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# Novel gas diffusion layer with water management function for PEMFC<sup> $\ddagger$ </sup>

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#### Abstract

The water management of the membrane electrode assembly (MEA) is a critical technology in developing a polymer electrolyte membrane fuel cell (PEMFC). To maintain the MEA with a satisfactory water content and distribution, a novel gas diffusion layer (GDL) was designed by inserting a water management layer (WML) between the traditional GDL and the catalyst layer of the PEMFC. A simulator was developed for the optimization of the GDL, where the water distribution in the electrode and the profile of the water transport in the polymer membrane could be predicted. For this purpose, the water vapor permeability of the GDL, which was the main variable, was investigated under conditions similar to the PEMFC. Furthermore, the cell performance of the PEMFC with the GDL was tested. The testing results indicated that the water management in the MEA could be significantly improved by using the WML. © 2004 Elsevier B.V. All rights reserved.

Keywords: Polymer electrolyte membrane fuel cell; Water management; Gas diffusion layer; Water transport; Cell performance

#### 1. Introduction

The polymer electrolyte membrane fuel cell (PEMFC) used for residential co-generation has been developed for the pre-commercial trial stage. For instance, the Osaka Gas Co., Japan, presented the possibility of operating 1 kW PEMFC systems for over 10,000 h [1]. Up to now, only a perfluorosulfonic polymer electrolyte, such as Nafion can be durable over that long a time in the PEMFC as the polymer electrolyte membrane [2]. The polymer electrolyte membrane in the PEMFC must be hydrated during operation, but flooding of the electrode must be avoided. Flooding and dehydration are well-known factors that negatively effect the performance of the PEMFC, and the temporary degradation effects of a flooded or dry cell are obvious [3,4]. Therefore, the water retention/removal properties of the PEMFC are important in maintaining a balance between good gas diffusion and high ionic conductivity of the electrode [5]. Especially, for commercial applications, proper water management must be achieved during the PEMFC operation.

Hydration of the polymer electrolyte in the PEMFC has been extensively studied [6–9]. The conventional design in-

volves supplying a water-saturated reactant gas, which is typically achieved by bubbling water in a humidifier. Water vapor injection and direct liquid water injection designs have also been recently presented. The water vapor injection design calls for a complex electrode and separator, possibly consisting of porous backing plates for continuous water addition along the flow channels. Components for this design would be more expensive to construct and difficult to operate [7]. The direct liquid water injection design is attractive because of its simplicity and efficiency. However, application of the water injection in a PEMFC was very difficult because excessive amounts lead to electrode flooding [8]. Others approaches such as a recirculation design would also be satisfactory, however, this method requires an external piece of equipment to recirculate the gas [9].

Of those humidification approaches, humidifying the reactant gas in advance is perhaps the most easy and reliable method to hydrate the polymer electrolyte. The inlet reactant gases are humidified by a humidification exchanger situated between the supplied and exhausted gases [10]. Considering the reasonable dimensions of the exchanger, a high dew point at the gas inlet cannot be expected. A lower humidification gas is fed, as shown in Fig. 1, there is a tendency that the relative humidity of the gas monotonously increases from the cell inlet to exit [11]. Therefore, when the relative humidity of the reactant gas at the cell inlet is higher, as shown in Fig. 1(a), it will be noted that liquid water may be present, and consequently, flooding of the electrode. On the other hand, as shown in Fig. 1(b), for a lower relative humidity of the inlet gas, drying-out of the polymer elec-

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Fig. 1. Distribution of the relative humidity in the electrode of PEMFC in the case of (a) high humidity of the inlet gas, and (b) low humidity of the inlet gas. The fuel gas and air were fed in the case of co-flowing to the anode and cathode, respectively.

trolyte near the cell inlet region may occur. Any improper water balance (either too wet or too dry) not only leads to performance losses, but also accelerates the degradation of the cell [12].

Therefore, proper water management is vital to ensuring the high performance and long life of PEMFC. In addition, better water management can reduce or eliminate the need for humidification. Water management can be gained by the design and operation of PEMFC [13]. The latter has been discussed regarding the pressure drop, temperature rise and counter-flow operation [14]. The former contains the design of the separator gas flow [15], the membrane electrode assembly (MEA) and the gas diffusion layer (GDL) [5,16–18]. Although the GDL has obtained less attention for optimization, it is one of the more important components in a PEMFC. The basic function of the GDL is to provide a homogenous distribution of the reactant gases for the reaction sites. With proper modification of the GDL, it is expected that additional functions, such as water management can be obtained, resulting in adequate hydration of the polymer electrolyte, and alleviating flooding of the electrode.

