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Investigation on the liquid water droplet instability in a simulated flow channel of PEM fuel cell

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Abstract

To investigate the characteristics of water droplets on the gas diffusion layer from both top-view and side-view of the flow channel, a rig test apparatus was designed and fabricated with prism attached plate. This experimental device was used to simulate the growth of a single liquid water droplet and its transport process with various air flow velocity and channel height. Not only dry condition but also fully humidified condition was also simulated by using a water absorbing sponge. The detachment height of the water droplet with dry and wet conditions was measured and analyzed. It was found that the droplet tends towards becoming unstable by decreased channel height, increased flow velocity or making a gas diffusion layer (GDL) dryer. Also, peculiar behavior of the water droplet in the channel was presented like attachment to hydrophilic wall or sudden breaking of droplet in case of fully hydrated condition. The simplified force balance model matches with experimental data as well.

Keywords: PEM fuel cell; Gas diffusion layer; Water droplet; Instability

1. Introduction

Fuel cells have been expected to be one of the clean technologies that can keep in step fundamentally with various environment regulation policies that are spreading worldwide, including the Kyoto protocol. The fuel cell has been well known as a source of clean alternative power. Especially, it is recognized that the proton exchange membrane (PEM) fuel cell is the most suitable fuel cell type for application to vehicles in terms of start ability, operating temperature, and quick response with load change etc. However, for the PEM fuel cell to be commercially viable, the efficiency and performance need to be much improved by proper engineering optimization and design work. In particular, the membrane of the PEM fuel cell needs to be well hydrated to maintain ionic conductivity for proper fuel cell operation. However, the formation and condensation of liquid water occurs within the PEM fuel cell by excessive humidification, and it can cause the accumulation of liquid water in the cathode gas diffusion layer (GDL) and electrode, which causes flooding. At high current densities, this can act as the main reason for lowering the cell performance. An optimal removal of produced water is very important under the conditions of maintaining adequate humidity inside the fuel cell. Therefore, effective water management is essential not only for improving the efficiency and performance of the PEM fuel cell, but also for robust operation and durability.

Recently, liquid water in the PEM fuel cell has received much attention in the literature due to its importance. Pekula et al. (2005) visualized water accumulation in an operating PEM fuel cell by using a neutron imaging technique [1]. They presented images of the water droplet distribution in the channel and the GDL under various operating conditions. The results showed that liquid water has a tendency to accumulate at specific locations within the fuel cell, and the channel-level liquid droplet velocity is not

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constant, changing substantially due to interactions with the flow channel walls and other droplets. Pasaogullari and Wang (2005) and You and Liu (2002) developed a multidimensional, two-phase, electrochemistry-transport coupled PEM fuel cell model and predicted that two-phase transport and flooding lowered fuel cell performance, due to reduced oxygen transport and active catalytic area [2] [3]. They also suggested important factors affecting cell performance and two-phase transport characteristics such as inlet stoichiometry ratio and humidification. However, the two-phase model has a limitation predicting the effects of liquid water accumulation in the channel because it was assumed that fluid in the channel existed in homogeneous two-phase flow and liquid water did not interfere with the gas phase transport. Yang et al. (2004) and Kim et al. (2005) reported direct visualization of liquid water transport in a PEM fuel cell made of transparent walls and under automotive conditions [4, 5]. It was observed that water droplets emerged from the GDL surface under oversaturation of water vapor in the gas phase, appeared only at preferential locations, and could grow to a size comparable to the channel dimension under the influence of surface adhesion. Liquid film formation on more hydrophilic channel walls and channel clogging were also revealed and analyzed. Water droplets in the channel were swept away periodically and each droplet was shifted at the critical size. Especially, a water droplet moved easily without contact with the land but slugs of liquid water appeared with attachment to the land during the growth of the water droplet. These studies provided evidence on the tendency of liquid water transport like sectional distribution and overall elimination characteristic but did not show detailed behavior characteristics of a single droplet.

