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Determination of effective water vapor diffusion coefficient in PEMFC gas diffusion layers

Jacob LaManna

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DETERMINATION OF EFFECTIVE WATER VAPOR DIFFUSION COEFFICIENT IN PEMFC GAS DIFFUSION LAYERS

By

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Partial Fulfillment of the
Requirements for the

Masters of Science

In

Mechanical Engineering

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ABSTRACT

Proton exchange membrane fuel cells have emerged as one of the leaders for the replacement of fossil fuel powered internal combustion engines. Water removal from the cell is one of the top concerns regarding fuel cell performance for transportation applications. During lower power output or high temperature operation, water removal in the vapor phase can dominate. The rate of water vapor diffusion through the porous cathode gas diffusion layer (GDL) of the fuel cell is limited by the porosity and tortuosity formed by the solid fiber matrix. In this work an experimental apparatus is designed to measure the rate of water vapor diffusion across the GDL to determine an effective diffusion coefficient. The effects of microporous layer (MPL) coating, GDL thickness, and polytetrafluoroethylene (PTFE) loading on the diffusion coefficient is demonstrated. Commercially available diffusion media are tested and include Mitsubishi Rayon Corp. Grafil U-105 series, SGL Sigracet® 25, 35, and 10 series, and Toray TGP-H-120 series.

Standard corrections, such as the Bruggeman correction, used in fuel cell literature are found to overpredict the effective diffusion coefficient for the GDL. The MPL was found to produce a significant resistance to water vapor diffusion due to its smaller pore diameters, lower porosity, and an increase in tortuosity. The GDL Grafil U-105 A produced a higher effective diffusion coefficient of $0.070 \text{ cm}^2/\text{s}$ compared to the SGL 25BC value of $0.063 \text{ cm}^2/\text{s}$. Confocal scanning laser microscope images indicated that the MPL for the Grafil U-105 A sample is possibly thinner, thus explaining some of the reduction in diffusion resistance. Thickness was found to have no influence on the effective diffusion coefficient for samples without MPL. PTFE causes a rapid decrease in effective diffusion coefficient from $0.095 \text{ cm}^2/\text{s}$ for TGP-H-120 0% PTFE to $0.024 \text{ cm}^2/\text{s}$ for TGP-H-120 40% PTFE. Comparison to other studies from the literature show good agreement with the present work thus validating the dynamic method for use in diffusion coefficient measurements in fuel cell diffusion media.

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NOMENCLATURE

Abbreviations

CL	Catalyst layer
CLSM	Confocal laser scanning microscope
GDL	Gas diffusion layer
ICE	Internal combustion engine
MFC	Mass flow controller
MPL	Microporous layer
PEM	Polymer electrolyte membrane
PTFE	Polytetrafluoroethylene
SLPM	Standard liters per minute

Symbols

A	Area $[m^2]$
C	Concentration $\left[\frac{mol}{m^3}\right]$
c_p	Specific heat $\left[\frac{J}{kg \cdot K}\right]$
d	Differential operator
D_{AB}	Diffusion coefficient $\left[\frac{m^2}{s}\right]$
D_{eff}	Effective diffusion coefficient $\left[\frac{m^2}{s}\right]$
D_H	Hydraulic diameter $[m]$
f	Friction factor

h	Convective coefficient $\left[\frac{W}{m^2} \right]$
h_m	Mass convective coefficient $\left[\frac{m}{s} \right]$
J	Diffusive molar flux $\left[\frac{mol}{s \cdot m^2} \right]$
j_D	Chilton-Colburn j-factor for mass transfer
j_H	Chilton-Colburn j-factor for heat transfer
k	Thermal conductivity $\left[\frac{W}{m \cdot K} \right]$
K_g	Overall mass transfer coefficient $\left[\frac{mols}{m^2 \cdot s} \right]$
K_y	Overall molar transfer coefficient $\left[\frac{m}{s} \right]$
l	Channel length $[m]$
L	Molar flow rate of dry air in channel 1 $\left[\frac{mols}{s} \right]$
Le	Lewis number
n	Constant used in Lewis Relation
Nu	Nusselt number
P	Pressure $[Pa]$
Pr	Prandtl number
Q	Volumetric flow rate or total discharge for Darcy's law $\left[\frac{m^3}{s} \right]$
q	Molar flux of water vapor across GDL $\left[\frac{mols}{s \cdot m^2} \right]$

R	Universal gas constant $\left[\frac{J}{K \cdot mol} \right]$
R_{tdot}	Total water vapor diffusion resistance $\left[\frac{s}{m} \right]$
Re	Reynolds number
Sc	Schmidt number
Sh	Sherwood number
St	Stanton number
t	GDL thickness $[m]$
T	Temperature $[K]$
$\bar{u}$	Average fluid velocity $\left[\frac{m}{s} \right]$
V	Molar flow rate of dry air in channel 2 $\left[\frac{mols}{s} \right]$
w	Channel width $[m]$
X	Ratio of moles of water to moles of air in channel 1 at position z $\left[\frac{mols_{water}}{mols_{air}} \right]$
Y	Ratio of moles of water to moles of air in channel 2 at position z $\left[\frac{mols_{water}}{mols_{air}} \right]$
z	Coordinate direction along channel length

Greek Symbols

α Thermal diffusivity $\left[\frac{m^2}{s} \right]$

Δ Change in a property

ε Porosity

κ Permeability $[m^2]$

μ Viscosity $\left[\frac{kg}{s \cdot m} \right]$

ρ Density $\left[\frac{kg}{m^3} \right]$

τ Tortuosity

ϕ Relative humidity [%]

Subscripts

0 z at channel entrance

1 Channel 1

2 Channel 2

l z at channel exit

y Coordinate direction across the GDL thickness

z Coordinate direction along channel length

1.0 INTRODUCTION

Polymer electrolyte membrane (PEM) fuel cells have emerged as one of the leading candidates to replace the internal combustion engine (ICE) for transportation. PEM fuel cells operate on hydrogen fuel and oxygen in the air to produce electricity through electrochemical reaction. Since water and heat are the only by-products of PEM fuel cell operation, they are seen as highly desirable for their potential to reduce carbon emissions. The smooth transition to fuel cell vehicles requires optimal performance of fuel cells to maintain the operational standards that consumers are accustomed to with current ICE powered vehicles.

PEM fuel cells operate by reacting hydrogen and oxygen in the presence of a platinum catalyst. As shown in Figure 1.1, seven layers make up the cross-section of one PEM fuel cell: anode bipolar plate, anode gas diffusion layer (GDL), anode catalyst layer (CL), PEM, cathode CL, cathode GDL, and cathode bipolar plate.

Figure 1.1: Schematic representation of a PEM fuel cell.

Hydrogen is delivered on the anode side and air on the cathode side through flow fields in each side's bipolar plate. From the flow fields, hydrogen and oxygen diffuse through the GDL to the CL on their respective sides. On the anode side, the catalyst splits the hydrogen molecules into hydrogen protons and electrons. The protons are transported through the PEM while the electrons must go through an external circuit due to the high electrical resistivity of the membrane. It is this external circuit that utilizes the electricity from the fuel cell to power the vehicle. After passing through the membrane, the hydrogen protons combine with oxygen and the electrons from the external circuit at the cathode CL to form water and heat.

Water must be maintained within certain levels to ensure proper fuel cell performance. The lower limit is defined as the minimum amount of water to maintain membrane hydration. Proper hydration of the membrane is critical as proton conductivity only occurs when the membrane is fully saturated with water. On the upper limit, too much water can saturate the cathode CL and GDL and reduce the pores available for reactant gas transport. This phenomenon of performance degradation caused by excess water within the cathode CL and GDL is referred to as flooding [1]. To prevent flooding, several treatments can be implemented to the GDL such as changing the thickness, adding a microporous layer (MPL), and/or altering the polytetrafluoroethylene (PTFE) treatment. Reduction of the GDL thickness reduces the water content that can be held by the GDL which improves shutdown purge times, while using a thicker GDL can increase temperatures at the catalyst layer resulting in less liquid water. MPL coatings are nano-scale carbon black powder and PTFE binder mixtures that are typically applied to the GDL/CL interface. The average pore diameter of the MPL is in the sub-micron range which limits the transfer of liquid water directly to the GDL. To increase the ability of the GDL to reject liquid water, the GDL can be treated with a PTFE coating that can range from 5-60% PTFE by weight. The PTFE increases the hydrophobicity of the GDL so that it will reject water more readily.

Water is transported through the porous GDL in either liquid or vapor form. Vapor phase transport is important to fuel cell operation as it does not inhibit reactant phase transport and it is thought to aid heat transfer through the heat pipe effect [2]. The heat pipe effect moves heat by

evaporating a working fluid at the hot end, driving vapor down the concentration gradient to the cool end where it then condenses. Reactant gas and water vapor diffusion through the GDL is limited due to the structure of the GDL. The GDL is typically a carbon fiber non-woven paper ranging in thickness from 200 μm to 400 μm . The carbon fibers are typically 8-10 μm in diameter and are randomly oriented with the length of the fiber oriented perpendicular to the through-plane direction. Pores are formed by the matrix of carbon fibers with average diameters of 30-40 μm as seen in Figure 1.2.

Figure 1.2: Through-plane direction image of GDL taken with a confocal laser scanning microscope (CLSM). Shown are the orientation of the fibers, binder and PTFE webbing, and the pores formed by the fiber matrix [3].

The pore structure that is formed by the GDL solid matrix gives the GDL a porosity that can range from 50 to 90%. This structure reduces diffusion area and increases the tortuosity of the diffusion path thus reducing the diffusion coefficient. The adjusted diffusion coefficient that accounts for the structure of the diffusion media is referred to as the effective diffusion coefficient. There are several correlations available to determine the effective diffusion coefficient for porous material, such as the Bruggeman correction [4]. This correction reduces

the diffusion coefficient based on a relationship to porosity. The Bruggeman correction is based on a packed bed of spherical beads and has not been proven for the fibrous structure of the GDL.

Several techniques have been developed in the textiles industry to determine effective diffusion coefficients for fibrous materials. Static methods include ASTM E 96 [5] upright and inverted cup methods and JIS L 1099 [6] desiccant inverted cup. These methods do not utilize any convective transfer enhancements at the surface of the test material. Dynamic methods include ASTM F 2298 [7] dynamic moisture permeation cell (DMPC) and ISO 11092 [8] sweating hot plate techniques. The DMPC method uses flowing gas streams on either side of the test specimen which removes issues with stagnant boundary layers. The sweating hot plate method combines concentration gradients with thermal gradients to simulate the interactions of the human body with outerwear. Although these methods can determine effective diffusion coefficients for materials similar to GDL, they do not reproduce typical fuel cell conditions, i.e. compression amounts, channel dimensions, and flow rates.

The objective of this work is to develop a test method for measuring the effective water vapor diffusion coefficients of the GDL. Samples tested will reflect typically used diffusion media from the automotive industry, i.e. standard thicknesses, PTFE coatings, and MPL coatings. The effects of MPL coating, thickness, and PTFE content on the effective diffusion coefficient will be elucidated. Diffusion coefficients collected experimentally with actual fuel cell diffusion media is critical to fuel cell modeling. Current models must rely on diffusion corrections such as the Bruggeman correction that have been determined to be inaccurate for diffusion media constructed of long, slender cylindrical rods [9] which is similar to GDL. Higher accuracy diffusion coefficients will allow for greatly improved model predictions that can give better insight into the operation of PEM fuel cells. These insights could lead to improved water management within the cell based on optimization of material sets, flow paths, and geometries.

2.0 LITERATURE REVIEW

2.1 Diffusion Correlations

Diffusion through porous media is a large field of research with applications in the petroleum industry, filtration, and textiles among others. The diffusion rate through porous materials can vary based on the pore structure, diffusion path length, and pore size, therefore diffusion rates differ from material to material. Diffusion through porous material is slower as the gases must diffuse in three-dimensions instead of one, resulting in a longer diffusion path. To account for the reduction in observed diffusion coefficients through porous media, an effective diffusion coefficient is utilized. This parameter accounts for the effects of porosity and tortuosity on diffusion rates.

The diffusion coefficient is specific to each gas pair and defines the ability of gas A to diffuse through gas B . Standard diffusion coefficients are defined for free stream diffusion where it is just the gases diffusing with no obstructions in the diffusion path. Diffusion is governed by Fick's Law of diffusion given in Equation (1.1).

$$J = -D_{AB} \frac{\partial C}{\partial x} \quad (1.1)$$

Where J is the diffusive molar flux given in $\text{mol}/\text{s} \cdot \text{m}^2$, D_{AB} is the diffusion coefficient in m^2/s or cm^2/s , and $\frac{\partial C}{\partial x}$ is the concentration gradient in the diffusion direction.

One of the most prevalent corrections in porous media and fuel cell research for determining effective diffusion coefficients is the Bruggeman correction [4] as shown in Equation (1.2).

$$D_{eff} = D_{AB} \epsilon^{1.5} \quad (1.2)$$

Where D_{eff} is the corrected effective diffusion coefficient with units of m^2/s , D_{AB} is the binary diffusion coefficient of gas A in gas B with units of m^2/s , and ϵ is the porosity of the material.

This correction was experimentally determined for the electrical conductivity of packed beds of spheres and electrolyte but due to the analogy between Ohm's Law and Fick's Law it can be extended to diffusion. It is assumed in this relationship that tortuosity is related to porosity by the power of 1.5.

Due to this limiting relation of tortuosity and porosity in the Bruggeman correction, other corrections have been developed such as the one discussed by Cussler [10] in Equation (1.3).

$$D_{eff} = D_{AB} \frac{\varepsilon}{\tau} \quad (1.3)$$

This correction is an improvement over the Bruggeman correction as it allows for values of tortuosity, τ , other than 1.5. Equations (1.2) and (1.3) are commonly used in fuel cell modeling but have not been verified for accuracy with respect to fuel cell diffusion media.

De La Rue and Tobias [9] experimentally investigated the validity of the Maxwell and Bruggeman approximations. An apparatus was developed to test the conductivity of dispersions of glass beads and electrolyte. Two chambers were utilized to compare measurements, one chamber of pure electrolyte and one with the precisely measured quantity of solids. Glass beads were tested over a range of diameters from 49-77 μm averages to 6100-6400 μm averages. Tests were also conducted with mixtures of glass beads and to test the validity of the corrections which were based on spherical materials, 3 mm polystyrene rods were also tested. The authors found that for narrow ranges of bead diameters the Maxwell correction predicted the conductivity of the solution the best. When there is a large distribution of bead diameters the Bruggeman correction is best. As for the polystyrene rods, which is most applicable to fuel cell diffusion media, the Bruggeman correction was found to not be applicable, especially when the rod length is much greater than the rod diameter.

