FLOW STRUCTURES AND FRICTIONAL CHARACTERISTICS ON TWO-PHASE FLOW IN MICROCHANNELS IN PEM FUEL CELLS

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ABSTRACT
This experimental paper presents a study of gas-liquid two phase flow in rectangular channels of 500µm x 45µm and 23.7mm long with different wall conditions of hydrophilic and hydrophobic surface, in order to investigate the flow structures and the corresponding friction factors of simulated microchannels of PEMFC. The main flow in the channel is air and liquid water is injected at a single or several discrete locations in one side wall of the channel.

The flow structure of liquid water in hydrophilic wall conditioned channel starts from wavy flow, develops to stable stratified film flow, and then transits to unstable fluctuating film flow, as the pressure drop and the flow velocity of air increase from around 10 kPa to over 100 kPa. The flow structure in hydrophobic channel develops from the slug flow to slug-and-film flow with increasing pressure drop and flow velocity.

The pressure drop for single phase flow is measured for a base line study, and the $fRe$ product is in close agreement with the theoretical value ($fRe=85$) of the conventional laminar flow of aspect ratio 1:11. At the low range of water injection rate, the gas phase $fRe$ product of the two phase flow based on the whole channel area was not substantially affected by the water introduction. However, as the water injection rate increases up to 100 µL/min, the gas phase $fRe$ product based on the whole channel area deviates highly from the single phase theoretical value. The gas phase $fRe$ product with the actual gas phase area corrected by the liquid phase film thickness agrees with the single phase theoretical value.

INTRODUCTION
In the past decades, Proton Exchange Membrane Fuel Cells (PEMFC) have been studied as effective portable power sources due to many advantages such as low operating temperature, high power density, and good turn on-and-off capability. Recently, the relation between performance of PEMFC and the gas routing channel size was studied and it was found that sub-millimeter channels for gas routing improved overall fuel cell performance [Cha et al.(2004)]. However, the use of microchannels causes complicated water management issues because of the increasing importance of interfacial forces such as surface tension as the channel size decreases.

Gas-liquid two phase flow in microchannels has been extensively studied in the scope of heat transfer including boiling and condensation, flow regime characterization, and pressure drop relations. Zhang et al (2002) characterized microchannel heat transfer phenomena in two phase flow. Flow regime and pressure drop in two phase flows was studied extensively by Kawahara et al (2002, 2003), Lee and Lee (2001), and Mishima and Hibiki (1996). They used liquid as the baseline transporting fluid, and gas was introduced to the liquid flow. Also, they tried to understand the flow phenomena of gas-liquid two phase flow in terms of mixed two phase parameters such as two phase multiplier and Chisholm parameter. This approach may be appropriate for air-water mixed two phase flow, but there is little physical justification for this approach in the separated and stratified two phase flow regime.

Gas liquid two phase flow in microchannels of PEM fuel cell can have diverse flow structures of gas liquid two phase flow. For example, near the downstream of the PEMFC microchannels the flow structure could be air water mixed flow due to the flooding of water inside the gas supply channels. However, farther upstream the flooding of gas supply channels does not happen and the two phase flow inside the channels shows different structure. To date, the study of two-phase flow in microchannels of PEMFC has mostly focused on transport phenomena in the Gas Diffusion Layer (GDL). Thus, there exists an urgent need for the study on two phase flow in PEMFC microchannels in order for a full model of a PEM fuel cell to be developed. For the development of predictive models of two-phase flow in microchannels, the characteristics of pressure drop and flow of gas and the liquid water transport phenomena should be studied under a broad range of conditions.
This experimental work investigates the flow structures in microchannels under different surface conditions - hydrophilic surface and hydrophobic surface, and then develops a model for two phase flow in microchannel under hydrophilic and hydrophobic surface.

**NOMENCLATURE**

- $f$  Darcy friction factor
- $Re$  Reynolds number
- $fRe$  $f*R*Re$ product
- $\rho$  density of air [kg/m³]
- $m$  mass flow rate of air [kg/s]
- $u$  air velocity [m/s]
- $\sigma$  cross sectional area ratio
- $Dh$  hydraulic diameter [µm]
- $dP$  pressure drop [kPa]
- $P$  pressure of air [kPa]
- $Qw$  water injection rate [µL/min]
- $T$  temperature [K]
- $G$  mass flux [kg/m²-s]
- $K$  pressure loss coefficient
- $R$  gas constant [J/kg-K]
- $L$  length [m]
- $V$  air velocity at inlet [m/s]
- $A$  cross sectional area of channel [m²]
- $Ma$  Mach number
- $CCM$  cubic centimeter per minute [cm³/min]
- $SCCM$  standard cubic centimeter per minute (at 25 degC, 1 Atm)
- $LPM$  liter per minute [L/min]