Therefore, to understand the concrete characteristics of a water droplet in the channel, analysis of single water droplet behavior on the GDL like development, instability and transform is required. Chen et al. (2005) investigated instability characteristics of a single water droplet with a simulated PEM fuel cell channel [6]. They showed correlations between instability of a single droplet and mean flow velocity, channel length, and GDL surface treatment. Droplet removal could be enhanced by increasing flow channel length or mean gas flow velocity, decreasing channel height or contact angle hysteresis, or making the GDL/GFC (gas flow channels) interface more hydrophobic. They also suggested simplified models which were developed for predicting the onset of instability leading to removal of water droplets at the GDL/GFC interface as in a PEM fuel cell. Similar to the work of Chen et al. (2005), Kumbur et al. (2006) confirmed that operational conditions, droplet height, chord length, channel size and level of surface hydrophobicity of the GDL directly affected the droplet instability, and they proposed additional parameters affecting the water droplet instability like Reynolds number and droplet aspect ratio (height/chord) [7]. They used 5mm × 4mm as a flow channel, but it was too large to simulate the real channel of a PEM fuel cell. A size of about 1mm × 1mm is generally used in the PEM fuel cell. Litster et al. (2006) employed a fluorescence microscopy technique for visualizing the transport of liquid water in unsaturated hydrophobic fibrous media and applied it to the gas diffusion layer of a PEM fuel cell [8]. They also proposed a new water transport scheme as the basis for developing improved models for water transport in hydrophobic gas diffusion layers. The water is not transported via a converging capillary tree as suggested in prior work and models. Rather, transport is dominated by fingering and channeling. That new hypothesis generally corresponded to the movement of liquid water with through-plane and growth of water droplets.

However, their experimental and analytical studies were focused only on growth and instability of the water droplet and performed only in a dry condition of GDL. In a real PEM fuel cell, GDL is often maintained at fully hydrated condition, and understanding of droplet behavior in the fully hydrated condition is required.

The objective of this work is to investigate the removal behavior of a single water droplet not only at dry condition but also fully hydrated. Therefore, the present work focuses on detachment and elimination of the water droplet and effects of a fully hydrated GDL surface.

2. Theory and experimental setup

Fig. 1 shows a control volume enclosing the single water droplet growing at the GDL surface. The water droplet becomes unstable when the surface tension between droplet and GDL surface equals the drag force by air flow. It can be represented as a nonlinear expression that is shown in Eq. (1). This expression is

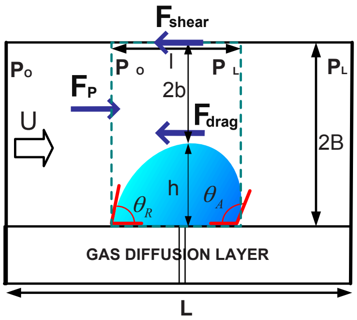


Fig. 1. Control volume enclosing the liquid water droplet.

referenced from a simplified model in [6].

$$F_p + F_{shear} + F_{drag} = 0$$
$$y\sqrt{1-y^2} - (\cot\theta_s)y^2 - \frac{12\mu U}{\pi\sigma\sin^2\theta_s} \times \frac{H(1+H)}{(1-\cos\theta_s)(1-H)^3 + (4B/L)H[1-(1-H)^3]} = 0 \quad (1)$$

where F_p is the pressure force on the droplet by pressure difference; F_{shear} is the shear stress on the top wall surface by flowing gas; F_{drag} is the viscous drag on the droplet surface by flowing gas; y = sin ($(\theta_A-\theta_R)/2$); θ_S is the static contact angle; H is the dimensionless droplet height; L is the flow channel length; U is the average air velocity; σ is the surface tension of water; μ is the viscosity of flowing gas.

The Bond number represents the ratio of a gravity force to a surface tension, and the gravity force is relatively critical when the number is larger than 2.5. In this experiment, the Bond number is smaller than unity, so the effects of gravity force to the water droplet behavior can be ignored.

