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Investigations on the double gas diffusion backing layer for performance improvement of self-humidified proton exchange membrane fuel cells

Im Mo Kong^a, Aeri Jung^b, Min Soo Kim^{b,*}

^a Korea Automotive Technology Institute, Republic of Korea

^b Department of Mechanical and Aerospace Engineering, Seoul National University, Seoul 08826, Republic of Korea

HIGHLIGHTS

- The performance of self-humidified PEMFCs can be improved with double GDBL.
- The effect of double GDBL on water retention capability and membrane hydration was investigated.
- In addition to HFR and EIS measurements, numerical analysis was conducted.
- Optimized design of double GDBL for self-humidified PEMFC was investigated.
- This study provides an inspiration on how to design the double GDBL.

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ABSTRACT

In order to simplify the system configuration and downsize the volume, a proton exchange membrane fuel cell (PEMFC) needs to be operated in a self-humidified mode without any external humidifiers. However, in self-humidified PEMFCs, relatively low cell performance is a problem to be solved. In our previous study, a gas diffusion layer (GDL) containing double gas diffusion backing layer (GDBL) coated by single micro porous layer (MPL) was introduced and its effect on the cell performance was evaluated. In the present study, the effect of the double GDBL was investigated by measuring high frequency resistance (HFR) and electrochemical impedance spectroscopy (EIS). In the experiments, the HFR value was remarkably reduced, while the diameter of semicircle of EIS was increased. It means that the membrane hydration was improved due to enhanced water retention capability of the GDL despite of interrupted gas diffusion. The result of numerical analysis also showed that the water retention capability of GDL can be improved with proper structure design of double GDBL. Based on the result, optimized design of double GDBL for water retention was obtained numerically. The result of this study provides useful information on the structural design of GDBL for self-humidified PEMFCs.

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1. Introduction

Hydrogen is the most abundant element in the universe and can be produced from a wide variety of primary energy sources using energy conversion technologies [1]. For this reason, hydrogen energy technologies are expected to play an important role in the future energy economy and fuel cells have received a significant attention due to its high electrical efficiency and clean exhaust energy conversion [2,3]. Among the various types of fuel cells, a proton exchange membrane fuel cell (PEMFC) has many advantages such as low operating temperature, quick response, and high fuel utilization efficiency [4]. Therefore, PEMFC system can be

* Corresponding author. E-mail address: minskim@snu.ac.kr (M.S. Kim).

http://dx.doi.org/10.1016/j.apenergy.2016.05.057 0306-2619/© 2016 Elsevier Ltd. All rights reserved. applied to various types of applications such as vehicle, building, and portable devices. Moreover, PEMFC-based CHP (Combined Heat and Power) systems can be used for domestic application and dairy farming [5,6].

In PEMFCs, the proton conductivity depends on the hydration level of the membrane and appropriate hydration of the membrane is an important factor for high cell performance [7]. In conventional fuel cell systems, external humidification systems containing membranes, bubblers, and injectors are used to humidify the supplied air. However, external humidifier increases system complexity and parasitic power loss in addition to increased manufacturing cost, weight and volume. For these reasons, many researchers have studied an operation of self-humidified PEMFCs despite of its reduced cell performance caused by insufficient hydration of membrane.





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Nomenclature			
P _c σ θ K ε J S	capillary pressure (Pa) surface tension (N m ⁻¹) contact angle (°) permeability (m ²) porosity Leverett J-function liquid water saturation	q D ^{eff} C _i D P T β	gas diffusion flux effective diffusivity gas concentration diffusivity pressure (Pa) temperature (K) effective diffusion factor
x M i	GDL thickness directional distance (µm) molecular weight of water (g mol ⁻¹) current density (A/cm ²)	Q A	liquid water remaining in GDL (mg/cm ²) active area (cm ²)
F ν k _κ d C δ	Faraday constant (C mol ⁻¹) kinematic viscosity (m ² s ⁻¹) Kozeny constant fiber diameter (m) integral constant thickness (µm)	Subscrip ref GDL GDBL MPL	ot reference original gas diffusion layer gas diffusion backing layer micro porous layer

To date, it has been demonstrated that stable operation of PEMFCs without external humidifier is possible and the cell performance can be improved by enhancing the self-humidification effect. In order to improve the self-humidification effect, some researchers developed advanced polymer electrolyte membranes by adding hydrophilic nanoparticles to the membrane [8–10] and/or catalyst layer [11–15]. However, since the hydrophilic particles act as electrical insulators, these methods cause reduced ion conductivity of the membrane [16].

