## **Chapter 3**

## Visible Imaging Measurement

The work described here details the application of visible imaging for studying water-droplet dynamics in the gas flow channels of proton exchange membrane (PEM) fuel cells. We will first describe our visible imaging system incorporated in a fuel cell test station in section 3.1. Next section, we will discuss performance test from a polarization curve of the PEM fuel cell designed in Chapter 1. The effect of drawing current to the water distribution in transient response and in steady state will be investigated in section 3.3. The process of water-droplet formation and removal in the flow channel and on the top surface of gas diffusion layer (GDL) will be presented in section 3.4.

#### 3.1 Visible Imaging System

Figure 3.1 shows schematic diagram for direct visualization measurement of a transparent fuel cell under operation. The experimental setup consists of a charge couple device (CCD) integrated into our in-house fuel cell test station. The test station used for operating a test fuel cell is composed of gas supplies, pressure regulators, mass flow controllers, an electronic load box and a computer based control interface which performs data acquisition. The test fuel cell as designed in Chapter 1 must be optically transparent for direct visualization. We use the poly-methylmethacrylate (PMMA) for a transparent window revealing the flow channels and the GDL top surface in the cathode area, where production of water occurs (seen in Figure 3.2). The active surface area of the cell is 6.25 cm<sup>2</sup>.



Figure 3.1: Schematic diagram of our visible imaging system incorporated in the fuel cell test station



Figure 3.2: The front view of the transparent fuel cell assembly.

In order to ensure a fully-hydrated membrane, it is important to operate the first-time membrane for at least 24 hours. For this thesis, the tested PEM fuel cell was operated at room temperature. Operation procedure of the PEM fuel cell started with a program that controls reactance gases into the cell. This step is called "open circuit". The voltage of the cell measured at this same time is called open-circuit voltage (OCV), which normally is more than 0.8 volt. After we performed open circuit for approximately 5 minutes, we slowly increased drawing current of the cell.

### 3.2 Performance of Proton Exchange Membrane Fuel Cell

Generally, the standard measurement of performance for PEM fuel cell systems is the polarization curve, which represents the cell voltage behavior against operating current density. Even though the theoretical voltage of a fuel cell is about 1.2 V, the actual cell voltage of the fuel cell is lower than the theoretical model due to species crossover from one electrode to the other through the electrolyte and internal currents. Typical voltage losses in a fuel cell are illustrated in Figure 3.3. The three major classifications of losses that result in the voltage drop from the OCV are activation polarization, ohmic polarization and concentration polarization [5]. The electrochemical behavior of our PEM fuel cell under standard conditions is shown in Figure 3.4. The shape of the polarization curve is similar to the one in Figure 3.3. The peak performance is at the power density of 0.8 Watt/cm<sup>2</sup>, and at the current density of 0.72 A/cm<sup>2</sup>. According to the polarization curve, the fuel cell is in the mass-transfer region. In this region, the performance is affected by water flooding in the flow channel and water content in the membrane electrode assembly (MEA).



Figure 3.4: polarization curve (●) and power curve (▲) for the tested PEM fuel cell.

#### 3.3 Water Distribution Response to Changes in Current Density

In this section, we observed the water distribution in the flow channels and the GDL top surface in the cathode area as we changed the operating current. The response to the change in transient and steady states are discussed below.

#### 3.3.1 Transient Response

The transient responses of the tested single fuel cell based on the polarization characteristics along with visualization images of the water distribution in the cathode flow channels were examined. The step change in current density was made according to the cell-current profile shown in Figure 3.5.



Figure 3.5: Current density profile adopted for the experiment on the transient response of the PEM fuel cell.

The time response of a PEM fuel cell is generally governed by three main processes [21]. The first process is the charge/discharge process of the electric double layer in the catalyst layer. The time constant describing the charge or discharge of the electrochemical double layer is estimated to be  $1.5 \times 10^{-7}$  seconds according to the normal parameters reported by Reference [22]. This value is small enough to be ignored in the dynamic modeling work done on automotive fuel cell. The second process is gaseous species transport in the GDL. Using the typical GDL thickness and effective gas diffusivity (based on a Bruggeman correlation), the time scale of the gas diffusion in the GDL is on the order of 0.1 - 1 second, as reported in Reference

[23]. The last process is membrane hydration/dehydration. The time constant associated with membrane hydration/dehydration is related to transient water transport.

It is believed that the transient water transport is mainly influenced by two main factors. The first factor is the membrane hydration/dehydration response to changes in the cell operating conditions. The second factor is electrode flooding resulting from excessive water produced under high current density operating conditions. These two factors are associated with complicated two-phase water transport processes inside the cell. Since these two factors are relatively slow processes, they have a dominant influence on the dynamic cell performance.

Figure 3.6 shows the voltage response to a step increase in the current density from 0.2 to 0.4  $A/cm^2$  for our PEM fuel cell. We observed that the cell responds to a step increase of the operating cell current density simultaneously, and reaches a new voltage level. It appears that the dynamic response of the cell voltage is almost instantaneous. However, the water distribution inside the fuel cell cannot quickly respond to the increase in operating cell current density as can be seen in Figure 3.7.



Figure 3.6: Dynamic response of the cell voltage with respect to a step current change from 0.2  $A/cm^2$  to 0.4  $A/cm^2$ .



Figure 3.7: Image of water distribution in the flow channels with time after a step increase in the current density from 0.2 to 0.4 A/cm<sup>2</sup>.