$$B_0 = \frac{\rho g D^2}{\sigma} \quad (2)$$

The experimental apparatus to measure the instability and detachment of single water droplet consisted of a simulated PEM fuel cell channel with water absorbing sponge, air supply system, water supply system, and optical systems is shown in Fig. 2. To simulate water droplet growth in the real PEM fuel cell, the water was pumped up through a hole by using a micro syringe pump (Harvard Apparatus 'Pump 11') at a constant rate of 0.2 $\mu\ell$/min. This water flow rate corresponds to a current density about 1A/cm^2 in a typical PEM fuel cell operation. The water which was pumped up through the hole formed a water droplet on the GDL. Dry or fully humidified air of ultra high purity was used and was controlled by a mass flow controller (MFC, Bronkhorst) and temperature controller (YOKOGAWA). Dry air was used in the experiment of using the dry GDL and fully humidified (RH 100%) air was used in case of using the fully hydrated GDL. To observe the water droplet behavior, SUGITOH zoom lens (TS-93001) and halogen lamp (150W) were applied. The water droplets were recorded at 0.5-second intervals and captured images had a resolution of 1280 × 1024 pixels. The droplet height and contact angle were measured with commercial software as depicted in Fig. 3. A water absorbing sponge was inserted under the GDL to compare the dry condition with the fully hydrated condition. The inserted wet sponge was maintained at fully hydrated condition with a continuous water supply and it kept the GDL hydrated.

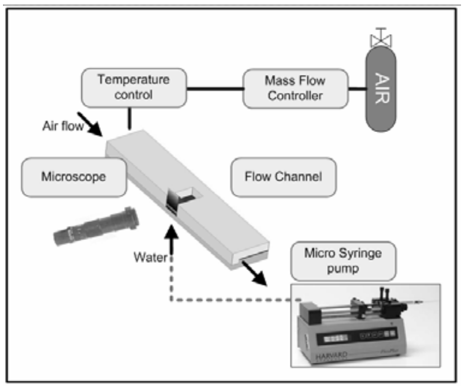


Fig. 2. Schematic representation of experimental apparatus.

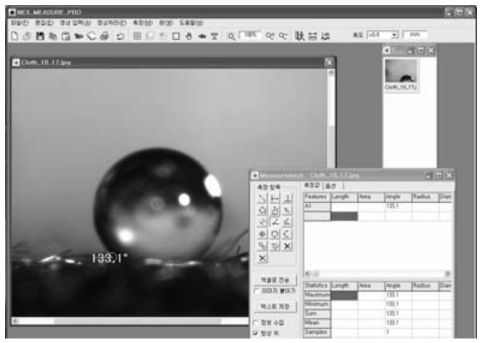


Fig. 3. NEX MEASURE software for the image capture and analysis.

The cross section of the air flow channel is shown in Fig. 4. The length of air flow channel was 20cm; a 0.4mm diameter hole was located in the middle of the channel for pumping the water. A BK7 window and

Table 1. Experimental conditions.

Channel height (mm)	1	2
GDL PTFE (wt %)	30	
Temperature (℃)	30	
Air humidity (%)	0 (with dry GDL)	100 (with wet GDL)
Air flow rate (LPM)	0.2-4	

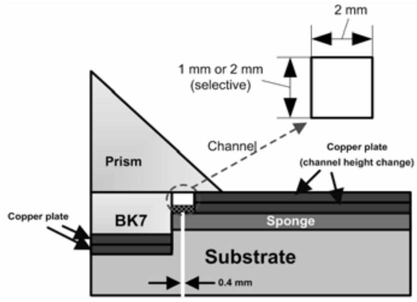


Fig. 4. Cross section of air flow channel.

prism were attached to the channel side to observe the water droplet with side view and top view simultaneously. Toray paper with 30 weight % Teflon® treatment was used as the GDL, and the temperature was maintained at 30 ℃ (start-up condition of PEM fuel cell). Efforts are presently underway to vary the temperature condition.

Experimental conditions are shown in Table. 1. Channel height, air flow rate, and humidity of air and GDL surface were taken as independent variables in order to discern conditions leading to droplet removal. Channel width was fixed as 2mm, and height was taken as 1mm and 2mm (typical channel size used in a real PEM fuel cell). Copper plates were stacked or removed to change the channel height. The air flow rate was varied from 0.2LPM (liters/min) to 4LPM. This air flow rate corresponds to a current density about from 0.1A/cm^2 to 1A/cm^2 in a PEM fuel cell operation which has an area of 330cm^2.