**Subscript**

- $in$  inlet
- $out$  outlet
- $abs$  absolute pressure
- $c$  contraction
- $e$  expansion
- $m$  mean value
- $r$  reduced area
- $1$  location 1
- $2$  location 2
- $3$  location 3

**EXPERIMENTAL APPARATUS**

**Test Structures**

For the purpose of simulation of the introduction of water into PEMFC microchannels, the test structures consist of a 500µm wide and 45µm deep microchannel and side slots allowing for distributed water injection. The channel is U-shaped as shown in Figure 1, with a total length of 23.7mm and a bend radius of 1.5mm. Water is introduced through side slots perpendicular to the gas flow. The microchannels are made by Deep Reaction Ion Etching (DRIE) process in a silicon wafer substrate, and covered by glass for optical access to water transport phenomena inside the microchannel.

**EXPERIMENTAL SET-UP**

The experimental setup comprises subsystems for air flow/pressure control and measurement, water flow control, and optical visualization.

Figure 2 shows a schematic diagram of the experimental setup. The control and measurement devices are controlled using Labview software. Building compressed air is supplied through a pressure regulator (ControlAir Type-500X), and oil and particle filters. The absolute pressure and temperature of the inlet air are measured at parallel connected Omega mass and volumetric flow meters (FMA-1603A, FMA-1620A) of different measurement range. The pressure drop across the test channel is measured using the differential pressure transducer (Validyne DP15-46, DP15-TL). To minimize pressure loss, we used 1/8inch ID supply tubing, which is very large compared to the hydraulic diameter of microchannel (Dh=82µm). At a specified absolute pressure of air at the inlet point, the flow
condition in the microchannel can be varied by the change of outlet condition using a needle valve at the channel exit.

Liquid water flow is introduced to the gas flow through the side slots as shown in Figure 1. Water supply is controlled by a Harvard Apparatus Pump 11 single syringe pump. Water flow rate from 1 µL/min to 200 µL/min is driven using different size of syringe. This range of water flow rate is appropriate for the sectional simulation of upstream-to-downstream gas supply channel of about 30 cm long PEM fuel cell channels. Distilled water is used for the purpose of simulation of water production in PEM fuel cells. The air and water are supplied to the channel at room temperature. The test structures have no heating source and stay at room temperature during the experiment.

For the flow structure study and investigation of liquid water film thickness in the microchannel, a Nikon TE2000U inverted epifluorescence microscope and a Roper Scientific 12 bit Cool SNAP ES CCD camera are used. Fluorescence measurement is used for the liquid water film thickness. For these experiments, Fluorescein dye is mixed with the water at a concentration of 0.5 milliMole of Fluorescein per Liter of water.

FLOW STRUCTURES

Hydrophilic Surface Microchannel Flow

By nature, the test channels are highly hydrophilic due to the silicon dioxide layer that deposits on the silicon channel surface, and because the cover glass surface is always hydrophilic. The typical flow configuration for most water and air flow rates studied is stratified flow in which the water flows in a thin film along the injection-side wall as shown in Figure 3. At low water flow rates and moderate air flow rates this film was quite thin. For water flow rates up to 10 µL/min, there is no significant change to the overall water film thickness, and essentially the full channel area is available for air flow.

<table>
<thead>
<tr>
<th>Water film</th>
<th>Water film</th>
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</tr>
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</table>
| Qw: 1 ul/min  
  dP₁₋₃: 56.9 kPa  
  Pin.abs = 3 Atm | Qw: 5 ul/min  
  dP₁₋₃: 57.0 kPa  
  Pin.abs = 3 Atm | Qw: 10 ul/min  
  dP₁₋₃: 56.9 kPa  
  Pin.abs = 3 Atm |

Figure 3. Hydrophilic microchannel flow structure under small water flow rate

For higher water flow rates, the water flow configuration is more complicated and dependent on the air flow rates. Figures 4a and c show that at a high water flow rate and small pressure drop corresponding to small air flow rate, the channel is almost flooded with water. The flow patterns shown are stable and repeatable. At larger pressure drops with higher air flow rates, the flow pattern returns to a thin film along the injection wall as shown in Figures 4b and d. However, in this case, the film thickness is sufficient to partially obstruct the air flow channel. Comparison of Figures 4a and c and 4b and d shows that varying the absolute pressure has little effect on the flow configuration or film thickness.

