design is primarily driven by the in-plane permeability. Also in the serpentine flow fields, the in-plane permeability may influence the performance of the cell (3). The performance of the PEM Fuel Cell is a function of permeability of the GDL in both the in-plane and the through-plane directions.

Yan et al. (5) concluded that the performance curve for PEM fuel cells is significantly higher when operating at a temperature of 60°C compared to its operation at 40°C. Additionally, Wang et al. (6) pointed out that an increased temperature decreases the surface tension, allowing liquid water to enter otherwise hydrophobic regions. This indicates that the liquid water saturation may be higher at higher temperatures and block the reactant transport, which would bring the in-plane permeability into greater relevance.

Although work has been done in studying the in-plane permeability of different GDL samples, the extent of characterization is limited and no studies have been conducted on the effect of temperature on the in-plane permeability. Current work is focused on...
understanding the effect of increasing temperature on the in-plane permeability. Prasad et al. (7) explored the in-plane permeability in their detailed experimentation. Their study focused on the variation of the in-plane permeability for GDL samples with changes in porosity.

Gostick et al. (8) also studied the permeability of the carbon paper widely used as GDL material. Their study looked at both in-plane and through-plane permeability and concluded that the in-plane permeability was significantly higher than the through-plane permeability for the different GDL samples tested in their experiments. They also showed that the anisotropic nature of the material caused the in-plane permeability in the two perpendicular directions to vary by up to a factor of 2. They proposed that there are viscous and inertial components of forces that play an important role in the study of permeability.

However, no work has been done on the effect of temperature on the permeability of GDL. Aruna (9) in 1976 investigated the effect of temperature and pressure on the absolute permeability of porous media. He used sandstones to emulate uniform and homogenous porous media. The results showed that there was no change in the permeability of nitrogen with temperature, though the permeability of water vapor through the porous media showed significant variation with temperature. Water vapor permeability varied up to 60%, as reported for a temperature range of 20°C - 150°C.

The current work investigates whether variations in permeability due to increase in temperature such as those observed in homogenous porous media hold true for inhomogeneous and anisotropic porous media such as the carbon paper being used as the GDL materials.

**Experimental Setup**

An experimental setup has been developed which allows humidified air to be pumped through the GDL at measured flow rates. The pressure drop across the GDL sample is measured to obtain the permeability of the material. The dry air is obtained from bottles of zero grade air which has less than 2 ppm (parts per million) of water vapor present. The flow rate and relative humidity are first measured before entering the humidifier, and again at the outlet. This assures that there are no leakages and that there is no condensation in the test section. Flow rates are measured with flow meters with flow range of 0 – 2 SLPM (standard liters per minute) and accurate to ±10 sccm (standard cubic centimeter per minute).

Figure 1 shows a schematic of the test section being used for this study. The test section has been designed to measure the permeability in the in-plane directions. The design has one inlet and one outlet channel, each 1 mm x 1.5 mm in cross section, with a length of 100 mm each adjacent to each other. The GDL sample is held between the two blocks providing a gas pathway through their cross-section. The inlet channel is fed by an 8 mm diameter manifold and an inlet header of 1 mm diameter. The exit header is also 1 mm diameter and the exit manifold is also 8 mm in diameter. Figure 2 is a photograph of the test section shown with thermocouples, pressure sensors and humidity sensors. The coolant channels are used to circulate water at set temperatures to maintain a constant
temperature in the test section. Thermocouples T2 to T6 are used to measure the temperature of the test section at different locations. Measurement readings are taken only after the temperature of the test section has reached a steady state. Thermocouple T4 reads the gas temperature of the incoming stream entering the GDL and is used to ensure that the inlet gas is at the same set temperature.

![Schematic representation of the test section](image1)

**Figure 1:** Schematic representation of the test section

![Photograph of test section in operation](image2)

**Figure 2:** Photograph of test section in operation
The flow within the channel is kept in the low Reynolds number range to maintain a laminar flow. The channel has a hydraulic diameter \( (D_h) \) of 1.2 mm. The flow rates used in the tests are 300 sccm, 400 sccm and 500 sccm. These correspond to the flow velocities \( (V) \) of 1.67 m/s to 2.78 m/s. These flow velocities represent the mid-range of flow velocities utilized in operating PEMFCs. These values are used with the viscosity \( (\mu) \) and density \( (\rho) \) of the humidified air in calculating the Reynolds number \( (Re=\rho V D_h/\mu) \). These numbers are given in table 1 for different test conditions employed. The tested Reynolds numbers are in the range of 90 to 160. In the case of minichannels or microchannels, any flow with a Reynolds number less than 1000 can be said to be in the laminar region (10).

