# FUELCELL2006-97041

# FACTORS IMPACTING LIQUID DROPLET INSTABILITY IN A PEFC FLOW CHANNEL

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#### **ABSTRACT**

To achieve optimal performance with minimal parasitic losses and degradation, the relationship between water removal parameters such as flow rate and the diffusion media (DM) surface properties must be clearly identified. An extensive experimental study of the influence of controllable engineering parameters, including surface PTFE (Teflon<sup>TM</sup>) coverage (ranging from 5% to 20% of wt.) and operational air flow rate, on liquid droplet deformation at the interface of the DM and the gas flow channel was performed. A new visualization technique was developed to better understand the droplet mechanisms with enhanced optical access of both side and top views of the flow channel of a simulated H2 PEFC. A telecentric lens and 5 mm by 5 mm prisms embedded in the flow channel side walls were used for the first time to measure droplet receding and advancing surface angles in an enclosed flow channel. The influence of channel air flow rate and emerging droplet size on droplet characteristics with varying PTFE content in the DM was investigated to identify the conditions under which the droplet tends toward an unstable state. The results indicate that operational conditions, droplet height, chord length, and level of surface hydrophobicity of the DM directly affect the droplet instability. At high flow rates, the surface hydrophobicity of the DM enhances the efficacy of droplet removal, and helps to avoid local channel flooding,

however at low flow rates, regardless of the amount of PTFE content, droplet instability (and removal) is unaffected by the DM surface PTFE content.

Keywords: flooding; PEFC; PTFE; droplet removal; contact angle; gas diffusion layer.

## 1. INTRODUCTION

Channel level water transport is a vital issue for maintaining high performance and low parasitic losses in a polymer electrolyte fuel cell (PEFC). Due to the complexity of the thin film porous media and small length scales involved, water transport phenomena in a PEFC is difficult to fully understand. Electrolyte hydration is essential for efficient proton conduction, since its conductivity is a strong function of water content. While the electrolyte membrane needs to be maintained in a fully hydrated condition to ensure high proton conductivity, excess liquid water may hinder the reactant transport to the catalyst sites, a phenomenon generally known as "flooding". Flooding results in high concentration and kinetic polarization losses in the cell. Therefore, proper water management is essential to maintain high cell performance.

Channel level flooding is commonly seen under low air flow rates, especially in the anode channel at low operating current density [1]. At higher flow rates, the cathode side is

more prone to accumulation of excess liquid water, due to the fact that water is transported from the anode side to the cathode side by electro-osmotic drag and electrochemical water generation also occurs on the cathode. Excess liquid water transported within the pores of the DM accumulates at the surface of the DM in the channel, forming small droplets. As the DM becomes saturated, these droplets increase in size, eventually coalescing into water slugs or films in the channel. These water slugs prevent reactants from flowing smoothly in the channel, increasing parasitic pressure loss, and also providing facilitated transport of ionic impurities, which can accelerate the degradation. On the anode, liquid blockage can cause voltage reduction and fuel starvation to the catalyst layer [2,3], which can lead to oxidation of carbon support and accelerated degradation of the ionomer. In order to avoid the partial coverage of the flow channel by liquid water, it is advantageous to understand the droplet behavior at the interface of DM and the flow channel in order to determine the most effective purge strategy. For example, the addition of hydrophobic additive to the DM can enhance water removal, but it also increases electrical and contact resistance between landing and DM, which is undesirable. Therefore, a proper balance between these effects must be obtained for the optimal performance.

