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Experimental and numerical modeling study of the electrical resistance of gas diffusion layer-less polymer electrolyte membrane fuel cells

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HIGHLIGHTS

- A coupled model was developed for corrugated-mesh fuel cells.
- Electrical, mechanical, and electrochemical factors were considered in the model.
- Considered material/contact resistance in analyzing electron pathway from CL to BPP.
- Electrical conductivity and rigidity of the MPL control the electron pathway.
- Internal resistance is reduced with mechanical pressure distribution.

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ABSTRACT

The gas diffusion layer (GDL)-less fuel cell composed of a corrugated-mesh shows low flooding performance even in the high current density region, since the gases are supplied more uniformly to the catalyst layer (CL) compared with the conventional fuel cells that utilize GDLs. On the other hand, the internal electrical resistance of the GDL-less fuel cell is higher than that of the conventional fuel cell, because the corrugated-mesh and the underlying microporous layer (MPL) have a low contact area with point contacts. This can greatly increase the resistance at the interface between the corrugated-mesh and MPL as well as that between the MPL and CL, compared to the conventional fuel cell where GDL can make a good contact with the MPL. In this study, the conductivities and the contact resistances of each material in the GDL-less fuel cell were measured under various mechanical compression pressures, and a coupled mechanical–electric–electrochemical model was developed to investigate the effect of electrical resistance on the fuel cell performance. We found that our model can simulate the GDL-less fuel cell well and the electric resistance contributes significantly to the polarization performance in the GDL-less fuel cell.

1. Introduction

Polymer electrolyte membrane fuel cells (PEMFCs), which convert the chemical energy of hydrogen directly into electrical energy, are regarded as promising alternatives and a clean power source for automotive, stationary, and portable applications [1–5]. The use of fuel cells for powering automotive devices requires a reduction in cost and size, as the current automotive fuel cells are not profitable and are too large, limiting the layout of the fuel cell system [6].

Generally, if the required power for a single cell can be produced with a smaller active area while keeping the voltage around 0.6 V [7], the multi-cell fuel cell stack can be made smaller and cheaper. Currently, most automotive fuel cells operate with a maximum current density of around 1.0–1.5 A cm^{-2} [7–9], which necessitates a large active area. Consequently, the size of catalyst coated membranes (CCM) containing polymer electrolyte membranes (PEM) and catalyst layers (CL) with precious metals, and gas diffusion layers (GDLs) is increased. The need for the additional

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materials leads to added expense and therefore, the need for larger active areas significantly increases the cost of the fuel cell stack. In addition, a GDL thickness of 90–200 μm for each electrode increases the length in the stacking direction and the volume of the GDL occupies about half of the total volume of the fuel cell stack. These factors pose cost and size-related limitations, for conventional fuel cell structures.

In order to solve these commercialization issues in automotive fuel cells, a novel fuel cell structure that enables high current density operation without the use of a GDL, is required. Increasing the current density of conventional fuel cells causes a sudden decrease in performance, which is known as cathode flooding. As the current density increases, more water is generated via the oxygen reduction reaction ($O_2 + 4H^+ + 4e^- \rightarrow 2H_2O$) at the cathode, and more water is transported from the anode to the cathode through the membrane via electro-osmotic drag [10–12]. The water at the cathode condenses and accumulates, thereby blocking oxygen diffusion and causing a sudden decrease in the cell voltage.

Simultaneously, increasing the current density causes the membrane to dry out at the anode. The membrane is dehydrated by the electro-osmotic drag from the anode to the cathode [13], in addition to the increased temperature from the electrochemical reaction and the ohmic heat. The proton conductivity of the membrane heavily depends on its water content, and the dehydration of the membrane causes a decrease in the proton conductivity and the cell performance.

