



# Performance enhancement of polymer electrolyte fuel cells by combining liquid removal mechanisms of a gas diffusion layer with wettability distribution and a gas channel with microgrooves



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## HIGHLIGHTS

- A novel PEFC with a combination of new types of GDL and gas channel was proposed.
- GDL with planer distributed wettability to keep oxygen diffusion paths was adopted.
- Gas channel with slanted micro-grooves to remove excess water from GDL was adopted.
- Current density limit and maximum power density were improved by the combination.
- The stability of the cell voltage was markedly improved.

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## ABSTRACT

Although polymer electrolyte fuel cells (PEFCs) are commercially available, there are still many problems that need to be addressed to improve their performance and increase their usage. At a high current density, generated water accumulates in the gas diffusion layer and in the gas channels of the cathode. This excess water obstructs oxygen transport, and as a result, cell performance is greatly reduced. To improve the cell performance, the effective removal of the generated water and the promotion of oxygen diffusion in the gas diffusion layer (GDL) are necessary. In this study, two functions proposed in previous reports were combined and applied to a PEFC: a hybrid GDL to form an oxygen diffusion path using a wettability distribution and a gas separator with microgrooves to enhance liquid removal. For a PEFC with a hybrid GDL and a gas separator with microgrooves, the concentration overvoltage of the PEFC was reduced, and the current density limit and maximum power density were increased compared with a conventional PEFC. Moreover, the stability of the cell voltage was markedly improved.

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## 1. Introduction

Moisture management is critical for improving the performance of a polymer electrolyte fuel cell (PEFC). At high current density, the generated water accumulates in the gas diffusion layer (GDL) and gas channels on the cathode side of the PEFC. The cell performance is greatly reduced by the excess water, which obstructs oxygen transport. To improve the cell performance, an effective removal of

the generated water is necessary. Past studies have examined methods to improve moisture control and liquid water removal performance on the cathode side to control the water accumulation in a PEFC. Approaches have included surface treatments, or finishing, of the GDL and the use of gas channels. For example, studies have considered using hydrophobic materials, such as polytetrafluoroethylene (PTFE) and fluorinated ethylene propylene (FEP), for the hydrophobic treatment of the GDL [1–4]. The effects of a microporous layer (MPL), PTFE content [5–7], and component fractions of hydrophobic and hydrophilic materials [8–10] on cell performance have also been examined. Moisture control using GDLs perforated with laser-cut holes [11,12] and multiple

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hydrophobic and hydrophilic MPLs [13,14] have also been tried. To enhance oxygen gas diffusion, control of the liquid water movement using a GDL with a different wettability in the planar direction has been considered [15,16]. Changes to the gas channel, such as channel configuration, have been examined for their effect on PEFC performance. Various flow channel types, such as parallel, serpentine, interdigitated, and hybrid channels, have been investigated [19–28]. Other studies related to the configuration of the flow channel have considered passive water removal by capillary droplet actuation [29] and channel wall wettability [30,31]. To increase the removal of liquid water, Okabe and Utaka [32] proposed arranging microgrooves inside gas channels and experimentally demonstrated the effectiveness of the microgroove arrangement. Koresawa and Utaka [33] applied microgrooves to a real PEFC and demonstrated enhancements to the current density limit. In the present study, two methods were applied to enhancing the performance of a real PEFC, i.e., combining the use of a GDL with a different wettability in the planar direction [15,16] and a microgroove arrangement inside a gas separator [32,33].

