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Analysis of the transient response and durability characteristics of a proton exchange membrane fuel cell with different micro-porous layer penetration thicknesses

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HIGHLIGHTS

- The GDL which has a large MPL penetration showed a better transient response.
- The transient response of the PEMFC was affected by the water balance of the GDL.
- A large MPL penetration balanced the capillary pressure gradient through the GDL.
- The carbon corrosion induced loss of the MPL penetration region.

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ABSTRACT

The optimal design of the gas diffusion layer (GDL) of proton exchange membrane fuel cells is crucial because it directly determines the mass transport mechanism of the reactants and products. In this study, the micro-porous layer (MPL) penetration thickness, which affects the pore size profile through the GDL, is varied as the design parameter of the GDL. The cell performance is investigated under various humidity conditions, and the water permeability characteristics are studied. In addition, the accelerated carbon corrosion stress test is conducted to determine the effect of MPL penetration on GDL degradation. GDLs with large MPL penetration thickness show better performance in the high-current–density region due to the enhanced management of water resulting from a balanced capillary pressure gradient. However, the loss of penetrated MPL parts is observed due to the low binding force between the MPL and the GDL substrate.

1. Introduction

The PEM fuel cell must overcome a few technical issues before it is commercialized in automotive applications. The primary goal is to develop PEM fuel cells that exhibit stable and instantaneous dynamic performance in response to drivers' power requirements. In addition, the enhanced durability of fuel cells to sustain stable long-term operation is required to meet consumer's needs [1–4].

The gas diffusion layer (GDL) is a key component of a PEM fuel cell that affects the cell's dynamic behavior and durability characteristics because it functions as a channel for mass transport [5,6].

However, the effects of the GDL on the transient response of PEM fuel cells have been overlooked in previous studies. Experimental researches on the dynamic behavior of PEM fuel cells have concentrated on phenomenal results induced by operating conditions [5,7–9]. The causes of the transient behavior of PEM fuel cells have not been systematically investigated.

In the case of dynamic modeling studies, the effects of the GDL on the transient response are treated simply, despite the fact that the GDL is the main component of mass transport in the PEM fuel cell. Although GDLs possess a variety of characteristics, a simplified expression for mass transport in the GDL is used [10,11]. In addition, dynamic two-phase models are in the early stages of development due to the complexity and difficulty associated with the mathematical analysis of dynamic gas–liquid behavior in porous medium [12–14]. Therefore, it is important to study the correlation between the transient response of the PEM fuel cell and the characteristics of the GDL by experimental approaches.

GDL degradation has been the focus of recent studies on the improvement of the durability of PEM fuel cells. To date, many studies have reported catalyst and membrane degradation under various conditions [15–21]. Recently, several papers have shown experimental results regarding the degradation of the GDL alone

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[22–25]. In particular, Ha et al. reported on changes observed in the characteristics of a degraded GDL, dividing the sources of these changes into an aging mechanism due to the dissolution effect of water [23] and a carbon corrosion effect due to electrochemical reactions [24]. Furthermore, Cho et al. demonstrated the effects of a mechanically aged GDL on the transient response of a PEM fuel cell [26]. The GDL, which was degraded by dissolution in water, induced a large voltage undershoot and unstable voltage response upon a sudden increase in load due to local water flooding in the substrate.

From these previous studies, it is clear that the characteristics of the GDL, the degradation of the GDL and the transient response of PEM fuel cells are closely connected. However, these issues have not been directly correlated. Indeed, the analysis of the effects of the GDL on the dynamic behavior of PEM fuel cells remains insufficient.

Moreover, the need to study GDL design to enhance performance and durability has increased. Through numerous studies, the hydrophobicity of substrates and the micro-porous layer (MPL), which is represented by the weight percent of PTFE, has been optimized [27–31]. Studies on the design of various types of GDLs have been recently reported [6,32,33]. However, they also overlook the effects on the dynamic behavior of PEM fuel cells. In addition, the correlation between the characteristics of the GDL and PEM fuel cell performance has not been clearly investigated. To enhance cell performance over a wide range of operating conditions, the mechanism of mass transport associated with the characteristics of the GDL and the effects of GDL design parameters on the transient response, as well as the steady-state performance, of fuel cells must be explored.