Due to the complex two-phase flow in the flow channel of PEFC, managing the transport of reactants and products through the flow channels is a challenging issue. The most common way to enhance liquid phase water removal from the channel and improve water management in the DM and catalyst layer is to use hydrophobic gas diffusion media. The naturally hydrophilic carbon fiber DMs are typically tailored by addition of hydrophobic material such as Polytetraflouroethylene (PTFE) during processing. To date, the fraction of hydrophobic additive used is generally determined through inefficient trialand-error testing. The existing literature [4-6] has also followed a phenomenological approach, and has yet to develop any clear rationale or fundamental knowledge of the basic transport processes of liquid droplets through the DM or at the interfacial boundaries between the DM and the flow channel. Additionally, few modeling or experimental studies on the droplet dynamics and removal at the interface of DM surface and flow channel have been presented in the literature. There have been a few qualitative direct visualization studies in the flow channel of the actual working fuel cell [7-9], but none have yet provided a fundamental understanding of how the transport of liquid droplet phenomena are related to the various parameters, including droplet formation, droplet size, air flow rate, surface tension and the hydrophobicity of the gas diffusion media. Several fundamental studies unrelated to fuel cells [10-13] observed the effects of droplet surface adhesion properties such as contact angle hysteresis, surface tension and roughness of surface, on transport phenomena. Even though these studies are not motivated by the problems present in an operational fuel cell, the physical phenomena of droplet interaction on different surfaces is similar to that occurring in fuel cell flow channels. Dussan et al. [10], Dimitrakopoulos and Higdon [11], Vafeei

and Podowski [12] and Lam et al. [13] have performed extensive studies (experimental and theoretical) on liquid droplet behavior over a range of surfaces and conditions. These studies are designed to explain the mechanisms of liquid droplet shear in the presence of the surrounding fluid shear, the displacement of fluid droplets from solid surfaces at different flow rates, the dependence of contact angle on the size of droplets on smooth surfaces, and the effects of liquid properties to contact angle hysteresis. While these studies are very helpful for understanding the droplet interaction on different surfaces, none of them provide sufficient information regarding effective water removal in the fuel cell flow channel under different operating conditions and DM surface hydrophobicities. Recently, Chen et al. [14] proposed a model related to the prediction of the onset of the liquid water droplet instability. This study draws important conclusions on droplet instability, based on a simplified analytical model. Chen et al. [14] concluded that droplet removal can be improved by increasing flow channel length and mean gas flow velocity, or decreasing channel height and contact angle hysteresis. However, there is still much work to be done. Additional parameters that are believed to be important for droplet instability are droplet size (chord length and height) and amount of PTFE surface coverage. In the present study, the effects of air flow rate, PTFE content and droplet size on droplet instability are investigated experimentally to understand the conditions leading to droplet removal.

#### 2. EXPERIMENTAL APPROACH

In order to investigate the droplet growth mechanism at the interface of the DM and flow channel, an experimental model cell was fabricated and mounted in a test stand that allowed for precise control of system parameters. While the non-operational model flow channel does not include electrochemical reaction, it allows simulation of the activity of a cell, and precise visual study of droplet formation and growth just above the DM, inside the flow channel.

The hydrogen PEFC flow channel experimental model consists of a rectangular flow channel (5 mm x 4 mm cross section), optically-accessible from two sides, with a third side consisting of a DM affixed to a feeder plate. Three different DMs with controlled hydrophobic treatment were utilized in this study (PTFE content equals to 5%, 10%, and 20% of wt.). Due to the extremely low liquid and gas flow rates (Reynolds numbers ~ O (100)), high precision equipment are used to control and monitor the air and water flow rate. A controlled liquid flow rate is achieved with a syringe pump apparatus. A combination of a regulator, throttling valve and mass flow meter are used to control the flow rate.

The model cell consists of two aluminum end blocks, an aluminum washer, a Lexan<sup>(TM)</sup> plate, an aluminum flow channel, prisms, an aluminum settling layer and a hole-pattern feeder plate. The desired flow rate of water simulating the electrochemical water generation is applied through the machined feeder plate by a syringe pump and a 0.177 mm

capillary tube. An air flow corresponding to operational fuel cell conditions is externally imposed into the channel through a pressurized gas system and heating system. The difference in pressure between the gas in the flow channel and liquid injected in the feeder plate assembly measured to increase the accuracy of the determination of the droplet pressure required to initiate two-phase flow in porous media from a static condition. Visual images of droplet formation and growth were acquired using a standard video microscopy setup with telecentric lens and strategically-placed prisms. A detailed schematic view of the experiment setup is shown in Fig. 1.