## 2. Summary of previous reports and objective relevance to this study

To improve the oxygen diffusion characteristics of liquid water, Utaka et al. [15] proposed a new configuration where two porous media with different wettabilities (i.e., hydrophobic and hydrophilic) were alternately arranged in the planar direction of a GDL (i.e., a hybrid configuration). Liquid water was moved from the hydrophobic medium to the hydrophilic medium due to capillary pressure originating from the difference in wettability. Voids in the hydrophobic medium were aligned in the direction of oxygen diffusion, enhancing oxygen diffusion. Water distribution profiles in the microporous media were visualized by X-ray computed tomography and oxygen diffusion characteristics were measured by Galvanic cell oxygen absorber apparatus [17,18] simultaneously, and the high oxygen diffusivity mechanisms were examined using model apparatus for the hybrid GDL, which was thicker (2.5 mm) than a conventional GDL.

Furthermore the hybrid configuration was applied to carbon paper used for GDLs [16]. The formation of oxygen diffusion paths was confirmed by X-ray radiography, where voids in the hybrid GDL were first formed in the hydrophobic regions and then spread to the untreated wetting region. The application of a hybrid GDL enhanced the oxygen diffusion characteristics. Although these results show the potential for a hybrid GDL, for the realization of actual power generation applications, liquid water should be effectively removed from the GDL.

At the same time, reduce the accumulation of liquid water on the GDL surface, Utaka et al. proposed an arrangement of thin microgrooves with axes tilted toward the surrounding air flow on the side walls, the upper wall inside the gas channel and isolated their effect by using model apparatus [32]. The water produced from the GDL was discharged along microgrooves facing the top of the GDL by capillary forces and air flow shear. Laser-induced fluorescence was used to measure the water velocity in the microgrooves. It was shown the microgrooves for an inclination angle of  $20^\circ$  were confirmed to be effective throughout a gas channel with a total length of 200 mm. Moreover, Koresawa and Utaka [33] demonstrated the effectiveness of microgrooves for an actual PEFC by applying the configuration to an actual PEFC with a straight gas channel and a total length of 200 mm. The PEFC with microgrooves showed a better performance than the conventional PEFC without microgrooves.

Thus, the combination of a hybrid GDL and a gas channel with microgrooves should facilitate a more effective removal of liquid

water. As shown in Fig. 1, an oxygen diffusion path in the hydrophobic region of the GDL was ensured by movement from the hydrophobic region to the hydrophilic region. However, liquid water easily accumulated in the hydrophilic region. The PEFC performance was improved by combining a gas channel with microgrooves and a hybrid GDL to prevent or minimize the accumulation of excessive liquid water on the GDL surface.

The objective of this study was to enhance the power generation of an actual PEFC by applying a hybrid GDL with planar distributed wettability and a gas channel with microgrooves.

## 3. Experimental apparatus and method

### 3.1. Experimental system

Pure hydrogen gas and air (approximately 78% nitrogen, approximately 21% oxygen, and approximately 1% argon) were supplied to the anode and cathode, respectively, from gas cylinders via a mass flow controller to control the flow rate. A humidifier was used to maintain water vapor saturation. To avoid cooling condensation, pipes from the humidifier to the PEFC were heated and covered with an insulating material. To operate the PEFC at a fixed temperature, separators of the anode and cathode were connected to a thermostatic bath, and water was circulated at a constant temperature. The cell voltage was measured with a data logger, and the cell resistance was measured with an LCR meter. The current load on the PEFC was changed in a stepwise fashion based on the electrical load. The results were used to determine the current-voltage characteristics and the cell resistance via the current density.

Fig. 2(a)–(c) show the entire PEFC apparatus, hybrid GDL and microgroove construction, respectively. To evaluate the differences in PEFC performance with and without microgrooves, a separator with a long gas channel was manufactured to mimic the application of this PEFC to an actual PEFC as shown in Fig. 2(a). The gas channel had a rectangular cross-section with width and height of  $d_g = h_g = 1.0$  mm and a length of 200 mm. Eleven gas channels were arranged in parallel. The membrane electrode assembly (MEA) of the PEFC had a reaction area of  $42$  cm<sup>2</sup> ( $21$  mm  $\times$   $200$  mm). For the GDL, carbon paper (Toray Industries, TGP-H-060) was hydrophobically treated and coated with an MPL. For the gaskets, a silicone rubber sheet was used on the anode side, and a silicone sponge sheet was used on the cathode side.