### TABLE 1. Calculation of Reynolds number for different flow velocities

<table>
<thead>
<tr>
<th>Flow Rate (sccm)</th>
<th>Flow Velocity (m/s)</th>
<th>Reynolds Number</th>
</tr>
</thead>
<tbody>
<tr>
<td>300</td>
<td>1.67</td>
<td>93.65</td>
</tr>
<tr>
<td>400</td>
<td>2.22</td>
<td>128.46</td>
</tr>
<tr>
<td>500</td>
<td>2.78</td>
<td>160.58</td>
</tr>
</tbody>
</table>

**Data Reduction**

Data is captured by the DAQ, which comprises of a SCXI 1000 chassis connected through a PCI 6221 card. Temperature is read through the SCXI 1102 modules with SCXI 1303 isothermal terminal. Eight E-type thermocouples are used for the measurement of temperature. Two are used at the inlet and outlet sections in the humidity chamber. Four thermocouples are used to obtain the temperature of the test section. Two thermocouples are placed at the inlet to the channel. The inlet gas is maintained at the desired temperature using a PID controller. A second SCXI 1102 module is used in conjunction with an SCXI 1300 voltage accessory for the acquisition of pressure data from the pressure sensors. Data from the humidity sensors are recorded using the SCXI 1302 feed through the terminal connected to the PCI card directly via the SCXI 1349 Connector Assembly.

The data is recorded in .lvm files which are standard output files from LabVIEW®. They are easily accessible with any standard spreadsheet software. Viscosity of the gases is a function of the temperature; therefore a curve fit is obtained to calculate viscosities at the different temperatures.

Darcy’s Law is commonly used to obtain the values of the permeability \( (K) \) for porous media. The same is used for the present data reduction as well. The mathematical formulation is given as

\[
\frac{Q}{A} = \frac{K \Delta p}{\mu L}
\]  

[1]

The volumetric flow rate \( (Q) \) is measured before the gas flows into the test section. The cross sectional area \( (A) \) is calculated from the compressed thickness of the GDL and the length of the channels. Viscosity \( (\mu) \) is calculated as a function of the temperature. Pressure drop \( (\Delta p) \) is measured across the GDL and also along the channel length. It is noticed that the channel pressure drop is insignificant compared to the pressure drop
across the GDL, and therefore may be neglected. The term L signifies the length of the path needed to be traversed by the gases through the porous media. In the current study, this length would be the distance between the two channels.

The uncertainty in the experimentally determined permeability is calculated using the following equation

\[ U = \sqrt{\left(\frac{B}{2}\right)^2 + \left(\frac{\sigma}{N}\right)^2} \]  \[ 2 \]

U is the uncertainty of the system, B is the bias errors and \( \sigma \) is the standard deviation associated with the measurement. N is the total number of individual measurements made.

All the errors associated with each reading are calculated and stored with each data point. These are then shown on the results plot as error bars. The uncertainty in the permeability varies from 7% to 10%.

Validation

Prasad et al. (7) investigated the effect of porosity on the in-plane permeability. For changing the porosity of the samples, they changed the hard stop shims used to control the compressed thickness of the GDLs. The more the GDLs were compressed, the thinner they became and their porosity decreased. In the present work, the PTFE hard stops were changed to accomplish different levels of compression. The PTFE comes in standard thicknesses of 4 mil (102μm), 5 mil (127μm) and 7 mil (178μm). These were used to obtain different compressed thicknesses for the GDL, based on the original porosity of 78%. Using the following relation, the changed porosity was estimated for each of the samples, where \( \varepsilon \) is the porosity and \( h \) is the thickness of the sample.

\[ \varepsilon = 1 - \frac{h_0}{h} (1 - \varepsilon_0) \]  \[ 3 \]

**TABLE 2.** Change in Porosity of Toray TGP-H 060 with hard-stops

<table>
<thead>
<tr>
<th>Thickness of PTFE Hard-stop (μm)</th>
<th>Compressed Porosity (%)</th>
<th>Percentage Thickness after Compression (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>102</td>
<td>58.86</td>
<td>53.47</td>
</tr>
<tr>
<td>127</td>
<td>67.09</td>
<td>66.84</td>
</tr>
<tr>
<td>178</td>
<td>76.49</td>
<td>93.58</td>
</tr>
</tbody>
</table>

Figure 3 shows the comparison of the current data with that of Prasad et al. (7). The figure also includes the data of Gostick et al. (8). Although the sample they used (Toray TGP-H 090) is not identical, it is very similar in its porosity, the only difference being the thickness of the sample. The data generated as part of the current work shows good agreement with both the available literature as average error is less than 10% and both data sets follow the same trend very closely.
Results and Discussion

Effect of temperature has been studied in five different samples from different manufacturers. Samples with similar properties have been selected to be able to compare their performance directly. Mitsubishi Rayon Corporation (MRC) has two samples of interest, a plain GDL sample (called baseline sample in this study), and one with 6% PTFE. The other samples come from Toray Inc. and Sigracet Corp. Toray TGP-H 120 series is tested using the samples with no PTFE and 10% PTFE respectively. SGL 10BA was tested from the portfolio of Sigracet Corp. None of the samples have any MPL coating. Table 3 gives a brief summary of the properties of the GDLs tested.

