Effects of fiber size and porosity in gas diffusion layers on performance of unitized regenerative fuel cells

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Abstract –Polymer electrolyte-based unitized regenerative fuel cells (URFCs) combine the fuel cell and electrolyser functions in one unit. This study focuses on the gas diffusion layers (GDLs) at oxygen side The GDLs of a URFC are responsible for an optimum humidification state and liquid water distribution in the catalyst layer during both the fuel cell mode and electrolysis mode. In our URFC experiments, titanium (Ti)-felt is used as a GDL of the oxygen electrode, which have different structural properties (porosity and fiber diameter). The relation between the properties of the GDL and the URFC performance was examined for both fuel cell and electrolysis operation modes. Experimental results showed that the cell with a Ti-felt GDL of 80μm fiber diameter had the highest round-trip efficiency due to excellent fuel cell operation under relatively high-humidity conditions despite degradation in performance in the electrolysis mode.

Keywords: proton exchange membrane, unitized regenerative fuel cell, gas diffusion layer, titanium felt, water management

1. Introduction

A polymer electrolyte membrane (PEM) electrolyser can split water into hydrogen and oxygen by the supplied DC power. And then, PEM fuel cell can make electric power from the reverse chemical reaction of PEM electrolyser. Unitized Reversible Fuel Cell (URFC) has both of functions as electrolyser and fuel cell in one unit at different modes. Therefore, URFC system, including hydrogen storage tank can be used for energy conversion and power supply system which behaves like a secondary battery as a self independent power source [1, 2].

One of key issue of the URFC is oxygen side gas diffusion layer (GDL) because the GDLs of URFC are responsible for an optimum humidification state and liquid water distribution to the catalyst layer during both operations of a fuel cell and an electrolysis. During fuel cell operation mode of a URFC, humidified gases humidify the membrane as same as general PEMFC. Condensed water from the fuel cell mode reaction may accumulate in the pores of oxygen side GDL and may prevent the oxygen transport to the catalyst sites [3, 4]. During electrolysis mode in a URFC, produced oxygen bubble from the electrolysis reaction may accumulate in the flow field through the GDL and may prevent the water transport.

Moreover, high corrosion resistance and effective mass transport of water and gas (oxygen or air) are essential for high performance and durability [5]. The GDLs of carbon material (carbon paper or carbon cloth) which is used at typical PEMFC are unsuitable for URFC due to high potential of the oxygen electrode during electrolysis mode. Carbon material tends to corrode rapidly from the cathodic potential [6-8]. Thus, for the GDL of the oxygen electrode, we use a titanium (Ti)-felt (nonwoven fabric) in which Ti fiber is bonded and sintered without adhesives.

The fully understanding of relation between liquid water transport and Ti-felt GDL characteristics is necessary in order to improve URFC performance but it has not been done in the previous work. The objective of the present work is to verify influences of the characteristic of Ti-felt GDL for the liquid water and oxygen gas transport in the
oxygen electrode of URFC.

In this paper, Ti-felt GDLs which have different structural properties (porosity and fiber diameter) were evaluated about water management ability.

2. Experiments

The configuration of the URFC with single-cell structure (Fig. 1) used here was the same as that for a PEMFC. The bipolar plate of the oxygen electrode side was titanium, and that of the hydrogen electrode side was carbon. Both bipolar plates had a parallel flow field that had 26 channels. The MEA used here was developed through collaboration between Takasago Thermal Engineering Co. and Daiki Ataka Engineering Co. Iridium oxide (IrO2) and platinum (Pt) mixed-electrocatalyst was used for the oxygen electrode, and Pt catalyst for the hydrogen electrode. The part C (MEA with an active area of 27cm²) and the GDLs were placed between the flow fields of both bipolar plates. The part A and B shows micrographs of the carbon paper (Toray 090) and Ti-felt (Bekinit) used for the GDLs. Both substrates had similar structure of an unwoven fabric made of fine fibers.

In this study, the U1 cell in Table I was considered the standard cell in which the Ti-felt GDL in the oxygen side had a fiber diameter (\( \phi \)) of 20\( \mu m \) and a porosity (\( \varepsilon \)) of 0.75. The Ti-felt GDL of the U2 and U3 cells had the same \( \varepsilon \) as the U1 cell but had different \( \phi \), namely, 40\( \mu m \) and 80\( \mu m \), respectively. The Ti-felt GDL in the U4 cell had the same fiber diameter as in the U1 cell but had different porosity as 0.50.