To prevent of air leakage, silicon rubber type gaskets were inserted between BK7 and copper plate, prism and copper plate, copper plate and substrate. Moreover, BK7 united with prism and leakage tests were performed after every new combination of components.

3. Experimental results

3.1 Characteristics of water droplet behavior

Generally, the water droplet sprung up from a particular point in each experiment, but sometimes, as shown in Fig. 5, a new droplet developed at another point nearby the initial point as the growing droplet of initial point stopped growing or shrunk (1mm of channel height, 4.2m/s of air flow velocity and dry condition). While the first water droplet was formed on the GDL, further progression of the second water droplet was restricted by the strong capillary pressure. But after the first droplet developed sufficiently, additional paths in the GDL developed slowly in the upward direction. And then, the local pressure of the second path decreased and the second water droplet sprung up to the surface as the local constriction was broken by slow development of liquid water in the GDL. These phenomena represent water in the GDL moving through selective paths, and paths have connectivity and interdependence with each other.

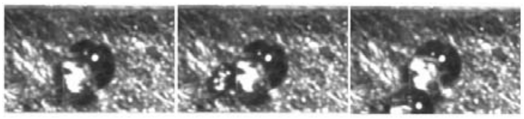


Fig. 5. Top-view of droplet emerging (Channel height=1mm, dry condition, air velocity=4.2m/s).

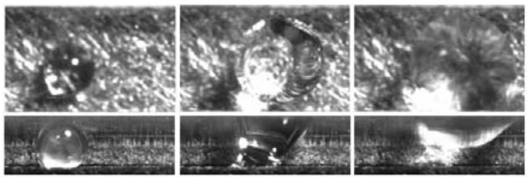


Fig. 6. Top-view and side-view of droplet attachment to the wall (Channel height=1mm, dry condition, air velocity=2.5m/s).

The captured images of water droplet growth with channel height of 1mm, air flow velocity of 2.5m/s and dry condition are shown in Fig. 6. In the case of low channel height, a droplet often attached to the top wall of channel which had hydrophilicity. The attachment to the hydrophilic wall causes a water film which has a large amount of surface tension. This water film cannot be eliminated easily, resulting in clogging of the channel. It is expected that hydrophobic treatment of channel wall will prevent this phenomenon.

A series of images that show water droplet removal by feeding from the other droplet with channel height of 1mm, air flow velocity of 5.8m/s and dry condition are given in Fig. 7. The first water droplet is grown to the critical size (first row images) and shifts in short

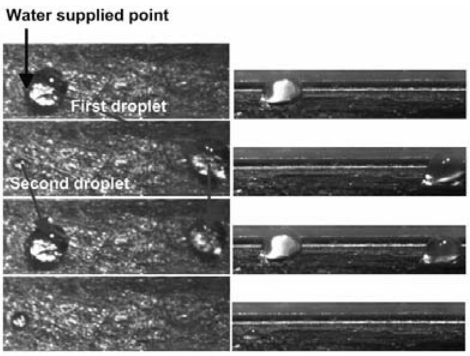


Fig. 7. Top-view and side-view of water droplet removal by feeding from the other droplet (Channel height=1mm, dry condition, air velocity=5.8m/s).

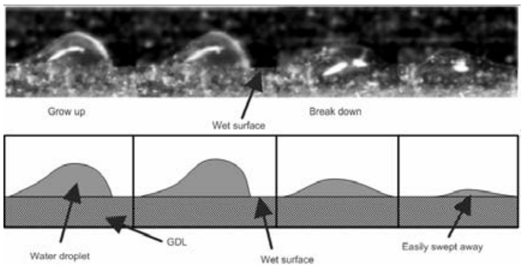


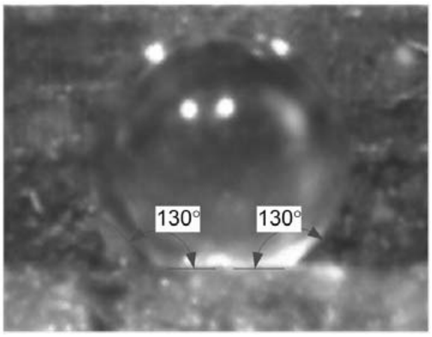
Fig. 8. Breaking of water droplet under fully hydrated condition droplet (Channel height=2mm, wet condition, air velocity=7m/s).