In the range of high pressure drop (which corresponds to high air velocity) and high water flow rate the stratified flow is unstable as shown in the image (e) of Figure 4. The thickness of water layer fluctuates although the water film always remains continuous. The initial impression is that the flow has become turbulent. However, the Reynolds number of the liquid phase flow is only around 37, and the Reynolds number for gas phase flow is around 900. Both Reynolds numbers are still in laminar flow range. For more physical understanding of this unstable flow, the phase interaction at the interface of gas-liquid phase and wall effects needs to be studied.

<table>
<thead>
<tr>
<th>Water film</th>
<th>Water film</th>
<th>Water film</th>
</tr>
</thead>
</table>
| (a) Pin.abs=2 Atm  
  dP₁₋₃ = 9.2 kPa | (c) Pin.abs=4 Atm  
  dP₁₋₃ = 8.97 kPa | |
| (b) Pin.abs=2 Atm  
  dP₁₋₃ = 96.2 kPa | (d) Pin.abs=4 Atm  
  dP₁₋₃ = 83.2 kPa | |
| (e) Pin.abs=4 Atm  
  dP₁₋₃ = 165.6 kPa | |

Figure 4. Hydrophilic Microchannel Flow Structures under the water flow rate of 100 µL/min

Hydrophobic Surface Microchannel Flow

In order for the initially hydrophilic test channels to be hydrophobic, the test channels are treated using a Self Assembly Monolayer (SAM) process known as Molecular Vapor Deposition (MVD). In contrast to the hydrophilic surface flow, the flow pattern in the hydrophobic surface channel shows different flow structures as shown in Figure 5. The flow pattern at low pressure drop and low air velocity is large slug flow along the wall of the water injection side. The Reynolds number of air flow has the range of 20 to 300 and that of the liquid water flow has the range below 30. In examining the water flow structure, it is important to keep in mind that the absolute pressure is falling along the length of the channel leading to a decreasing density and an increasing velocity of the air from inlet to outlet. The effect of temperature change on the density and viscosity of the air is negligible in this experimental condition. The images in Figure 5 show that the slug size is decreasing in both length and width with the increase of differential pressure drop and air velocity in the channel. The contact angle of water slug is different between the leading
edge and the trailing edge, due to the movement driven by static pressure difference between the trailing and leading edge, by the dynamic pressure of air, and by the air-water interface frictional force. The flow structure at high water flow rate and high pressure drop has a tendency to partly become a stratified film flow as developed on the hydrophilic surface channel flow.

![Image]

Figure 5. Flow structures in the hydrophobic surface channel with different water introduction rate and pressure drop at the same absolute pressure of air (2Atm) at the inlet of channel.

PRESSURE DROP ANALYSIS

Flow Model for Hydrophilic Surface Channel Flow

Before constructing a model for the two phase flow, a general model for entire test apparatus is needed to indicate important locations for the pressure drop analysis. As shown in Figure 6, points 1 and 3 are the channel entrance and exit points respectively. The pressure drop $P_{1-3}$ is the desired output of both the measurements and model. Point 2 indicates the water injection point and two phase flow starts from this point.

![Diagram]

Figure 6. Schematic drawing of test channel and key locations.

Inlet and outlet points can be assumed to be reservoirs due to the large diameter of tube compared to the channel hydraulic diameter. Temperature and pressure measurements are taken at these points. Room temperature air is supplied at the inlet to the channel. The temperature remains nearly uniform due to the high heat conductivity of silicon substrate and low temperature variation of air flow. For instance, the temperature variation of air under adiabatic wall condition is less than 1 degree Celsius in this experimental flow velocity range. Under the condition of isothermal wall the temperature variation is even smaller than that of adiabatic wall condition. From experimental measurements at the inlet and outlet, conditions at point 1 and 3 can be obtained by relations for sudden contraction and sudden expansion internal flow with correction for compressibility effects. The relations can be expressed as

\[ dP_{in-out} = dP_{in-1} + dP_{1-3} + dP_{3-out} \]  
\[ dP_{in-1} = \frac{1}{2} \frac{G^2}{\rho_1} \left( K_1 + \frac{1}{\sigma^2} + 2 \left( \frac{\rho_2}{\rho_1} - 1 \right) \right) \]  
\[ dP_{3-out} = \frac{1}{2} \frac{G^2}{\rho_3} \left( K_3 - \frac{1}{\sigma^2} + 2 \left( \frac{\rho_3}{\rho_3} - 1 \right) \right) \]  
\[ G = \frac{\rho_3}{\rho_3} \rho_1 \right) \]  
\[ \rho_1 = \frac{P}{RT} \]  

Entrance pressure drop and exit pressure drop are composed of irreversible pressure loss, reversible pressure drop and pressure drop by acceleration due to compressible flow expansion, respectively. Using the ideal gas law, the desired pressure at inlet and outlet can be obtained under the constant temperature assumption. In this experimental range of air velocity which is lower than 50 m/s, the pressure drop by incompressible component, $K$ and $\sigma$, is negligibly small compared to the total pressure drop ($P_{in} - P_{out}$) which is higher than 150 kPa. Also, compressibility effect on the pressure drop at inlet and outlet is negligible due to the low Mach number. However, the compressibility effect on the density through the whole channel is significant and this effect is taken into account for the pressure drop as expressed in Equation 6.

