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Effect of Cross-Flow on Liquid-Water Distribution: An In-Situ High-Resolution Neutron Radiography Study

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Abstract

Liquid-water management in polymer-electrolyte fuel cells (PEFCs) remains an area of ongoing research. To enhance water removal, certain flow-fields induce cross-flow, or flow through the gas-diffusion layer (GDL) via channel-to-channel pressure differences. While beneficial to water removal, cross-flow comes at the cost of higher pumping pressures and may lead to membrane dehydration and other deleterious issues. This paper examines the impact of cross-flow on component saturation levels as determined through in-plane high-resolution neutron radiography. Various humidities and operating conditions are examined, and the results demonstrate that cell saturation levels correlate strongly with the level of cross-flow rate, and lower GDL saturation levels are found to correlate with an increase in permeability at higher flow rates. Effective water removal is found to occur at channel-to-channel pressure gradients greater than the measured breakthrough pressure of the GDL, evidence that similar liquid-water transport mechanisms exist for under-land area transport as in transverse GDL flow.

Keywords: Fuel cells, Neutron Radiography, Permeability, Liquid-water saturation, Gas-diffusion layer, Cross-flow, Breakthrough Pressure

1. INTRODUCTION

Polymer-electrolyte fuel-cell (PEFC) systems have the potential to improve energy efficiency and storage capabilities for mobile and grid-level applications in the near future. In PEFCs, the electrode structure is composed of a catalytic layer supported by porous gas-diffusion layers (GDLs) where multiphase reactant/product transport and electrical conduction occur. Flow-fields, consisting of lands and channels, deliver and remove gases and water to and from the GDL. This produced liquid water can contribute to performance issues if not properly handled. Numerous studies have shown the importance of water-management strategies during start-up/shutdown and operation where lower cell temperatures or high current densities may lead to liquid build-up [1-13].

Mechanisms of liquid-water transport vary depending on flow-field design. In interdigitated and serpentine flow-field designs, pressure gradients between adjacent channels induce convective transport through the GDL, which is termed cross-flow [14, 15]. Benefits of cross-flow include increased reactant concentrations near the catalyst layer due to convective transport and reduced water content under lands. While cross-flow inducing flow-fields have been shown to produce higher performance compared to diffusion dominated parallel designs, higher pumping losses due to flow through GDL consume a larger percentage of net power [16], thereby limiting their use. PEFC humidification also has a large impact on cell performance where there is a balance between membrane dehumidification and catalyst-layer (CL)/GDL flooding [17]. Performance sensitivity to inlet stream humidification in interdigitated flow-field cells has been discussed in earlier work where higher humidity led to increased overpotential; however, detailed experimental analysis was not undertaken [18]. Understanding the behavior of water in the membrane and GDL regions at various cross-flow rates and inlet humidity will help define next generation water-management strategies.

Neutron radiography (NR) has been used successfully to quantify channel and membrane-electrode-assembly (MEA) liquid-water content [19]. Trabold and Owejan compared serpentine and interdigitated

flow-field performance using through-plane methods [20, 21]. While useful for overall cell characterization, through-plane imaging does not resolve the critical transverse MEA water content. In-plane high resolution ($< 25 \mu\text{m}$) techniques, in conjunction with specially designed differential test cells, can resolve such water profiles [22, 23]. Over the past several years in-plane NR has been used to characterize PEFC water-transport mechanisms related to phase-change-induced (PCI) flow, diffusion flow, humidity effects, pressure effects, surface hydrophobicity and microporous-layer (MPL) validation [24-29]. While these studies have added greatly to the understanding of liquid-water content in PEFCs, the effects of cross-flow on the liquid-water distribution has yet to be characterized. Such information is pertinent since many designs incorporate cross-flow inducing geometries.

Recently Taira and Liu analyzed in-situ effective permeability due to cross-flow in PEFC highlighting its sensitivity to humidity and land widths as well as characterizing non-Darcy effects [30], yet the water content remained unknown. This study builds on the previous works by imaging the MEA liquid-water behavior with different flow rates **under parallel and interdigitated configurations** in order to provide a more fundamental understanding of the relationship between liquid-water behavior and cross-flow. Herein, in-plane, high-resolution NR is used to measure trends crucial to understanding interactions between convective gas flow and liquid-water transport within the MEA in order to provide cell modelers and designers with valuable insight into the complex multi-phase physics.