distance (second row images). During the shift, it shrinks to a size below the critical point because of water absorption of the GDL surface and evaporation by dry air, and stops moving. On the other hand, a new water droplet is formed at the water supplied point and moves toward the first droplet after growing to the critical size (third row images). The new droplet collides with the first droplet and they are eliminated together (fourth row images). As can be known in this process, the water droplet which stops growing due to the interruption in water supply cannot be eliminated without feeding the other droplet. In other words, a growing water droplet that is not developed into a critical size can be removed by feeding another droplet and it causes the periodic elimination of water droplets in the channel.

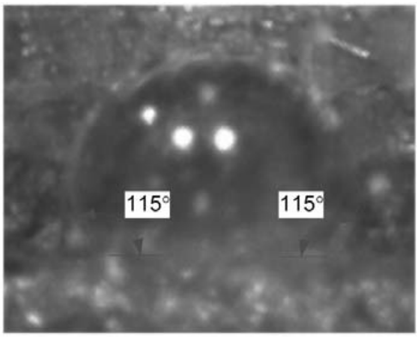
Especially, in the wet condition, sudden breaking of the water droplet was observed. Captured images of the growing water droplet with channel height of 2mm, air flow velocity of 7m/s and the wet condition are shown in Fig. 8. (Drawn pictures are added for convenience of recognition.) When the growing water droplet reached a particular surface which was fully hydrated, the droplet was attached to the wet surface and broken down. Compared to the droplet attachment to the hydrophilic wall, attachment to the wet surface does not form a thick film or large droplet size that brings channel clogging. Therefore, in a real PEM fuel cell, this phenomenon is expected to improve the elimination of water droplets and clogging in the channel.

3.2 Effect of wet surface

Sample images of water droplets in dry condition and fully hydrated GDL with fully humidified air condition (wet condition) are shown in Fig. 9. The heights of water droplets were approximately 1mm and contact angle was measured by NEX MEASURE software. Static contact angles with water droplet heights are shown in Fig. 10. As droplet height increases, the static contact angle increases and theoretically, the contact angle when the droplet height equals zero is defined as the static contact angle at that condition. The static contact angle provides a standard of instability of water droplet. As a static contact angle increases, the contact area of the water droplet to the surface decreases and the droplet becomes more unstable. The static contact angle of the water droplet in the dry condition (125°) is larger than that in the wet condition (110°) as represented in Fig. 10. It means that the water droplet tends to be stable in the wet condition. A difference of 15° cannot be ignored because the static contact angle difference between 30wt % Teflon® treated and untreated is 20° and it causes measurable disparity [9]. Actually, effects of the difference to the droplet instability could be confirmed by analyzing droplet detachment. Air flow velocity with dimensionless droplet height of detachment is shown in Fig. 11. The air flow velocity was calculated from air flow rate to compare the conditions easily. Detachment droplet height (dimensionless, h/2B) is reduced for increased air flow velocity. And in a dry condition rather than a wet condition, a droplet is detached easily. This stability difference with hydration level is minimized at the condition of high air flow velocity. It also can be confirmed that a smaller channel height makes the water droplet unstable. Fig. 12 shows the dimensionless droplet height to the contact angle hysteresis by calculating from Eq. (1). The line is a boundary between the stable and unstable area of the water droplet. Upper region of the line is the unstable region, and the lower



(a) Dry condition



(b) Wet condition

Fig. 9. Images of static contact angle of water droplet.