The white part in the flow channels represents the area of water distribution monitored soon after a step increase in the current density started to 15 seconds later. At 5 seconds, which is on the order of time constant for the gaseous transport through the GDL, a noticeable change in the water distribution in the flow channel has not observed. The water content in the flow channel starts to increase in addition to an enhancement in water production, which results from high current density operation at about 10 seconds after the load changes. This can be confirmed by comparing the water distribution in the marked area of Figure 3.7 at 10 seconds with that at 5 seconds.

#### 3.3.2 Steady State Response

We also studied the water distribution as a function of the drawing current when the cell reached steady state. All photographs in Figure 3.8 were captured at 20 minutes after the current of the cell were drawn at 0.2, 0.4, and 0.6  $A/cm^2$ . It can be observed that the white color along the flow channel become brighter as the current increased, which indicates that the flow channel is full of water buildup. As shown in Figure 3.8, the variation of the current density yields only a small increase in the water accumulation at the cathode, which implies that the increase in the water outflow that is caused by humidification and reaction is no more than that of the water outflow from the cell.





Figure 3.8: Photographs of water buildup at the cathode with different current densities (a) at current density of 0.2  $A/cm^2$ , (b) at current density of 0.4  $A/cm^2$ , (c) at current density of 0.6  $A/cm^2$  after drawn current 20 min.

# **3.4 Characteristics of Droplet Formation and Water Removal at Fixed Current Density**

In this section, the recorded photographs were analyzed on how the primary water droplets are formed, grown and transported along the flow channels.

#### 3.4.1 Droplet Formation Process

At a fixed current density of 0.4 A/cm<sup>2</sup>, droplet formation process in the flow channel of the PEM fuel cell was captured by the CCD camera described in the followings. Figure 3.9(left) is the image of flow channel at the start of operation when water-droplet has not yet present. After 5 seconds into operation (Figure 3.9(right)), water droplets from the electrochemical reaction start to form, initially, around the u-bend of channel. It suggests that flooding may happen first in that particular region which can be attributed to the distribution of gas in the channels. The distribution of gas is denser at the bend than at the inlet and the outlet, which is capable of more reactions and thus more water production at the bend.



Figure 3.9: Photographs of empty flow channel (left), and water-clogged flow channel after 5 seconds into operation time (right).

Furthermore, a sequence of water droplet images forming in the channel is shown in Figure 3.10 with an interval of 5 minutes between each frame. It can be stated from these images that water droplet in contact with the sidewall, the so-called land-touching droplet, become visible after 5 minutes into the operation. At 10 minutes passed, there is a new droplet appearing at the gas outlet. At 15 minutes passed, the land-touching droplet continues to appear in the flow channel. By that time, one land-touching droplet near the second bend of channel grows and expands until contacting each side to completely block the flow channel.



Figure 3.10: Droplet formation in the flow channel for transparent cell at current density of 0.4 A/cm<sup>2</sup>.

#### 3.4.2 Water Removal Process

In order to observe the characteristics of water removal process, the image of the flow channel for the cell assembly was recorded starting at 20 minutes after the current density at 0.6 A/cm<sup>2</sup> was drawn. Figure 3.11 shows a sequence of water droplet images removing from the flow channels with an interval of 5 minutes between each frame. At the beginning of the water removal process (at 20 minutes), there is already water accumulation in the flow channel and the bend of channel. At 25 minutes, it can be observed that the water droplet appear in the channel (especially at the U-bend of the flow channel). Only a few of water droplets is removed out of the fuel cell. As seen in the later pictures, the droplets start to become smaller in size. The droplets are probably pushed out of the cell by the gas flow as a result of increased cross-section area in the direction of the gas flow.

Figure 3.12 reveals the water accumulation at the cathode after water removal process at 45 minutes. As show in Figure 3.12(b), there is still certain amount of water left on the channel walls, seen as bright color beside the channel walls. This is due to the wall adhesion effects, and this part of water remains difficult to be removed. The second bend of flow channel (Figure 3.12(c)) is clogged by the land-touching droplet. The produced water can cause flooding in every part of the channel. If a serpentine channel is clogged by the produced water, the reactant gas cannot be supplied beyond the location of the clog, and therefore decreases effective electrode area. As a result, the cell performance is deteriorated

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Figure 3.11: Visualization of water removal process in the flow channel for transparent cell at current density of  $0.6 \text{ A/cm}^2$ .





Figure 3.12: (a) Photographs of water buildup in the flow channel at current density of 0.6 A/cm<sup>2</sup>, (b) enlargement of part of picture (a) at position 1, (c) enlargement of part of picture (a) at position 2.

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The visible imaging together with the fuel cell test station is implemented in an effort to deepen understanding the dynamic processes in terms of water transport characteristics within an operating PEM fuel cell. The transparent cell designed in Chapter 1 with the PMMA window on the cathode side is subject to visualization during the operation conditions. A preliminary experiment was performed to study the transient response of the cell with respect to load changes. We found that the cell voltage responded almost instantaneously to a change in drawing current, while the water content in the flow channel and on the gas diffusion layer GDL surface could not as quickly respond. Under the conditions tested in this study, it took about 10 seconds to alter the water distribution in the flow channel as a result of load changes. When the cell reached steady state for each drawing current, we observed a slightly increase in water accumulation for higher current densities. The process of water-droplet formation and removal in the flow channel and on the top surface of the GDL was investigated at a fixed current density. At 5 seconds after drawn current, water started to accumulate and build up, particularly at the bend of channel where the reactant gas was denser, i.e., more reactions. Land-touching droplets appeared after 5 min from drawing the current, and some of them grew and expanded until became water column. The removal process sped up when the water droplets became smaller in size; some droplets eventually were removed from the cell. However, the water droplet at the corner of the bend of channel was hardly removed out of the cell due to wall adhesion effects. The inefficient water removal process could cause clogging and flooding resulting in lower cell performance.



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