<table>
<thead>
<tr>
<th>Sample Name</th>
<th>Thickness (μm)</th>
<th>PTFE Content (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MRC – Baseline</td>
<td>205</td>
<td>0</td>
</tr>
<tr>
<td>MRC – Baseline with PTFE</td>
<td>205</td>
<td>6</td>
</tr>
<tr>
<td>Toray TGP-H 120%</td>
<td>370</td>
<td>0</td>
</tr>
<tr>
<td>Toray TGP-H 10%</td>
<td>370</td>
<td>10</td>
</tr>
<tr>
<td>SGL 10BA</td>
<td>400</td>
<td>5</td>
</tr>
</tbody>
</table>

Figure 4 presents the plots of the in-plane permeability of 95 percent humid air for the different GDL samples and its variation with temperature. The five GDL samples tested have been presented in the individual plots. As seen below, all the samples show an increasing trend in the permeability with an increase in temperature, irrespective of the
GDL manufacturer. The temperature of the incoming gases and the test section are both set at the temperatures indicated.

Figure 4: Effect of temperature on the in-plane permeability of different GDL samples
Table 4 gives the details of the tested compression and porosity for each of the different samples investigated in this study. This shows that the percentage compression for all the samples is within a 5% range while the compressed porosity varies significantly. This also explains some of the difference in permeability which is noted between the different GDL samples.

**TABLE 4. Change in Porosity and Compression of tested GDL samples**

<table>
<thead>
<tr>
<th>Sample Name</th>
<th>Original Thickness (μm)</th>
<th>PTFE Hard Stop used (inches)</th>
<th>Original Porosity (%)</th>
<th>Compressed Porosity (%)</th>
<th>Percentage Thickness after Compression (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>MRC – Baseline</td>
<td>205</td>
<td>7</td>
<td>86.73</td>
<td>86.73</td>
<td></td>
</tr>
<tr>
<td>MRC – Baseline with PTFE</td>
<td>205</td>
<td>7</td>
<td>86.73</td>
<td>86.73</td>
<td></td>
</tr>
<tr>
<td>Toray TGP-H 120 0%</td>
<td>370</td>
<td>12</td>
<td>78</td>
<td>73.29</td>
<td>82.38</td>
</tr>
<tr>
<td>Toray TGP-H 12010%</td>
<td>370</td>
<td>12</td>
<td>78</td>
<td>73.29</td>
<td>82.38</td>
</tr>
<tr>
<td>SGL 10BA</td>
<td>400</td>
<td>13</td>
<td>88</td>
<td>85.46</td>
<td>82.55</td>
</tr>
</tbody>
</table>

Figure 4(a) shows the increase of in-plane permeability to be up to 58% with increase in temperature from 23°C to 82°C in the case of the plain baseline sample. Figure 4(b) represents the variation observed in the case of baseline with 6% PTFE. It shows an increase of about 20% between 23°C to 82°C. Toray TGP-H 120 without any PTFE content shows an increase of 8% while the same with 10% PTFE impregnation shows an increase of 9%. SGL 10BA has an increase of between 24% for the same temperature range. It is seen that for all the samples tested, permeability increased with temperature.

There are two possible explanations for this increasing trend observed in permeability with temperature. Firstly, the observed phenomenon in the GDL, which is an anisotropic fibrous material, is observed to be similar to that observed for homogeneous porous media. Most studies on permeability agree that pore area and fiber diameters constituting the material of the porous media are the two parameters that directly affect the permeability (11). However, neither pore area nor fiber diameter are affected to any great extent at temperatures of up to 80°C; hence we have to look for other explanations for the current findings.

The other explanation concerns the effect of temperature on the mean free path of the gas molecules. At higher temperatures, the molecules have a longer mean free path and travel more freely, thus introducing lower pressure drops to be observed for the same porosity. This means that the energy of the molecules increases at the higher temperatures and thus it is easier for them to pass through the porous material (12). Although the increased viscosity of gases at higher temperatures is expected to account for these changes, the effect introduced due to small pore sizes is perhaps not accounted completely in the case of porous media with microscopic pore structures. This aspect needs to be further investigated to provide a more robust explanation for the observed effect of increased permeability with temperature.

**Conclusions**

An ex-situ test setup was developed for the investigation of the in-plane permeability of five GDL samples used in the Proton Exchange Membrane Fuel Cell. The tested GDL
samples include Toray TGP-H 120 with no PTFE and 10% PTFE loading, SGL 10BA and two samples from MRC, one with 6% PTFE and the other with no PTFE content. The current work evaluates the in-plane permeability of the GDL and studies the effect of temperature.

It has been observed that with an increase in temperature, the in-plane permeability increases for all tested samples varying between 8 – 58% for a temperature increase from 23°C to 82°C. This increase is similar to that observed for homogeneous porous media reported in the literature (12).

The current work provides experimental values of permeability of humid air at different temperature for different GDLs used in practical PEMFC systems. This is expected to improve the accuracy in modeling the transport processes within the PEMFC.

Acknowledgments

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References