MEASUREMENT OF THE DISTRIBUTIONS OF LOCAL CURRENT DENSITY AND LOCAL IONIC RESISTANCE OF PEFCS

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ABSTRACT

An experimental apparatus for a PEFC was constructed with which the local ionic resistance distribution can be measured as well as the local reaction current distribution, using segmented current collectors and an AC resistance meter. The apparatus presented a clear view on the current distribution and its causes: hydration and dehydration of the membrane and the oxygen consumption along the gas streams. The effects of flow direction, gas stoichiometry and humidification were examined.

INTRODUCTION

In a standard fuel cell experiment, the primary interest is focused on its voltage-current relationship because it indicates the overall performance of the cell. To analyze the properties of the cell in detail, however, this relationship is not relevant because the current density is not uniform in most cases. One of the main causes of this nonuniformity is that the reactant gases flown on the electrode are consumed gradually along the way. In addition, water, the reaction product, is accumulated and affects the conductivity of the membrane and the gas diffusion. To obtain local information of a cell, several attempts have been made recently. Gottesfeld et al. applied a printed circuit board on segmented current-collectors and two load boxes and obtained the current of each segment by introducing the current from a targeted segment to one load box while the rest of the current was introduced to the other load box [1]. Stumper et al. used graphite blocks as shunt resistors contacting with the back of a flow-field/current-collector [2]. Wieser et al. applied a Hall sensor to obtain the local current density [3]. In either case, although the current density distributions were successfully obtained, the causes of the distributions were still only speculated and more detailed local information has been craved.

In this study, we introduce a simple but effective method to determine the local ionic resistance as well as the local current density. Ionic resistance of a whole fuel cell is usually measured by the current interruption method or the AC impedance method and IR correction is carried out to expose other overpotentials: reaction and concentration overpotentials. This approach, however, can be misleading because the local resistance also can be nonuniform and the local current density distribution does not necessarily correspond with the local resistance distribution. Therefore, determining the local ionic resistance along with the local current density is an essential first step for the performance analysis of a cell.
EXPERIMENTAL

Principle of Measurement

Using a segmented flow-field/current-collector, it is relatively simple to obtain the local current flowing into each segment by directly measuring the current using a cramp type current meter or using a shunt resistor and a DC voltmeter. Figure 2 shows the schematic diagram of the system using the latter. The DC voltage drop in the shunt resistor is measured with a DC voltmeter. Using the Ohm’s law, the DC current, \( I_d^i \), flown to the segment \( i \), is determined by:

\[
I_d^i = \frac{V_d^i}{R_s}
\]  

[1]

where \( V_d^i \) is the measured DC voltage drop in the shunt resistor; and \( R_s \) is the resistance of the shunt resistors.

To obtain a local resistance, an AC impedance technique is necessary. In this system, an AC voltage of 1 kHz is applied between the cathode and the anode. The AC current, \( I_a^i \), flown to the segment \( i \), is also determined by:

\[
I_a^i = \frac{V_a^i}{R_s}
\]  

[2]

where \( V_a^i \) is the measured AC voltage drop in the shunt resistor. Assuming this segmental AC current is inversely proportional to the local cell resistance of the area that the segment is facing, the local cell resistance, \( R_i \), for the segment \( i \), is determined by:

\[
R_i = \frac{V_a^i}{I_a^i} = \frac{V_a^i}{V_d^i} \quad [3]
\]

where \( V_a^i \) is the applied AC voltage between the cathode and anode. In this calculation, \( R_s \) is assumed to be small enough comparing to the cell resistance, \( R_i \). Even if the diffusion layer, a carbon cloth, is not segmented, the in-plane resistance of the carbon cloth is usually large enough if \( R_s \) is selected properly.

If an AC voltmeter and a DC voltmeter or an AC current meter and a DC current meter, are available for each one of the segments, continuous measurement can be carried out. Otherwise, a switch box is necessary to select the shunt resistor to be measured one by one and some interval is needed to finish a set of measurement.

A further simpler instrumentation, which we employed, can be used. Instead of using an AC power supply and an AC voltmeter separately, an AC resistance meter is applicable, which applies a preset AC current between the two current-terminals, measures the AC voltage between the two voltage-terminals and displays the calculated
result, *i.e.* the measured AC voltage drop / the preset AC current, as the measured resistance. In this application, the two current-terminals were connected with the cathode and the anode. The two voltage-terminals were also connected with the cathode and the anode firstly to measure the resistance of the whole cell area. The AC terminal voltage can be obtained by:

\[ V_a^i = I_a^i R^i \]  \[4\]

where \( I_a^i \) is the AC current preset in the AC resistance meter; and \( R^i \) is the measured resistance. Then the two voltage-terminals of the AC resistance meter were switched to the both ends of each shunt resistor one after another. The AC voltage drop in each shunt resistor is determined by:

\[ V_a^i = I_a^i R_s^i \]  \[5\]

where \( R_s^i \) is the reading of the AC resistance meter. Using Eqs. \[4\] and \[5\], Eq. \[3\] becomes:

\[ R_t = R_s \frac{R^i}{R_s^i} \]  \[6\]