### 3.2. Structure of hybrid GDL

A hydrophobically treated carbon paper GDL (Toray Industries, INC. TGP-H-060) with an MPL coating on one side was used. The

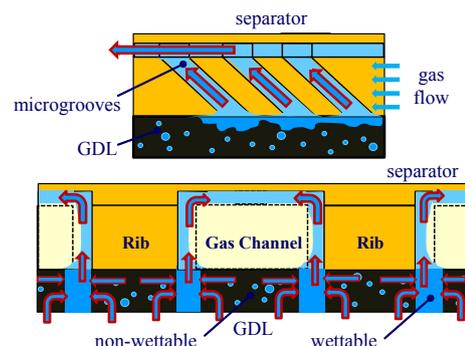
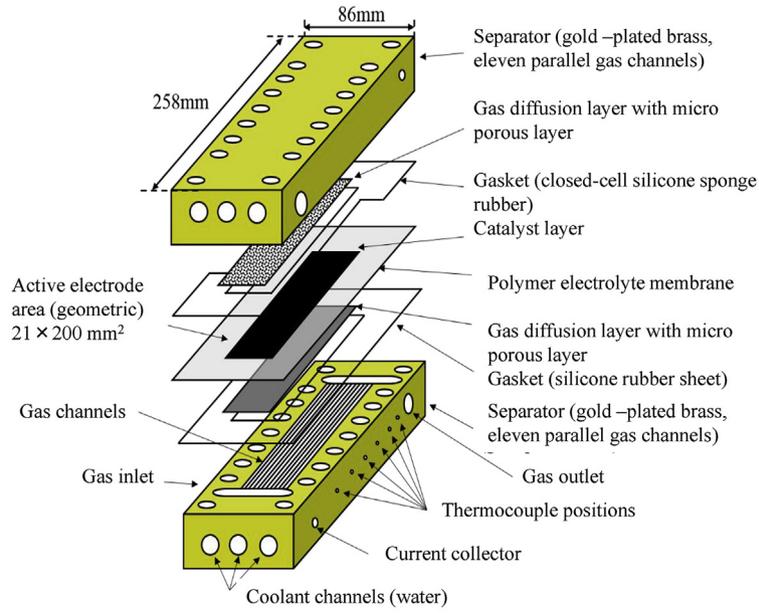


Fig. 1. Schematic of water movement with combined hybrid GDL and microgrooves.

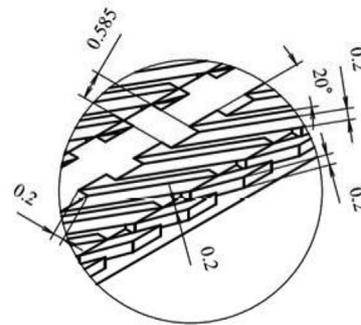
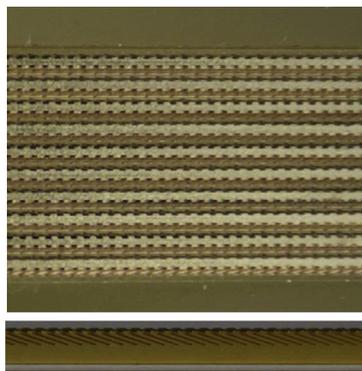


(a) Polymer electrolyte fuel cell apparatus



(b)

Hybrid GDL with planer distributed wettability



(c) Separator with microgrooves and details of microgroove construction

Fig. 2. Schematics of PEFC apparatus, hybrid GDL with planar distributed wettability and gas channel with microgrooves.