2. EXPERIMENTAL PROCEDURE & SETUP

Cell – Neutron imaging was conducted at the National Institute of Standards and Technology (NIST) Center for Neutron Research with a high spatial resolution of $\sim 15 \mu\text{m}$. **A differential test cell was designed and constructed with 1 cm long channels and 1 mm channel width, height and land dimensions. A 2.25 cm^2 active area was achieved** to simulate a portion of a larger cell while being capable of in-plane radiography. Additionally, the cell can operate in either parallel or interdigitated flow-field configuration on the cathode via the manipulation of external manifold valves to allow for saturation measurements

under a zero cross-flow rate condition [16]; Figure 1 shows the cell highlighting key components. Due to the small feature size, the manifold housings were 3D printed using a high-temperature ABS plastic. Manifolds are 6.25 times the cross-sectional area of a single channel to reduce pressure drop and flow maldistribution. The bipolar plate flow-fields were machined out of 6061-T6 aluminum for neutron penetrability and plated with a thin multilayer containing nickel, gold and rhodium for corrosion protection. A series of Teflon gaskets were used to seal the manifold, bipolar plate, and MEA interfaces. Guide pins were included between each section to ensure proper alignment of channels and manifold holes. The anode electrode was operated under parallel configuration for all testing. The NIST test station was used to control cell and gas temperatures, relative humidity (RH), gas flow-rate, and current density. Cell temperature was maintained using 1/8" diameter cartridge heaters and t-type thermocouples installed directly into both the anode and cathode bipolar plates from the sides. Manifolds and Swagelok components were insulated in order to eliminate gradients. The MEA consisted of SGL 10 BC on the cathode and SGL 35 BC GDL on the anode (~420 μm and ~325 μm uncompressed thickness respectively), each with 5% PTFE wt.-loading and microporous layer (MPL), hot pressed to a Gore SELECT membrane (~55 μm thick). Compression was set to approximately 10% of total uncompressed GDL thickness, on the cathode a thickness of ~380 μm was measured under the lands via the radiographs.

Test Parameters – All data was acquired at 70°C cell and gas temperatures, and an overall test matrix is shown in Table 1. The high current density (HCD) 0.600 A cm⁻² and the low current density (LCD) 0.150 A cm⁻² settings were held constant while cathode inlet flow rates were stepped ranging from 100 to 500 sccm. The HCD setting is closer to a PEFC current density at maximum power density rather than a limiting current density (LCD) which can be well over 1.0 A cm⁻² [16]. This resulted in air mass fluxes,

$\dot{q}$, through each land GDL area of 0, 0.45, 0.82, 1.18, 1.55, 1.90, and 2.27 g cm⁻²-s⁻¹. The anode H₂ flow rate was maintained at 900 sccm for all tests. The cell was purged thoroughly and operated for 15 minutes before every test run. Each case consisted of 15 min of data collection; three images (300 sec

exposure time each) were taken during this period. For the zero cross-flow rate images the cathode was operated in parallel formation. In this mode, the channel gas velocity was set to match the average channel velocity in the $0.45 \text{ g cm}^{-2}\text{-s}^{-1}$ interdigitated case to make results comparable. Cathode inlet and outlet pressures were measured directly at manifold *Inlet A* and *Outlet B* (see Figure 1), respectively, for each flow condition at a sample rate of 10 Hz. In the cathode, high and low inlet RH conditions were tested, respectively (80% RH (HRH) and 50% RH (LRH)); the anode inlet RH was maintained at 80%. Cell alignment along the beam path (azimuthal) was verified to 1/10 degree accuracy to eliminate boundary effects in the images; further, the NIST beam is collimated to minimize spreading.