2.2 Diffusion Coefficient Measurement and Prediction in Fuel Cells

Water vapor diffusion and diffusion in general for fuel cell diffusion media has not been extensively investigated in literature. It is more common to find bulk flow, Darcy's law based

permeability studies such as Ihonen *et al.* [11] and Gostick *et al.* [12]. In these types of studies, experimental equipment is developed to push a determined volumetric flow of dry nitrogen or dry air through the GDL in the through-plane or in-plane direction. Differential pressure is measured across the GDL to determine the permeability of the diffusion media. Several numerical models exist that calculate diffusion corrections for fibrous materials like the GDL such as by Tomadakis and Sotirchos [13], Mezedur *et al.* [14], and Nam and Kaviany [15]. These models calculate the diffusion coefficients based on cylindrical geometries and therefore give better predictions than the spherical based Bruggeman correction. Experimental validation of these models is lacking in the literature.

Baker *et al.* [16] utilized a combination of in situ and ex situ testing to determine oxygen and water vapor diffusion coefficients. Two samples were tested, Toray TGP-H-060 untreated paper without MPL and SGL 25BC with MPL. Limiting current in situ testing was used to determine the oxygen diffusion coefficient and an ex situ setup was designed and used to test for the water vapor diffusion coefficient.

Limiting current testing used by Baker *et al.* for diffusion coefficients is the process of running the cell to the mass transport limiting regime and using an equation based on oxygen consumption and Fick's law to determine the diffusion coefficient. The downside to this type of testing is determining how much of the mass transport resistance originates from the diffusion media and what originates from the electrodes. The pore structure within the electrodes is too small to be governed by Fick's law of diffusion and is instead governed by Knudsen diffusion. In situ testing allows for both samples of GDL to be tested. A 5 cm² fuel cell was used to allow for small pressure drops along the length of the channel. The small pressure drops and the use of high stoichiometries of 12/31 at 2 A/cm² allowed for uniform oxygen concentrations along the length of the channel. Stoichiometries of 12/31 are not seen in typical operating fuel cells. Oxygen diffusion rates were found to give D/D_{eff} values of 4.29.

The authors' ex situ setup for testing water vapor was similar in design to standard cup methods. The apparatus consisted of a water chamber above the test sample with desiccant below the

sample. Fifteen samples of Toray, 30 mm in diameter, were layered in between sintered metal plates that allowed for compression variation. The samples were stacked to lower the mass flux through the samples to allow use of the cup method which are not typically applicable for highly permeable materials. SGL 25BC was not tested because of the presence of the MPL which would not facilitate stacking of layers. Humidity sensors calibrated with a 2 point calibration curve were placed on either side of the diffusion media to determine the concentration gradient across the test samples. Tests were allowed to run for 24 hours before the desiccant was reweighed to determine the amount of water absorbed. Diffusion rates were determined for a range of compressions and were found to give D/D_{eff} of 2.75 for 0% compression and D/D_{eff} of 4.5 at 30% compression.

Quick *et al.* [17] developed an ex situ setup to examine the influences of PTFE loading, carbon black/PTFE loading, homogeneity of PTFE loading, and microporous layer on the water transport coefficient of GDL. All of the diffusion media tested was provided by Freudenberg Fuel Cell Component Technology KG. The experimental setup consisted of a horizontally oriented modified 50 cm² fuel cell with straight channels on the gas side and a single serpentine channel on the water side. Water was fed from a reservoir to the water side where it was wicked into a hydrophilic polyamide material. The water would then fully saturate a complete catalyst coated membrane (CCM). Air was passed on the gas side to remove water that would transport through the diffusion media from the CCM. Water removal quantities were determined by measuring the weight of the reservoir.

A water transport coefficient was developed by Quick and coworkers for this study instead of a diffusion coefficient since water could possibly transport through the GDL in both vapor and liquid phases. MPL coatings were found to reduce the water transport due to the reduction in pore size and increased resistance of the MPL. PTFE impregnation composition was found to have a more significant influence on water transport when compared to PTFE impregnation weight. PTFE impregnation weight was found to decrease water transport with increasing weight percentage due to decreases in porosity as expected. The nano-scale of the carbon black added to

PTFE to alter the composition was found to have a significant effect as the particles significantly reduced the air permeability of the GDL.

Oxygen diffusion coefficients for the GDL was determined by Zamel *et al.* [18] using a Loschmidt diffusion cell setup as described by Astrath *et al.* [19]. A Loschmidt cell consists of two cylindrical chambers attached by a ball valve. Nitrogen and water vapor were filled in the upper chamber and oxygen and water vapor in the lower chamber. The tertiary system was found to be a binary system as the water vapor did not diffuse due to equal concentrations on either side. Temperature and PTFE loadings effects were investigate by varying temperature from 25 to 80°C with a Toray TGP-H-120 0% PTFE GDL and varying the PTFE content of the TGP-H-120 from 0 to 40% weight content at constant temperature. The GDL was placed just above the ball valve which was considered the zero datum of the test apparatus.

The authors found temperature to have a negligible effect on the oxygen diffusion coefficient through the GDL. The diffusion coefficient only increased from 0.05 cm²/s at 25°C to 0.065 cm²/s at 80°C. PTFE treatment was found to linearly decrease the diffusion coefficient with increasing levels of PTFE content. The authors explained this decrease as a result of the reduction of porosity from the addition of more solids in the diffusion media. The results were compared to predictions by Bruggeman (1935), Neale and Nader (1973), Tomadakis and Sotirchos (1993), Nam and Kaviani (2003), and Das *et al.* (2009) and were found to be significantly lower than the predictions.

Stumper *et al.* [20] developed an in situ testing method call MEA Resistance and Electrode Diffusivity (MRED). The tests were conducted with a single Ballard MK 9 fuel cell with a segmented cathode flow field for current distribution measurements. Oxygen diffusion coefficients were determined by flooding the cathode with O₂ and then closing the inlet and outlet valves on the cathode side. A constant current was then pulled with hydrogen flowing on the anode side. Current was allowed to decay to zero thus facilitating the calculation of diffusion rates based on theoretical oxygen consumption. The effective diffusion coefficient for oxygen through the entire cathode electrode was determined to be 6.1×10^{-7} m²/s.

In situ testing using limiting current was also conducted by Beuscher [21] to test for oxygen diffusion coefficients. A 25 cm² fuel cell with triple serpentine channels running low cathode stoichiometry of 1.2 was utilized for the tests. Pressure drop through the cell was closely monitored to determine accurate concentrations through the cell as pressure has a significant effect on concentration. The setup was found to be sensitive enough to detect the addition of a second GDL and change in carrier gas from nitrogen to helium. It was also found that the GDL only accounts for 21% of the mass transport resistance with the remaining coming from the catalyst layer and ionomer.

An electrochemical diffusimetry method for analyzing reduction in diffusion coefficients was developed by Kramer *et al.* [22] and used by Flückiger *et al.* [23] to test GDL. This method takes advantage of the analogy between electrical conductivity and diffusion to determine diffusion coefficients. Resistance measurements are taken for a pure electrolyte sample to form a baseline then the resistance is measured for electrolyte saturated GDL. Fixtures were developed to test both through-plane and in-plane diffusion that allowed for up to 20 MPa compression. Stacks of 4-8 samples were used to test in-plane diffusion and stacks of 10-20 samples were used for through-plane testing. Toray TGP-H-060 0%, 20%, and 40% PTFE content and SGL 24 and 25 series samples were tested. The SGL series diffusion media was found to give a more isotropic diffusion coefficient while the Toray series was found to be anisotropic. Increases in PTFE content was found to decrease the diffusion coefficient due to the decrease in porosity and increase in tortuosity. Due to the manufacturing differences between the Toray series and SGL series, the authors concluded that there is structure dependence for the diffusion coefficient.

2.3 Additional Methods for Measuring Diffusion Coefficients

Water vapor effective diffusion coefficients are tested in several industries ranging from construction materials, polymer science, and textiles. To gain a broader understanding of diffusion coefficient testing, literature was reviewed from the previous mentioned industries. It was found that testing of construction materials represented scales much larger than what will be considered in this research [24] or tested impermeable materials such as concrete [25] and

therefore will not be considered. Polymer testing also typically deals with impermeable membranes [26] and therefore will not be considered as there are no bulk flow considerations with impermeable membranes. The textiles industry will be the focus of relevant literature with testing methods that can be categorized as static, semi-dynamic, and dynamic.

Static methods include standards such as ASTM E 96 [5] upright and inverted cup and JIS L 1099 [6] desiccant inverted cup methods. These methods do not use any active convection and the diffusing gas must overcome the stagnant boundary layers on either side of the test material [26]. Dolhan [27] compared the results obtained with the static Canadian General Specifications Board (CGSB) [28] control dish method and the semi-dynamic DND method developed in the author's lab. The control dish method consists of six cups separated in two groups, one group with a cover fabric and the other without. Each cup within the two groups is filled with different amounts of water to vary the air column beneath the test material. The six cups are placed within a humidity controlled chamber and the water remaining in the cup at the end of the test is used to determine the diffusion resistance of the test material. The DND method utilizes a dry gas stream to actively remove the permeate from the test sample.

To compare the methods the authors determined the test method accuracy and advantages and disadvantages. Accuracy was tested by measuring air gap diffusion resistance in both setups. Air gaps were obtained by gluing fabric onto both sides of varying thickness metal spacer rings. The DND method was found to produce data with significantly less scatter when compared to the control dish method. Large error and the scatter seen in the control dish method was determined to originate from the measurements required in the method and the calculations used to extract the diffusion coefficient. The control dish method was found to be an improvement over the ASTM E 96 method as the E 96 method produces poor results for textiles due to the low resistances to water vapor found in typical textiles. The DND method is an advantage over any of the cup methods as it does not required a highly condition atmosphere for proper diffusion measurements.

A comparison of static and dynamic methods was conducted by McCullough *et al.* [29]. Static methods tested were the ASTM E 96 upright and inverted cup method [5] and the JIS L 1099 [6] desiccant inverted cup method while dynamic methods included ASTM F 2298 [7] dynamic moisture permeation cell and ISO 11092 sweating hot plate methods [8]. The JIS L 1099 method measures the diffusion coefficient by layering the test sample between two PTFE films with a water reservoir on one side and a potassium acetate desiccant on the other. The desiccant is weighed before and after the 15 minute long test. ASTM F 2298 is a test cell where gas channels are placed on both sides of a test specimen. The top channel has 95% RH air flowing through it while the bottom channel has 5% RH air flowing through it to produce the water vapor concentration gradient across the test fabric. ISO 11092 tests the evaporative resistance of a material by heating the water side of the fabric to produce a temperature and concentration gradient across the fabric. Dry air is passed on the top side of the material to remove the diffused moisture for measurement.

To compare the five different methods, McCullough *et al.* tested 26 different fabrics that represented four different categories of breathable fabric types. Each method was used to test each fabric and the results for all 26 samples were averaged for each test. These averaged values were used to compare the methods. Spearman rank order correlation tests were used to determine statistically significant relationships between the test methods.

The authors found that the method with the lowest transfer rates was the upright cup followed by the dynamic moisture permeation cell, inverted cup, and the desiccant cup. The sweating hot plate method measures an opposite parameter and therefore is not directly comparable. The Spearman ranking showed that the dynamic moisture permeation cell was significantly correlated to the upright cup method while the hot plate method was closely inversely correlated with the inverted cup method. The dynamic moisture permeation cell was also shown to obtain results faster than the upright cup method and it allows for alteration of the humidity gradients and humidity ranges.

Semi-dynamic methods typically have one static side and one dynamic side on the test specimen. Chalier *et al.* [30] developed a semi-dynamic system for testing aroma diffusion rates through paper coated with wheat gluten. The system developed by the authors places a small vial with-in a larger bottle with two connection ports. The vial held the aroma and was sealed with the testing material on the upper surface. Pure nitrogen was pumped into one connection of the bottle to provide a sweep gas on the external surface of the test material. The nitrogen and aroma mixture exited the second connection of the bottle where it entered a gas chromatograph for analysis. This type of setup allows for better containment of the gas to be diffused which is important with non-water based test gases.

Gibson *et al.* [31] developed a fully dynamic method call the dynamic moisture permeation cell. This setup is what was developed into the ASTM standard F 2298 Water Vapor Diffusion Resistance and Air Flow Resistance of Clothing Using the Dynamic Moisture Permeation Cell [7]. The method was developed to test the moisture diffusion resistance of hydrophobic fabrics used for protective clothing.

The authors' setup uses dry nitrogen as the carrier gas for the experiments. Bubblers are utilized to saturate the gas streams with controls allowing saturation levels from 0% to 100% relative humidity. The gases enter the test section in a parallel flow format and are given time to develop before diffusion takes place. Channel dimensions and flow rates were designed to maintain velocities of 0.5 m/s or higher to minimize the effects of boundary layer resistances. The test section and bubblers are placed within an insulated and temperature controlled chamber to ensure isothermal operation.

Determination of the diffusion coefficient is found using simple equations that apply to either the top or bottom channels shown in Equation (1.4) taken from ASTM F 2298 [7].

$$R_{dot} = \frac{A(\Delta\phi)}{Q(\delta\phi)} \quad (1.4)$$

Where R_{dot} is the total diffusion resistance, A is the transfer area, $\Delta\phi$ is the relative humidity difference between the top and bottom channels at the section inlet, Q is the volumetric flow rate through one of the channels, and $\delta\phi$ is the relative humidity change in the bottom channel between inlet and outlet.

The authors investigated the results of running the method in a counter-flow arrangement. This flow orientation would give a more constant and uniform concentration gradient along the entire length of the test sample which should induce higher rates of diffusion. They found no significant differences between flow orientations for materials with low water vapor transfer rates. Flow orientation was found to be a good measure of boundary layer resistance.

Gibson *et al.* also investigated the relationship of the dynamic moisture permeation cell with the ISO 11092 [8] sweating hot plate method. A group of materials was tested with both methods and the results were compared. It was found that the dynamic moisture permeation cell correlates very well with the sweating hot plate method.