GDL size was the same as the reaction area, i.e., 21 mm × 200 mm. Fig. 2(b) shows an example of the hybrid GDL used in this study. Hydrophobic-treated and untreated regions were alternately arranged in a linear fashion at 1.0 mm intervals to facilitate liquid water discharge into the hydrophilic region. The microgrooves were tilted at a 45° angle to the gas flow direction to avoid contacting bridging between neighboring wettable areas. The liquid water moved diagonally backward to the channel wall and was siphoned to the microgrooves. A hybrid GDL was used on the cathode side, and a GDL with uniform hydrophobic treatment was used on the anode side.

### 3.3. Structure of gas channel with microgrooves

Fig. 2(c) shows a partial schematic view of the gas channel with microgrooves. Microgrooves having their axis tilted 20° to the gas flow and a square cross-section of 1.0 × 1.0 mm<sup>2</sup> were arranged on both side walls and the upper wall at 0.4 mm intervals. These microgrooves were connected to a single microgroove arranged at the center of the upper wall. The width and depth of the microgrooves were both 0.2 mm. Because a small amount of liquid water accumulated upstream of the gas channel, the microgrooves were arranged 50–200 mm from the gas channel inlet. A gas channel with

microgrooves was used as the cathode separator, and a gas channel without microgrooves was used as the anode separator. The separators were made from brass. The ribs and channels of the separator were separately fabricated and assembled after coating with gold. The gold-coated surfaces were confirmed to be hydrophilic.

### 3.4. Determination of clamping pressure of MEA

Because the power generation of a PEFC depends on the clamping pressure [33–35], the appropriate clamping pressure was first determined. To examine the effect of the clamping pressure on the power generation, experiments where the clamping pressure was varied under constant conditions of a 70 °C cell temperature, a 70% relative humidity, and an air flow velocity of 6.0 m/s. The relations between the current density and cell voltage and between the power density and current density were examined. Because the maximum power density was obtained at a clamping pressure of 1.23 MPa, the standard clamping pressure was determined to be 1.23 MPa.

Table 1 lists the conditions for the gas channel and the hybrid GDL determined in the above experiments. To evaluate the PEFC performance, experiments were carried out with a variable air flow velocity. The air velocity was calculated from the air flow rate prior to humidification. The utilization and stoichiometry were calculated as the standard flow rate of the supply gas at a power density of 2.0 A/cm<sup>2</sup>.

To evaluate the performance of the PEFC combining the hybrid GDL and the gas channel with microgrooves, four configurations were investigated with different air velocities:

- (1) A normal PEFC (a conventional separator without microgrooves and a GDL with uniform hydrophobic coating);
- (2) A hybrid PEFC (a hybrid GDL and a conventional separator without microgrooves);
- (3) A PEFC with microgrooves (a separator with microgrooves and a GDL with uniform hydrophobic coating); and
- (4) A PEFC with microgrooves and a hybrid GDL (a separator with microgrooves and a GDL with wettability distribution).

## 4. Experimental results and discussion

### 4.1. Effects of cell temperature and relative humidity

The power generation characteristics of a PEFC are known to be affected by the cell temperature and relative humidity. To

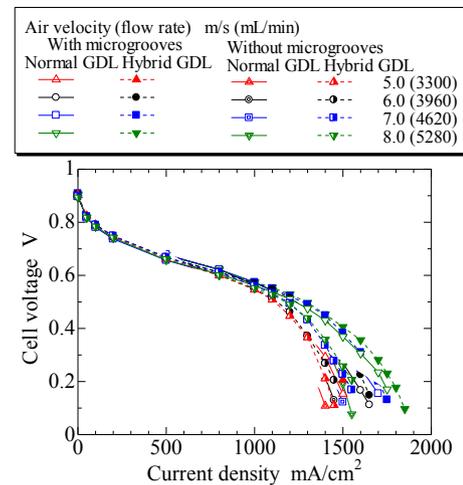


Fig. 3. Effects of microgrooves and hybrid GDL on cell performance (Current density vs. cell voltage curve).