The developing flow region is short compared to the full length of channel, so the full length of fully developed flow approximation is made for the pressure drop analysis.

The pressure drop from point 1 to 3 is composed of pressure drop of single phase flow region (point 1 to 2) and that of two phase flow region(point 2 to 3). For the hydrophilic surface, the two phase flow can be approximately modeled as stratified liquid film flow as depicted in Figure 7.

![Diagram]

Figure 7. Hydrophilic surface channel flow model

The pressure drop of two phase flow region (P2-P3) can be obtained by the subtraction of the pressure drop of the single phase flow region (P1-P2) from the total pressure drop (P1-P3). As will be shown below, the experimental $fRe$ product of single phase flow is in very close agreement with the theoretical $fRe$ product ($fRe=85$) for laminar flow in a rectangular channel of 1:11 aspect ratio. Therefore, the pressure drop of single phase
flow region (P1-P2) can be determined using the single phase flow relation as expressed by Equation (6). The single phase pressure drop is composed of frictional pressure drop and pressure drop by acceleration due to compressible flow expansion.

\[
dP_{1-2} = \frac{1}{2} G^2 \left( 2 \left( \frac{\rho_1}{\rho_2} - 1 \right) + f \frac{L}{D_h} \frac{\rho_2}{\rho_1} \right) \tag{6}
\]

\[
\frac{1}{\rho_w} = \frac{1}{L} \int \rho \, dx \geq \frac{2}{\rho_1 + \rho_2} \tag{7}
\]

\[
f = \frac{85}{Re} \tag{8}
\]

A simple model is proposed to determine the pressure drop in the two-phase flow region for the stratified flow regime. In this model, the liquid water is assumed to play no role other than to restrict the flow area available to the air as suggested in Figure 8. We assume that the no-slip, zero velocity boundary condition is appropriate for the air at the air/water interface. Therefore, as the water film thickness grows, the air flow is restricted to a smaller cross sectional area, and the air friction factor based on the full channel cross sectional area increases. The assumptions appear to be valid for the parameter range where the water layer thickness remains relatively small. Given the known water flow rate and the measured water film thickness, the average liquid water velocity can be determined. In all cases, the water velocity is one to two orders of magnitude less than the gas velocity. Also, at low water injection rate where the film is very thin, the water has essentially no effect on the measured friction factor as will be seen below.

Figure 8. Detailed schematic drawing for two phase flow region

To test the simple model, the water film thickness must be measured over a range of water flow rates and air flow parameters. The measured film thickness is plotted in Figure 9 as a function of pressure drop under different water injection rates. For the wavy flow regime which occurs at low air velocity, the average thickness along the length of the channel is plotted. The interesting phenomena on the film thickness are that the film thickness sharply decreases with pressure drop at the low range of pressure drop. In other words, the film thickness is very sensitive to the pressure drop at low pressure drop range. However, the flow and pressure drop measurements have higher uncertainty at low pressure drop range, and such flow and pressure drop measurement are directly related to friction factor calculation. However, in the high pressure drop range, the film thickness is a weak function of pressure drop. In this high pressure drop range, the model can be accurately tested due to not only the weak dependence of the film thickness and increased certainty on the flow measurement.

Figure 9a. water film thickness vs air inlet velocity based on nominal channel size

PRESSURE DROP MEASUREMENT RESULTS

Single Phase Flow

Single phase flow experiments were conducted to investigate the channel frictional characteristics as a baseline for each test structure. The experimental results are in close agreement with the theoretical friction factor \((fRe=85)\) for laminar flow in an 1:11 aspect ratio as shown in Figure 10. The friction factor measurements are highly uncertain due to the high relative uncertainty in the mass flow rate measurements in this flow rate range. Except the low pressure drop range, the \(fRe\) product is close to 85. From this result, microchannel flow of this size (500µm x 45.5µm) is shown to have the same frictional characteristics as conventional laminar flow for \(Re\) up to at least 700. This result was expected, and confirms our measurement accuracy. Based on this result, frictional characteristics of single phase flow region of the two phase channel flow is assumed to be the same as single phase flow. This allows us to determine the pressure drop of two phase flow region for the two phase frictional analysis.
Hydrophilic Surface Two phase flow

The two phase flow in the hydrophilic channel can be divided into two regimes in terms of the gas frictional characteristics: thin water film flow regime and thick water film flow regime.