**Experimentation**

An MEA having an active area of 13 cm\(^2\) was used. The anode flow-field/current-collector was segmented to electrically separated 10 small segments according to its flow pattern (Fig. 2). The MEA was prepared using techniques similar to those used by Wilson and Gottesfeld \[4\]. The catalysts were house-made carbon-supported platinum and the loadings are 0.4 mgPt/cm\(^2\) for the both electrodes. An ETEK diffusion layer was used for the both sides.

An AC resistance meter (Hioki 3560), a switch box and a DC voltmeter were controlled and monitored with a PC for automatic control and data acquisition. A DC power supply was used for the FC operation and controlled under the galvanostatic mode. The gas flow rates for the both electrodes were automatically adjusted to corresponding values to the current density using mass flow controllers such that a predetermined gas stoichiometry be maintained even when the current density was varied.

Firstly the cell was operated under the condition that fully humidified oxygen and fully humidified hydrogen were introduced to the cell at 80°C at much higher flow-rates than necessary. The local current density and the local ionic resistance were obtained and the nearly uniform distributions were confirmed. Then the cathode gas was switched to air and the humidification and the gas stoichiometry for the both sides were changed to more realistic conditions. Besides, two types of gas flow direction: co-flow and counter flow, were employed.
RESULTS AND DISCUSSION

Flow Direction

Figures 3 and 4 respectively show the IV & IR characteristics and the current-density and ionic-conductivity distributions when the cell was operated under the non-humidified condition with an air and hydrogen stoichiometry of 1.25 changing the directions of the gas flows: counterflow and co-flow. In the case of co-flow, the ionic conductivity at the gas inlet area (cell 10-7) was too low (high resistance) to allow a meaningful current density in this area. Nevertheless, in the mid area (cell 6-4), the conductivity increased because of produced water, resulting in a large current density. In the gas outlet area (cell 3-1), although the conductivities were high, the current density decreased because of the oxygen depletion effect. On the contrary, in case of counterflow, the conductivities were rather homogeneous. In this case, a dry gas introduced into the area where the other gas humidifies the membrane with produced water [5] and, therefore, no area is excessively dried. As a result, the current density seemed to depend only on the oxygen concentration, which was decreased in the air outlet area.

Gas Stoichiometry

Gas stoichiometry for the both electrodes was varied from 1.25 to 3.0. The IV and IR characteristics and the current and ionic conductivity distributions are shown in Figs. 5 and 6 respectively. At a high gas stoichiometry of 3.0, although the ionic conductivity is low throughout the membrane due to the dehydration, the current density is relatively uniform and high. On the contrary, at a low gas stoichiometry of 1.25, in spite of the uniform and higher conductivity, the current density is drastically decreased from the inlet to the outlet of the air.

Gas Humidification

The gas stoichiometry was fixed to 1.25 and the gas humidification was varied. The results are shown in Figs. 7 and 8. The conductivity was low near the air inlet for low humidification conditions because of membrane dehydration and the current density distribution reflected this conductivity distribution.

CONCLUSION

The ionic resistance distribution was successfully measured as well as the current density distribution with the system described in this paper. Under realistic operation conditions: i.e. low humidification and low gas stoichiometry, the current density was heavily nonuniform and this nonuniform current density was attributed quantitatively to the membrane dehydration effect and the O₂ concentration effect. As shown in these results, this system can be used as a very powerful tool to reveal the real situation inside the cell and can give us various hints to improve the performance of PEFCs.
REFERENCES

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Fig. 1 Circuit diagram of the measuring system.

Fig. 2 Schematic image of the segmented current collector. Segmented Cell numbers are shown in the figure. The total active area is 13 cm².
Fig. 3 Comparison between co-flow and counter-flow: IV characteristic and overall ionic conductivity. Cell temperature was 80°C. Air and hydrogen were not humidified. Air and hydrogen stoichiometries were 1.25.

Fig. 4 Distributions of current density and ionic conductivity at 0.5V. The same condition with Fig.3. Ionic conductivity is employed instead of ionic resistance because the resistance could be too large to confine the graph with an appropriate y-axis range.
Fig. 5 Stoichiometry dependence of the cell performance. Stoichiometries for the anode and the cathode were equal.

Fig. 6 Stoichiometry dependence of the distributions of current density and ionic conductivity.
Fig. 7 Relative humidity (RH) dependence of the cell performance.

Fig. 8 Relative humidity dependence of the distribution at 0.3V.