Huang [32] describes design and use of the ISO 11092 [8] sweating hot plate method. This method utilizes a porous center heater and an outer guard heater to reduce heat losses. The porous heater is saturated with liquid water and attached to a reservoir to maintain saturation. Heat is applied to evaporate the water and produce the concentration gradient needed to drive water vapor through the test material that is placed on top of the heater. Air is then blown across the top of the test material to remove the moisture that has diffused through. Vapor diffusion resistance is calculated based on the assumed full saturation at the heater, 40% relative humidity of the sweep gas, and the water consumption rate from the reservoir.

The author found that there are several difficulties with this method in obtaining consistent results. Airflow direction is critical as vertical airflow will penetrate the membrane and increase heat and mass transfer. Water feed to the heater can be misleading as too much water causes heater flooding and sample floating while too little will cause dry out and under-prediction of the

diffusion resistance. Also, bubbles or wrinkles existing in the test material interface with the heater will cause overestimation of resistance as the air pockets act as insulators.

Additional methods include permeate side vacuum cells [33] which are similar in design to the dynamic moisture permeation cell but instead of supply gas to the permeate side a vacuum is applied to remove the gas for analysis. Also, photoacoustic methods have been developed by Tomas *et al.* [34] to detect water vapor permeability in ceramics.

2.4 Objectives

After investigating the current literature, several issues with current methods of testing diffusion coefficients for fuel cells were identified. The validity of commonly used corrections, both general and numerically developed for fuel cells, has not been extensively investigated through experimentation for the wide range of common diffusion media. This validation is critical as research indicates [9] that general corrections are not accurate for fibrous materials. Key parameters from general literature for diffusion coefficient test section design have been identified as differential pressure across test sample, boundary layers on membrane gas interfaces, and feed and permeate concentration levels.

It has been identified that experimental data for the water vapor diffusion coefficient through the GDL based on a wide range of diffusion media is lacking. This work will address this gap in the literature by developing a system capable of measuring the water vapor diffusion coefficient quickly and efficiently while utilizing current fuel cell geometries. Data produced will allow for extrapolation throughout the typical operation range for automotive PEM fuel cells. Diffusion media tested will represent state-of-the-art technology currently used in fuel cells.

2.5 Approach

An ex situ setup will be developed based on proven dynamic testing methods from the textile industry and will use standard fuel cell channel dimensions and flow rates. Fuel cell geometry is

chosen so as to impose the same boundary conditions on the GDL/channel interface as seen in a fuel cell. The setup will allow for compression of the GDL to typical compression ranges. Testing will be conducted on one GDL sample at a time and will not be averaged over a stack to avoid possible interfacial influences.

The test setup will have one single, straight gas channel on either side of the GDL to reduce convective flow through the GDL. Convective flow through the GDL has been identified as a concern for obtaining accurate values of D_{eff} as the flow will turn the passive diffusion process into an active convective mass transfer process. It is common to find the use of serpentine gas channels in the literature, but these flow fields allow convective flow through the GDL flowing across the channels because of the pressure drop along the length of the channel. Similarly, multi-channel test sections could also induce convective flow through the GDL between channels due to differences in pressure drop caused by variances in GDL intrusion or machining variability.

An ex situ setup will be used instead of an in situ fuel cell to reduce the many unknowns associated with actual fuel cells. In the use of limiting current to determine diffusion coefficients it is necessary to uncouple the diffusion resistance of the GDL from the diffusion resistance of the catalyst layer. This can be a complex task as the catalyst layer pore structure and size is greatly different than the GDL. Time is also an issue with in situ testing as assembling the cell can be more complex than a ex situ setup and there is the requirement to condition the cell after every assembly and any length after shutdown.

3.0 EXPERIMENTAL SETUP

3.1 System Introduction

An experimental setup is developed to measure the effective water vapor diffusion coefficient for GDL. This setup is designed to allow for variation of humidity levels, flow rates, and process temperatures of both gas streams independently. The setup is divided into four subsystems consisting of the test section, air delivery system, humidifier, and chilled water loop. Basic schematic of system layout with representation of channel water vapor concentrations is shown below in Figure 3.1.

Figure 3.1: Schematic of test section where 1) bottled ultra zero grade air, 2) mass flow controllers, 3) humidifier, 4) sensors and test section, 5) exit flow meters, 6) backpressure control valves, 7) cross-section of test section

3.2 Test Section

The test section consists of modified hardware previously developed under a General Motors sponsored project and the Master's thesis of John Borrelli [35]. Two independent flow streams are directed through two 1 mm square channels with the GDL sample as the interfacial wall between the channels. This configuration forms a parallel mass exchanger as seen in Figure 3.2.

Concentrations of water vapor are represented by the dashed lines in Figure 3.2 where the lines will approach each other like the fluid temperatures in a parallel flow heat exchanger.

Figure 3.2: Representation of test section as parallel flow mass exchanger.

3.2.1 Channels

To form the flow fields of the mass exchanger, a single 254 mm, 1 mm square channel is machined into a polycarbonate block. Channel dimensions are chosen based on common fuel cell flow field dimensions [36]. Dimensions are at the upper limit of common geometries to reduce the pressure drop along the length of the channel to prevent bulk flow of air through the GDL. Polycarbonate was chosen for its low thermal conductivity so as to thermally isolate the channels from the metal compression hardware, ability to add flow visualization if necessary, and its machinability. Flat bottom wells with 1/8 inch NPT pipe taps provide the inlet and outlet headers on the backside of each channel block. Air enters and exits each channel through a 1 mm diameter circular transition header oriented at 45°. The header transitions the flow to the channels from the pipe fittings on the back side of the polycarbonate blocks. Design drawings for the channels can be found in Appendix A. Figure 3.3 displays the pipe fitting transition to the channels.

Figure 3.3: Pipe fittings on back side of polycarbonate block transitions to the square channels through 1 mm diameter circular ducts oriented at 45°.

3.2.2 Compression hardware

To ensure proper sealing and to compress the GDL to simulate actual fuel cell conditions, the channels are housed within compression hardware. This hardware allows uniform compression along the channel length with compression values up to 1000 kPa. Exact compression is achieved by compressing the test section to the desired compression rate in a tensile/compression tester and then bolting the top plate into position. The test section was developed under a previous General Motors sponsored project and modifications were made to allow inlet and outlet fittings to be attached to the channel pieces. The modifications consist of the U-shaped cuts at the ends of the top, middle, and bottom plates that allow the tee-fitting for temperature measurement and flow entry to attach to the channel pieces. Figure 3.4 identifies the components in the hardware, missing from view are the side plates that connect the upper and lower end blocks. All plates are made from stainless steel and the springs ensure even compression.

Figure 3.4: Exploded view of compression hardware: 1) Top plate, 2) Polycarbonate channel blocks, 3) O-rings, 4) PTFE spacer, 5) GDL sample, 6) Compression plate, 7) Compression springs, and 8) Bottom plate.

3.2.3 Humidity measurements

To measure the water vapor content of the air streams, relative humidity sensors are used. The relative humidity sensors selected for the test setup are Honeywell HIH-4602-L-CP sensors. These sensors operate on the change in capacitance the water vapor in air causes on the sensor

membrane. Factory 2-point calibrations are packaged with the sensors and give factory rated accuracy of $\pm 2\%RH$. The sensors require a 5V excitation and output 0 to 5V.

Humidity measurements are taken at the inlets and outlets of the two streams. The measurement locations are placed as close to the inlet and outlet sections as possible. Although the sensors are not integrated within the channels, relative humidity is assumed constant as long as dew point is maintained below testing temperatures so as condensation does not occur. Thermocouples are placed near the humidity sensor to allow for temperature correction of the relative humidity measurements. This is necessary due to the strong temperature dependence of relative humidity.

Plastic chambers are used to adapt the humidity sensors meant for PCB mounting to the air flow tubing. The chambers are constructed from polypropylene for its thermal and electrical insulating properties. Inlets to the chambers are placed above the outlets to allow drainage of any condensed water so as to protect the humidity sensor from becoming saturated by liquid water. Caps on the chamber hold and seal the sensor in place as shown in Figure 3.5.

Figure 3.5: Cross-sectional and exploded images of humidity measurement chambers:
 1) chamber, 2) sensor pocket plate, 3) humidity sensor, and 4) sensor sealing cap plate

3.2.4 Temperature measurements

E-type thermocouples were chosen because of their high voltage change per degree change. Eight temperature measurements are taken on the test section: one thermocouple for each humidity sensor for temperature correction of humidity levels, one thermocouple for each inlet, and one thermocouple for each outlet. The inlet and outlet thermocouples were placed in the transition header as close to the channel as possible. Figure 3.6 shows the position of the humidity sensor thermocouples with respect to the humidity sensors and Figure 3.7 shows the placement of the inlet and outlet thermocouples with respect to the channels.

Figure 3.6: Position of thermocouples for temperature correction of humidity measurements

Figure 3.7: Thermocouple position in inlet and outlet headers with respect to channels

3.2.5 Differential pressure measurements

To mitigate bulk flow, one differential pressure sensor measures the differential pressure between the two channels 1 cm after the channel entrance and a second sensor measures differential pressure between the channels 1 cm before the channel exit. Two Honeywell 176PC14HD2 low pressure silicon differential pressure sensors are utilized for their ± 1740 Pa range. Bulk flow sensitivity on diffusion coefficients is discussed later in section 6.3. The flow lengths and cross-sectional dimensions are identical for the two channels between the differential pressure sensors which results in identical pressure drops along the flow length so if both differential pressures are minimized it is assumed there are no further deviations in the channels. Differential pressure is minimized with a combination of methods. To minimize the entrance differential pressure, flow rates are adjusted at the mass flow controllers located upstream. Exit pressures are minimize with needle valves placed on the exits of the channels. These valves allow adjustment of system backpressures. Placement of the mass flow controllers and needle valves are shown in detail in Figure 3.8.

3.2.6 Test section sealing

The test section is sealed with the use of o-ring gaskets. Both polycarbonate channel blocks have o-ring grooves machined around the parameters of the blocks. The o-rings are designed to compress flush with the polycarbonate blocks so that the o-rings do not carry compression load or alter the compressed thickness of the GDL. To ensure the proper compressed thickness of the GDL, PTFE membrane spacers are used. These PTFE membranes are hard stops that only compress 20% at the tested compression loads.

3.3 Air Delivery System

The air delivery system provides the air that forms the water vapor concentration gradient across the GDL. This system regulates pressure and flow rates of ultra zero grade air provided in gas cylinders. Ultra zero grade air is utilized for its water content of less than 2 ppm and extremely low levels of particulates, trace gases, and other contamination. Figure 3.8 outlines a schematic representation of the air delivery system.

Figure 3.8: Diagram of air system layout showing typical and optional flow patterns. Key: Reg – Regulator, FCV – Flow Control Valve, TI – Temperature Indication, MI – Humidity Indication, PI – Differential Pressure Indication, FI – Flow Rate Indication.

Flow rate is controlled by regulators attached to a manifold on the gas cylinder and mass flow controllers before the inlet to the humidifier. Two Unit UFC-1100 mass flow controllers (MFC) are utilized in conjunction with a Brooks Instrument 0154E control box. The regulators on the gas cylinder provide 70 to 100 kPa of pressure to the MFC's. Flow rate is verified at the exit of the test section with two Omega FMA-1816 electronic flow meters. All flow controllers and meters have a measurement range of 0 to 2 slpm. This range is chosen to reflect common fuel cell gas channel velocities. Exit flow rate verification is required to determine leakage and as a secondary method to ensure minimized bulk flow. At 25°C and the flow rates investigated, the water vapor in fully saturated air has less than a 0.9% change in flow rate and therefore if significant bulk flow exists, it would be detectable through discrepancies between the two exit flow meters.

3.4 Humidifier

Water vapor content of the gas streams is controlled with an Arbin DPHS-D50 dew point fuel cell humidifier. The humidifier is capable of fully saturating two independent gas streams with flow rates up to 50 slpm. Humidity level is controlled by setting the dew point temperature of the humidifier. Arbin rates the accuracy of humidity to $\pm 1.0^{\circ}\text{C}$ with respect to the dew point temperature. Deionized water is supplied to the humidifier's boilers from an external tank to

provide the water vapor for the gas streams. The air streams are saturated in the humidifier by passing through the system's boilers. In the boilers, the air must pass through the temperature controlled deionized water therefore picking up moisture until saturation at the temperature of the water.

3.5 Chilled Water System

Due to oscillations in temperature in the humidifier boilers caused by the built in temperature controllers, chilled water must be used to control the temperature of the boilers. Temperature control of the boilers utilizing this method provides a very stable and constant temperature which is important to maintain the same humidifier outlet relative humidity being supplied to the test section. Water is supplied from a connection with the lab domestic water supply in an open loop configuration. The system has to provide a minimum of 125 sccm of water to each boiler during operation. A Lauda RE120 chiller is added inline to control the inlet water temperature to between 21 and 23°C (dependant on operational temperature). The chiller is capable of removing up to 0.43 kW from its internal water bath and can control temperature to an accuracy of $\pm 0.2^{\circ}\text{C}$. To prevent contamination of the distilled water used in the chiller, an Alfa Laval flat plate heat exchanger is placed into the loop. A schematic of the chilled water system is given in Figure 3.9.

Figure 3.9: Diagram of chilled water system.

3.6 Data Acquisition

A National Instruments cDAQ-9172 compact USB DAQ chassis is used for all sensor measurements for the experimental setup. Eight thermocouple measurements are taken with two NI 9211 cards, the humidity measurements are recorded with a NI 9205 card, and the pressure sensors are recorded with a NI 9206 card. Data collection uses a setup specific LabVIEW GUI. Data is collected at 1000 Hz and data points are averaged for a user defined period of time to reduce noise before being outputted to the display screen or written to file. The LabVIEW program is capable of outputting Microsoft Excel spreadsheets that includes all temperature, relative humidity, and pressure readings for user specified durations.

4.0 ANALYTICAL MODEL

An analytical model is required to determine the effective diffusion coefficients from the collected experimental data since it is not possible to measure the diffusion coefficient directly. The model is capable of taking the averaged inlet and outlet relative humidities and temperatures and determines the effective diffusion coefficient. This section goes through the derivation of the analytical model.

4.1 Model Assumptions

1. Steady state
2. Isothermal
3. No chemical reactions
4. Binary system of water vapor and air
5. Ideal gas
6. No advection/bulk flow through GDL
7. Laminar flow
8. Diffusion is not considered along the length of the channel
9. Diffusion only occurs in the GDL between the channels
10. No condensation occurs

4.2 Validation of Key Assumptions

4.2.1 Validation of Assumption 6

Advection is neglected due to the lack of differential pressure across the GDL to drive bulk flow. The basis for this is governed by Darcy's law shown in Equation (4.1).

$$Q = \frac{-\kappa A}{\mu} \nabla P \quad 4.1$$

Where Q is the total discharge in m^3/s , κ is the permeability of the material, μ is the viscosity of the fluid, and ∇P is the pressure gradient across the porous material. As shown in Equation (4.1) bulk fluid flow does not occur without a driving pressure gradient.