In this study, the water management function was achieved by employing a material (generally polytetrafluoroethylene (PTFE) powder mixed with carbon black, etc.), which was dry-dispersed on a traditional GDL (generally, a type of carbon paper), and then sintered at high temperature. The water management layer (WML) was situated between the carbon paper and the catalyst layer of the PEMFC. In addition, the WML was non-uniform designed with gradients in the structures between the cell inlet and outlet, which may provide some additional interesting possibilities of the PEMFC [13].

To help the design of the novel GDL, a computer simulator was developed for the calculation of the water distribution and water transport of the PEMFC. The main variable of the simulator was the water vapor permeability of the GDL, which was characterized using a special apparatus constructed in house, under conditions similar to the operation of an actual PEMFC. Based on the simulation results, the optimal GDL was obtained. Cell performances of the PEMFC using the optimal GDL and the traditional GDL were tested and compared under atmospheric pressure at 80 °C.

#### 2. Experimental

The novel GDL was prepared by coating a composite layer onto the traditional GDL of a carbon paper using a dry process. The composite was prepared by dispersing PTFE powder into nano-sized carbon black. The GDL was heated at 350 °C for 10 min under atmospheric conditions, which allowed us to remove the dispersion agents from the PTFE, and to sinter the PTFE into the WML. Scanning electron microscope observations identified that the WML was a microporous layer with the PTFE powder being uniformly dispersed in the carbon black. The micropore size of the GDL was characterized by porosimetry using a mercury intrusion porosimeter. Changes in the composition and types of conductive particles and PTFE powder, properties of the WML, such as hydrophobic and hydrophilic, characterized by the contact angle method, were monitored. Electric resistance measurements indicated that there was little difference of the electric resistance between the novel GDL and the carbon paper substrate.

In order to numerically evaluate the PEMFC performance with the novel GDL, a computer simulator was developed by considering the electrochemical process, mass and energy conservation equations. The simulator was used to analysis the water distribution and water transport of the PEMFC. The main variable component of the simulator was the water vapor permeability of the GDL, which was detected as the follows.

Fig. 2 shows the special apparatus constructed in-house for the measurement of the water vapor permeability of the GDL. The measurement conditions were similar to that of a PEMFC. The GDL was placed horizontally in the apparatus. Below the GDL, a water bath with a temperature controller was placed. The distance from the water surface to the GDL was kept at about 10 cm. The environment be-



Fig. 2. Simulated apparatus for the water vapor permeability measurement of the gas diffusion layer.

low the GDL was considered to be water-saturated at 80 °C. Above the GDL, a stainless steel separator with dimensions of 3 cm  $\times$  15 cm was placed. The depth and width of the gas flow channels on the separator were 0.47 and 1.0 mm, respectively. The supplied gas flowed between the separator and GDL. The gas flow rate was 0–5.0 l/min, and controlled by a flowmeter. The exit gas was cooled about to 0 °C by a condensator. The condensed water was collected in a flask that was weighted. By using this procedure, the water vapor permeability of the GDL was examined, and the relative humidity of the exit gas at 80 °C was calculated. The results were used as input data of the simulator for the calculation of the relative humidity distributions in the catalyst layers, and the water transport through the polymer membrane. Based on this simulation, the optimized GDL was prepared.

The performance of the fuel cell containing the optimized GDL was tested using the Fuel Cell Performance Testing Equipment (Chino Corp., Japan). This system was equipped with humidifiers for the reactant gas, an electronic load (Kikusui Electronics Corp., Japan) for the electric current controlling, a personal computer for the equipment monitoring and the data output, and a single  $3 \text{ cm} \times 15 \text{ cm}$  PEMFC connected to the test equipment. MEA consisting of a catalyst-coated membrane and two GDLs was incorporated into the fuel cell for testing. The PEMFC was operated under atmospheric pressure at 80 °C.