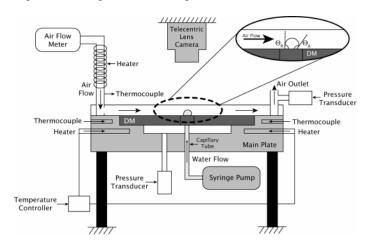


Figure 1. Schematic view of experimental setup

# 2.1 Method of Approach

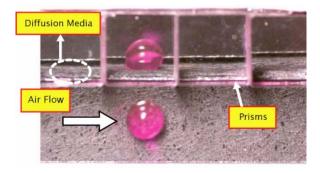
Three DMs with different levels of hydrophobic treatment (PTFE content 5%, 10%, and 20%) were utilized in these experiments. PTFE was assumed to be sprayed homogenously throughout the surface of DM, although microscopically, variations occur. Contact angles at the interface of three different DMs and gas flow channel were measured at 60 °C at air flow rates ranging from 0 to 5500 ml/min with an increment 440 ml/min. Liquid water was injected with a predetermined flow rate 0.023 ml/min to the DM through the 0.177 mm capillary tube. This water flow rate (0.023 ml/min) is used to simulate a current density 1 A/cm<sup>2</sup> at an equivalent cathode stoichometry of two and an active DM surface area of 4 cm<sup>2</sup>. Unhumidified air at 60 °C was supplied to the inlet of flow channel with a controlled flow rate. For each PTFE content, images at each operating Re number (based on channel hydraulic diameter) were captured and saved to allow for analysis of the droplet growth rate and deformation. Each image was carefully processed, and advancing and receding contact angles were measured by using IMAGE TOOL® software. Although the high magnification camera and telecentric lens were used to minimize the distortion on each image, each angle was measured five times and then averaged in order to minimize the measurement errors. The error associated with the contact angle measurements is  $\pm 2.8^{\circ}$ .

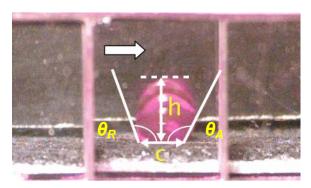
## 3. RESULTS AND DISCUSSION

# 3.1 Contact Angle Interpretation

Knowledge of surface tension is critical for modeling the effective water removal from the channel, since the drag force required for removing a liquid water droplet from the DM surface depends on the interfacial interaction of water molecules and carbon fibers or PTFE on the surface of the DM. The contact angle is a measure of the amount of wetting of the DM by a liquid. It is directly related to the interfacial energy of solid, liquid and vapor phases along the three phase boundary. The contact angles depend both on the base material of surface and surface morphology (roughness). Therefore, investigating the contact angles on different surfaces provides information on the energy of the surface of the interest [16]. However, due to the complexity and small length scale involved in an operating fuel cell, this interface in an enclosed channel is not easily accessible.

Two dynamic contact angles are seen when the liquid droplet is under the influence of air shear flow in the channel. These angles are defined as the dynamic advancing angle and dynamic receding angle ( $\theta_A$  and  $\theta_R$  in Fig. 2). These angles can be interpreted as the measure of the ability of the droplet to resist the drag force. The difference between advancing and receding contact angle (contact angle hysteresis,  $\Delta$ ) is a key parameter in determining the adhesion energy and the instability of the droplet deformed by the air flow [12-16].





where h is droplet height, c is droplet chord length,  $\theta_A$  is advancing angle,  $\theta_R$  is receding angle.

Figure 2. Captured image from the channel

In this study, it was experimentally observed that contact angle hysteresis ( $\Delta$ ), depends on channel air flow rate (Reynolds Number), droplet aspect ratio (height over chord ratio), surface treatment of the gas diffusion media (PTFE content) and surface roughness. Therefore a functional form of the contact angle hysteresis ( $\Delta$ ) can be written as:

$$\theta_A - \theta_R = \text{Contact angle hysteresis } (\Delta)$$
  
 $\Delta = \text{func } (Re, c, h, \text{PTFE, Roughness})$  (1)

Once all the data for three different PTFE cases (5%, 10%, and 20%) were collected, contact angle hysteresis ( $\Delta$ ) versus h/c ratio was plotted at different Reynolds numbers, and experimental data for 5% PTFE DM sample is shown in Fig. 3. As it is seen from the Fig. 3, there is scattering in experimental data, especially at low contact angle hysteresis ( $\Delta$ ). Therefore, a statistical approach was utilized to correlate and understand the behavior of the experimental data, since contact angle measurement by itself was not sufficient to describe any functional dependence due to the complex intermolecular forces and difficulties in measuring the surface angles. Kwok et al. [15] explained that obtaining meaningful contact angles for the determination of solid surface tensions is a difficult task due to the effects of swelling, chemical composition (inert) and roughness on contact angle measurements, and concluded that statistical tools may help to improve the meaning of measured contact angles to minimize the measurement errors.