determine the appropriate cell temperature and relative humidity for the air supply gas, experiments were carried out by individually varying the cell temperature and relative humidity under the base conditions of a 70 °C cell temperature, 70% relative humidity, 6.0 m/s air flow velocity, and 1.23 MPa clamping pressure. The appropriate cell temperature and relative humidity were determined by experiments with the cell temperature and the relative humidity as the independent variables. The maximum power densities were obtained at a cell temperature of 80 °C and a relative humidity of 50%, and the maximum current densities were obtained at a cell temperature of 70 °C and a relative humidity of 70%. Thus, a cell temperature of 70 °C and a relative humidity of 70%, which were the conditions at which the maximum power density was obtained, were selected as the standard conditions.

### 4.2. Effects of air velocity on cell performance

Figs. 3 and 4 show the experimental results when the supply air velocity was changed from 5.0 m/s to 8.0 m/s. Fig. 3 shows the relationship between the current density and cell voltage, and Fig. 4 shows the relationship between the power density and cell voltage. The results for the normal PEFC, hybrid PEFC, PEFC with microgrooves, and PEFC with microgrooves and a hybrid GDL were

Table 1  
Experimental conditions.

Gas channel and gas flow	Flow design	Parallel
	Gas channel width (mm)	1.0
	Gas channel depth (mm)	1.0
	Rib width (mm)	1.0
	Inclination angle of microgrooves (°)	20
	Width of microgrooves (mm)	0.2
	Depth of microgrooves (mm)	0.2
	Active electrode area (geometric) (cm <sup>2</sup> )	2.1 × 20
	Clamping pressure (MPa)	1.23
	Cell temperature (°C)	70
	Relative humidity (%)	70
	Gas velocity (flow rate)	
	Hydrogen m/s (mL/min)	1.2 (800)
	Hydrogen utilization (stoichiometry)	0.80 (1.25)
Hybrid GDL	Air m/s (mL/min)	5.0 (3300), 6.0 (3960), 7.0 (4620), 8.0 (5280)
	Air utilization (stoichiometry)	5.0 m/s: 0.46 (2.17), 6.0 m/s: 0.38 (2.60) 7.0 m/s: 0.33 (3.04), 8.0 m/s: 0.29 (3.47)
	Width of stripes of non-wettable and wettable (mm)	Approximately 1.0
	Stripe angle of inclination to flow direction (°)	45

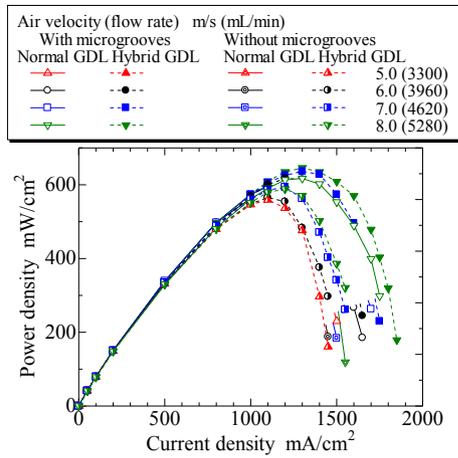


Fig. 4. Effects of microgrooves and hybrid GDL on cell performance (Power density curve).

compared. The highest performance level was achieved by the PEFC with microgrooves and a hybrid GDL. For this configuration, the upper limit of the current density, maximum power density, and curve of the power density shifted toward a higher current density. These phenomena could be explained by the following reasons. First, in the hybrid GDL with a wettability distribution, an oxygen diffusion path was formed in the hydrophobic region by the movement of liquid water from the hydrophobic region to the hydrophilic region; this finding was previously supported in earlier studies [15,16]. Second, the liquid water that accumulated in the hydrophobic region was removed through the microgrooves. Near 1000 mA/cm<sup>2</sup>, the concentration overvoltage becomes large due to the increased generation of liquid water; at this point, significant differences among the different PEFC configurations start appearing. The PEFC with microgrooves performed better than the normal PEFC, which agreed with the results of a previous study [33]. When a PEFC with microgrooves and a hybrid GDL was used, the increase in performance was large. This was due to the presence of a hybrid GDL, which facilitated a high oxygen diffusivity when containing liquid water and affected the PEFC performance. There were small differences in the current density limit and maximum power density between the normal and hybrid PEFCs. When only the hybrid GDL was applied an oxygen diffusion path was ensured, allowing for liquid water to easily accumulate in the hydrophilic region of the GDL. The liquid water in the hydrophilic region of the GDL was effectively removed when the hybrid GDL was combined with a separator containing microgrooves. This enhanced the cell performance.