Because the GDL consists of two main parts, a gas diffusion backing layer (GDBL) referred to as the substrate and the MPL, which is coated on the substrate, each design parameter must be investigated individually. In manufacturing process of the GDL, the carbon particles of the MPL penetrate through the substrate when the MPL is coated on the substrate. Thus, it is important to understand how the MPL penetrates the substrate to allow for better mass transport, strength and durability because the penetration thickness determines the pore distribution profile throughout the GDL, which induces mass balance characteristics inside of the GDL. Additionally, aging characteristics and their effect on the performance of PEM fuel cells must be investigated.

Therefore, the objectives of this research were to analyze the effects of MPL penetration thickness on the transient response of a PEM fuel cell and to analyze its effects on the degradation characteristics of the cell. By analyzing the transient voltage response with a step change in current, the variation in the dynamic performance of the fuel cell as a function of the design and degradation of the GDL was characterized. An accelerated carbon corrosion stress test was conducted to study the aging of the GDL independently. Furthermore, the correlation between the GDL design and the dynamic performance of the PEM fuel cell were analyzed by experiment and by well-defined theory. In this study, it was determined how MPL penetration affects mass balance in the GDL and consequently the transient response of the PEM fuel cell and how the MPL penetration region is degraded by carbon corrosion; the variation in the transient response of the PEM fuel cell was also investigated.

2. Experiment

2.1. GDLs

Three samples with different MPL penetration thicknesses, as shown in Fig. 1a, were fabricated. The same paper-type substrate (or gas diffusion backing layer (GBDL)), measuring 280 μm in thickness, was used for all samples. A 110 μm thick MPL was coated on the substrate. MP-A showed a small region of penetration. MP-B was controlled to have an intermediate penetration thickness of 100 μm, and MP-C showed thick penetration into the substrate. Approximately 50% of the substrate was filled with MPL in the case of MP-C. Because excessively deep penetration will block the mass transport of reactant gases, through a preliminary study, the thickness of MP-C was defined as the maximum penetration thickness allowed for the retention of a balanced MPL and substrate structure. All GDLs studied had 10 wt% and 23 wt% PTFE loading for the substrate and MPL, respectively.

2.2. Degradation of the GDL by carbon corrosion

To investigate the effect of MPL penetration thickness on the degradation characteristics of the GDL and the corresponding effect on the transient response of a PEM fuel cell, an accelerated carbon corrosion test was performed because our research team reported that the carbon corrosion of the GDL occurs mainly in the MPL penetration region [24].

Extensive carbon corrosion occurs when a potential of 1.45 V is applied to a PEM fuel cell under start-up, shut down, and local fuel starvation conditions [17,24]. A three-electrode corrosion setup was assembled to apply a precise potential on the GDL independently. The GDL was the working electrode, and a Pt mesh and Ag/AgCl were used as the counter electrode and reference electrode, respectively. A 0.5 M solution of sulfuric acid was used as the electrolyte. A potentiostat (Solartron analytical 1255B LF Frequency Response Analyzer and 1480A Multistat) was used to apply a potential to this system. A corrosion time of 96 h was determined, corresponding to 5000 h of PEMFC operation [24].

After corrosion, various properties of the GDL were assessed: contact angle, weight, thickness, pore distribution, FESEM image and change in water permeability. The specifications of the measurement instruments are detailed in references [23,24] and the following chapter.

2.3. Fuel cell and test station

As shown in Fig. 2, a 25 cm^2 unit cell with a parallel serpentine channel (1 mm × 1 mm) was used to investigate the effects of structural differences in the GDL on the cell performance. A fresh GORE™ PRIMEA® 5710 catalyst-coated membrane was used for each test. The designed and degraded GDLs were assembled to both sides of the membrane. High purity air and hydrogen were humidified by the membrane type humidifier. The electric loader controlled the load changes, and a data acquisition board from National Instrument was used to obtain fast and reliable voltage transient responses during the load changes. All test protocols, including activation immediately after assembling the cell and purging, were performed in the same way in every case. Detailed experimental apparatus was shown in Refs. [5,26].

In addition, to evaluate the characteristics of the designed GDL, the water permeability was measured. A GDL sample measuring 5.0 cm in diameter, with the MPL side facing upward, was inserted into a cylindrical chamber. Water was continuously supplied to the chamber until water passed through the sample. The change in the hydrostatic head, which corresponds to a change in pressure, and the amount of water drained were obtained every second. The water permeability was evaluated by monitoring the mass flow rate as a function of pressure. The experimental apparatus is described in detail in reference [34].

The pore distribution of the GDL was also measured using mercury porosimetry (Autopore IV 9500, Micromeritics USA). Under constant pressure, the pore diameter filled by mercury is inversely proportional to pressure. Thus, the pore volume, pore distribution,