Table 1 Specification of constitution parts of O₂ side GDL

<table>
<thead>
<tr>
<th>Cell</th>
<th>Fiber diameter</th>
<th>porosity</th>
</tr>
</thead>
<tbody>
<tr>
<td>U1</td>
<td>20( \mu m )</td>
<td>75%</td>
</tr>
<tr>
<td>U2</td>
<td>40( \mu m )</td>
<td>75%</td>
</tr>
<tr>
<td>U3</td>
<td>80( \mu m )</td>
<td>75%</td>
</tr>
<tr>
<td>U4</td>
<td>20( \mu m )</td>
<td>50%</td>
</tr>
</tbody>
</table>

The stoichiometric ratio of the supplied pure hydrogen and oxygen in the air was kept at 1.43 and 2.50, respectively, and the humidification temperatures of both were changed arbitrarily but were always the same for a given test.

When the operating current density (\( i \)) of a fuel cell mode is higher than zero, the cell voltage deviates from the open-circuit voltage and keeps decreasing due to cell polarization represented by the overpotential [9]. We summarize three types of overpotential: activation overpotential (\( \eta_{act} \)), concentration overpotential (\( \eta_{conc} \)), and ohmic overpotential (\( \eta_{ohm} \)). Thus, the polarization curve of a fuel cell mode of URFC can be described as

\[
V_{cell} = V_{OCV} - \left[ \eta_{act} + \eta_{conc} + \eta_{ohm} \right]
\]

where \( V_{cell} \) is the cell voltage at a given \( i \), and \( V_{OCV} \) is the open-circuit voltage.

3. Results and discussion

The current-voltage (\( i-V \)) characteristics for both operation modes of a URFC were evaluated in this study by using a single cell with oxygen-side GDLs whose substrates were Ti-felt that had different fiber diameter \( \phi \) (20, 40, or 80\( \mu m \)), porosity \( \varepsilon \) (0.75 or 0.50).

3.1 Electrolysis performance

During the electrolysis, because carbon paper cannot be used as the GDL of the oxygen electrode side, only cells with Ti-felt GDL for the oxygen side were used for the electrolysis performance test. Figure 2 shows the effect of different fiber diameter and porosity by plotting the \( i-V \) characteristics. In Fig. 2 (A), the i-V characteristic of U1 is compared to those of U2 and U3 which have bigger fiber diameter GDL. The i-V curve of U1 (\( \phi = 20\mu m \) ) shows better performance than that of U2 (\( \phi = 40\mu m \) ) or U3 (\( \phi = 80\mu m \) ) even in higher current density region, and the voltage difference between U1 and U2/U3 is about 30mV at 0.93 A/cm².
Figure 2. Current density ($i$) and voltage ($V$) characteristics during electrolysis mode. (A) effect of fiber diameter ($\phi$) of Ti-felt GDL U1 ($\phi = 20\mu$m), U2 ($\phi = 40\mu$m), and U3 ($\phi = 80\mu$m) cells, and (B) Effect of porosity of Ti-felt GDL ($\epsilon$) for the oxygen electrode with U1 ($\epsilon=0.75$), U4 ($\epsilon=0.50$), cell temperature ($T_{cell}$) was 80°C.

However, we could not observe any noticeable difference of the performance between U1 and U4 which have different porosity GDLs in Figure 2 (B).

3.2 Effect of humidification temperature on fuel cell performance

Figure 3 shows the $i$-$V$ characteristics of U1 during the fuel cell mode at various humidification temperatures of the gas ($T_{fuel}$) when the cell temperature ($T_{cell}$) was 80°C. The $T_{fuel}$ was the same for both hydrogen and air (between 60 to 80°C) and controlled by a bubble humidifier.

![Figure 3](image)

Figure 3. Characteristics of current density ($i$) and voltage ($V$) during fuel cell operation of U1 cell at various humidification temperature of gases (H$_2$ and air) ($T_{fuel}$) from 60°C to 80°C, when the cell temperature is 80°C. In the U1 cell, the hydrogen side GDL is carbon paper treated with 10 wt% PTFE, and the oxygen side GDL is Ti-felt without PTFE treatment.