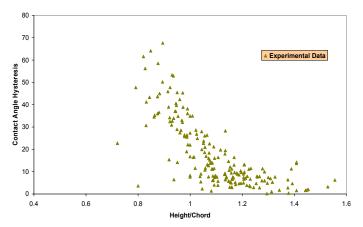


Figure 3. Contact angle hysteresis ( $\Delta$ ) versus height over chord ratio for 5% PTFE DM sample (experimental data)

A multi dimensional linear regression model was chosen to correlate contact angle hysteresis ( $\Delta$ ) with the relevant non-dimensionalized experimental parameters such as Re,  $c/d_c$ ,  $h/d_c$  where  $d_c$  is the hydraulic diameter of the flow channel. The contact angle hysteresis ( $\Delta$ ) also depends on surface treatment (PTFE content) and surface roughness, but surface roughness of the DM is not a controlled parameter in these experiments. Therefore, in our regression model, contact angle hysteresis ( $\Delta$ ) is functionally defined as:

$$\Delta = A + B \cdot \text{Re} + C \cdot (c/d_c) + D \cdot (h/d_c)$$
 (2)

The chord length (c) and height of the droplet (h) are non-dimensionelized by  $d_c$ , and a different multi-dimensional linear regression model was defined for each PTFE content. The three multi-dimensional linear regression fits were derived for three different PTFE contents, based on the extensive experimental data taken. These are:

$$\Delta_{\%PTFE} = -11.2 + 0.0156 \text{Re} + 139 (c/d_c) - 86 (h/d_c) \quad R^2 = 74.1\%$$

$$\Delta_{\%AOPTFE} = 6.02 + 0.018 \text{Re} + 177 (c/d_c) - 163 (h/d_c) \quad R^2 = 75.1\%$$

$$\Delta_{\%2OPTFE} = -6.52 + 0.0214 \text{Re} + 145 (c/d_c) - 964 (h/d_c) \quad R^2 = 82.1\%$$
(3)

Comparison of experimental and predicted contact angle hysteresis data based on experimentally measured droplet chord length and height, for 5% PTFE DM sample is shown in Fig. 4.

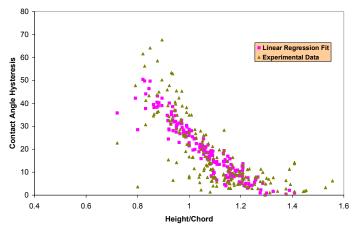


Figure 4. Comparison of experiment data and linear regression data for 5% PTFE DM sample

Since the experimental results indicated that contact angle hysteresis ( $\Delta$ ) depends on roughness, air flow rate, droplet size (droplet aspect ratio) and surface treatment of DM (PTFE content or surface tension), these effects were separately investigated in this study and are reported in the following sections.

# 3.2 Effect of Roughness

Roughness can be interpreted as the measure of the disturbances on the surface due to the disarrangement and misalignment of the carbon fibers on the DM surface. Generally, the average roughness, Ra, is defined as the integral of the absolute value of the roughness profile (Eq. 4). Ra is the most commonly used roughness parameter to identify the level of the roughness of the surface [17].

$$Ra = \frac{1}{L} \int_{0}^{L} |r(x)| dx \tag{4}$$

It is expected that roughness directly affects the line tensions and three phase boundary; therefore it may lead to errors in contact angle measurement and interpretations. Kwok et al. [15] also stated that there is no general criterion to quantify the level of roughness at which roughness has no effect on contact angle measurements. To evaluate the effect of roughness on contact angle measurements, scanning electron microscopy (SEM) pictures of cross sectional views of DM samples were taken and the cross sectional SEM image of 20% DM sample is shown in Fig. 5. As seen from Fig. 5, maximum disturbance on the DM surface is around 35 µm in a 4 cm long DM sample, which gives Ra values less than 8.75 x  $10^{-4}$ . Therefore effect of roughness was neglected in this present study and it was assumed that PTFE content in DM samples has no effect in terms of roughness within the range of PTFE of the DMs tested in this study, although PTFE particles on carbon fibers may cause disturbances which can affect the droplet and fiber interface and contact angle.

