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Effect of Compression Thickness on Performance of Gas Diffusion Layer of Direct Methanol Fuel Cells

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This study considers the performance of a direct methanol fuel cell (DMFC) unit for the various compression ratios of a gas diffusion layer (GDL) on the cathode side. A performance of fuel cells is significantly influenced by the compression ratio of the GDL. The compression ratio of the GDL can be directly controlled by the concentration compression force at the central area of the cell and gasket thickness. The balancing of the compression ratio is an important factor to increase the unit cell performance of DMFCs. In this study, the performance test of the unit cell for varying compression ratio of a GDL on the cathode side is carried out. We describe how the compression ratio affects the stack performance by the variation of compression force for two different kinds of current collectors, which directly compresses the central area of the unit cell with varying gasket thickness. This study also suggests a method to show the effect of mass transport loss by amplitude of voltage fluctuation and the unit cell relationship between the pressure drop information and the peak power of the unit cell.

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1. Introduction

A fuel cell is one of the promising electric conversion devices. There are various type of fuel cells, like solid oxide fuel cell(SOFC), proton exchange membrane fuel cell(PEMFC) and molten carbonate fuel cell(MCFC).1,2,26,34,35 The PEMFC consists of a pressure plate, insulator, current collector, separator, gasket and a membrane electrode assembly (MEA). In particular, the MEA which consists of the gas diffusion layer (GDL), catalyst layer and ion conducting membrane, is an essential element of fuel cells. A direct methanol fuel cell (DMFC) is a type of PEMFC that uses methanol as fuel. The basic operation of a DMFC consists of fuel as a water/methanol liquid and is supplied into the flow channel on the anode side of the separator. The DMFC diffuses through the anode GDL until it reaches the anode catalyst layer where it is converted into protons and electrons. The electrons are transported to the cathode via an external circuit and the protons are transported to the cathode through the electrolyte membrane. On the cathode side, the oxidant diffuses through the cathode GDL until it reaches the cathode catalyst layer. The oxygen is consumed by the protons and electrons transported from the anode, and liquid water is generated. All of the transport phenomena involve transportation loss that eventually reduces the performance of the fuel cells. Therefore, the transport paths must be

designed carefully. This goal is achieved by careful control of the key material characteristics and the physical factors in a fuel cell such as the crossover effect,^{1-3,34} temperature effect,^{4,5} humidity and differential pressure effect,^{4,6} and compression force effect of the GDL.⁷⁻³² The optimization of the key components such as a GDL is critical to improve the performance of the DMFCs. Many studies have recently focused on the understanding of GDL characteristics. Gurau⁸ and Lim⁹ studied PEMFC performance with respect to the hydrophobic polymer content of a GDL. Their findings showed the possibility of performance control using hydrophobic polymer contents. Litster¹⁰ presented the visualization of the water transport in the GDL of a PEMFC using fluorescence microscopy. Pasaogullari^{10,11} analytically studied the transport phenomena of the water and electron in a GDL. Kong,¹³ Jordan,^{14,15} Lee¹⁶ and Chu¹⁷ conducted various studies to verify the effect of the morphology variation of GDL by the versatile manufacturing method or treatment procedures. Besides the optimization of the material characteristics of the GDL, physical factors like compression pressure, compressed thickness, temperature distribution, and mass transport rate should be optimized to improve the stack performance without any change of materials. Hottinen¹⁹ conducted research on temperature distribution. The effect for the degree of compression on the GDL was investigated.¹⁸⁻³²

Like this, the compression force and control of material contents of the



GDL for optimization of the compression ratio have been the focus of many studies. Many researchers controlled compression ratio using an assembling force of stack or a compression of testing jig. In this study, we controlled compression ratio with an alternative method. The compression ratio of GDL is optimized by the variation of the compression force according to the design of the stack structure and the variation of PTFE gasket thickness. This is effective for preventing over-compression and sealing of the stack. We consider the effect of the compression ratio on the unit cell performance. The performance test of the unit DMFC is applied according to the variation of the compression ratio of the GDL at the cathode side by two different kinds of thicknesses of the current collectors. The compression using current collector which has valid thickness has some advantages. Previous method like using gasket thickness cannot avoid corrosion and contamination problems due to expose with fuel. Moreover, this method maintain valid compression during end-plate bending condition due to tensile force from assemble rod. This structure is acting like a disk spring. This condition directly affects the compression ratio at the central area of the GDL while various thicknesses of the PTFE gaskets are performed. We also use the amplitude of voltage fluctuation of the DMFC unit for quantization of mass transport loss, which shows what the unit DMFC relationship is between the pressure drop and the peak power.