#### 3. Results and discussion

# 3.1. Novel gas diffusion layer with water management function

The carbon black in a PEMFC is generally used as the support material for the dispersion of the noble metal particles due to its high corrosion resistance and electric conductivity. The effects of the carbon black on the PEMFC performance has been reported by Uchida et. al. [19]. The electrochemical reaction site is a three-phase interface, where reactant gas, hydrogen protons and electrons are transported through the hydrophobic pore, the hydrated electrolyte, and the carbon black, respectively. Usually, the PTFE particle in the catalyst layer forming the hydrophobic pore for the transport of the reactant gas can also prevent the water va-

por from condensing and gathering, thus, avoiding flooding of the electrode.

With regard to the special structure of the carbon black and the high hydrophobic properties of the PTFE, a WML mainly composed of these two materials was designed. It is expected that by inserting the WML into a PEMFC, the problems of drying-out of the polymer electrolyte and the flooding of the electrode could be simultaneously solved [20]. As shown in Fig. 3(a), the WML was inserted between the traditional GDL and the catalyst layer of the PEMFC. A similar design, in the terms of a three-layer gas diffusion electrode, has also been studied [1,21-24]. The properties of the WML, such as the composition, thickness, porosity and hydrophobicity were gradually changed along with the gas flow direction, so that the effective water management of MEA becomes possible [13,23-25]. The advantages of providing in-plane non-uniformity and gradients in the fuel cell structure and conditions, especially, varying the electrochemical activity characteristics in and across the plane of the electrode to accommodate variations in the gas concentration and flow, water flux and temperature variation have been discussed by Wilkinson and St-Pierre [13]. The water distribution in the catalyst layer is considered by the profiles shown in Fig. 3(b). For comparison, the water distribution in the catalyst layer of a traditional PEMFC was also plotted as the broken curve. The relative humidity of the traditional PEMFC monotonously increases, resulting in drying-out of the polyelectrolyte near the inlet region. However, in the PEMFC of this study, the relative humidity of the



Fig. 3. Construction and relative humidity distribution in the electrode of the developed PEMFC. (cathode side).



Fig. 4. Specific volume distribution in the gas diffusion layer prepared from WML coated carbon paper. Insert shows the specific volume distribution of the WML at the range of  $0.01-10 \,\mu$ m.

catalyst layer was considered to fluctuate in the high region of 80–100%. As a result, the polymer electrolyte between the anode and cathode catalyst layers can maintain its hydrated state and thus its high proton conductivity during the PEMFC operation.

The water management function of the GDL results from the micropore structure and the hydrophobic/hydrophilic nanoseparation of the WML. As shown in Fig. 4, the typical specific pore volume distribution of the GDL was arbitrarily divided into two regions with a boundary at about a 10 µm pore radius, identified as micropores in the area between 0.01 and 10  $\mu$ m, and the macropores having a size between 10 and 100 µm. The macropores were located in the carbon fiber of the carbon paper, while the micropores were situated among the carbon black aggregates and agglomerations. The micropores in the WML were used as the gas channels for the reactant gas moving to the catalyst reaction site. However, with proper PTFE treatment, the micropores probably hinders the formation of larger water drops, thus avoiding flooding of the electrode. In other words, water transport in the WML can be effectively controlled. Moreover, the WML on the carbon paper can play the role of obstructing the catalyst particles getting into the pores of the carbon paper [26].

#### 3.2. Water vapor permeability of the novel GDL

The water vapor permeability of the GDL was measured using the special equipment shown in Fig. 2, as a function of the flow rate of the feed gas. The conditions of the measurement were similar to that of the actual PEMFC during operation at 80 °C and atmospheric pressure. The effects of the parameters of the GDL, such as thickness of the carbon paper and the WML, types of carbon black in the WML, on the water vapor permeability of the GDL were detected.