In a previous study, we achieved a cell voltage of 0.45 V at 3.0 A cm^{-2}, without experiencing a significant voltage drop even at a relatively high current density of over 1 A cm^{-2}, by utilizing a corrugated-mesh in the flow channel located directly on the microporous layer (MPL), without using a GDL. We also measured the cell performance with conventional flow fields using a GDL, in which case a severe voltage drop was seen starting at a relatively low current density of 0.4 A cm^{-2} [14,15]. From the results of the study, we concluded that the corrugated-mesh flow channel can supply the reactant gases uniformly to the CL, with no significant indication of flooding up to 3.0 A cm^{-2} in the polarization curve. On the other hand, the GDL-less fuel cell with a corrugated-mesh flow channel shows a larger high-frequency resistance (HFR) compared to the cells having conventional flow fields with a GDL. Owing to the fewer contact points between the corrugated-mesh and the MPL, the electron has to be conducted in the in-plane direction of the MPL and CL, which can cause a larger HFR, since the MPL and the CL have lower conductivity and are thinner than the GDL. In addition, the electron travel pathway in this case is longer than that in conventional fuel cells. Furthermore, two different MPLs composed of carbon black (CB-MPL) and silver flakes (SF-MPL) were tested in the GDL-less fuel cell. The SF-MPL, which is three orders of magnitude more conductive than the CB-MPL, showed a similar HFR as the CB-MPL and both of them showed a higher HFR than the conventional fuel cell. This implies that the difference in the conductivity of the MPL alone cannot account for the HFR difference between these fuel cells, and hence not only the conductivity, but also the contact resistance of the corrugated-mesh|MPL and the MPL|CL interfaces may have a strong effect on the total HFR under highly non-uniform compression pressure distribution conditions. Although the contact resistance heavily depends on the contact pressure and the contact pressure distribution in turn, is determined by the elasticity modulus and the thickness of the MPL [16], stainless steel MPL (SS-MPL) was used instead of the SF-MPL to investigate the effect of the modulus of the MPL. The SS-MPL showed a lower HFR compared to the SF-MPL, implying that a stiffer MPL can distribute the compression pressure and reduce the contact resistance of the MPL|CL interface. However, there is no theoretical model accounting for the electrical resistance issues in the GDL-less fuel cells, and we realized that stress–electrical–electrochemistry simulations would be required to gain a better understanding of the effect of electrical conductivity and contact resistance on the cell performance.

Some investigations have been conducted to understand the effect of electrical resistance on the performance of the conventional fuel cells. Meng et al. [17] developed a three-dimensional numerical model for PEMFCs under single-phase and isothermal conditions with electron transport in the CL, GDL, and bipolar plate (BPP). They found that the current density distribution and the cell performance were influenced by both the oxygen concentration distribution by diffusion through the GDL as well as the voltage distribution by electron conduction through the GDL. In addition, they also found that at lower current densities, the electron transport is more dominant, whereas at higher current densities, gas diffusion is more dominant. Zhou et al. [18] developed a similar model to investigate the effect of anisotropic conductivity in the GDL, and they concluded that the effect of the electrical resistance of the GDL is negligible for the typical values of GDL conductivity seen in real systems, because gas transport in the GDL and CL are dominant. Sun et al. [19] performed similar investigations, incorporating a two-dimensional model to verify the channel-land geometric ratio, and they found that while a narrow land can supply the gases more efficiently and uniformly, they also increase the electrical resistance for the long electron pathway in the GDL. Taymaz et al. [20] developed a three-dimensional model incorporating a mechanical deformation effect on the porosity and contact resistance of the GDL|BPP interface. They found that an increase in the compression pressure causes a degradation in the fuel cell performance owing to a lower diffusivity in the GDL under compression, despite the lower contact resistance. They considered the balance between the gas diffusion and electron transport in the in-plane direction of the GDL. On the cathode side, air is provided through the channel on the BPP and the air diffuses from the GDL to the surface of the CL. In particular, in the under-land region, the air diffuses in the in-plane direction of the GDL, which results in an oxygen concentration distribution along the in-plane direction of the GDL. This is referred to as air management in the fuel cell [21]. On the other hand, the electrons on the cathode side are provided from the land on the BPP and the electrons are transported through the GDL to the surface of the CL. The voltage decreases owing to the IR drop along the in-plane direction of the GDL during the electron transport from the under-land region to the under-channel region. Although the effects of both air and electron transport have been investigated extensively, all the studies have been performed on conventional fuel cells and it has been concluded that the air management issues influence the fuel cell performance more than the electron transport issues. On the other hand, in the GDL-less fuel cell, air appears to be distributed more uniformly, whereas the electron transport appears to be poor through the MPL, which has a lower thickness and lower conductivity compared to the corresponding values for the GDL [15]. Furthermore, the contact resistance of the corrugated-mesh|MPL interface appears to be high owing to the low contact area. In addition, the contact resistance of the MPL|CL interface also appears to be high, since the soft MPL does not allow an even distribution of the compression load on the CL.

In this study, we measured the mechanical and electrical properties of corrugated-mesh, MPLs, CL, and the GDL. In addition, we also developed a three-dimensional coupled model incorporating mechanical, electrical, and electrochemical considerations for GDL-less fuel cells under single gas, single phase, and isothermal conditions, and validated the model with the fuel cell performance results obtained experimentally for conventional and GDL-less fuel cells. In addition, we also studied the influence of