A gas diffusion layer (GDL) also plays an important role on water management in PEMFCs. Conventional GDLs are composed of single gas diffusion backing layer (GDBL) and single microporous layer (MPL) [17–20]. In order to enhance the water retention capability and self-humidification effect of GDLs, some researchers [21-28] investigated double MPL composed of hydrophobic layer and hydrophilic layer to retain more amount of water produced by the fuel cell reaction. However, although they observed performance improvements under certain conditions, the use of hydrophilic MPL easily causes flooding by pore blockage even under low humidity conditions since the MPL has micro-scale pores and low gas permeability [29-31]. On the other hand, the GDBL has a higher porosity and thickness than the MPL [17–20]. For this reason, we have suggested double GDBL structure and evaluated its effect on self-humidification by measuring the cell performance [32]. As a result, it was found that the cell showed a better performance with the modified GDL containing double GDBL under low humidity conditions compared with the conventional GDL containing single GDBL. However, in the previous study, the enhanced water retention capability of GDL was not verified clearly and numerical evaluation was not carried out. Therefore, in the present study, the effect of the GDL containing double GDBL on water retention capability was evaluated experimentally by measuring electrochemical characteristics such as high frequency resistance (HFR) and electrochemical impedance spectroscopy (EIS). In addition, the GDL was investigated numerically in order to provide detailed information. In this study, one-dimensional steady-state model based on a capillary pressure-saturation relationship [32-34] was introduced. Optimal property arrangement of each GDBL for water retention was also investigated to provide useful information for performance enhancement.

2. Experimental apparatus

Fig. 1 shows a schematic design of two GDLs adopted in this study. The GDLs prepared in the present study were the same with

the GDLs evaluated in the previous study [32]. The conventional GDL, SS-GDL, was composed of single MPL and single GDBL. For the GDBL of SS-GDL, the porosity was 0.90 and thickness was 305 ± 6 μ m. Next, the modified GDL, SD-GDL, was composed of single MPL and double GDBL (GDBL-1 and GDBL-2). The SD-GDL was prepared by stacking single GDBL (GDBL-2) with MPL coated GDBL (GDBL-1). For SD-GDL, The porosities of GDBLs were 0.88 for the GDBL-1 and GDBL-2 was 309 ± 4 μ m, whereas the thickness of GDBL-1 and GDBL-2 was 120 ± 5 μ m. Then, the thickness of the GDBL from the total thickness of the GDL. The total thickness of the GDL. The total thickness of the GDL was 120 ± 5 μ m. Then total thickness of the GDLs were 348 ± 4 μ m for SS-GDL and 355 ± 3 μ m for SD-GDL. More detailed information on the prepared GDL is available in our previous study [32].

Fig. 2 shows the schematic diagram of the experimental setup used for the electrochemical measurements.

The single cell used in this study was composed of electrically insulated end plates, graphite bipolar plates with flow channels, Teflon-type gaskets, and a commercial membrane electrode assembly (MEA) and GDLs. The MEA (CNL Energy, Korea) was composed of Nafion-211 and a catalyst layer (0.4 mg of Pt/cm²) with an active area of 9 cm² for both the anode and cathode sides.

In order to supply reactant gases to the single cell, mass flow controllers (HI-TEC MFC, Bronkhorst, Netherlands) were installed for hydrogen and air. Also, an electric loader (PLZ 664WA, Kikusui, Japan) and an impedance meter (KFM2150, Kikusui, Japan) were installed for the electrochemical measurements. The installed bubble-type humidifiers were only used for the activation of the single cell. For the electrochemical measurements, dry air and hydrogen were supplied to the fuel cell without passing through the humidifier by controlling the valves.

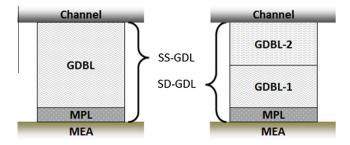


Fig. 1. Schematic design of the GDLs located between MEA and channel.

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