ABSTRACT

An important function of the gas delivery channels in Proton Exchange Membrane (PEM) fuel cells is the evacuation of liquid water created at the cathode. The resulting two-phase flow can become an obstacle to reactant transport and a source of parasitic losses. The present work examines the behavior of two-phase flow in 500 µm x 500 µm x 60 cm channels with distributed water injection through a porous carbon paper wall to gain understanding of the physics of flows relevant to fuel cell water management challenges. Flow regime maps based on local gas and liquid flow rates are constructed for experimental conditions corresponding to current densities between 0.5 and 1 A/cm² and stoichiometric coefficients from 1 to 4. Flow structures are analyzed along the entire length of the channel. It is observed that slug flow is favored to plug flow at high air flow rates and low liquid flow rates. Stratified flow dominates at high air flow rates and low liquid flow rates. Stratified flow dominates at high air flow rates and low liquid flow rates. Stratified flow dominates at high air flow rates and low liquid flow rates. Along the axial flow direction, the flow regime consistently transitions from intermittent to wavy to stable stratified flow. This progression is quantified using a parameter of flow progression which characterizes the degree of development of the two-phase flow toward the stable stratified condition. This parameter is discussed in relation to fuel cell operating conditions. It provides a metric for analyzing liquid water removal mechanisms in the cathode channels of PEM fuel cells.

INTRODUCTION

The management of water in a proton exchange membrane (PEM) fuel cell is a performance-limiting concern in these carbon-free energy-conversion devices. The performance of the membrane depends on ample humidification. However, liquid water in the microporous layer, gas diffusion layer (GDL), and/or gas delivery channels impedes the flow of reactant gases to catalyst sites and cripples cathodic reaction rates. Two-phase flow phenomena have been identified as concerns for flooding-induced performance degradation, cell voltage hysteresis, increased parasitic pumping losses, and the absence of predicted performance improvements [1-5]. While microchannels are proposed for use in PEM fuel cells with advantages of increased convective transport, flexibility of geometric design, and decreased resistive path lengths, these benefits have not been observed in practical systems [4]. One explanation for the unrealized performance improvements is an increased propensity toward flooding in smaller channels, where surface tension becomes significant compared with other fluidic forces. The present work provides fundamental insight into the physics of two-phase flow in geometries common to practical fuel cells which is critical to their performance.

Some two-phase flow characterization has been developed for in-situ fuel cell applications. In fuel cell components lacking optical access, neutron imaging, magnetic resonance imaging, and X-ray tomography allow for identification of liquid water through opaque surfaces. Neutron imaging is a promising technique for real-time imaging of liquid water in an operating fuel cell [8-10]. This technique is limited to two dimensions; delicate analysis is necessary to distinguish flow structures between the anode and cathode. X-ray tomography and
magnetic resonance imaging have been used to create three dimensional maps of water concentration in the membrane and GDL layers with limited resolution [11, 12].

Opaque bipolar plates, layers of porous materials with various surface properties, and non-uniformity of water production rate and location are obstacles to the analysis of flow regimes in fuel cell channels. Fuel cells with modified geometries allow optical access into the gas distribution channels and have been used to correlate the appearance of liquid water with increasing in gas pressure in the channels and voltage drops in the fuel cell [13-19]. These fuel cells may demonstrate atypical reaction distribution because the electrical pathways are distorted when conductive bipolar plates are replaced with non-conductive glass plates for optical access. Much insight can be drawn from these studies in the correlation between liquid water and fuel cell performance; however the precise flow conditions at any particular location in the cell are difficult to quantify due to non-uniformities in reaction distribution and subsequent uncertainties in local flow rates.