4.2.2 Validation of Assumption 7

Flow rates tested result in Reynolds numbers in the range of 500 to 800 as shown in Table 4.1, well within the laminar regime.

Table 4.1: Calculated Reynolds numbers for tested flow rates.

Q [sccm]	Re
500	524.3
625	655.4
750	786.4

4.2.3 Validation of Assumption 8

The influence of diffusion down the length of the channel was determined by calculating the diffusion down the channel and comparing it to the diffusion across the GDL. Transfer rates were determined using Fick's Law given in Equation (2.1). The concentration gradient for the down-the-channel case was determined with the inlet and outlet relative humidities. Relative humidities for the two channels were averaged to determine the concentration gradient across the GDL. The worst case was determined to be for the Toray TGP-H-120 40% 500 sccm test case. For this case the diffusion down the length of the channel was found to represent 0.35% of the diffusion through the GDL thus showing the influences of diffusion down the length of the channel to be negligible.

4.2.4 Validation of Assumption 9

Diffusion over the land areas is neglected due to the higher diffusion resistances in these areas and the lack of a driving concentration gradient.

4.3.5 Validation of Assumption 10

Temperatures are maintained above the saturation point of water vapor thus reducing the likelihood of condensation. To verify that water vapor does not condense within the test section, a mass balance is performed. A plastic sheet is placed in the test section in lieu of a GDL sample and the relative humidities are measured. Water vapor balance is validated when the inlet and

outlet relative humidities for each channel match to within error. During standard testing, inlet and outlet relative humidities are summed together to ensure mass balance.

4.3 Mass Transfer Model

An analytical model is developed to convert the data obtained from experimentation to the desired diffusion coefficient. To model the diffusion across the GDL, a differential element is taken from the cross-section of the channel/GDL system as shown in Figure 4.1.

Figure 4.1: Governing differential element and control volume.

The in-flows and out-flows for the differential element are identified and summed together in Equation (4.2).

$$LX|_z - LX|_{z+\Delta z} - q\Delta zw = 0 \quad (4.2)$$

Where L is the molar flow rate of dry air in channel 1, X is the ratio of moles of water to moles of air in channel 1, q is the molar flux of water vapor across the GDL, and w is the channel width. Equation (4.2) can then be written in differential form, shown in Equation (4.3).

$$L \frac{dX}{dz} = -qw \quad (4.3)$$

Equation (4.5) is found by substituting the definition of q , Equation (4.4), into Equation (4.3) where K_y is the overall molar transfer coefficient and Y is the ratio of moles of water to moles of air in channel 2.

$$q = K_y \left(\frac{X}{1+X} - \frac{Y}{1+Y} \right) \quad (4.4)$$

$$L \frac{dX}{dz} = -K_y w \left(\frac{X}{1+X} - \frac{Y}{1+Y} \right) \quad (4.5)$$

As there are two unknowns, X and Y , it is necessary to develop a second governing equation to make this a well posed problem. A control volume is drawn around the channel/GDL system and conservation of mass is applied yielding Equation (4.6).

$$LX_0 + VY_0 = LX + VY \quad (4.6)$$

Where X_0 and Y_0 are the initial mole ratios for channel 1 and 2 respectively, L and V are the molar flow rates of dry air for channel 1 and 2 respectively, and X and Y are the mole ratios at any position z . It is now possible to solve Equation (4.6) for Y and substitute this into Equation (4.5). Boundary conditions of X_0 , Y_0 , and X_l are used to govern the system where X_l is the exit mole ratio for channel 1. Use of X_l as a boundary condition arises from solving the differential equation in terms of X and with re-derivation Y_l , the exit mole ratio for channel 2, can be used as boundary condition. Values for the boundary conditions are obtained from the experimentally collected data. Using U-substitution, it is possible to integrate the differential equation and solve it resulting in Equation (4.7).

$$\frac{L}{2wl} \left\{ \left[\left(1 + \frac{X_0 + Y_0}{2} \right)^2 \ln(X_0 - Y_0) - \frac{1}{8}(X_0 - Y_0)^2 \right] - \left[\left(1 + \frac{X_0 + Y_0}{2} \right)^2 \ln(2X_l - X_0 - Y_0) - \frac{1}{8}(2X_l - X_0 - Y_0)^2 \right] \right\} = K_y \quad (4.7)$$

A full step-by-step solution of the differential equation can be found in Appendix B. The effective diffusion coefficient can be found using the definition of the overall molar transfer coefficient in Equation (4.8).

$$K_y = \frac{1}{\frac{1}{h_{m,1}} + \frac{t}{D_{eff}} + \frac{1}{h_{m,2}}} \quad (4.8)$$

K_y has units of $\text{mol}/\text{m}^2\text{s}$ and must be converted to m/s to extract the diffusion coefficient with units of m^2/s . Equation (4.9) converts K_y to K_g which has units of m/s .

$$K_g = K_y \frac{RT}{P} \quad (4.9)$$

Where R is the universal gas constant, T is system temperature, and P is the system pressure. Now, rearranging Equation (4.8) with the definition of Equation (4.9) it is possible to solve for the effective diffusion coefficient in Equation (4.10).

$$D_{eff} = \frac{t}{\frac{1}{K_g} - \frac{1}{h_{m,1}} - \frac{1}{h_{m,2}}} \quad (4.10)$$

Where $h_{m,1}$ and $h_{m,2}$ are the mass convection coefficients for the two channels, t is the thickness of the GDL, and D_{eff} is the effective diffusion coefficient. The above equations are solved in Microsoft Excel.

4.4 Lewis Relation and Coupling of Heat and Mass Transfer

The Lewis Relation is used to determine the mass convective coefficients based on thermal convective coefficients by comparing the Chilton-Colburn j-factors [37] for heat transfer (Equation (4.11)) and mass transfer (Equation (4.12)).

$$j_H = \frac{Nu}{Re Pr^{(1-n)}} = St Pr^n = \frac{f}{2} \quad (4.11)$$

$$j_D = \frac{Sh}{Re Sc^{(1-n)}} = St_m Sc^n = \frac{f}{2} \quad (4.12)$$

Where Nu is the Nusselt number, Re is the Reynolds number, Pr is the Prandtl number, St is the Stanton number, Sh is the Sherwood number, Sc is the Schmidt number, and St_m is the mass transfer Stanton number. Equation (4.13) is formed when equating Equation (4.11) with Equation (4.12).

$$St Pr^n = St_m Sc^n \quad (4.13)$$

Equation (4.14) is found by inserting the definitions for the non-dimensional numbers in Equation (4.13).

$$\frac{h}{\rho c_p \bar{u}} \left(\frac{c_p \mu}{k} \right)^{2/3} = \frac{h_m}{\bar{u}} \left(\frac{\mu}{\rho D_{eff}} \right)^{2/3} \quad (4.14)$$

Simplifying Equation (4.13) yields the Lewis Relation, Equation (4.15), where Le is defined as α/D or the ratio of thermal diffusivity to mass diffusivity.

$$\frac{h}{h_m} = \rho_c Le^{2/3} \quad (4.15)$$

From Equation (4.15) and utilizing $Le = 0.851$ for zero saturation and $Le = 0.841$ for full saturation from Kusuda [38] it is possible to find the mass convective coefficient. As seen in Equation (4.15), the mass convective coefficient is dependent on an accurate thermal convective coefficient.

4.5 Heat Transfer Properties

To determine an accurate thermal convective coefficient, an accurate Nusselt number is necessary. The Nusselt number must be for square ducts with heat transfer on only one side (see Figure 4.2) and simultaneously developing laminar flow to match the experimental setup.

Figure 4.2: Channel cross-section showing insulation on three wall surfaces and heat transfer on one wall surface.

From Shah and London [39] and Wibulswas [40], the applicable mean Nusselt number was found to be 3.075. This number was determined by multiplying the simultaneously developing Nusselt number of 3.75 by 0.82 to account for the reduction in heat transfer surfaces from four to one. This ratio was determined by using the fully developed Nusselt number tables for varying numbers of transfer surfaces.

The thermal convective coefficient is found using the definition of the convective coefficient in Equation (4.16).

$$h = \frac{kNu}{D_H} \quad (4.16)$$

Where k is the thermal conductivity of the fluid and D_H is the hydraulic diameter.

4.6 Data Processing

Experimental data is used as the inputs to the model. Collected data files from LabVIEW are opened in Excel and the temperatures and relative humidities are averaged for each measurement location over the recorded time. The eight average temperatures are averaged together to determine the system temperature. The volumetric flow rate, average inlet and outlet relative humidities for each channel, average system temperature, atmospheric pressure, and GDL thickness are entered as inputs into designated cells in the Excel workbook as shown in Figure 4.3.

Dry air flow rate	0.5	slpm	} Model Inputs
Inlet RH channel 1	96.15	%	
Outlet RH channel 1	67.54	%	
Inlet RH channel 2	5.54	%	
Outlet RH channel 2	34.15	%	
System temperature	24.89	°C	
Barometric pressure	100950	Pa	
GDL thickness	0.000225	m	
diffusivity	8.18E-06	m ² /s	} Model Output

Figure 4.3: Model input/output display showing example inputs and calculated diffusion coefficient.

The relative humidities must be converted to molar ratios of water vapor to dry air to be valid inputs to the model. Conversion begins with the calculation of the water vapor partial pressure based on the saturation pressure at the system temperature. The water vapor partial pressures are then used to calculate the humidity ratio for each channel as shown in Figure 4.4.

Saturation Pressure	3148.371658 Pa
System Temperature	298.04 K
Mass flow rate of air in channel 1	1.00417E-05 kg/s
Mass flow rate of air in channel 2	1.00417E-05 kg/s
Inlet water vapor partial pressure in channel 1	3027.159349 Pa
Outlet water vapor partial pressure in channel 1	2126.410218 Pa
Inlet water vapor partial pressure in channel 2	174.4197899 Pa
Outlet water vapor partial pressure in channel 2	1075.168921 Pa
Inlet humidity ratio channel 1	0.019228334 kg _w /kg _a
Outlet humidity ratio channel 1	0.013383719 kg _w /kg _a
Inlet humidity ratio channel 2	0.001076542 kg _w /kg _a
Outlet humidity ratio channel 2	0.006695932 kg _w /kg _a

Figure 4.4: Section of spreadsheet tool used to convert the measured relative humidities to humidity ratios based on the system temperature.

Conversions are finalized when the humidity ratios are multiplied by the ratio of molar weight of air to the molar weight of water as shown in Figure 4.5. The last input to be converted for the model is the volumetric flow which is converted to a molar flow rate for dry air.

Initial moles of water to moles of air in channel 1	X_0	0.030916899	mol _w /mol _a
Initial moles of water to moles of air in channel 2	Y_0	0.001730952	mol _w /mol _a
Final moles of water to moles of air in channel 1	X_i	0.021519445	mol _w /mol _a
Air molar flow rate channel 1	L	0.000346671	mol/s
Air molar flow rate channel 2	V	0.000346671	mol/s
Channel width	W	0.001	m
Channel length	I	0.254	m

Final form of
model inputs

Figure 4.5: Final conversions to obtain molar ratios of water to air and molar flow rates.

The numbers obtained in Figure 4.5 are the final form for the inputs to the model and are used to calculate the diffusion coefficient. On the diffusion coefficient calculation page of the workbook the conversions from the conversion sheet are entered into the differential equation. K_y is calculated from the differential equation and then converted to K_g before D_{eff} is calculated. Equation (4.9) is used to calculate D_{eff} and allows for variation of the convective coefficient based on flow conditions as shown in Figure 4.6.

Main Equation					
$\frac{L}{\beta W l} \left\{ \left[\gamma \delta \ln(X_0 \beta - \alpha) + \left(\frac{\delta}{\beta} - \frac{L\gamma}{V\beta} \right) (X_0 \beta - \alpha) - \frac{L}{2V\beta^2} (X_0 \beta - \alpha)^2 \right] - \left[\gamma \delta \ln(X_i \beta - \alpha) + \left(\frac{\delta}{\beta} - \frac{L\gamma}{V\beta} \right) (X_i \beta - \alpha) - \frac{L}{2V\beta^2} (X_i \beta - \alpha)^2 \right] \right\} = K_y$					
Definitions					
$\alpha = \frac{L}{V} X_0 + Y_0$	α	0.032647851	hm1	0.0693 m/s	Convection coefficients
$\beta = 1 + \frac{L}{V}$	β	2	hm2	0.0712 m/s	
$\gamma = 1 + \frac{\alpha}{\beta}$	γ	1.016323926			
$\delta = 1 + \alpha - \frac{L\alpha}{V\beta}$	δ	1.016323926			
	K_y	0.727901256	Calculation and conversion of final results. Diffusion coefficient displayed on front panel.		
	K_g	0.017866962			
	D_{eff}	8.18352E-06			

Figure 4.6: Main calculation spreadsheet of model.

All calculations and conversions are automated within the workbook once the averaged experimental data has been entered. The model calculates the effective diffusion coefficient in units of m^2/s .

5.0 TESTING PROCEDURE

5.1 Test Cases

Test cases are selected to test the effects of MPL coatings, GDL thickness, and PTFE content on the water vapor diffusion coefficient. The three test variables represent the most significant changes between different GDL samples. Each test sample is tested under three flow rates of 0.500, 0.625, and 0.750 slpm at 25°C. The three flow rates are chosen as the best compromise between flow control hardware accuracy and reduction in differential pressure. Three flow rates are chosen to ensure that there are no flow rate influences on diffusion which should be the case for laminar flow.

Twelve GDL samples are utilized to test the three test variables. Mitsubishi Rayon Corporation (MRC) Grafil U-105 series and SGL Sigracet series nonwoven diffusion media are used to test the effects of MPL as both series are available with and without MPL. SGL series will test the effects of thickness as the series is available in three different thicknesses that are manufactured with the same process and coated with the same MPL. Identical manufacturing processes and MPL coatings are necessary as different manufacturing process can result in different microstructures that can change the diffusion properties of the material. Toray paper will test the effects of PTFE content as it is available in untreated, 10, 20, and 40% PTFE content by weight.