Fig. 5 shows the current density limit and maximum power density under 0.2 V cell voltage. The air velocity showed similar qualitative effects, i.e., the current density limit and maximum power density increased with the air velocity. However, an increase in the air velocity was found to more significantly affect the increase in the current limit density when the separator was patterned with microgrooves than without. This was because the increase in the air velocity increased the air shearing force on the liquid water within the microgrooves, which improved the discharge of the liquid water within the microgrooves. With regard to the maximum power density, the highest power density was obtained by the PEFCs with microgrooves and a hybrid GDL for all air velocities. Among the power densities obtained by the other configurations, no significant differences were observed. At an 8.0 m/s air velocity, the new PEFC design with microgrooves and a hybrid GDL improved the critical current density at a cell voltage of

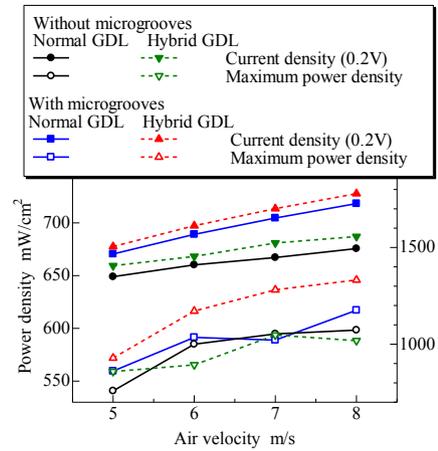
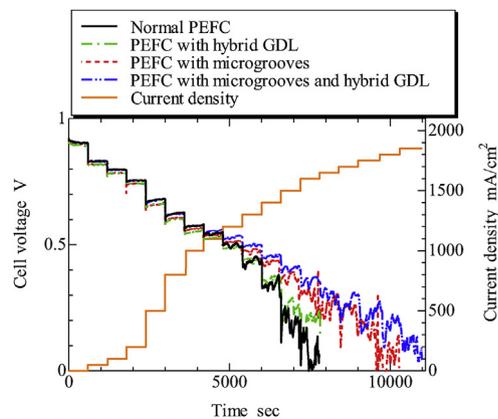


Fig. 5. Effect of air velocity on maximum power and critical current densities for application of hybrid GDL and microgrooves.

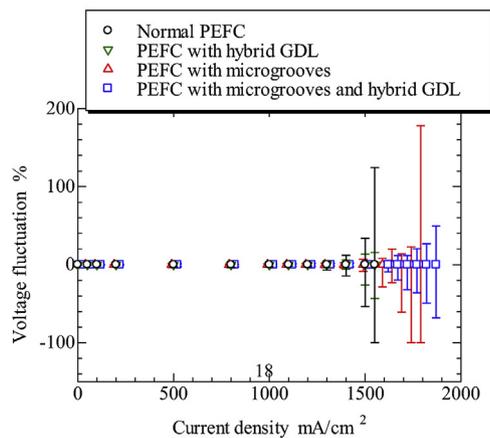
0.2 V by approximately 19.5% relative to that of a normal PEFC (from 1490 mA/cm<sup>2</sup> to 1780 mA/cm<sup>2</sup>). Similarly, the maximum power density was improved by approximately 8.0% from 598 mW/cm<sup>2</sup> to 646 mW/cm<sup>2</sup>.