Absent the electrochemistry of an operating fuel cell, ex-situ visualization experiments can quantify the local flow rates of both phases. Experiments with a porous GDL wall have been used to simulate the GDL-channel interaction of a fuel cell for mini-channels. Single-phase gas flow profiles have been examined in small channel sections using particle image velocimetry in order to characterize the interaction of the channel and porous layer and the flow around corners [20-22]. Su et al. studied flow in 5 mm square cathode channels by injecting water into the channels through a carbon paper layer; the large channel dimensions were chosen to reduce the effect of surface tension on the resulting flow [23]. Others have examined droplet departure from a GDL into a gas channel using short channel segments [24, 25]. Two-phase flow patterns are characterized in microchannels for lab-on-a-chip and microchannel cooling applications [26-29], however these studies do not replicate the distributed liquid water introduction, long channel lengths, and porous boundary conditions of a PEM fuel cell.

This study bridges the gap between in-situ experiments on operating fuel cells and ex-situ two-phase flow experiments in different geometries. Metered amounts of liquid water are introduced into the channel through the porous carbon paper gas diffusion layer which flanks one channel wall, thereby allowing for the characterization of flow regime transitions according to known flow rates of both phases. Unlike visualization of operating fuel cells, this setup allows for precise control of the liquid and gas flow rates and injection conditions. Compared with other ex-situ experiments, this study incorporates channels with smaller cross-sectional dimensions than many previous fuel cell related studies. Compared with fundamental two-phase flow studies in microchannels, this study better replicates the characteristics of fuel cell channels, including longer channel lengths and distributed water introduction. This study provides insight to key two-phase flow structure transitions of importance for performance-critical water management in PEMFC channels.

**NOMENCLATURE**

\[
\begin{align*}
i & : \text{current density} \\
Q & : \text{volumetric flow rate} \\
U & : \text{superficial velocity} \\
\lambda & : \text{stoichiometric ratio} \\
\text{Subscripts} \\
\lambda & : \text{air (gas phase)} \\
w & : \text{water (liquid phase)}
\end{align*}
\]

**EXPERIMENTAL METHOD**

A single serpentine channel replicates flow conditions associated with fuel cell gas delivery channels. A channel with a 500 x 500 µm square cross-section is machined in acrylic with a 60-cm long serpentine layout incorporating five long segments and four short turns. The fourth wall of the channel is comprised of a 2 mm wide, 190 µm thick ribbon of porous carbon paper GDL which follows the channel geometry and fits tightly into a latex gasket seal. The GDL paper is pressed against the channel structure with an acrylic slab using 44 bolts for even distribution of pressure. Water is injected into the channel at eight locations using a multisyringe pump (Harvard Instruments) via 500 µm diameter holes through the acrylic slab. Figure 1 is a schematic of the structure and its cross-section.

This study bridges the gap between in-situ experiments on operating fuel cells and ex-situ two-phase flow experiments in different geometries. Metered amounts of liquid water are introduced into the channel through the porous carbon paper gas diffusion layer which flanks one channel wall, thereby allowing for the characterization of flow regime transitions according to known flow rates of both phases. Unlike visualization of operating fuel cells, this setup allows for precise control of the liquid and gas flow rates and injection conditions. Compared with other ex-situ experiments, this study incorporates channels with smaller cross-sectional dimensions than many previous fuel cell related studies. Compared with fundamental two-phase flow studies in microchannels, this study better replicates the characteristics of fuel cell channels, including longer channel lengths and distributed water introduction. This study provides insight to key two-phase flow structure transitions of importance for performance-critical water management in PEMFC channels.

**Figure 1.** Cross-section of channel assembly.

Various injection configurations were considered before discrete liquid injection was chosen. Other injection...
geometries incorporating reservoirs or channels behind the GDL wall suffer from the preferential flow of air through the GDL pores and small channels. Liquid water cannot be made to completely fill injection reservoirs when a pressure gradient is applied across the device; air is unavoidably entrained into these cavities.

Toray GDL papers (TGPH-090) with and without 10 wt% Teflon impregnation are utilized in order to examine the effect of GDL hydrophobicity on the flow observed in the channels. Similar tests are performed on both samples.