Table 5.1 displays the samples to test and indicates whether the sample is coated with MPL, weight percentage of PTFE treatment, thickness, and tested hypothesis. Testing is performed in groups for comparison purposes as shown in Table 5.1. MRC Grafil U-105 with and without MPL is tested first, followed by the SGL series tested in thickness pairs to compare MPL effects first, with the last group being the Toray series.

Table 5.1: GDL sample data and tested hypothesis.

test groups	GDL	MPL coating	Thickness (μm)	PTFE (wt%)	Tested Hypothesis
1	Grafil U-105 A	Y	200	9	MPL
1	Grafil U-105 B	N	208	7	MPL
2	SGL 25BC	Y	225	5	MPL/Thickness
2	SGL 25BA	N	190	5	MPL/Thickness
3	SGL 35BC	Y	325	5	MPL/Thickness
3	SGL 35BA	N	300	5	MPL/Thickness
4	SGL 10BC	Y	420	5	MPL/Thickness
4	SGL 10BA	N	400	5	MPL/Thickness
5	Toray 120 - plain	N	370	0	PTFE
5	Toray 120 - 10%	N	370	10	PTFE
5	Toray 120 - 20%	N	370	20	PTFE
5	Toray 120 - 40%	N	370	40	PTFE

The MRC Grafil U-105 diffusion media is supplied by General Motors Electrochemical Energy Research Laboratory with GM proprietary MPL coating applied. SGL samples with MPL are coated with MPL applied by the manufacturer. All PTFE coatings are applied by the manufacturers.

5.2 Testing Procedure

Testing is started at the lowest flow rate of 0.5 slpm and allowed to run until the humidifier temperatures are stabilized (typically 30 minutes). Once the humidifier temperatures are stabilized and the inlet humidities to the test section are steady, data is recorded. LabVIEW collects all eight temperature measurements, four humidity measurements, and two differential pressure measurements for thirty seconds before writing the collected data to an Excel file.

The flow rates are tested in ascending order and allowed to reach steady-state before data is recorded. Flow rates and needle valves are adjusted around the desired setpoints until differential pressure between the channels is reduced to zero within the precision of the differential pressure sensors. Differential pressure is also monitored by exit flow rate meters, larger differential pressure values will result in differences in exit flow rate quantities. Steady-state is determined by scrolling charts in LabVIEW that plot the relative humidity measurements. Humidity,

temperature, and pressure readings are monitored and steady-state is assumed when these readings remain constant over a two minute period.

5.3 Leak Mitigation

Leak testing is conducted after the test section is disassembled and reassembled for the next round of testing. This testing is necessary as the test section is completely disassembled and the new gasket material may not provide a complete seal. Testing for leaks is conducted by blocking the outlets to the test section with fitting caps and pressurizing the system to approximately 14 kPa which is greater than operational pressure. Gas leak detecting liquid is applied to all fittings and along the polycarbonate mating face to detect leaks. If bubbles are detected on fittings, the fittings are tightened to prevent leakage. If tightening the fitting does not stop leakage, additional Teflon tape is applied to the fitting. In the event of leakage on the polycarbonate blocks, effort is taken to reduce it as much as possible by adding additional compressive force to the setup or reassembly in the event of a severe leak.

6.0 UNCERTAINTY ANALYSIS

6.1 Calibration

Calibrations are performed for all three sensor types: temperature, pressure, and humidity. Calibration methodology is explained here.

6.1.1 Thermocouple Calibration

Calibration of the E-Type thermocouples is conducted using a two-point method referencing the freezing and boiling points of distilled water. The freezing point reference is achieved by submersing the tips of the thermocouples in a vessel containing packed shaved ice and water. Boiling water is produced in a small stainless steel vessel where the water is allowed to reach a rolling boiling. Accurate boiling temperatures are determined from barometric pressure readings. Three measurement files, each containing recorded data for ten seconds, are taken for both calibration temperatures. The measurement files are individually averaged and inputted into a data reduction spreadsheet where linear regressions can be formed for the measured temperatures versus the actual temperatures. These regressions are used to correct the measured data in LabVIEW.

6.1.2 Differential Pressure Sensor Calibration

The differential pressure sensors are calibrated using an Omega DPI610 pressure calibrator. Pressure measurements are taken from -1700 Pa to 1700 Pa in 350 Pa increments. LabVIEW is used to measure and log sensor output voltages during calibration. The measured voltages are entered into Excel and compared to calibration pressures determined by the DPI610. From this data a linear regression is calculated for the sensor and the coefficients for the regression are used in LabVIEW to convert measured voltages to pressure in Pascals.

6.1.3 Humidity Sensor Calibration

Humidity sensors are calibrated with a Vaisala HM70 humidity and temperature transmitter with HMP76 probe with NIST traceable calibration. A chamber is used to calibrate all four sensors simultaneously and is described in more detail on pages A-6 and A-7 in Appendix A. The chamber positions the four sensors around the Vaisala calibrator probe. Simultaneous calibration

is utilized to ensure consistent and repeatable calibrations between the four sensors. LabVIEW is used to record the sensor voltages for ten seconds. Six reference relative humidities of 2, 36, 44, 51, 66, and 92% are used to achieve accurate calibrations. The Vaisala calibrator can record data for one minute intervals and can download the recorded data to a computer. Software is then used to extract the reference humidities from the calibrator which are then compared to the sensor voltages to determine the sensor calibration curves. These curves are entered into the LabVIEW program to convert the measured sensor voltages to relative humidities.

6.2 Diffusion Coefficient Uncertainty

To determine the error in the diffusion coefficient measurement, measured sensor errors must be carried through the solution of the differential equation. The uncertainty for each measurements can be found using Equation (6.1).

$$U = 2\sqrt{\left(\frac{B}{2}\right)^2 + \left(\frac{\sigma}{\sqrt{N}}\right)^2} \quad (6.1)$$

Where U is the measurement uncertainty, B is the accuracy of the sensor, σ is the measurement standard deviation, and N is the number of independent samples. Once measurement uncertainties are determined it is possible to combine the uncertainties in a weighted average for the differential equation. Uncertainty induced strictly by measurement error for the calculated diffusion coefficient is found to be 8.0%. Additional uncertainty induced by possible bulk flow is discussed and added to the 8.0% uncertainty to give the total error. Bulk flow uncertainty is not contained here as system differential pressure is not a property used by the differential equation in the calculation of the diffusion coefficient.

6.3 Bulk Flow Sensitivity

Diffusion measurements in a dynamic apparatus such as used here are susceptible to bulk flow influences. Due to the independently driven flows and pressure drops along the channel lengths, differential pressure between the channels can exist. If this differential pressure becomes significant enough, the induced bulk flow across the GDL dominates the much smaller diffusion

component. Bulk flow in the same direction of diffusion will result in a higher calculated diffusion coefficient while bulk flow opposing diffusion will reduce the calculated diffusion coefficient.

An in-depth analysis into the influences of bulk flow on the calculated diffusion coefficient was performed. Mass transfer was calculated for an assumed effective diffusion coefficient of $0.10 \text{ cm}^2/\text{s}$ and channel water vapor concentrations based on 95% and 0% relative humidity. Bulk flow was calculated based on Darcy's law for given differential pressures and GDL permeabilities. Water content was calculated for the air transfer by bulk flow based on the channel's mixing ratio that the air originated in. This additional water content was then added to the water vapor transferred by diffusion to calculate an influenced diffusion coefficient. Table 6.1 displays the estimated influences of bulk flow for GDL with and without MPL. The left side of Table 6.1 shows that the small pore diameter and low permeability of the MPL reduces the influence of bulk flow for GDL with MPL compared to without shown on the right. With the given accuracy of the pressure sensors of $\pm 5 \text{ PA}$, additional error must be added to the diffusion coefficient. For samples without MPL, an additional 14.24% error must be combined with the error based on the differential equation. Samples with MPL accrue an additional 0.62% error for a total of 8.62% error. This setup is primarily intended to test more common fuel cell diffusion media that are coated with a MPL due to the greater accuracy obtained.

Table 6.1: Error in diffusion coefficient induced by pressure driven bulk flow for GDL with MPL (left) and without MPL (right) with bulk flow in diffusion direction.

DP [Pa]	Total transfer [kmol/s]	Influenced diffusion coefficient [m ² /s]	Percent error
0.1	9.16E-10	1.00E-06	0.01
0.2	9.16E-10	1.00E-06	0.02
0.3	9.16E-10	1.00E-06	0.04
0.4	9.16E-10	1.00E-06	0.05
0.5	9.16E-10	1.00E-06	0.06
0.6	9.16E-10	1.00E-06	0.07
0.7	9.16E-10	1.00E-06	0.09
0.8	9.16E-10	1.00E-06	0.10
0.9	9.16E-10	1.00E-06	0.11
1	9.16E-10	1.00E-06	0.12
2	9.17E-10	1.00E-06	0.25
3	9.17E-10	1.00E-06	0.37
4	9.17E-10	1.00E-06	0.50
5	9.17E-10	1.01E-06	0.62
6	9.17E-10	1.01E-06	0.75
7	9.17E-10	1.01E-06	0.88
8	9.18E-10	1.01E-06	1.00
9	9.18E-10	1.01E-06	1.13
10	9.18E-10	1.01E-06	1.26
15	9.19E-10	1.02E-06	1.89
20	9.20E-10	1.03E-06	2.54
40	9.23E-10	1.05E-06	5.19
60	9.26E-10	1.08E-06	7.96
80	9.29E-10	1.11E-06	10.87
100	9.33E-10	1.14E-06	13.91
150	9.41E-10	1.22E-06	22.18
200	9.49E-10	1.32E-06	31.57
250	9.57E-10	1.42E-06	42.33
300	9.65E-10	1.55E-06	54.77
350	9.73E-10	1.69E-06	69.32
400*	9.82E-10	1.87E-06	86.56

DP [Pa]	Total transfer [kmol/s]	Influenced diffusion coefficient [m ² /s]	Percent error
0.1	9.17E-10	1.00E-06	0.25
0.2	9.17E-10	1.01E-06	0.51
0.3	9.17E-10	1.01E-06	0.77
0.4	9.18E-10	1.01E-06	1.02
0.5	9.18E-10	1.01E-06	1.28
0.6	9.18E-10	1.02E-06	1.54
0.7	9.19E-10	1.02E-06	1.80
0.8	9.19E-10	1.02E-06	2.07
0.9	9.19E-10	1.02E-06	2.33
1	9.20E-10	1.03E-06	2.60
2	9.23E-10	1.05E-06	5.31
3	9.26E-10	1.08E-06	8.15
4	9.30E-10	1.11E-06	11.12
5	9.33E-10	1.14E-06	14.24
6	9.36E-10	1.18E-06	17.52
7	9.40E-10	1.21E-06	20.96
8	9.43E-10	1.25E-06	24.59
9	9.46E-10	1.28E-06	28.41
10	9.50E-10	1.32E-06	32.44
15	9.66E-10	1.57E-06	56.52
20*	9.83E-10	1.90E-06	89.87

* Calculated diffusion coefficient becomes larger than free stream coefficient after last point listed.

7.0 RESULTS AND DISCUSSION

This section will give an overview of the experimental results obtained with the apparatus purposed in this work along with discussions on the observed effects of MPL, thickness, and PTFE loading on the effective diffusion coefficient. Results are compared to other works for validation and discussion.

7.1 EXPERIMENTAL RESULTS

Validation of the experimental apparatus was conducted before full scale testing was completed. A Grafil U-105 A GDL sample was used to test measurement repeatability for samples with MPL. Repeatability testing was conducted over three days with full equipment shutdowns between each point. Five points were collected at 500 sccm over the three day period. Data points show good repeatability with all points falling within error along the linear trend line shown in Figure 7.1.

Figure 7.1: Repeatability study using a Grafil U-105 A sample. Five individual tests at 500 sccm flow rates performed over a three day period.

Repeatability was also conducted for samples without MPL utilizing a Grafil U-105 B sample. Testing for the no MPL sample was conducted over two days with a total of three points at 500

sccm being taken. The data points for the Grafil U-105 B sample show good agreement across the three data points taken as shown in Figure 7.2.

Figure 7.2: Repeatability study using a Grafil U-105 B sample without MPL. Three individual tests at 500 sccm flow rate performed over a two day period.

After the test setup was shown to give repeatable data for both types of samples, the full range of samples was tested. The Grafil sample pair was tested first, followed by the SGL samples in series pairs, and finally the Toray series was tested. Experimental results pertinent for the model are given in Table 7.1 and Table 7.2. The tables list the sample thickness, test flow rate, inlet relative humidity for both channels, outlet relative humidity for the dry channel, system temperature, atmospheric pressure, individual test case calculated effective diffusion coefficient, and the averaged effective diffusion for all test cases of the sample.

Table 7.1: Experimental results Part 1.

	Thickness [μm]	Sample Test Number	Flow Rate [scm]	Wet Channel Inlet Humidity [%]	Dry Channel Inlet Humidity [%]	Dry Channel Outlet Humidity [%]	Dry Channel Humidity Change [$\Delta\%$]	Temperature [$^{\circ}\text{C}$]	Atmospheric Pressure [Pa]	Calculated Diffusion Coefficient [cm^2/s]	Sample Averaged Diffusion Coefficient [cm^2/s]
Grafil U-105 A	210	1	500	92.28	7.18	34.82	27.64	25.11	102000	0.065	0.070
		2	500	92.55	7.51	35.93	28.42	25.05	102000	0.070	
		3	500	95.54	7.97	36.56	28.59	24.38	103000	0.068	
		4	500	94.44	10.21	44.85	34.64	24.65	103000	0.063	
		5	500	92.70	7.06	35.55	28.49	24.94	103000	0.072	
		6	625	93.03	6.41	32.52	26.11	24.92	103000	0.075	
		7	750	94.60	5.83	29.56	23.73	24.58	103000	0.074	
Grafil U-105 B	200	1	500	93.75	7.14	37.67	30.53	24.67	103000	0.093	0.095
		2	500	93.31	6.99	37.20	30.21	24.86	102470	0.091	
		3	500	92.87	6.85	36.91	30.06	24.93	102470	0.091	
		4	625	93.27	6.12	33.65	27.53	24.79	103000	0.098	
		5	750	92.15	5.33	30.17	24.84	24.92	103000	0.100	
SGL 25BC	225	1	500	94.86	7.10	33.75	26.65	24.55	101450	0.060	0.063
		2	625	94.63	6.10	29.97	23.87	24.62	101450	0.064	
		3	750	94.27	5.39	26.66	21.27	24.68	101450	0.065	
SGL 25BA	190	1	500	95.02	7.00	39.2	32.20	24.35	101760	0.101	0.102
		2	625	94.34	6.05	34.91	28.86	24.57	101760	0.102	
SGL 35BC	325	1	500	97.71	7.82	35.38	27.56	25.03	101290	0.091	0.091
		2	500	98.83	7.75	34.85	27.10	24.63	100910	0.084	
		3	625	97.74	7.02	31.50	24.48	25.09	101290	0.094	
		4	750	97.67	6.13	28.52	22.39	25.18	101290	0.095	
		5	750	97.73	6.00	27.81	21.81	25.25	101290	0.093	

Table 7.2: Experimental results Part 2.