The curve for $T_{fuel} = 80°C$ at $i = 0.1$ to $0.2A/cm^2$ and those for $T_{fuel} = 60°C$ and $T_{fuel} = 65°C$ at $i = 0.3$ to $0.45A/cm^2$ show a rapid decrease in $V$. The decrease in fuel cell performance at $T_{fuel} = 80°C$ was caused by a rapid increase in $\eta_{conc}$. This increase in $\eta_{conc}$ was caused by a common phenomenon of flooding in GDL by supplied high humidity and produced water that blocks the fuel distribution in pores of the oxygen-side GDL. When $T_{fuel} = 60$ and 65°C, the $\eta_{ohm}$ was relatively high due to an increase in cell resistance caused by insufficient water supply to the membrane. In contrast, the curves for $T_{fuel} = 70°C$ and $T_{fuel} = 75°C$ do not show such a rapid decrease, but rather a relatively good performance (as evidenced by a steady decrease) until a high $i$. Comparison of the $i$-$V$ at $T_{fuel} = 70$ and 75°C reveals that the lower $\eta_{conc}$ at $T_{fuel} = 70°C$ yielded better $i$-$V$ characteristics than that at $T_{fuel} = 75°C$. However, $\eta_{ohm}$ at $T_{fuel} = 75°C$ was less than that at $T_{fuel} = 70°C$. This reversal in $\eta_{conc}$ and $\eta_{ohm}$ at $T_{fuel} = 70$ and 75°C indicates a critical point of “dry” and “wet” conditions during the fuel cell mode. The $\eta_{ohm}$ is an index of hydration state of the membrane and strongly depends on $T_{fuel}$ but the electrical resistance is insignificant. The cell impedance was almost constant at the entire range of current densities, and was 0.148 Ohm$^2$ and 0.098 Ohm$^2$ at 70 and 75°C of $T_{fuel}$, respectively. Because $\eta_{act}$ and $\eta_{ohm}$ were the same at both temperatures regardless of the properties of the oxygen electrode GDL, in order to verify the relationship between the properties of the Ti-felt GDL and the fuel cell performance, we focused only on the $i$-$V$ characteristics and $\eta_{conc}$ at $T_{fuel} = 70$ and 75°C.

3.3 Effect of fiber diameter of GDL on fuel cell performance

The effect of fiber diameter $\phi$ of Ti-felt GDL on fuel cell performance was also evaluated based on $i$-$V$ characteristics.

![Figure 4](image)

Figure 4. Comparisons of different fiber diameter of the Ti-felt GDL ($\phi$) of the oxygen electrode in the current density ($i$) - voltage ($V$) characteristics of fuel cell operation using U1, U4, and U5 cells at the humidification temperature of (a) 70°C and (b) 75°C, when the cell temperature is 80°C.
Figure 4 shows polarization behavior for the U1, U2, and U3 cells, where Ti-felt GDL with $\phi = 20$, 40, and 80μm, respectively, was used for the oxygen electrode (Each Ti-felt GDL had a porosity of 0.75). The $i$-$V$ characteristics for the U1, U2, and U3 cells showed no significant difference when $T_{\text{fuel}}$ = 70°C. However, the fuel cell performance improved with increasing $\phi$, with U3 ($\phi = 80\mu m$) showing the highest and most stable fuel cell performance when $T_{\text{fuel}}$ = 75°C. The $\eta_{\text{conc}}$ for U3 was relatively low and stable up to a high $i$ (>400 mA/cm$^2$).

3.4 Effect of GDL porosity on fuel cell performance

The effect of porosity $\varepsilon$ of Ti-felt GDL on fuel cell performance was evaluated based on the $i$-$V$ characteristics. Figure 5 shows the polarization behavior for the U1 and U4 cells, in which Ti-felt GDLs with $\varepsilon = 0.75$ and 0.50, respectively, were used for the oxygen electrode (Each Ti-felt GDL had the same $\phi = 20\mu m$). The fuel cell performances of U1 and U4 were almost the same when $T_{\text{fuel}}$ = 70°C. However, a lower $\varepsilon$ of Ti-felt GDL (U4, $\varepsilon = 0.50$) yielded better performance compared to a higher $\varepsilon$ (U1, $\varepsilon = 0.75$) when $T_{\text{fuel}}$ = 75°C. The better performance up to a high $i$ (>400 mA/cm$^2$) at wet conditions when $T_{\text{fuel}}$ = 75°C represented by the $i$-$V$ characteristics for U4 was mainly due to a lower $\eta_{\text{conc}}$.

![Figure 5](image)

Figure 5. Comparisons of different porosities of the Ti-felt GDL ($\varepsilon$) of the oxygen electrode in the current density ($i$) - voltage ($V$) characteristics of fuel cell operation using U1 and U6 cells at the humidification temperature of (a) 70°C and (b) 75°C, when the cell temperature is 80°C. The Ti-felt GDL of the oxygen electrode in these two cells is not treated with PTFE, and its fiber diameter is 20μm.