Two-phase flow conditions simulate the operating conditions of a PEM fuel cell. Figure 2 shows the experimental setup delivering air and water to the channel structure. Upstream air pressure is held constant at a gage pressure of 1 atm. Downstream pressure is regulated with a needle valve at the exit in order to establish the desired flow conditions. Inlet and differential pressures are measured using diaphragm differential pressure sensors (Validyne). Air flow rate is measured with flow meters located in-line upstream of the test section (Sensirion thermal mass flow meter and Omega laminar-passage air flow meter). Liquid injection is metered with a syringe pump (Harvard PHD 2200) fitted with a multiple syringe rack holding eight identical glass syringes (Hamilton Gastight 1 mL).

Experimental conditions correspond to reasonable operating conditions for PEM fuel cells as enumerated in Table 1. The overall cell current determines the liquid flow rate, while the stoichiometric ratio determines the ratio of air to liquid flow rates. In these experiments, water is injected at rates equivalent to current densities, $i$, between 0.5 and 2 A/cm$^2$. Air flow rates are calculated based on equivalent stoichiometric ratios, $\lambda$, from 1 to 4. Fuel cell equivalents are calculated based on an effective land area equal to the channel area. An osmotic drag coefficient of 0.5 molecules of water per proton is assumed and considered in the calculation of the equivalent current density for a given water flow rate. These conditions represent typical operating conditions for a PEM fuel cell.

![Figure 2. Schematic of Experimental Setup.](image)

![Figure 3. Image decomposition. The entire channel length is captured in a series of eight overlapping regions as illustrated on the top photo. Each region includes three channel segments, resulting in 20 channel segments for flow regime observation. The bottom image is one of these channel segments.](image)

<table>
<thead>
<tr>
<th>$i$ [A/cm$^2$]</th>
<th>0.5</th>
<th>1</th>
<th>2</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Q_{water}$ [µl/min]</td>
<td>33.6</td>
<td>67.3</td>
<td>134.6</td>
</tr>
<tr>
<td>$Q_{water per injection}$</td>
<td>4.2</td>
<td>8.4</td>
<td>16.8</td>
</tr>
<tr>
<td>stoichiometric coefficient, $\lambda$</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>0.088</td>
<td>0.176</td>
<td>0.352</td>
</tr>
<tr>
<td>2</td>
<td>0.176</td>
<td>0.352</td>
<td>0.704</td>
</tr>
<tr>
<td>3</td>
<td>0.264</td>
<td>0.528</td>
<td>--</td>
</tr>
<tr>
<td>4</td>
<td>0.352</td>
<td>0.704</td>
<td>--</td>
</tr>
<tr>
<td>$Q_{air}$ [SLPM]</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Between experiments, dry air flows through the channel for a minimum of 30 minutes to dry the GDL of accumulated water from the previous test in order to provide consistent starting conditions. The air flow condition is established by setting the downstream needle valve before water is introduced into the channel. Then liquid water injection begins. Pressure and air flow rates are recorded while images of the channel are captured using a 1392 x 1040 pixel CCD camera (Nikon Coolsnap ES). Sequential images are collected to create movies of the flow in the channel. As illustrated in Figure 3, that the entire channel is visualized in eight images. Analysis is performed on twenty channel sections (4 per serpentine segment) extracted from these images.
RESULTS AND DISCUSSION

Flow observation

Two phase flow in the channel is observed at 20 locations along its length for each flow condition. As expected, the structure of the liquid water depends upon the properties of the channel wall, the amount of water in the channel, the air flow rate, and to some extent, the distribution of the water injection. While there are general trends in the progression of flow regimes and the conditions under which they are observed, a strong stochastic element exists due to the complicated fluidic interactions in the serpentine geometry with a textured, porous wall with non-uniform surface properties.