	Thickness [μm]	Sample Test Number	Flow Rate [sccm]	Wet Channel Inlet Humidity [%]	Dry Channel Inlet Humidity [%]	Dry Channel Outlet Humidity [%]	Dry Channel Humidity Change [$\Delta\%$]	Temperature [$^{\circ}\text{C}$]	Atmospheric Pressure [Pa]	Calculated Diffusion Coefficient [cm^2/s]	Sample Averaged Diffusion Coefficient [cm^2/s]
SGL 35BA	300	1	500	95.12	5.95	38.62	32.67	24.83	101150	0.156	0.148
		2	500	97.42	5.98	38.82	32.84	24.36	101930	0.140	
SGL 10BC	420	1	500	93.42	6.04	30.41	24.37	24.63	99900	0.093	0.096
		2	625	92.75	5.25	26.99	21.74	24.73	99900	0.098	
SGL 10BA	400	1	500	96.49	5.37	35.54	30.17	24.67	101800	0.138	0.146
		2	625	96.54	4.70	32.29	27.59	24.69	101800	0.153	
Toray 120 - plain	370	1	500	96.97	5.77	31.87	26.10	24.53	101390	0.086	0.089
		2	500	95.82	5.62	31.50	25.88	24.83	101760	0.086	
		3	625	95.19	4.80	27.51	22.71	24.98	101760	0.089	
		4	750	95.04	4.23	24.91	20.68	25.02	101700	0.095	
Toray 120 - 10%	370	1	500	97.01	5.66	28.86	23.20	24.71	101420	0.066	0.068
		2	500	92.47	5.53	27.57	22.04	25.89	101229	0.066	
		3	625	96.38	4.74	25.06	20.32	24.84	101420	0.069	
		4	750	95.09	4.10	22.09	17.99	25.14	101420	0.072	
Toray 120 - 20%	370	1	500	94.28	5.89	27.30	21.41	25.45	101290	0.060	0.063
		2	625	93.79	4.80	23.47	18.67	25.52	101290	0.062	
		3	750	93.46	4.19	21.22	17.03	25.58	101290	0.068	
Toray 120 - 40%	370	1	500	96.47	5.83	18.54	12.71	24.87	101600	0.025	0.026
		2	625	96.01	4.90	15.61	10.71	24.87	101600	0.025	
		3	750	95.76	4.30	13.95	9.65	24.93	101600	0.027	

Experimental results obtained follow expected trends with MPL coated samples transferring less water vapor than samples uncoated and increasing levels of PTFE decreasing the amount of water vapor transfer. Toray samples all transferred less water vapor than the comparable SGL 10BA and behaved much more like samples with MPL coatings. The reduction in the effective diffusion coefficient can be elucidated from the permeability specifications for the different layers. Permeability values for bulk flow as listed in Table 7.3 show that the value for TGP-H-120 0% is 2.5 times less than the comparable SGL 10BA. Also, the MPL sample like behavior can be explained by the permeability value that is closer to the MPL sample values than any of the SGL BA samples. Visible inspection of the Toray samples gives indication of the lower porosity and permeability as it is not possible to see through even the plain Toray sample when held up to the light while SGL 10BC, which is thicker than TGP-H-120, offers visibility through the sample.

Table 7.3: Permeability values for listed GDL samples.

Sample	Permeability* [cm ³ /cm ² ·s]
SGL 25BC [41]	1
SGL 25BA [41]	210
SGL 35BC [42]	1.5
SGL 35BA [42]	170
SGL 10BC [43]	1.45
SGL 10BA [43]	85
Toray TGP-H-120 Series [44]**	34.9
* All values taken from manufacturer data sheets, see Appendix C for details.	
** Toray permeability converted from ml·mm/cm ² ·hr·mmAq based on SGL pressure drop of 304 Pa for comparison purposes.	

The low permeability of Toray is best explained by the differences in manufacturing processes. SGL manufactures the Sigracet GDL series as one layer in a dry-laid process whereas Toray uses a wet-laid [45] base layer, TGP-H-030, stacked together to make the

different thicknesses, in this case 4 layers to make TGP-H-120. These individual layers could induce higher diffusion resistances at the interfacial zones depending on how the layers are bonded together. The differences in manufacturing techniques also causes differences in bonding and sintering of the fiber matrix which can alter the pore structure and tortuosity of the diffusion media.

7.2 EFFECTS OF MPL

Microporous layer coatings provide significant resistance to diffusion due to the reduction in pore size when compared to the pore size of GDL. The typical pore size for MPL range between 0.02-0.5 μm while the GDL has pores in the range of 1-100 μm [17]. Grafil U-105 series and SGL 25 series diffusion media can be directly compared because of their similar thicknesses. SGL 25BA and Grafil U-105 B, both without MPL, were found to have similar effective diffusion coefficients of $0.10 \pm 0.02 \text{ cm}^2/\text{s}$ and $0.10 \pm 0.02 \text{ cm}^2/\text{s}$, respectively. Differences were determined for SGL 25BC and Grafil U-105 A, both with MPL, with effective diffusion coefficients of $0.063 \pm 0.005 \text{ cm}^2/\text{s}$ and $0.070 \pm 0.006 \text{ cm}^2/\text{s}$, respectively.

Samples of SGL 25BC and Grafil U-105 A were imaged with a confocal laser scanning microscope (CLSM) to determine possible morphological differences between the two diffusion media. From the CLSM images obtained from the samples, it appears that the MPL on the Grafil U-105 A sample is thinner than the layer on the SGL 25BC sample. It is possible to see visible fibers protruding from the MPL coating on the Grafil sample as seen in Figures 7.3 and 7.4. Although no visible fibers can be seen in the SGL 25BC images, Figures 7.5 and 7.6, cracks and other surface imperfections can be seen. Due to the proprietary nature of the two microporous coatings it is not possible to draw definitive conclusions to the different performances of the layers. This is an area that can be explored in future work.

Figure 7.3: Low magnification image of Grafil U-105 A MPL with visible fibers using a CLSM.

Figure 7.4: High magnification CLSM image of Grafil U-105 A MPL showing visible fibers and surface imperfections.

Figure 7.5: Low magnification CLSM image of SGL 25BC MPL showing surface imperfections.

Figure 7.6: High magnification SLCM image of large crack in surface of SGL 25BC MPL.

If the MPL coating of the Grafil sample is thinner, it would provide less diffusion resistance than the thicker coating applied to the SGL sample resulting in a higher effective diffusion coefficient. The effective diffusion coefficient for the SGL 25BC sample could be lower than what was observed here as the cracks in the MPL provide a lower resistance path for diffusion to occur. Figure 7.6 is a high magnification image of a crack in the SGL MPL showing that the crack transverses through the entire thickness of the MPL. These cracks and imperfections in the MPL can hinder proper water management as the cracks allow liquid water to transport directly from the catalyst layer to the GDL whereas it would normally have to diffuse in the vapor phase through the small pores of the MPL. The effects of these imperfections and their influence on diffusion could be addressed in future work.

7.3 EFFECTS OF THICKNESS

Thickness effects were determined to be negligible for the SGL series without MPL. No effects were expected for the SGL BA series diffusion media because of the high air permeability for all samples as listed in Table 7.3. Figure 7.7 compares the three GDL samples without MPL. As seen in the figure, all three samples fall within error of each other suggesting that no effect from thickness exists. A greater number of tests is necessary to conclude this and will be pursued in future work.

Figure 7.7: Comparison of the three SGL BA series GDL without MPL coating.

The SGL BC series with MPL did demonstrate minor thickness effects with the thinnest sample, 25BC, giving a lower effective diffusion coefficient of $0.063 \pm 0.005 \text{ cm}^2/\text{s}$ compared to the values of $0.091 \pm 0.008 \text{ cm}^2/\text{s}$ and $0.096 \pm 0.008 \text{ cm}^2/\text{s}$ for SGL 35BC and SGL 10BC, respectively.

Figure 7.8: Comparison of the three SGL samples with MPL coating.

When the MPL is coated onto the GDL it can imbibe into the GDL pores thus making its influence towards the total combined effective diffusion coefficient greater. This effect is less significant in thicker GDL if the imbibition distance is equal to the thinner material as the combined coefficient has less influence from the MPL. Figure 7.9 displays the relative ratios of influence for the MPL in thin and thick GDL. The thin GDL on the left would have an MPL/GDL combination to GDL ratio of approximately 1:1 whereas the thicker sample would be 1:3. MPL effects for SGL 35BC and 10BC could have been lessened by the extra thickness enough to allow similar effective diffusion coefficients even with the increase in thickness between the samples.

Figure 7.9: Ratio of MPL/GDL combination to plain GDL for two GDL thicknesses, approximately 1:1 for left sample and 1:3 for right sample, showing the reduction in MPL influence on diffusion resistance for thicker samples.

7.4 EFFECTS OF PTFE

PTFE loading was found to have a dramatic effect on the effective diffusion coefficient. Figure 7.10 shows the decreasing trend in effective diffusion coefficient for increasing levels of PTFE loading.

Figure 7.10: Decreasing trend in effective diffusion coefficient with increasing weight percentage of PTFE.

The decrease in effective diffusion coefficient is caused by the reduction in porosity, listed in Table 7.4, and increase in tortuosity. Diffusion path length increases as PTFE can form

webbing that constricts or completely choke off pores making it necessary for water vapor to diffuse more three-dimensionally to find the best paths through-plane.

Table 7.4: Toray TGP-H-120 porosity with changing PTFE loading [46].

Sample	Porosity
TGP-H-120 0%	76.3
TGP-H-120 10%	73.9
TGP-H-120 20%	69.6
TGP-H-120 40%	61.6

7.5 EXPERIMENTAL COMPARISON TO THE LITERATURE

Validation of the experimental apparatus is performed through comparison to values from the literature. Experimental values for the Toray series are compared to the values obtained by Flückiger *et al.* [23], Figure 7.11, and Zamel *et al.* [18], Figure 7.12.

Figure 7.11: Good agreement is shown with the work performed by Flückiger *et al.* [23].

Comparison is performed with Toray series GDL.

Figure 7.12: Agreement shown with work performed by Zamel *et al.* [18]. Comparison is performed with Toray series GDL.

As shown in Figures 7.11 and 7.12, good agreement with the values from literature is achieved. This exhibits the validity of the dynamic method for testing diffusion coefficients in fuel cell gas diffusion layers. Comparisons of SGL results to the literature are not performed as identical SGL samples have not been tested in the literature. Work has been performed with different variants of the same SGL series used here and would not allow for a 1-to-1 comparison.

7.6 COMPARISON TO CORRECTIONS FROM THE LITERATURE

As stated earlier, there are several corrections for calculating the effective diffusion coefficient that are typically used in fuel cell modeling. The empirically based Bruggeman correction [4] is given again in Equation (7.1) where ε is porosity.

$$D_{eff} = D_{AB} \varepsilon^{1.5} \quad (7.1)$$

Tomadakis and Sotirchos [13] modeled the effective diffusion coefficient for randomly oriented fibers and developed the correction given in Equation (7.2) where ε_p is the percolation threshold and α is an empirical constant.

$$D_{eff} = D_{AB} \varepsilon \left(\frac{\varepsilon - \varepsilon_p}{1 - \varepsilon_p} \right)^\alpha \quad (7.2)$$

Mezedur *et al.* [14] modeled diffusion through catalytic materials to determine a correction for effective diffusivity. This correction is given in Equation (7.3).

$$D_{eff} = D_{AB} \left(1 - (1 - \varepsilon)^{0.46} \right) \quad (7.3)$$

To test the accuracy of these corrections, they are compared to the experimental results obtained in this study. Corrections are only compared with GDL samples without MPL due to the presence of Knudsen diffusion in the MPL. The effective diffusion coefficient calculated for the samples with MPL include Knudsen diffusion and therefore do not strictly represent Fickian diffusion. Figure 7.13 compares the experimental results obtained for the SGL series GDL with the three corrections. The corrections are plotted across the full range of porosity from 0 to 1 while the experimental results are plotted at the respected porosity. All three corrections overestimate the effective diffusion coefficient. The Bruggeman correction estimates the highest value given that the correction was developed using spherical objects to form the solid matrix. The correction developed by Mezedur *et al.* produced the estimation with the closest agreement with the experimental results.

Figure 7.13: Comparison of SGL series, 5% by weight PTFE loading and without MPL coating, to common fuel cell diffusion corrections.

The three corrections are plotted out for the entire possible range of porosity, from 0% to 100%, and compared to the Toray series testing in Figure 7.14. Once again it is shown that the corrections overestimate the effective diffusion coefficient. The Bruggeman correction produces the highest estimate over the porosities typically seen in fuel cell diffusion media, approximately 50% to 90%, while the correction by Mezedur *et al.* produces the lowest estimation over this range.

Figure 7.14: Comparison of correction values to experimental results obtained with Toray series diffusion media.

The overestimation of effective diffusion coefficients by the three corrections allow for an overestimation of fuel cell performance in numerical models. An increase in the water vapor diffusion coefficient would allow for a higher mass flux of water vapor to transverse the GDL from CL to gas channel thus helping to prevent flooding conditions from occurring. Although models such as Tomadakis and Sotirchos [13] and Mezedur *et al.* [14] account for random and patterned fiber orientations, they neglect the interaction of binder and PTFE loading.

8.0 CONCLUSION

An apparatus has been designed and developed to measure the effective water vapor diffusion coefficient for typical diffusion media. The setup allows precise control of the water vapor concentration gradient induced across the test sample while removing as many unknowns typically present in in situ testing. Water vapor concentration gradients are controlled with humidifiers in independent flow streams on either side of the GDL. Effective diffusion coefficients were measured to determine the effects that MPL coatings, GDL thickness, and PTFE loading have on diffusion. To test these variations in GDL manufacturing, several commercially available GDL were selected and include MRC Grafil U-105 series, SGL Sigracet® 25, 35, and 10 series, and Toray TGP-H-120 in 0%, 10%, 20%, and 40% PTFE loading by weight.