Image Post-Processing – The radiographs acquired at NIST were processed using an in-house MATLAB code that implemented the following analysis. Neutron attenuation follows the Lambert-Beer Law therefore the raw radiograph optical density (OD) can be related to water thickness [31]

$$OD = \ln \left(\frac{I_{wet} - I_{dark}}{I_{dry} - I_{dark}} \right) = \sigma_1 t + \sigma_2 t^2 \quad (1)$$

where I_{wet} and I_{dry} are wet and dry reference images and I_{dark} is the dark field image used to account for background radiation. The constants σ_1 and σ_2 are obtained from calibrating to a known set of water thicknesses and were determined to be $0.374 \pm 0.005 \text{ mm}^{-1}$ & $-0.0086 \pm 0.0004 \text{ mm}^{-2}$ respectively. To convert OD to water thickness, t_{H_2O} , the following equation was used:

$$t_{H_2O} = -\sqrt{\frac{OD}{\sigma_2} + \frac{\sigma_1^2}{4\sigma_2^2}} - \frac{\sigma_1}{2\sigma_2} \quad (2)$$

Liquid water volume, V_{H_2O} , is then equal to $t_{H_2O} \cdot A_P$ where A_P is the representative spatial area of a single pixel, in this case approximately 0.0025 mm². Examples of processed radiographs are displayed in Figure 2, each image is the average of the three images captured during each case.

3. RESULTS & DISCUSSION

3.1 Liquid-Water Distribution

Liquid-water thickness across the MEA is plotted in Figure 3 for the 0, 0.82, 1.55 & 2.27 g cm⁻²-s⁻¹ air mass-flux cases. As reported in previous literature, the highest water content occurs near the membrane/cathode CL interface since this is where the oxygen reduction reaction predominately occurs [22, 29, 32, 33]. The twin peaks at both the anode and cathode GDL/flow-field interfaces are present in many prior NR studies' data and have been attributed to water vapor condensation. The cathode water decreases with increased cross-flow rate due to a larger pressure differential within the GDL and also, to a lesser degree at these flow rates, convective forces. Additionally, since the RH is less than 100%, a higher rate of water vapor is removed by the air stream. Varying cathode conditions impacted the anode water content as the drier cathode conditions can induce water migration from anode to cathode. In the anode GDL region, the water content is higher than in the cathode, except for the HCD-HRH parallel tests (Figure 3a), due to lack of cross-flow. Both HCD-HRH and HCD-LRH parallel configuration cases also have markedly higher residual water amounts closer to the cathode CL, evidence that cross-flow affects pore bound water throughout the thickness of the GDL.

3.1.1 Humidification and Current Density Effects

In addition to reduced GDL water, membrane humidification was observed to decrease at high flow rates (Figure 3). Membrane drying may be prevented by running at a higher inlet RH as both the HRH cases

showed significantly greater membrane and CL water content at increased cross-flow rates compared to the LRH cases, while still showing reduced GDL saturation. Cases with increased water presence from reaction and elevated inlet humidity (HCD and HRH) showed higher sensitivity to changes in cross-flow. Additionally, as anode humidity was held constant, the higher liquid-water content present in the cathode caused a steeper water-concentration profile, reducing the amount of water transferred from anode to cathode, resulting in more water present in the anode and raising the entire curve. Further, there is overall less liquid present in the LCD cases as expected since there is a lower rate of product water formation. The anode side showed higher sensitivity to reaction rate as the LCD cases exhibited less liquid water compared to the HCD cases, potentially due to a reduction in water transport from cathode to anode CL. Cell designers should be aware that cross-flow, while effective at removing water from the GDL, may impact performance by dehydrating the membrane. Maintaining proper hydration may require additional humidification of reactant gasses, an additional energy input to the PEFC system above and beyond the extra pumping losses.

3.1.2 GDL Land vs. Channel Water

Cathode data was separated into channel and land GDL water profiles, depicted in Figure 4. Prior to the peak in land GDL signal, the water content falls lower than the channel GDL water content. This is probably due to the increased presence of cross-flow beneath the lands. The rise in signal at the GDL/flow-field interface in both data sets may be due to water buildup at the land walls and on the GDL surface (e.g., PCI flow). Similar behavior was observed in previous studies that altered the wettability of channel coatings which attributed similar profiles to hydrophilic conditions, as the spikes were not present under hydrophobic wall conditions [26]. The contact angle of liquid water on the rhodium was measured at approximately 35° , highly hydrophilic, which would lead to some of the observed increased water signal. Cross-flow inducing flow-fields may improve water removal in hydrophilic surface conditioned flow-fields as GDL water content decreased significantly compared to the parallel case. Though results show that land and channel profiles converge in the membrane and CL region despite differences in GDL

water content between land and channel based positions, our resolution may not be high enough to draw definitive conclusions.