In general, more water is seen in the channels of the structures constructed with GDL containing Teflon. This difference may be the results of differences in flow structure and liquid water velocity due to differences in the surface tension at the liquid solid interface may or this observation may indicate that more liquid water flows through the GDL with no Teflon impregnation so that less water is forced into the channels. There was no significant difference in the pressure vs. flow rate curves for the Teflon and no Teflon cases. Water introduction caused a very slight decrease in flow rate for a given pressure drop compared to a dry channel for both samples.

structures occurring under relatively higher and lower air flow conditions. Black arrows between images represent common transitions between flow regimes as more liquid is encountered in the channel. The dry channel transitions to intermittent flow where water moves through the channel in discrete packets, droplets, slugs, or plugs. Intermittent flow becomes wavy flow when these packets of water travel down the channel as an abrupt thickening of a liquid film on the side walls of the channel. Finally, a stratified flow ensues when the sidewall film becomes steady and no significant oscillations are observed at the interface. Stratified flow exists most commonly at the furthest downstream locations, while slug/plug flow is rarely observed downstream of the first several injection locations.

This trend suggests that stratified flow is hydrodynamically preferred to intermittent flow regimes, provided there is enough liquid flowing in the channel to maintain such a film. When a slug or plug travels the three-phase interface line moves down the channel, so viscous, inertial, and pressure forces must continuously overcome surface tension forces at the triple point line. Once the liquid forms a film along a channel wall, this force is eliminated in the flow direction and the gas-liquid surface area is minimized. The water is propelled by the pressure gradient in the channel and shear forces from the air.

The flow conditions chosen for this study correspond to typical operating conditions for a PEM fuel cell. It is expected that a similar progression of flow regimes occurs in operating fuel cells with equivalent surfaces. Based on these observations, the precise location of flow regime transitions in an operating fuel cell will depend strongly on the geometry and layout of the channels. For example, flooding of the channel is not observed under any flow conditions, likely due to the single channel and sealed GDL configuration which does not exhibit the type of dead spots which can occur at corners or between parallel channels in standard fuel cells. The single channel configuration maintains pressure gradients and flow paths for the entire channel length.

Flow Regime Definition

For the purpose of this analysis, we describe the observed flow regimes with three labels: plug, slug, and stratified flow. Figure 5 shows representative images of each flow regime. A main distinction between plug and slug flow regimes and stratified flow regimes is the steadiness of the interfaces. Plug and slug flow show transient variation of the air-liquid-solid interfaces. Stratified flow is steady; the position of the interface does not vary with time for given operating conditions. Wavy flow has a constant liquid-solid interface at the channel wall, but a varying liquid-gas interface.

Plug Flow – Liquid droplets fill the channel, touching both channel walls, sometimes resting on the GDL. This flow regime is very unstable in these experiments. Plug flow quickly dissolves into slug or stratified flow.
Slug Flow – Water droplets touch only two or three of the channel walls, allowing a passage for air flow around the slug.

Stratified Flow – A steady liquid film flows on one or both side walls of the channel. The GDL is visibly dry in the center of the channel. The thickness of these films varies dramatically. The same image could also represent a corner flow at an intersection of two channel walls.

Variation of flow structures with flow rate
Most two-phase flow studies organize the flow regimes onto regime maps according superficial gas and liquid velocities. This parameterizes the fluidic conditions which yield particular flow structures and transitions. In this study, the distributed water introduction along the length of the channel requires that the flow regime map consider the local flow rate of liquid after each injection location. Consequently, each experimental condition provides information at eight vertically-aligned locations in flow rate space. Furthermore, in this complicated geometry, flow structures often change spatially between injection points, such that a single experimental condition may result in multiple flow regimes for a particular air and local water flow rate, depicted as overlapping points on the regime map. The choice of experimental conditions also creates overlap of data points from different channel locations experiencing identical flow rates. Despite these complications, flow regime maps provide insight into the range of flow regimes which may occur under given operating conditions. Figures 6 and 7 plot observed flow regimes against local air and water flow rates. The local water flow rate at a given position is the product of the number of liquid injection points upstream from the imaging location and the rate of water introduction per injection point. The volumetric air flow rate is determined from the inlet mass flow rate and pressure.