- MPL coatings were found to have a significant resistance towards diffusion with values of effective diffusion coefficients of the GDL/MPL pair 1/2 to 1/3 that of just the GDL by itself. The MPL coating for the Grafil series was found to be thinner than the SGL MPL with visible fibers protruding through the top surface of the MPL. SGL MPL was shown to have surface defects and cracks that transverse the entire layer. The imperfections in both the Grafil series and SGL series can result in preferential transport locations and reduction in the effectiveness of the MPL for water management.
- Thickness was found to have a negligible influence on the effective diffusion coefficient measurement. Variation in the GDL/MPL combined diffusion coefficient occurred in the thinnest SGL sample, 25BC, and is due to the greater influence the MPL has when combining the two resistances. Care must be taken in the measurement of compressed thickness to calculate the correct effective diffusion coefficient for the tested diffusion media.
- An increase in PTFE loading has a marked reduction in the measured effective diffusion coefficient. Toray TGP-H-120 0% achieved an effective diffusion coefficient of $0.095 \text{ cm}^2/\text{s}$ while TGP-H-120 40% only achieved an effective diffusion coefficient of $0.024 \text{ cm}^2/\text{s}$. The rapid reduction in effective diffusion

coefficient with increasing levels of PTFE loading is indicative of a reduction of porosity and pore diameter and an increase in tortuosity.

- Experimental data is compared to other experimental works from the literature. Good agreement is shown with these works thus validating the dynamic method utilized in this work.
- Experimental values for SGL 25BA, 35BA, and 10BA and the Toray series are compared with correction factors developed by Bruggeman [4], Tomadakis and Sotirchos [13], and Mezedur *et al.* [14]. All three corrections were found to overpredict the effective diffusion coefficient. The correction by Mezedur *et al.* produces the closest fit, within experimental error, to the SGL data. All models significantly overpredict the Toray series over the entire range of PTFE loading.
- Effective water vapor diffusion coefficients are measured experimentally for a wide range of current commercially available diffusion media. These coefficients are measured at standard temperature and pressure and can be extended to other temperatures and pressures with standard corrections for temperature and pressure. The experimental coefficients show the overprediction of diffusion and fuel cell performance determined by numerical models due to the use of common corrections. It is found that it will be necessary to pay close attention to diffusion coefficients when modeling water transport in the GDL to calculate appropriate fuel cell performance from numerical models.

9.0 SUGGESTIONS FOR FUTURE WORK

9.1 Channel Air Velocity Reduction

As bulk flow has been shown to influence the diffusion coefficient measurements, especially for GDL without MPL, it will be important to reduce this influence. Bulk flow is governed by the differential pressure across the GDL and thus differential pressure will be the focus area for reduction of bulk flow. Pressure within the channels is affected by the air velocity in the channel. Two approaches can be used to obtain lower air velocities, first, lower flow rate mass flow controllers would allow for lower flow rates, hence lower velocities, and better control resolution. Second, utilizing a multichannel setup to divide the flow evenly between channels will reduce the flow rates through each channel by the number of channels.

9.2 Temperature Effects

It is desired to test the influence of temperature variation on diffusion to verify if the standard temperature correction for binary diffusivities is still applicable to diffusion through a solid matrix. In order to maintain uniform test section temperature at temperatures above ambient, an environmental chamber is required to ensure isothermal operation. This approach will maintain an even temperature for the test section whereas heaters mounted to the test section would cause temperature gradients and fluctuations induced by the heater controller. The chamber should allow for temperatures to range from ambient up to typical operational conditions around 80°C where little liquid water typically exists in the cell. Several temperatures should be tested between 20°C and 80°C to determine if changes in diffusion coefficients occur in a linear fashion.

9.3 Analytical Model Improvements

The analytical model could be made more robust to increase accuracy. To help mitigate influences of bulk pressure and with better pressure sensors, the model could account for bulk flow. An input could be added to calculate bulk flow based on pressure gradient and remove that mass transfer from the calculations for diffusion coefficient. The pressure gradient could be based on an average of the inlet and outlet differential pressures or based on a linear gradient from inlet to outlet based on the differential pressures at each end of the

channel. It is also possible to include developing flow and entrance region effects in more detail in the model as currently it is only included in the calculation of the Nusselt number for determining the convection coefficient.

9.4 Through-plane GDL Temperature Gradient

Another un-captured condition in operating fuel cells that may affect the water vapor diffusion coefficient is the presence of a temperature gradient from the catalyst layer to the gas channels. This gradient could induce higher diffusion rates by producing a heat pipe effect within the GDL. To test this effect, new hardware would need to be developed that would reduce the thermal conductance of the system, provide thermal isolation between the two halves, and allow independent temperature control of the two halves. If the heaters are designed properly it would also be possible to test a thermal gradient from inlet to outlet of the channels with the temperature increases towards the channel exit as this is a typical condition of actual fuel cells.

9.5 Compression Effects

It is desirable to know if water vapor diffusion coefficients change with GDL compression. A setup should be developed that allows controlled compression based on compression distance as opposed to compression force. Controlled compression distance will allow for accurate compression percentages that could vary from zero to 50% compression. A range of compression percentages should be tested to determine linearity of change in diffusion coefficients.

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APPENDIX A

DESIGN PACKAGE

TOLERANCES		R.I.T	
X.X ± 0.1			
X.XX ± 0.01		CHANNEL_PI	
X.XXX ± 0.001		A	
≤ ± 1°		INCH	SHEET 1 OF 1
JACOB LAMANNA			

SCALE 0.750

Dimensions for channel and alignment pins

Channel depth 0.390 inches
Alignment pins 0.125 diameter, 0.150 deep, flat bottom

SCALE 0.750

TOLERANCES X.X ± 0.1 X.XX ± 0.01 X.XXX ± 0.001 < ± 1°		R.I.T		
JACOB LAMANNA	A		ORING_CHANNEL	
		INCH		SHEET 1 OF 1

Dimensions for o-ring gasket groove

Notes:
All dimensions for centerline of oring gland
Gland depth 0.050" to 0.054"
Gland width 0.084" to 0.089"

TOLERANCES X.X ± 0.1 X.XX ± 0.01 X.XXX ± 0.001 < ± 1°		R.I.T	
JACOB LAMANNA	A	ORING_CHANNELC	
SCALE 0.750	INCH	SHEET 1 OF 1	

Dimensions for pipe taps and headers

TOLERANCES		R.I.T	
X.X ± 0.1			
X.XX ± 0.01			
X.XXX ± 0.001			
JACOB LAMANNA		A	ORING_CHANNEL B
		INCH	SHEET 1 OF 1

SCALE 0.750

SECTION A-A

SCALE 0.500

TOLERANCES X.X $\pm$ 0.1 X.XX $\pm$ 0.01 X.XXX $\pm$ 0.001 $\angle \pm 1^\circ$		R.I.T	
JACOB LAMANA	A		
ORING_CHANNEL		SHEET 1 OF 1	

- Notes:
- A: All 1 inch ports provide mounting locations for humidity sensors
  - B: Hole tapped for 1/4 inch NPT connection fitting
  - C: Holes tapped for #8 cap screws to provide clearance for connection fitting

TOLERANCES
X.X $\pm$ 0.1
X.XX $\pm$ 0.01
X.XXX $\pm$ 0.001
 $\angle$ $\pm$ 1°

R.I.T

JACOB LAMANNA

CALIBRATOR

INCH

SHEET 1 OF 1

SCALE 0.750

- Notes:
- A: Calibration Chamber
  - B: Calibrator Probe
  - C: Humidity Sensors
  - D: Humidity Sensor Plates
  - E: Rubber Band Supports
  - F: Connection Fitting
  - G: Cap Screw Leg
  - H: Flow Outlet

TOLERANCES		R.I.T	
X.X ± 0.1			
X.XX ± 0.01		BRATOR_CROSS_SECT ON	
X.XXX ± 0.001		A	INCH
< ± 1°		SHEET 1 OF 1	
JACOB LAMANNA			

SCALE 1.000

APPENDIX B DIFFERENTIAL EQUATION SOLUTION

Solution of system of differential equations defined by Equations (1) and (2).

$$L \frac{dX}{dz} = -K_y w \left(\frac{X}{1+X} - \frac{Y}{1+Y} \right) \quad (1)$$

$$LX + VY = LX_0 + VY_0 \quad (2)$$

L, V, X_0 , and Y_0 are known, solving for Y.

$$Y = -\frac{L}{V}X + \frac{L}{V}X_0 + Y_0 \quad (3)$$

Let,

$$\alpha = \frac{L}{V}X_0 + Y_0 \quad (4)$$

Subbing (4) into (3)

$$Y = -\frac{L}{V}X + \alpha \quad (5)$$

Subbing (5) into (1)

$$L \frac{dX}{dz} = -K_y w \left(\frac{X}{1+X} - \frac{\left[-\frac{L}{V}X + \alpha \right]}{1 - \frac{L}{V}X + \alpha} \right) \quad (6)$$

Let,

$$A = \frac{X}{1+X} + \frac{\frac{L}{V}X - \alpha}{1 + \alpha - \frac{L}{V}X} \quad (7)$$

Summing both fractions together from (7),

$$A = \frac{X\left(1 + \alpha - \frac{L}{V}X\right) + (1 + X)\left(\frac{L}{V}X - \alpha\right)}{(1 + X)\left(1 + \alpha - \frac{L}{V}X\right)} \quad (8)$$

Expanding (8),

$$A = \frac{X + \alpha X - \frac{L}{V}X^2 + \frac{L}{V}X - \alpha + \frac{L}{V}X^2 - \alpha X}{(1 + X)\left(1 + \alpha - \frac{L}{V}X\right)} \quad (9)$$

Cancelling like terms and simplifying (9),

$$A = \frac{X\left(1 + \frac{L}{V}\right) - \alpha}{(1 + X)\left(1 + \alpha - \frac{L}{V}X\right)} \quad (10)$$

Subbing simplified Equation (10) into (6),

$$L \frac{dX}{dz} = -K_y w \left[\frac{X\left(1 + \frac{L}{V}\right) - \alpha}{(1 + X)\left(1 + \alpha - \frac{L}{V}X\right)} \right] \quad (11)$$

Separate variables in (11)

$$\left[\frac{(1 + X)\left(1 + \alpha - \frac{L}{V}X\right)}{X\left(1 + \frac{L}{V}\right) - \alpha} \right] dX = \frac{-K_y w}{L} dz \quad (12)$$

Integrate each side as indefinite integrals, limits will be added after integration. dX limits $X_0 \rightarrow X_1$, dz limits $0 \rightarrow 1$.

Setup u substitution,

$$u = X\left(1 + \frac{L}{V}\right) - \alpha \quad (13)$$

$$X = \frac{u + \alpha}{\left(1 + \frac{L}{V}\right)} \quad (14)$$

$$du = \left(1 + \frac{L}{V}\right) dX \quad (15)$$

Rewrite dX portion of (12) with u substitution definitions,

$$\int \frac{\left(1 + \frac{u + \alpha}{\left(1 + \frac{L}{V}\right)}\right) \left(1 + \alpha - \frac{L}{V} \left[\frac{u + \alpha}{\left(1 + \frac{L}{V}\right)}\right]\right) \frac{du}{\left(1 + \frac{L}{V}\right)}}{u} \quad (16)$$

Simplifying (16),

$$\int \frac{\left(1 + \frac{\alpha}{\left(1 + \frac{L}{V}\right)} + \frac{u}{\left(1 + \frac{L}{V}\right)}\right) \left(1 + \alpha - \frac{L}{V} \frac{\alpha}{\left(1 + \frac{L}{V}\right)} - \frac{\frac{L}{V} u}{\left(1 + \frac{L}{V}\right)}\right) \frac{du}{\left(1 + \frac{L}{V}\right)}}{u} \quad (17)$$

Let $\beta = 1 + \frac{L}{V}$,

$$\int \frac{\left(1 + \frac{\alpha}{\beta} + \frac{u}{\beta}\right) \left(1 + \alpha - \frac{L}{V} \frac{\alpha}{\beta} - \frac{Lu}{V\beta}\right) \frac{du}{\beta}}{u} \quad (18)$$

Let $\gamma = 1 + \frac{\alpha}{\beta}$ and $\delta = 1 + \alpha - \frac{L\alpha}{V\beta}$,

$$\int \frac{\left(\gamma + \frac{u}{\beta}\right) \left(\delta - \frac{Lu}{V\beta}\right) \frac{du}{\beta}}{u} \quad (19)$$

Expanding terms,

$$\frac{1}{\beta} \int \frac{\gamma\delta - \frac{L\gamma}{V\beta}u + \frac{\delta}{\beta}u - \frac{L}{V\beta^2}u^2}{u} du \quad (20)$$

Integrating,

$$\frac{1}{\beta} \left[\gamma\delta \ln u + \left(\frac{\delta}{\beta} - \frac{L\gamma}{V\beta} \right) u - \frac{L}{2V\beta^2} u^2 \right] + c \quad (21)$$

Dropping the constant as it has no physical meaning, substituting the definition of u back in, and adding the limits of integration as defined earlier,

$$\frac{1}{\beta} \left[\gamma\delta \ln(X\beta - \alpha) + \left(\frac{\delta}{\beta} - \frac{L\gamma}{V\beta} \right) (X\beta - \alpha) - \frac{L}{2V\beta^2} (X\beta - \alpha)^2 \right]_{X_0}^{X_l} \quad (22)$$

Apply the limits if integration to (22),

$$\begin{aligned} & \frac{1}{\beta} \left[\gamma\delta \ln(X_l\beta - \alpha) + \left(\frac{\delta}{\beta} - \frac{L\gamma}{V\beta} \right) (X_l\beta - \alpha) - \frac{L}{2V\beta^2} (X_l\beta - \alpha)^2 \right] \\ & - \left[\gamma\delta \ln(X_0\beta - \alpha) + \left(\frac{\delta}{\beta} - \frac{L\gamma}{V\beta} \right) (X_0\beta - \alpha) - \frac{L}{2V\beta^2} (X_0\beta - \alpha)^2 \right] = \frac{-K_y w l}{L} \end{aligned} \quad (23)$$

Assuming $L=V$ as is the case for this study, and applying this to the definitions of α , β , γ , and δ , the definitions simply as follows,

$$\alpha = X_0 + Y_0 \quad (24)$$

$$\beta = 2 \quad (25)$$

$$\gamma = 1 + \frac{X_0 + Y_0}{2} \quad (26)$$

$$\delta = 1 + \frac{X_0 + Y_0}{2} \quad (27)$$

Applying Equations (24) – (27) to (23) and solving for K_y ,

$$\frac{L}{2wl} \left\{ \left[\left(1 + \frac{X_0 + Y_0}{2} \right)^2 \ln(X_0 - Y_0) - \frac{1}{8}(X_0 - Y_0)^2 \right] - \left[\left(1 + \frac{X_0 + Y_0}{2} \right)^2 \ln(2X_l - X_0 - Y_0) - \frac{1}{8}(2X_l - X_0 - Y_0)^2 \right] \right\} = K_y \quad (28)$$

Equation (28) gives the final form used for the Microsoft Excel calculations.