Figure 11. Air inlet velocity and pressure drop relations under different water injection rates. Re for gas phase: 30 ~ 1000

For high water flow rates where the film thickness is significant, the simple reduced area model was applied using the measured film thickness. Figure 14 presents the corrected measurements of the $fRe$ product, showing good agreement with the theoretical value. Recall that at high pressure drops and high water flow rates, the structure of the water film became unstable resulting in a fluctuating film thickness. Using the average film thickness in the model produced excellent agreement with the theoretical value for friction factor. These results confirm our assumption that there is no significant interfacial interaction between the water and air, only a change in the available air passage area.

On the other hand, in small pressure drop range the friction factor highly deviates from the theoretical value. This deviation may partly come from the flow measurement error. In order to increase the flow measurement accuracy, two kinds of flow meters are used: 10CCM for low flow range and 1LPM for high flow range. By use of the 10CCM flow meter the flow measurement uncertainty can be reduced to below 10 % in flow rate at the range of very low pressure drop.

Additional uncertainty at low pressure drop comes from the film thickness measurement. As shown in Figure 9, the film thickness is very sensitive to the pressure drop at the low range of pressure drop. A small error in pressure drop measurement...
sharply changes the film thickness value, and it sharply changes the gas phase effective area. For the purpose of the film thickness measurement, fluorescence imaging is used and the measured liquid flow boundary is dependent to the threshold value of the intensity chosen. Also, in microchannel flow the viscosity and surface tension may be changed by the addition of a small amount of fluorescent dye. This may be a factor in the poor agreement with theory.

In addition to the measurement uncertainties, in this small pressure drop range under high water flow rate the two phase flow pattern is wavy and very thick water flow as shown in Figure 4, but the film thickness is taken as the averaged thickness. This different flow regime is another reason for the deviation even though this flow is still in laminar region. The air flow in this flow regime behaves more like a series of compressible nozzle and diffuser flows, not like plane wall and constant area flow. Therefore, a more detailed analysis appears to be necessary in this flow regime.

**Hydrophobic Surface Two Phase Flow**

In the hydrophobic channel, the flow velocity is highly changed by small water injection. The mass flow variation is significant even in very small water introduction rate and the variation is comparable to that of high water introduction rate condition in hydrophilic surface channel flow. This is because the water flow is not the film flow, but a kind of slug flow, which obstructs the air flow as shown in Figure 4. Figure 16 and 17 shows that just 1 µL/min of water introduction to air flow gives a significant change in the air velocity and friction factor. However, as the pressure drop and air velocity increase, the driving force to water slug increases. Hence, the slug is faster and smaller as captured in Figure 5. A More interesting point is that the water flow in a hydrophobic surface channel at high air velocity looks similar to a mixed regime of hydrophilic and weak hydrophobic surface channel flow. Hence, as the averaged air velocity increases, the hydrophobic surface flow can be modeled in the same way as the stratified flow on a hydrophilic surface channel. For the analysis of two phase flow in the low air velocity range, the water slug behavior in hydrophobic surface channel should be first understood including the effect of surface tension, static and dynamic pressure force and dynamic movement of water slug.
SUMMARY AND CONCLUSIONS

This experimental study examined the frictional characteristics of simulated two-phase flow in microchannels of PEM fuel cell to see the effect on gas friction of different liquid water introduction rate. The two phase flow structure depends on the surface condition of the channel (hydrophobicity), on the liquid water flow rate and on the pressure drop and air velocity. For the hydrophilic surface flow of water, the flow can be modeled as stratified film flow for liquid phase, and the gas phase friction is only a function of effective gas phase area. The interfacial effects at the air-water interface do not play a key role in this condition of two phase flow. However, in the low pressure drop range and low air velocity range the wavy and thick film flow includes more complex behavior and factors beyond what the present two phase model can explain. At the high range of water flow rate, air velocity and pressure drop, the water flow develops into unstable fluctuating film flow, but it still has the same frictional characteristics as the stable stratified film flow. The interfacial interaction of the two phases does not substantially affect the air friction.

Under the hydrophobic surface of microchannel the water flow behavior is basically different from hydrophilic surface microchannel flow, and it needs different model for the analysis of friction. However, as the air velocity and pressure drop increases, the effect of hydrophobicity on the water flow decreases and the flow of this range can be modeled as stratified two phase flow of hydrophilic surface flow.

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