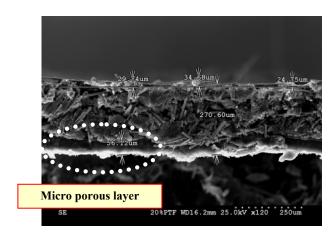


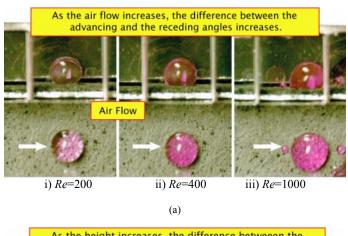
Figure 5. SEM picture of the cross sectional view of 20% PTFE DM

# 3.3 Effect of Air Flow Rate

Figure 6-a shows a sequence of captured images of the same size droplets under different air flow rates. It is clearly seen that contact angle hysteresis ( $\Delta$ ) increases with flow rate and the droplet tends towards an unstable condition. To investigate the air flow rate effects on contact angle hysteresis ( $\Delta$ ), air flow rate versus contact angle hysteresis ( $\Delta$ ) was plotted by using the linear regression data for 5%, 10% and 20% PTFE DMs at a specified droplet size (chord length = 1.7 mm and height = 1.9 mm) and the output of the 5% PTFE DM sample is shown in Fig. 7. It was observed that as the velocity of the air increases, the drag force increases and the droplet deforms along the flow direction, causing an increase in contact

angle hysteresis ( $\Delta$ ). As flow rate increases, the hysteresis increases and the surface energy (adhesion energy) between DM and water gradually becomes insufficient to resist increasing drag force. As a result, at some point, droplets will detach from the DM surface and will form an annular film, or roll over the DM surface in the direction of flow.

These results indicate that imposing high air flow rates into the flow channel increases the contact angle hysteresis, thereby causing the droplet to move towards an unstable condition. Thus, imposing high flow rates may enhance the liquid water droplet removal from the flow channel; however one consequence is that parasitic losses in a fuel cell system also increase with air flow rate.



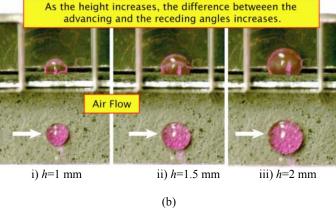


Figure 6. Captured images of a) droplets with same height under different air flow rates, b) droplets with different height under same air flow rate

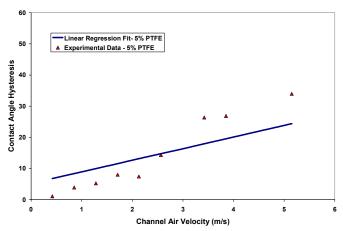


Figure 7. Contact angle hysteresis (Δ) versus air velocity at a constant droplet chord length (1.7 mm) and height (1.9 mm) for 5% PTFE DM sample

# 3.4 Effect of Droplet Size (Aspect Ratio)

Another important parameter affecting the droplet instability under different operating conditions is droplet aspect ratio (droplet height to chord ratio). In this study, it was experimentally observed that the droplet size has a significant influence on the droplet instability, since any instability is closely related to the drag force, and drag force is related the droplet size. Figure 6-b shows the captured images of three droplets of different aspect ratio (different height over chord ratio) subjected to a constant air flow rate. As seen from Fig. 6b, the larger droplets deform more in response to the same shear flow. A larger deformation means that the droplet experiences a greater difference between advancing and receding contact angles ( $\Delta$ ) to resist increased drag. As the droplet spreads over the DM surface, its chord length increases more than its height, and the surface tension force (proportional to chord) becomes more dominant than the drag force acting on the droplet surface (proportional to h<sup>2</sup>). Therefore, the ratio of droplet height to droplet chord length (droplet aspect ratio) is useful in describing droplet behavior.