APPENDIX C
GDL MANUFACTURER TECHNICAL SPECIFICATION DATA
SHEETS

SGL 25 Series.....	C-2
SGL 35 Series.....	C-4
SGL 10 Series.....	C-6
Toray TGP-H Series.....	C-8

SIGRACET®

GDL 24 & 25 Series Gas Diffusion Layer

After the introduction of our 200-micron thick GDL series GDL 20/21 in 2002, SGL Group has been further improving the series with respect to production yield and product variability. We have introduced our newest grades, GDL 24 and GDL 25, to the commercial market. SGL Group has invested heavily in this product development and has been contributing resources from a number of different and cross-functional parts of the organization. The result has been the launch of our new grades GDL 24/25.

Users have confirmed the excellent match of the properties:

- Greatly improved homogeneity inter-lot and intra-lot.
- The overall variability has been shown to be better than $\pm 10\%$ (2 sigma)

- Greatly improved surface flatness
- Greatly reduced occurrence of faulty spots on a roll
- Greater roll length
- Best performance under various operating conditions

The above list is not sacrificing any of the list of properties which makes SGL Group's GDL unique:

- Produced and shipped as a continuous roll good
- Utilizing low-cost nonwoven processes
- Hydrophobic treatment of the substrate to the desired level by coating the bulk surfaces with PTFE
- Coating of material grades with SGL Group's unique microporous layer for better membrane contact, water management and mechanical protection
- Concept of lower and higher porosity (GDL 24: lower, GDL 25: higher porosity) to react with cell humidity level.

The introduced product series GDL 24/25 is intended to combine the required production robustness and the excellent performance properties. Stack developers and assemblers are benefiting from the greatly improved characteristics.

Properties of SIGRACET® GDL 24/25

Property	Unit	GDL 24 BA	GDL 24 BC	GDL 25 BA	GDL 25 BC
Thickness ▲	mil	7.5	9.3	7.5	9.3
	µm	190	235	190	235
Areal Weight	oz/ft²	0.18	0.33	0.13	0.28
	g/m²	54	100	40	86
Porosity	%	84	76	88	80
Air Permeability ■	cm³/(cm² · s)	60	0.6	210	1.0
Electrical Resistance (through plane) ●	mΩcm²	< 10	< 12	< 10	< 12

▲ Under 0.25 N/cm², sample diameter 13 mm

■ Gurley model 4118, 300 cc, 0.1 sq in orifice

● 2-point measurement, circular (25 mm diam.) gold-plated contacts under pressure of 10 bar

Broad Base. Best Solutions.

Grades

We supply SIGRACET Gas Diffusion Layers in roll form in lengths of 75 +/- 25 m. Our standard roll width is 45 cm, but we can split parent rolls into multiples thereof, i. e. 2 x 22.5 cm wide rolls, 3 x 15 cm wide rolls, etc. Sheets stamped to a specified geometry are also available if so desired. Substrate PTFE loadings are available from 0 to 30 wt%, but 5 wt% is standard.

AA	YA		YC		
Y	A	B	C	D	E
% PTFE	0	5	10	20	30

GDL "AA" is our plain substrate with no value-added post-processing.
GDL "BA" is our hydrophobized substrate with a 5 wt% PTFE loading.
GDL "BC" is our hydrophobized substrate (5 wt% PTFE) with our standard Microporous Layer (MPL) on one side.

By courtesy of Nuvera

Unique Characteristics

By courtesy of Hydrogenics

Our GDL 24/25 nonwovens are of a 2-dimensional structure designed to have lower thickness, higher bending stiffness, lower compression set, and lower compressibility than GDL 10.

GDL 24/25 is a further development towards thinner materials maintaining the properties of the GDL 20/21 series. GDL 24/25 has also greatly improved tolerances (thickness and areal weight) and a lower anisotropy factor between the machine and transverse directions.

On request, we will advise you on maximum compression loads for use in bonding to Catalyst-Coated Membranes (CCMs) or assembling stacks. Gasket design also needs to take the compression behavior into account.

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This information is based on our present state of knowledge and is intended to provide general notes on our products and their uses. It should therefore not be construed as guaranteeing specific properties of the products described or their suitability for a particular application. Any existing industrial property rights must be observed. The quality of our products is guaranteed under our "General Conditions of Sale".

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SIGRACET®

GDL 34 & 35 Series Gas Diffusion Layer

After the introduction of our 300-micron thick GDL series GDL 30/31, SGL Group has been further improving the series with respect to production yield and product variability. We have introduced our newest grades, GDL 34 and GDL 35, to the commercial market. SGL Group has invested heavily in this product development and has been contributing resources from a number of different and cross-functional parts of the organization. The result has been the launch of our new grades GDL 34/35.

Users have confirmed the excellent match of the properties:

- Greatly improved homogeneity inter-lot and intra-lot.
- The overall variability has been shown to be better than +/- 10% (2 sigma)

- Greatly improved surface flatness
- Greatly reduced occurrence of faulty spots on a roll
- Greater roll length
- Best performance under various operating conditions

The above list is not sacrificing any of the list of properties which makes SGL Group's GDL unique:

- Produced and shipped as a continuous roll good
- Utilizing low-cost nonwoven processes
- Hydrophobic treatment of the substrate to the desired level by coating the bulk surfaces with PTFE
- Coating of material grades with SGL Group's unique microporous layer for better membrane contact, water management and mechanical protection
- Concept of lower and higher porosity (GDL 34: lower, GDL 35: higher porosity) to react with cell humidity level.

The introduced product series GDL 34/35 is intended to combine the required production robustness and the excellent performance properties. Stack developers and assemblers are benefiting from the greatly improved characteristics.

Properties of SIGRACET® GDL 34/35

Property	Unit	GDL 34 BA	GDL 34 BC	GDL 35 BA	GDL 35 BC
Thickness ▲	mil	11.0	12.4	11.8	12.7
	µm	280	315	300	325
Areal Weight	oz/ft²	0.28	0.46	0.18	0.36
	g/m²	86	140	54	110
Porosity	%	83	75	90	80
Air Permeability ■	cm³/(cm² · s)	45	0.35	170	1.5
Electrical Resistance (through plane) ●	mΩcm²	< 11	< 14	< 12	< 15

▲ Under 0.25 N/cm², sample diameter 13 mm

■ Gurley model 4118, 300 cc, 0.1 sq in orifice

● 2-point measurement, circular (25 mm diam.) gold-plated contacts under pressure of 10 bar

Broad Base. Best Solutions.

Grades

We supply SIGRACET Gas Diffusion Layers in roll form in lengths of 75 +/- 25 m. Our standard roll width is 45 cm, but we can split parent rolls into multiples thereof, i. e. 2 x 22.5 cm wide rolls, 3 x 15 cm wide rolls, etc. Sheets stamped to a specified geometry are also available if so desired. Substrate PTFE loadings are available from 0 to 30 wt%, but 5 wt% is standard.

AA	YA		YC		
Y	A	B	C	D	E
% PTFE	0	5	10	20	30

GDL "AA" is our plain substrate with no value-added post-processing.
GDL "BA" is our hydrophobized substrate with a 5 wt% PTFE loading.
GDL "BC" is our hydrophobized substrate (5 wt% PTFE) with our standard Microporous Layer (MPL) on one side.

By courtesy of Viessmann

By courtesy of P21

Unique Characteristics

Our GDL 34/35 nonwovens are of a 2-dimensional structure designed to have lower thickness, higher bending stiffness, lower compression set, and lower compressibility than GDL 10.

GDL 34/35 has also greatly improved tolerances (thickness and areal weight) and a lower anisotropy factor between the machine and transverse directions.

On request, we will advise you on maximum compression loads for use in bonding to Catalyst-Coated Membranes (CCMs) or assembling stacks. Gasket design also needs to take the compression behavior into account.

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SIGRACET®

GDL 10 Series Gas Diffusion Layer

SIGRACET Gas Diffusion Layers (GDLs) are "graphitized" carbon fiber-based nonwovens, either papers or felts, specifically designed to transport reactant gases into and excess liquid product water out of the electrocatalyst layers of Proton Exchange Membrane (PEM) fuel cells.

The GDL 10 series is our first-generation baseline material, which is produced using a novel technique unique to SGL Group. This material has an open pore structure, good mechanical strength and high

electrical conductivity, and is targeted for use in moderate-to-high humidity and higher current density operating environments. GDL 10 types can also be used with both low or high stack temperatures and in both ambient or pressurized gas conditions. GDL 10 is well suited to stationary and portable power systems. The GDL 10 product line has also been demonstrated successfully as either a cathode GDL or anode Liquid Diffusion Layer (LDL) in Direct Methanol Fuel Cell (DMFC) applications.

GDL 10 substrates are produced in-house by SGL Group and are ready for "post-processing". Our continuous value-added processing steps to the substrate consist of a bulk impregnation of the fiber matrix with PTFE and our standard carbon/PTFE-blended Microporous Layer (MPL) applied to one side. The end result is a "finished GDL" ready to be inserted directly into PEM stacks or to be bonded to Catalyst-Coated Membranes (CCMs) for producing Membrane Electrode Assemblies (MEAs).

Properties of SIGRACET® GDL 10

Property	Unit	GDL 10 BA	GDL 10 BB	GDL 10 BC
Thickness ▲	mil	15.7	16.3	16.3
	µm	400	420	420
Areal Weight	oz/ft²	0.28	0.41	0.44
	g/m²	85	125	135
Porosity	%	88	84	82
Air Permeability ■	cm³/(cm² · s)	85	3	1.45
Electrical Resistance (through plane) ●	mΩcm²	< 12	< 15	< 16

▲ Under 0.45 N/cm² (5 psi), sample diameter 30 mm

■ Gurley model 4118, 300 cc, 0.1 sq.in orifice, 304 Pa, sample diameter 30 mm

● 2-point measurement, circular (25 mm diam.) gold-plated contacts 100 N/cm²

Broad Base. Best Solutions.

Grades

We supply SIGRACET Gas Diffusion Layers in roll form in lengths of 75 +/- 25 m. Our standard roll width is 45 cm, but we can split parent rolls into multiples thereof, i. e. 2 x 22.5 cm wide rolls, 3 x 15 cm wide rolls, etc. Sheets stamped to a specified geometry are also available if so desired. Substrate PTFE loadings are available from 0 to 30 wt%, but 5 wt% is standard.

AA	YA	YC					
Y	A	B	C	D	E		
% PTFE	0	5	10	20	30		

By courtesy of SFC

GDL "AA" is our plain substrate with no value-added post-processing.

GDL "BA" is our hydrophobized substrate with a 5 wt% PTFE loading.

GDL "BB" or "BC" is our hydrophobized substrate (5 wt% PTFE) and with our standard Microporous Layer (MPL) on one side.

Unique Characteristics

Our GDL 10 nonwovens are truly 3-dimensional in fiber orientation. They are more compressible and resilient when compared with typical 2-dimensional papers. On request, we will advise you on maximum compression loads for use in bonding to Catalyst-Coated Membranes (CCMs) or assembling stacks. Gasket design also needs to take the compression behavior of GDL into account.

The resilience of our GDL 10 is useful in maintaining uniform electrical contact by compensating for the dimensional changes of other components such as membranes that swell during stack operation.

As with other continuously manufactured webs, properties in the machine and transverse directions are different; the anisotropy factor is typically 4 to 6.

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This information is based on our present state of knowledge and is intended to provide general notes on our products and their uses. It should therefore not be construed as guaranteeing specific properties of the products described or their suitability for a particular application. Any existing industrial property rights must be observed. The quality of our products is guaranteed under our "General Conditions of Sale".

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Toray Carbon Fiber Paper "TGP-H"

Gas Diffusion Layer

Toray TGP-H is a Carbon-Carbon Composite Paper which has been used for PAFC and PEFC for decades and has proven durability. Its high mechanical strength, conductivity and gas permeability are suitable for use as a Gas Diffusion Layer (GDL) in fuel cell applications.

Characteristics

- High strength
- Excellent gas permeability and low electrical resistivity
- Good handling
- Minimal electrochemical corrosion

Structure

- TGP-H is made of PAN Carbon Fiber "TORAYCA" featuring high tensile strength and high modulus. Fibers are firmly connected by carbon.
- SEM (Scanning Electron Micrography) images on the right.

Surface

Cross section

Standard Sizes

- Thickness: TGP-H-030, 0.11mm
TGP-H-060, 0.19mm
TGP-H-090, 0.28mm
TGP-H-120, 0.37mm
Thicker materials available on request.
- Dimensions: 15"x15", 400mm x 400mm, 500mm x 500mm and 800mm x 800mm.
800mm x 800mm board for TGP-H-030 is not available.

Basic Data

Properties	Unit	TGP-H-030	TGP-H-060	TGP-H-090	TGP-H-120
Thickness	mm	0.11	0.19	0.28	0.37
Bulk density	g/cm ³	0.40	0.44	0.44	0.45
Porosity	%	80	78	78	78
Surface roughness Ra	μm	8	8	8	8
Gas permeability	ml·mm/(cm ² ·hr·mmAq)	2500	1900	1700	1500
Electrical resistivity					
through plane	mΩcm	80	80	80	80
in plane	mΩcm	—	5.8	5.6	4.7
Thermal conductivity					
through plane (room temp.)	W/ (m·k)	—	(1.7)	(1.7)	(1.7)
inplane (room temp.)	W/ (m·k)	—	21	21	21
inplane (100°C)	W/ (m·k)	—	23	23	23
Coefficient of thermal expansion					
in plane (25–100°C)	×10 ⁻⁶ /°C	-0.8	-0.8	-0.8	-0.8
Flexural strength	MPa	40	40	40	40
Flexural modulus	GPa	8	10	10	10
Tensile strength	N/cm	—	50	70	90

•The above data are experimental values and are not guaranteed.