Figure 6. Characteristics of current density ($i$) and round-trip efficiency ($\eta_{\text{RT}}$) of electrolysis and fuel cell operation with U1, U3, and U4 cells at the humidification temperature during fuel cell operation of (a) 70 and (b) 75°C when the cell temperature of both operation 80°C.

3.5 Round-trip efficiency of URFC

The overall performance of a URFC during the electrolysis mode and fuel cell mode was evaluated here based on the round-trip efficiency ($\eta_{\text{RT}}$) for fuel cells with Ti-felt GDL with different $\phi$ and $\varepsilon$ used as the oxygen electrode. Figure 6 shows the $i$ and $\eta_{\text{RT}}$ for U1, U3, and U4 (as discussed in Sections 3.3 and 3.4). The $\eta_{\text{RT}}$ discussed here was calculated from the efficiency of cell voltage during electrolysis ($\eta_{\text{ELY}}$) and fuel cell ($\eta_{\text{FC}}$) at each $i$ as follows:

$$\eta_{\text{RT}}(i) = \eta_{\text{ELY}}(i) \times \eta_{\text{FC}}(i) = \frac{V_{\text{FC}}(i)}{V_{\text{ELY}}(i)}$$

where $V_{\text{ELY}}$ and $V_{\text{FC}}$ are the cell voltage at each $i$ during electrolysis mode and fuel cell mode, respectively, and $\eta_{\text{ELY}}$ and $\eta_{\text{FC}}$ are defined in terms of the thermo-neutral voltage ($V_{\text{tn}}$) as follows:

$$\eta_{\text{ELY}}(i) = \frac{V_{\text{tn}}}{V_{\text{ELY}}(i)}$$

$$\eta_{\text{FC}}(i) = \frac{V_{\text{tn}}}{V_{\text{FC}}(i)}$$

where $V_{\text{tn}} = 1.481V$. When $T_{\text{fuel}}$ = 75°C, however, U3, which showed the lowest $\eta_{\text{conc}}$ during the fuel cell mode, exhibited the highest $\eta_{\text{RT}}$ despite a loss of about 30mV in the electrolysis mode (Sec. 3.1). The $\eta_{\text{RT}}$ for U4 was higher than that for U1 but lower than that for U3 at high $i$ (> 400 mA/cm$^2$). The $\eta_{\text{RT}}$ of U1, U3, and U4 at $i = 740$ mA/cm$^2$ was 28, 34, and 33%, respectively, when $T_{\text{fuel}}$ = 75°C, and the $\eta_{\text{RT}}$ of U3 when $T_{\text{fuel}}$ = 75°C was the highest among all cells and conditions.
4. Conclusions

The effect of the properties of Ti-felt GDLs on the liquid water and oxygen gas transport in the oxygen electrode of a URFC were evaluated using Ti-felt of different structural properties (porosity and fiber diameter). The cell performances for both the fuel cell and electrolysis operating modes were evaluated based on the \(i-V\) characteristics. Based on our results, conclusion about relation between the properties of Ti-felt GDLs and the cell performance are as follows:

**URFC in electrolysis mode**

- When the fiber diameter of Ti-felt GDL of the oxygen electrode is small as 20\(\mu m\), the difference of porosity does not cause any noticeable effect on the cell performance.

- When we use the Ti-felt with larger fiber diameter as 40 and 80\(\mu m\) as the oxygen electrode GDL, the cell performance is degraded at high current density region compared to the standard cell with Ti-felt of 20\(\mu m\).

**URFC in fuel cell mode**

- The ohmic overpotential depends only on the humidification temperature (relative humidity) of gas regardless of the properties of the GDL substrate. Thus, the cell performance at each respective humidification temperature is mainly determined by the concentration overpotential.

- When the humidification temperature (\(T_{\text{fuel}}\)) is relatively low at 70\(^\circ\)C and the operation condition is dry during fuel cell mode, the properties of GDL do not significantly affect to the \(i-V\) characteristics.

- When the \(T_{\text{fuel}}\) is relatively high at 75\(^\circ\)C and the operation condition is wet, Ti-felts with larger fiber diameter or lower porosity are excellent because different pore distribution must give the effect of securing the path for gas during the water discharge.

**URFC in Round-trip efficiency**

- When the cell temperature at the both operations of fuel cell and electrolysis is the same as 80\(^\circ\)C, the cell with Ti-felt GDL of larger fiber diameter as 80\(\mu m\) shows the highest round-trip efficiency as a whole because of excellent fuel cell operation under high-humidity conditions in spite of the performance degradation in the electrolysis mode.

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References