Figure 8 shows the quantitative relationship between contact angle hysteresis ( $\Delta$ ) and h/c, based on a regression of the experimental data at Re=1000 for different PTFE contents, where Reynolds number is based on channel hydraulic diameter ( $d_c$ ). Contact angle hysteresis ( $\Delta$ ) is shown to be directly affected by the height to chord ratio.

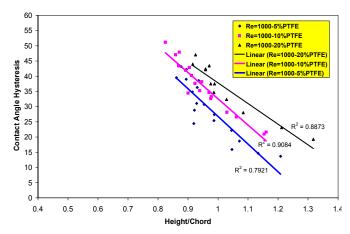


Figure 8. Contact angle hysteresis (Δ) versus height to chord ratio at a constant *Re*=1000 for different PTFE contents

#### 3.5 Effect of PTFE Content

Lastly, the effect of the amount of the surface coverage of PTFE on droplet removal was investigated. To date, empirical studies have been inconclusive regarding the best DM structure for two-phase flow management at the channel level, and to the authors' knowledge, no empirical correlation relating the surface tension to the PTFE coverage of DM has been reported.

Experiments were conducted with three different PTFE samples of DM carbon paper (PTFE content of 5%, 10%, and 20%). The droplet behavior at these different PTFE loading DMs was carefully investigated, in order to obtain a reasonable estimate of the droplet instability as a function of PTFE content. The change in these experimentally-measured surface contact angles was analyzed for the three different hydrophobic DMs to observe the effects of the PTFE loadings on the contact angle hysteresis, ( $\Delta$ ).

A linear regression was used to compact the data and plot contact angle hysteresis ( $\Delta$ ) versus height over chord ratio at a constant air flow rate (Re=1000) on different PTFE loading DMs (Fig. 8). At a specified height to chord ratio, the maximum contact angle hysteresis occurred in DM with 20% PTFE content and minimum contact angle hysteresis occurred in DM with 5% PTFE content. Physically, the surface adhesion force is reduced by rendering the DM surface more hydrophobic. As a result, at higher PTFE loadings of the DM (decreased average adhesion energy of the water molecules onto the carbon fiber, binder and PTFE substrate), the droplet deforms more readily, causing high contact angle hysteresis. Hence, the liquid water droplets located on a high PTFE loading surface tend to be more unstable, and the drag force required to remove the water droplets of a given size decreases.

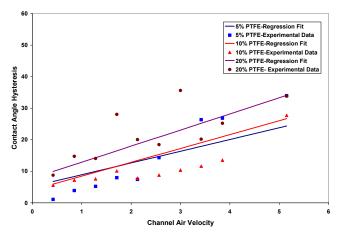


Figure 9. Contact angle hysteresis (Δ) versus air velocity at a constant droplet chord length (1.7 mm) and height (1.9 mm) for different PTFE content

# 3.6 Impact of PTFE Content

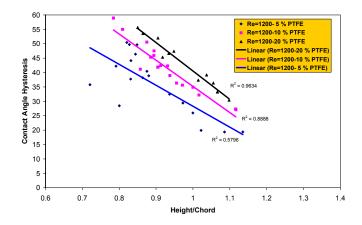
In order to develop a new design criterion for effective water removal from the flow channel, an optimal balance between the amount of PTFE content of the DM and air flow rate must be considered. Engineering consequences of increasing PTFE content and air flow rate include material cost, increase in electrical resistance of the DM, and the increase in parasitic losses.

The linear regression fits for each DM samples at specified chord length and height (c=1.7 mm and h=1.9 mm) are plotted and shown in Fig. 9. As the air velocity decreases from 5 to 2 m/s, the difference in contact angle hysteresis ( $\Delta$ ) between 5%, 10% and 20% PTFE decreases.