Figure 6. Flow regimes as a function of water and air flow rates and superficial velocities for channels with 10 wt% Teflon GDL. The results of ten experimental conditions are compiled here. The multicolored lines of constant Qwater/Qair ratio show the groups used for analysis of progression toward stratified flow which is discussed in the following subsection.

Figure 7. Flow regimes as a function of water and air flow rates and superficial velocities for channels with 0 wt% Teflon GDL. The results of ten experimental conditions are compiled. The stratified flows which occur at low water flow rates are very thin.

Figure 6 shows that for the samples containing Teflon, intermittent (slug and plug) flow regimes occur for low water injection rates. This corresponds to the first several injection points along a channel. Plug flow occurs at low air flow rates,
but slugs exist at the same flow parameters, because these plugs are quickly broken into slugs as they travel down the channel. At higher air flow rates, low liquid flow rates yield slug flow. For all air flow rates, stratified flow is the dominant flow regime at high water flow rates.

In Figure 7, the samples without Teflon also exhibit stratified flow at the highest liquid flow rates as well as at the highest air flow rates. Intermittent flows are observed at moderate liquid flow rates; plug flow occurs at only under the lowest air flow and slugs occur at moderate air flow. At the lowest water flow rates, stratified flow is indicated however the films at these low water flow rates are very thin. These could likely be labeled as another category, such as dry or corner flows. Considering this, the map in Figure 7 is very similar to that of Figure 6, only shifted to a higher liquid flow rate. This similarity supports the observation that more liquid water appears in the channels with Teflon coating. If liquid water preferentially flows through the GDL in the more hydrophilic case, this would effectively shift the regime map for flow occurring in the channel exclusively.

*Flow regime progression*

Based on the observation that the liquid structure evolves along the length of a channel in a relatively consistent progression from dry to intermittent to wavy to stratified flow, a *parameter of flow progression* is defined to quantify an average development of flow in the channel toward a steady stratified regime. Each flow regime is arbitrarily assigned a value according to its degree of development toward stratification. A dry channel is assigned a value of 0, a stratified flow regime is given a value of 1. Intermediate flow regimes, including intermittent and wavy flows, are worth \( \frac{1}{2} \). The average of these values for a particular flow condition constitutes its parameter of progression for that condition. Using this scale, this parameter also serves as a rough measure of the most likely flow regime present under given flow conditions.

The average can be defined over any flow condition of interest. In this case, the parameter of progression is averaged over the ratio of flow rates of liquid and gas flow. This average is chosen because it is a well-defined parameter at any given location in the channel, but evolves along the channel length. It corresponds to a diagonal line on a flow regime map, as shown in Figure 6. It also seems to have some physical relevance to the transition between slug and stratified flow, based on this figure. The choice of flow rate ratio as an averaging parameter for the 0 wt% Teflon channel is less evident based on the location of flow regime boundaries on that map. Another parameter may be better suited to this sample.

In terms of fuel cell operating conditions, the ratio of water production along an entire channel to air flow rate directly relates to the stoichiometric ratio of the air flow. When considering distributed liquid introduction, the ratio of liquid to gas flow rate increases along the channel for a given stoichiometric ratio. In order to capture this subtlety in the analysis of flow progression, averages are taken for similar values of \( Q_{lw}/Q_a \) only from tests at the same stoichiometric ratio. Each point in a stoichiometric series represents a position further down the channel, after an additional water injection location. The results of this analysis are presented in Figures 8 and 9.

**Figure 8.** Parameter of Progression for flow in samples with 10 wt% Teflon GDL.

**Figure 9.** Parameter of Progression for flow in samples with non-teflonated GDL.
lingers below a value of 1 for the sample with no Teflon in the GDL.