Overall water reduction was calculated by normalizing the total GDL water volume, V , at each air mass flux to the zero cross-flow rate. These trends are plotted in Figure 5 for the two complete data sets. There is a diminishing return on cross-flow rate as flux exceeding approximately $1.50 \text{ g cm}^{-2} \text{ s}^{-1}$ did not significantly reduce water in the HRH cases, while lower cross-flow rates led to higher water content. For the PEFC, there may exist an optimal cross-flow rate, where the diminishing returns of increased water removal are exceeded by the associated losses due to parasitic pumping power. These results do not take into account cross-flow's effect on the electrochemical reaction and so comments reflect only water removal capacity and not net performance effects.

3.2 Effective Permeability Trends

Gas flow through the GDL at lower velocities is governed by Darcy's law,

$$-\nabla p = \frac{\mu}{k_{eff}} v \quad (3)$$

where p is the pressure, μ is the viscosity, k_{eff} is the effective permeability and v is the velocity. To account for the geometry of the flow-field, the Darcy expression can be integrated across a

land width, L , from the inlet pressure (p_i) to the outlet pressure (p_{out}), yielding [30]

$$p_i^2 - p_{out}^2 = \frac{2RTL}{M_w} \frac{\mu}{k_{eff}} (q) \quad (4)$$

where R is the ideal gas constant, T is the temperature, q is the mass flux, and M_w is the

molar weight of the mixture equivalent to $M_w = \sum_{i=1}^N M_i x_i$, where M_i and x_i are the molar

weight and ratio of each species respectively. Viscosity, μ , for the humid cases is approximated by a semi-empirical expression for mixtures [34]:

$$\mu = \frac{\sum_{i=1}^N \frac{x_i \mu_i}{\sum_{j=1}^N x_j \Phi_{ij}} \quad (5)$$

where Φ_{ij} is a dimensionless number determined by

$$\Phi_{ij} = \frac{1}{\sqrt{8}} \left(1 + \frac{M_i}{M_j} \right)^{-1/2} \left[1 + \left(\frac{\mu_i}{\mu_j} \right)^{1/2} \left(\frac{M_i}{M_j} \right)^{1/4} \right]^2 \quad (6)$$

Plots of cathode pressure differential and permeability using these equations and test results are presented

in Figure 6(a) and (b). **While not depicted**, linear trends were observed for the $p_{\text{c}}^2 - p_{\text{out}}^2$ term in Eq. 4

with respect to q which validate the assumption that flow rates were low enough to neglect any inertial term and assume a Darcy relationship. Permeability values were measured to be in the realm of, albeit lower due to in-situ testing, those found in literature which report around $\sim 2 \times 10^{-11} \text{ m}^2$ [35].

3.2.1 GDL Saturation

Permeability is sensitive to GDL saturation as increasing liquid-water content reduces volume available to gas flow. Just as liquid-water content is reduced with increased cross-flow rate in Figure 6(c), permeability is seen to increase as less water resides in the GDL. GDL liquid-water saturation, s , ranges from 0 to 1 and can be calculated by

$$s = \frac{V}{\phi V_{GDL}} \quad (7)$$

where V_{GDL} is the total volume occupied by the GDL and ϕ is the porosity of the GDL (~82%).

While overall permeability does not change significantly, there is some increase at higher flow rates ($>1.50 \text{ g cm}^{-2} \text{ s}^{-1}$) where water removal is enhanced.

3.3 Liquid-Water Transport Mechanisms

To understand the nature of liquid-water transport via cross-flow between channels, transverse liquid water/GDL breakthrough pressure was measured ex-situ as described previously [36] using SGL 10BA GDL (5 wt-% PTFE), which is simply an SGL 10BC GDL lacking an MPL. Breakthrough pressure is a measure of the maximum capillary pressure that must be overcome in order for liquid water to flow through a GDL; it is often used to characterize a GDL's resistance to water removal [37]. Figure 7(a) displays the time-series data of liquid-water pressure as it is forced through the sample GDL. Flow does not occur until the maximum is overcome, which is designated the breakthrough pressure. It is interesting that at cross-flow rates where water is most effectively removed from the GDL, the pressure difference between inlet and outlet channels (Fig. 7 (b)) is slightly greater than the measured breakthrough pressure ($\approx 1.70 \text{ kPa}$). Liquid water removed from the land regions must traverse GDL pores between the inlet and outlet channels, which can only occur if the convective and pressure forces acting on liquid-filled pores overcomes the capillary forces. Since the inertial term was found to be negligible for this range of cross-

flow (Section 3.2), the effect of convective shear forces on internal GDL liquid water is probably minimal. Therefore, the pressure is the dominate mechanism of liquid-water transfer from inlet to outlet channels. This may explain the diminishing return on cross-flow rate; once breakthrough pressure is overcome liquid transport can occur and the majority of water is effectively removed via outlet channels.