Fig. 5 shows typical results for the water vapor permeability of (a) the carbon papers (traditional GDLs with different thicknesses), and (b) the novel GDLs (with different WML thicknesses) and at 80 °C, as a function of the gas flow. As expected, the relative humidity of the outlet gas decreases



Fig. 5. Water vapor permeability through (a) the carbon paper with different thicknesses (( $\bigcirc$ ) 180 µm, ( $\square$ ) 280 µm, and ( $\triangle$ ) 350 µm), and (b) the novel GDL with different WML thicknesses (( $\textcircled{\bullet}$ ) 20 µm, ( $\blacksquare$ ) 50 µm, and ( $\bigstar$ ) 100 µm).

with the increase in the gas flow rate. Especially, the relative humidity of the novel GDL, which was coated with WML, decreased from 64 to 18% when the gas flow rate increased from 0.5 to 5.0 l/min. It was surprising that neither the thickness of the carbon paper nor the thickness of the WML had an effect on the water vapor permeability. In the entire flow rate region, the relative humidity of the outlet gas of the novel GDL was about 30% lower than that of the carbon paper. The decreased water vapor permeability of the novel GDL is very advantageous for the PEMFC, which can keep the polymer electrolyte in the MEA from drying out.

Fig. 6 shows the effect of the different carbon blacks (Vulcan XC72R from Cabot Corp. and Ketjenblack EC-600JD from Akzo Nobel, Inc.) in the WML on the water vapor permeability of the GDL. The characteristics of the carbon blacks are shown in Table 1. Although the particle sizes of the carbon blacks are almost the same, the Ketjenblack has a much larger specific surface area than that of the Vulcan XC72R, due to its micropore structure in the primary parti-



Fig. 6. Effect of carbon black in the WML on the water vapor permeability of the novel GDL.

Table 1 Characteristics of the carbon black

Carbon black	Producer	Particle size (nm)	Specfic surface area (m <sup>2</sup> /g)
Vulcan XC72R	Cabot Corp.	30	254
Ketjenbalck	Akzo Nobel, Inc.	34	1270
EC-600JD			

cles. As shown in Fig. 6, the relative humidity of the GDL from Ketjenblack EC-600JD was slightly lower than that from the Vulcan XC72R. This may be due to the stronger water vapor absorptivity of the Ketjenblack EC-600JD with its larger specific surface area and micropores in its primary particles. Water absorbed by the WML acted as a barrier for the water vapor permeation through the GDL.

#### 3.3. Simulation of water management in PEMFC

The results of the water vapor permeability studies of the GDL were input into the developed simulator so that the relative humidity distribution in the catalyst layer and the water transport in the polymer electrolyte membrane could be predicted. The simulation was performed under the follow conditions. A PEMFC with the above mentioned MEA and carbon separator with the conventional parallel flow channels was simulated to be operated at 80 °C and atmospheric pressure. Reformed fuel (content 80% hydrogen and 20% carbon dioxide) and air, with a humidification temperature of 60 °C, were simulated to be fed to the PEMFC in the case of co-flowing. The utilization efficiencies of the hydrogen and oxygen were 70 and 40%, respectively. The current density of the PEMFC was fixed at  $0.4 \text{ A/cm}^2$ .

In Fig. 7 is shown the comparison of the relative humidity distribution in the PEMFC cathode electrode between (a) the novel GDL, and (b) the traditional GDL, only PTFE-treated carbon paper. The novel GDL was obtained by coating WML on the carbon paper from the cell inlet to center, while no WML was coated from the center to the outlet. The simulation results showed that the water distribution in the cata-



Fig. 7. Simulation results of the relative humidity distribution in the cathode of PEMFC (a) with WML and (b) without WML.



Fig. 8. Water transport in the polymer electrolyte membrane of the PEMFC (a) without WML and (b) with WML.

lyst layer was largely influenced by the WML. Namely, the WML coating on the GDL increases the relative humidity in the catalyst layer near the cell inlet where the polymer electrolyte had originally a tendency to dry out, while the non-coating of the WML on the GDL lowers the relative humidity in the catalyst layer near cell outlet where the electrode had originally a tendency to be flooded. Thus, over the whole catalyst layer, the water vapor could be controlled to the proper level, and the flooding of the cathode could be effectively avoided.