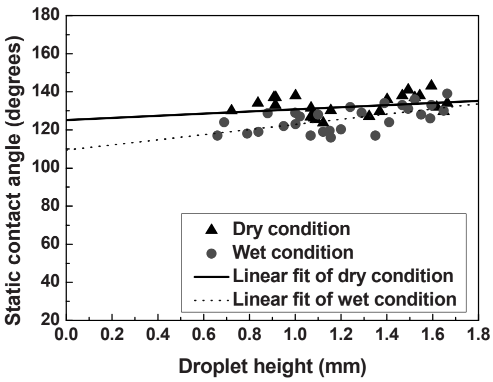


Fig. 10. Static contact angle with droplet height.

region is stable region; therefore, droplet instability can be estimated with Fig. 12. As the line drops downward, the area of the stable region decreases, that is, a water droplet of that condition is relatively unstable. For precise comparison, matching the experimentally measured data (droplet height according to contact angle hysteresis) to the simplified model prediction was performed, but the gap between experimental results and model is quite big as shown in Fig. 12. This is attributed to under-estimating the drag on the droplets and the shear force on the walls due to the geometry simplifications. Though this is a discrepancy, a graph of the simplified model is useful for a grasp of droplet instability tendency. As shown in Fig. 12, the unstable region becomes large for increased static contact angle and air flow velocity. As well, minimization of the stability difference with static contact angle at the condition of high air velocity can be verified by the graph.

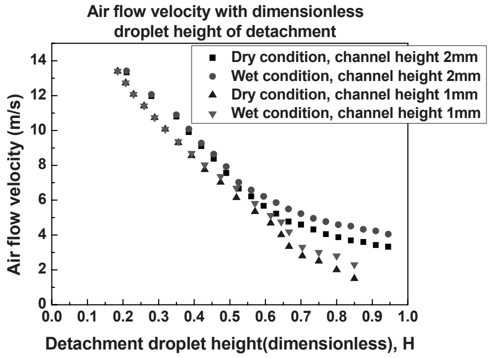


Fig. 11. Air flow velocity with dimensionless droplet height of detachment.

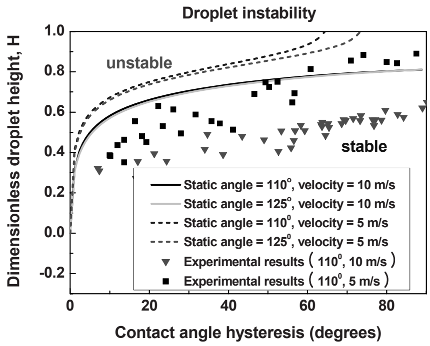


Fig. 12. Instability diagram of liquid water droplet.

4. Conclusions

A water droplet in the PEM fuel cell was visualized with a simulated channel, and the formation, transform and instability of the water droplet with dry and wet conditions were investigated. Analyses of detachment droplet height of the water droplet with various conditions were performed, and removal characteristics of water droplets were examined by assaying captured images. The experimental result was validated with a simplified model, and the simplified model could be the criterion for estimation of tendency. The main results about water droplet instability and increased are summarized as follows.

(1) The static contact angle of a water droplet has a larger value in case of dry condition (dry condition : 125°, wet condition : 110°). As the static contact angle increases, the water droplet tends to become unstable, and this is proved simultaneously by model and experimental results. The unstable region becomes large with larger static contact angle, but the tendency decreases with high velocity of air flow.

(2) A water droplet of the same size is prone to be detached and removed at smaller channel height under identical velocity of air flow. An optimized channel design is needed that satisfies both the good removal of liquid water and reasonable pressure drop.

(3) The water in GDL moves in selective paths, and the water formed in a channel occasionally attaches to the hydrophilic wall or is removed by feeding other droplets.

(4) Breaking of water droplet was observed at fully hydrated condition. A growing water droplet is broken down suddenly with attachment to the wet surface of GDL. This breaking of the droplet brings on the improvement of water elimination in the channel of a fuel cell.

Acknowledgment

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Nomenclature

B	:	Half height of the flow channel (mm)
Bo	:	Bond number
D	:	Droplet height (mm)
H	:	Dimensionless droplet height ; h/2B
L	:	Flow channel length, (mm)
Re	:	Reynolds number
U	:	Average air velocity (mm s^{-1})
y	:	Sin ($(\theta_A-\theta_R)/2$)
θ_A	:	Advancing contact angle (degree)
θ_R	:	Receding contact angle (degree)
Δ	:	$\theta_A - \theta_R$ (degree)
θ_S	:	Static contact angle (degree)
σ	:	Surface tension (dyn cm^{-1})
μ	:	Viscosity (g cm^{-1} s^{-1})

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