5. CONCLUSIONS

High-resolution in-plane neutron radiography has been used to investigate the effect of cross-flow on liquid-water saturation in an operating PEFC. By measuring liquid-water content at various cross-flow rates and cell conditions, a further understanding of complex multiphase interactions is gained. The key findings in this study are the following:

Cross-Flow Rate: Increasing levels of cross-flow rate through the cathode GDL reduced MEA water substantially. Membrane hydration was found to decrease at elevated cross-flow rates which should be taken into consideration to avoid dehydration. Slightly less water was observed under lands than under channels due to flow occurring from inlet to outlet channels; however, image noise from plating attenuation makes details hard to verify. If hydrophilic surface conditions are an aspect of the design of a PEFC, cross-flow inducing flow-fields may improve water-removal capability.

Humidity & Current Density: Increased relative humidity (RH) caused elevated water content in the membrane and cathode CL. Controlling RH may be an effective method to preventing membrane dehydration at higher levels of cross-flow rate since the high relative-humidity cases had more water than the low relative-humidity ones. The current density appeared to have a larger impact on anode water content than humidity, possibly due to electro-osmosis and water back transport.

Effective Permeability & Saturation: As cross-flow increased, permeability trended upward and saturation decreased. Mechanisms of water removal via cross-flow may be related to breakthrough pressure since

pressure differences between inlet and outlet channels where water removal was maximal were slightly higher than the measured breakthrough pressure for SGL 10 BA.

For designers and modelers of PEFCs in which cross-flow exists, these results will be valuable in understanding the nature of liquid-water distribution throughout a PEFC. This will further improve water-management strategies via materials selection and flow-field design, as well as humidification and flow-based techniques. Future work incorporating cell performance is currently being pursued to link observed trends with electrochemical behavior.

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TABLES & FIGURES

Table 1: Testing matrix for the interdigitated configuration cathode conditions (SGL 10 BC)

Test #	Label	Current Density [A cm ⁻²]	RH [%]
1	HCD-HRH	0.600	80
2	HCD-LRH	0.600	50
3	LCD-HRH	0.150	80
4	LCD-LRH	0.150	50

Figure 1: Overview of differential test cell. Cathode manifold valves can be used to switch between parallel and interdigitated flow (shown in interdigitated configuration). **Open valves result in parallel flow. Closed valves result in interdigitated flow where higher pressure inlet gas, P_H (red), must travel through the GDL to reach lower pressure outlet channels, P_L , (blue).**

Figure 2: Example in-plane radiographs of the flow-field channels for two humidity conditions at the same mass flux showing liquid-water distribution. Land regions are masked with black.

Figure 3: Water-thickness data across the MEA thickness. The *Zero Cross Flow* condition refers to the parallel configuration test.

Figure 4: Cathode water thickness separated into channel GDL and land GDL regions. The *Zero Cross Flow* condition refers to the parallel configuration test.

Figure 5: Total cathode GDL water volume normalized to the zero cross-flow (parallel) case as a function of air mass flux through a single land-GDL area.

Figure 6: (a) Pressure difference between cathode inlet and outlet channels, (b) Effective permeability across lands, (c) GDL saturation. All are a function of the air mass flux through a single land-GDL area.

Figure 7: (a) Breakthrough pressure measured ex-situ through a sample of SGL 10BA GDL (5% PTFE wt., $\sim 390 \mu\text{m}$ thick). Liquid water was injected through a $310 \mu\text{m}$ diameter hole at $0.05 \mu\text{L s}^{-1}$. Method details are available in the following reference [36]. (b) Cathode GDL saturation as a function of channel-to-channel pressure gradient, vertical dotted line designates measured breakthrough pressure.

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