Fig. 8 shows the water transport through the polymer membrane in the PEMFC simulated using the abovementioned conditions. For the traditional PEMFC with carbon paper as the GDL (Fig. 8(a)), due to the osmosis-electric, water transport with protons produced a higher water potential at the cathode, although the back diffusion of water from cathode to anode happened, the net water transport in the whole PEMFC was from the anode to the cathode, resulting in the drying-out of the polymer electrolyte. The drying-out of the polymer electrolyte was more serious in the cell inlet region. On the other hand, as shown in the Fig. 8(b), there was a different water transport profile in the PEMFC due to the inserted WML. In the area of the WML inserted near the cell inlet, the net water transport was from the cathode to the anode. This meant there was more water for electro-osmosis of protons on the anode side, resulting in a higher membrane water content and thus a higher membrane conductivity. However, in the area near the cell outlet without WML, due to the hydrogen consumed and thus an increase in the water content, the net water transport direction was changed towards to cathode side, so that the water flooding of the electrode is effectively avoided.

By this simulation, the optimum GDL was obtained by correlating the WML on the carbon paper, such as distribution, pore size, hydrophobicity, thickness and the components of the WML. In addition, the gas flow channel (wider and deep) in the anode and cathode separators was also optimized using the simulator.



Fig. 9. Cell performance of the PEMFC with and without WML at the different humidification temperature of (a) 70  $^{\circ}$ C, and (b) 40  $^{\circ}$ C.

#### 3.4. Performance of the PEMFC with the novel GDL

Using the simulator, the optimal GDL was prepared. Fig. 9 shows the cell performance of the PEMFC installed with the optimal GDL. For comparison, results of the PEMFC installed with carbon paper (without WML) were also plotted. The PEMFC was operated at 80 °C at atmospheric pressure. Reformate fuel and air, with the same humidification temperature of (a) 70 or (b) 40 °C, were fed into the PEMFC. The utilizations of the hydrogen and oxygen were 70 and 40%, respectively. The gas flow rate and cell current density were controlled by a computer program. The cell current density was swept at a rate of  $0.05 \text{ A/cm}^2 \text{ min}$ . The cell voltage and electric current read from the electric load were collected by a personal computer.

Fig. 9(a) shows the cell performance of the PEMFC at the humidification temperature of 70 °C. The cell performance of the PEMFC with WML was clearly higher than that of the PEMFC without the WML, especially in the high current density region where the cell voltage of the PEMFC without the WML drastically decreased. It was mainly a result from the drying-out of the polyelectrolyte in the cell inlet region [12]. Due to a better water management, i.e., minimizing the electrode flooding and optimizing the polyelectrolyte hydration, the cell voltage of the PEMFC with WML gradually decreased with the increase of the current density [18]. The higher cell voltage of the PEMFC with the WML in the low current density region was also considered as a result of the decreased contact resistance between the GDL and the catalyst layer. That is, nano-sized carbons black in the WML makes a good electrical contact between the catalyst layer and the carbon fiber of the carbon paper. Therefore, the WML plays the role not only for water management in the MEA, but also for decreasing the inner electric resistance of the PEMFC.

Fig. 9(b) shows the cell performance of the PEMFC at the humidification temperature of 40  $^{\circ}$ C, which was quite lower than the cell temperature. A similar performance improvement was obtained. However, it was surprising that even with such a lower humidification temperature, the PEMFC inserted with the WML could be steadily operated up to 0.8 A/cm<sup>2</sup>, and that it had the relatively higher cell voltage

of 0.65 V at the current density of 0.4 A/cm<sup>2</sup>. In this case, the system efficiency of the PEMFC would significantly increase due to the very low required power for the humidifier. On the other hand, with the traditional GDL of carbon paper, the relatively dry reactant gas caused the membrane to severely dehydrate and the cell performance significantly decreased. In fact, the cell voltage was not stable, and after 1 h, the PEMFC was difficult to run even at the lower current density of 0.1 A/cm<sup>2</sup>. Therefore, with the novel GDL, a lower humidified reactant gas makes it possible for the PEMFC to operate with a higher performance. Further studies such as the WML preparation, structure and composition, and long-term stability of the PEMFC using different humidified reactant gases should be performed in detail.

## 4. Conclusions

A novel GDL with the function of water management in an MEA has been designed, simulated and tested. The WML coated on the carbon paper could make the water distribution in the MEA more uniform and could effectively avoid any drying-out of polyelectrolyte and flooding of the cathode, resulting in a high PEMFC performance. The performance enhancement of the PEMFC with the WML appeared to be more pronounced for a relatively low humidified reactant gas. Such a result suggests that the WML may allow one to develop a PEMFC which needs a low humidification or zero humidification by effectively designing the WML.

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