### 4.3. Stability of output voltage

The hybrid GDL was expected to reduce voltage fluctuations induced by the liquid water through the formation of oxygen diffusion paths in the hydrophobic region of the GDL. Microgrooves were expected to reduce the voltage fluctuation induced by the accumulation of liquid water inside the gas separator by removing the liquid water on the GDL surface. To confirm these effects, variations in the cell voltage over time for the four GDL and separator conditions (i.e., normal PEFC, hybrid PEFC, PEFC with microgrooves, and PEFC with microgrooves and a hybrid GDL) were considered, as shown in Fig. 6. The experimental conditions under which the highest power density was obtained were a clamping pressure of 1.23 MPa, a cell temperature of 70 °C, a relative humidity of 70%, a hydrogen velocity of 1.2 m/s, and an air velocity of 8.0 m/s. As shown in Fig. 6(a), at a low current density, the different PEFC combinations showed little differences in cell voltage variations over time. However, at a high current density of over 1300 mA/m<sup>2</sup>, the hybrid GDL and microgrooves considerably reduced the fluctuations in the cell voltage. Fig. 6(b) shows the fluctuations from the average voltage as error bars. The voltage fluctuation rates were calculated from the average cell voltage  $V_{ave}$ , the minimum cell voltage  $V_{min}$ , and the maximum cell voltage  $V_{max}$ . Thus,  $(V_{min} - V_{ave})/V_{ave}$  [%] is the relative difference between the minimum and average voltages, and  $(V_{max} - V_{ave})/V_{ave}$  [%] is the relative difference between the maximum and average voltages. If  $V_{ave} - V_{min}$  and  $V_{max} - V_{ave}$  are small, the cell voltage is steady. The hybrid GDL alone was found to stabilize the cell voltage when compared with the normal PEFC and the PEFC with hybrid GDL. The microgrooves were also found to stabilize the cell voltage compared with a normal separator without microgrooves. The microgrooves allowed the liquid water to be rapidly discharged without an unsteady accumulation on the GDL surface. Thus, enhancing the liquid water removal was important for enhancing the cell voltage stability. The combination of the hybrid GDL and microgrooves in a PEFC was found to considerably enhance the stability of the cell voltage at high current densities. To reduce the effects of the concentration overvoltage and cell voltage fluctuation induced by liquid water, this combination was an effective



(a) Time variation of cell voltage with current density



(b) Normalized cell voltage fluctuations

Fig. 6. Output fluctuations.

approach. The hybrid GDL enhanced the oxygen diffusion by forming oxygen diffusion paths, and the microgrooves within the separator enhanced the liquid removal from the GDL surface.

## 5. Conclusions

In this study, two functions proposed in previous papers were combined and applied to a PEFC, i.e., a hybrid GDL with oxygen diffusion paths and wettability distribution and a gas separator with microgrooves, which enhanced liquid removal. This structure was adopted for the gas channel of an actual PEFC with an effective length of 200 mm. Improvements in the PEFC performance were experimentally examined. Based on the results, the following conclusions were obtained.

- (1) The PEFC with microgrooves and a hybrid GDL reduced the concentration overvoltage and enhanced the current density limit and maximum power density compared with the normal PEFC, hybrid PEFC, and PEFC with microgrooves. At an air velocity of 8.0 m/s, the PEFC with microgrooves and a hybrid GDL improved the current density limit and maximum power density by approximately 19.5% and 8.0%, respectively, when compared with the normal PEFC.
- (2) The PEFC with microgrooves and a hybrid GDL reduced the cell voltage fluctuation induced by the generated liquid water and improved the stability of the cell voltage. This was

due to an oxygen diffusion path formed by the movement of liquid water through the hybrid GDL and the discharge of liquid water from the GDL surface and the gas channels.

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