For the teflonated GDL, the lowest stoichiometry exhibits the slowest transition to stratified flow with $Q_w/Q_g$. The higher the stoichiometric ratio, the faster the onset of stratified flow with increasing water/air ratio. This hints that the air flow rate plays an important role in creating a liquid film on the channel wall.

For the non-teflonated GDL, the trends are less clear. The three lower stoichiometries approach stratified flow conditions quickly, while the highest stoichiometry remains in an intermediate regime, even at the highest flow rate ratio. This condition occurs because very thin films are prevalent at low liquid flow rates in the non-teflonated samples. This method assigns a value of zero to these thin-film flows which keeps the average value low. As previously stated, the ratio of water to air flow rates may not be the appropriate averaging parameter to describe the probable flow regime for these samples.

Modifications to this technique including further subdivision of flow regime designations and analysis of other appropriate flow conditions for averaging could improve this metric for flow characterization and regime prediction. This could be interesting in the development of alternative coordinates for mapping two-phase flow based on relevant physical parameters.

**Flow Transitions**

This study reveals a variety of transitions between flow structures as they advect down the channel. A parametric analysis of some of these flow transitions can provide fundamental understanding into the physics of interaction between these flows.

![Image of stratified flow transition from films on two channels walls to a film on one channel wall by means of a liquid bridge occurring downstream of a water injection point.](figure10)

Stratified flows – One of the most prominent and consistent transitions is a change in structure of stratified flow. Stratified flow regimes tend to progress consistently from occuring on two channel walls to occurring on a single wall. Several types of conditions serve as catalyst for this transition, including the presence of a stationary liquid bridge resting on the GDL surface, a water injection point which also wets the GDL, or a corner. Figure 10 is an image of one such transition point, a stationary liquid bridge. These liquid bridges may form where local variations in the surface properties of the GDL favor the accumulation of water.

![Analytical solutions to the momentum equations for simplified stratified flow in a rectangular channel, as shown in Figure 11. provide insight into a hydrodynamic advantage that single-wall stratified flow has over two-walled stratified flow.](figure11)

Analytical solutions to the momentum equations for simplified stratified flow in a rectangular channel, as shown in Figure 11, provide insight into a hydrodynamic advantage that single-wall stratified flow has over two-walled stratified flow. The pressure gradient within a channel of a particular dimension is determined by the ratio of flow rates of air and liquid phases (which is proportional to the stoichiometric ratio) and scales with the absolute magnitude of the flow rate (proportional to current density). After non-dimensionalization, the ratio of pressure gradient to volume flow rate of air is a function of the ratio of water to air volume flow rate (equivalent to fuel cell stoichiometry) and aspect ratio of the channel. The pressure gradient necessary to sustain equivalent flow rates of both gas and liquid in identical square channels is between 4 and 15% lower for stratified flow on a single-wall compared with stratified flow on two walls. Additionally, a single wall stratified flow maintains half the interface area between phases, so surface energy is also minimized.

Similar analyses of other flow transitions will provide valuable insight into physics governing the two-phase flow in these microchannels.

**CONCLUSION**

An experimental system is developed to study two-phase flow in fuel cell microchannels by ex-situ visualization under known liquid and gas flow rates. Flow structures relevant to PEM fuel cells are identified and labeled in order to construct regime maps and calculate a parameter of progression. These tools provide insight into the flow conditions which govern the appearance of particular flow regimes. In general, a progression from intermittent to stable stratified flow is observed as more liquid is introduced along the channel length. For flow conditions with higher air flow rates, the stratified film forms quickly and liquid plugs do not span the channel. The absence of liquid flooding in the channel is attributed to the single-serpentine flow structure which eliminates alternative air
flow paths, shortcuts, and dead zones. Analysis of the physics of flow regime transitions provides key understanding which can be applied for optimization of fuel cell microchannels.

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