To assess the relative significance of PTFE content and air flow rate on the stability of the droplet, several experiments were conducted. The variation in contact angle hysteresis on different PTFE content of the DM was compared at different air flow rates (Re=1200, 900, 600, and 300). The results are plotted and shown in Figs. 10-11. As the Reynolds number decreases from 1200 to 600, the difference in contact angle hysteresis ( $\Delta$ ) between 5%, 10% and 20% PTFE decreases. The contact angle hysteresis ( $\Delta$ ) differs significantly at higher flow rate such as Re=1200 and Re=900, however at low flow rates; regardless of amount of PTFE content, the contact angle hysteresis ( $\Delta$ ) is similar. This means that the influence of PTFE content on contact angle hysteresis is more dominant at high air flow rate regime. However, under low air flow conditions, water removal from the channel is unaffected by the surface PTFE content, and therefore high PTFE content is not necessary, or desirable since PTFE additive will have the additional effect of increasing thermal and electrical contact resistance.

An analytical model based on a channel level macroscopic force balance was developed for an emerging droplet in the flow channel in order to validate the current experimental observations and identify the conditions under which the droplet tends towards an unstable state as a function of

engineering parameters. This combined theoretical and experimental study will be published in a separate publication.



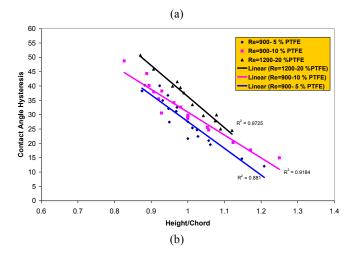


Figure 10. Contact angle hysteresis (Δ) versus height to chord ratio for different PTFE a) *Re*=1200 b)*Re*=900

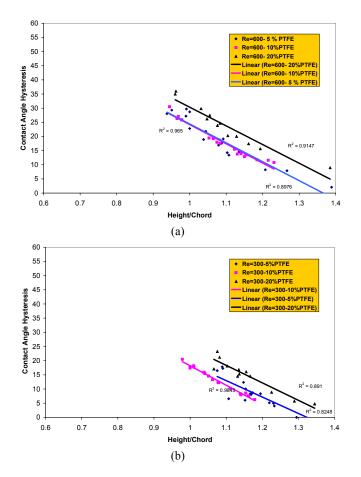


Figure 11. Contact angle hysteresis (Δ) versus height to chord ratio for different PTFE a) *Re*=600 b)*Re*=300

## 4. CONCLUSIONS

Employing simultaneous microscopic visualization of both the top and side views of water droplet growth in a flow channel model designed to simulate a flow channel of a PEFC, the droplet growth and deformation mechanisms under an imposed shear have been analyzed over a range of realistic operating conditions. The experiments were designed to provide a quantitative understanding of liquid water droplet removal at the interface of the gas diffusion layer and gas flow channel. It was observed that operational conditions, droplet size and surface properties of gas diffusion media such as hydrophobicity directly affect the level of droplet deformation, and therefore influence the condition for the droplet removal. Important conclusions that can be drawn for this experimental study include: i) imposing high air flow rates into the flow channel increases the contact angle hysteresis, promoting droplet removal. However, increasing air flow rate is known to increase parasitic losses, so optimization of flow rate should be considered within the context of the other operating conditions. ii) Droplet instability is sensitive to the droplet size (aspect ratio). iii) Using higher PTFE loadings of the DM promotes increased deformation of the droplet, causing high contact

angle hysteresis, due to the decreased surface interfacial tensions of the water molecules onto the carbon fibers. However, under a relatively low air flow condition, droplet instability (and removal) is unaffected by the surface PTFE content, and so at low air flow rate operations such as in anode, high PTFE content is not necessary, or desirable, since increased surface PTFE increases electrical resistance and material cost. Currently, a detailed theoretical modeling of this study is under development and it will be presented in a separate publication.

# **ACKNOWLEDGEMENTS**

This research is supported by NSF Grant # CTS-0414319. The authors would like to thank M. Edson, S. O. Ural, M. Khandelwal, S. Y. Yoon, E. Corbin and T. Chorman for their help during this study. The authors are also grateful to Dr. K. S. Chen and Dr. U. Pasaogullari for their helpful comments and discussions.

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