2. Methodology

Fig. 1 shows the schematic diagram of the single cell of a DMFC. E-TEK's DMFC membrane electrolyte assemblies (MEA, 12D-W) with a 50 cm^2 active area is used as a performance test in this research. 12D-W MEAs are manufactured by using DuPont[™] Nafion® 117 and carbon cloth GDLs. The catalyst loading on the anode side is 4.0 mg/ cm² platinum Ruthenium Black, while the catalyst loading on the cathode side is 4.0 mg/cm² platinum black. The total thickness of the MEA is about 1.17 mm. To prevent over-compression of GDL, polytetrafluoroethylene (PTFE) skived sheets are used for the unit cell gasket. The PTFE gaskets on the cathode side are prepared with various thicknesses from 150 μ m to 300 μ m. The flow fields of the air and fuel sides have serpentine channels and the open ratio between the channel and land width is 0.923. The current collectors, which are the middle insert type in the insulator, was designed to prevent corrosion. There is a large gap between the manifold and the current collector. It is also easy to align with a single cell or stack and to control the compression ratio using the thickness of the current collector with a PTFE gasket.

The two types of thicknesses for the current collectors are manufactured to show the compression ratio effect. The thickness of the thin current collector and the groove of the insulator is 1.5 mm. Fig. 1 (b) shows the schematic of installed current collector. Thickness of the thicker current collector and the groove of the insulator is 1.7 mm. The insulators are made of epoxy glass and are used for isolation of electricity. The pressure plates have a thickness of 20 mm and are manufactured with duralumin. The assembly torque of the unit cell is 80 kgf-cm. The stack specifications of these features are shown in Fig. 1.

The test station consists of a fuel supply, air supply and control parts. The methanol solution is supplied by using the metering pump. The pump concentration is controlled by using the calibrated FC6 Methanol Concentration-Density Meter, which has an accuracy of



(a) Schematic diagram of the DMFC single cell



Fig. 1 Single cell design of the DMFC

 $\pm 0.3\%$ w/w. The fuel is 1M methanol solution and the fuel stoichiometric proportion is about 10 M for decoupling the fuel side effect. Air is supplied by microbial fuel cells (MFC) with an accuracy of 0.1%. The humidity of the air was controlled by a bubbler-type humidifier. The relative humidity is fixed at 60%. Temperature is regulated by using a proportional-integral-derivative (PID) controller, and the operating temperature is 60°C. Agilent N3300A DC electronic load and Agilent E3633A power supply are used for this work. The voltage and current data are acquired from National Instruments (NI) compactDAQ. The data on the temperature and pressure between the unit cell inlet and outlet are also obtained by using NI compactDAQ.

3. Results

3.1 Control the Compression Ratio by Current Collectors

To verify the effect of the compression ratio in the central area of the DMFC unit, the experiment is implemented by varying the thickness of the current collectors. The thicknesses of the current collectors are 1.5 mm and 1.7 mm and the assembly pressure is applied equally. Because the structure of the current collector is positioned in the middle part of the insulator, stress for the central area of the unit cell is concentrated. The performance could differ by the varying thickness of the current collector. Fig. 2 shows the unit cell performance for the varying thickness of the current collectors. The stoichiometric ratio of the air and fuel is fixed at 10 for decoupling the stoichiometric effect related with the amount of supplying the fuel and



Fig. 2 Unit cell performance with respect to the thickness of the current collector

air. The operating temperature is about 60°C and the relative humidity is 60%. The maximum power in the case of the 1.5 mm current collector thickness is about 30 W. The maximum power in the case of the 1.7 mm current collector thickness is about 73 W. The open circuit voltage (OCV) and activation loss seem to have approximately the same case. However, this result shows that the ohmic loss has a large difference for the performance between the two cases because of the different thickness of the current collectors. GDL is a porous material that has several important functions such as the transfer of the current and delivery of the fuel from the bulk site to the catalyst layer. The porosity of GDL is varied when the unit cell is assembled. Then, the performance of the unit cell differs according to varying porosity. This reflects the ohmic loss decreases and performance increases when the central area of the MEA using the 1.7 mm current collector thickness has more compression ratio than the other current collector thickness samples. An increasing compression ratio is an effective way for decreasing ohmic loss. However, it is ineffective for decreasing mass transport loss. It is difficult to transfer the oxidizer on the catalyst layer by decreasing the porosity of the GDL.

To show the effect of the mass transport with respect to the compression ratio, the unit cell is tested in terms of time with varying air flow rate and the varying thicknesses of the current collectors. Fig. 3 shows the results of the transient variation for the voltage varying the air flow rate. The unit cell is controlled with a constant current density of 100 mA/cm², and by varying the air flow rate. It means both of cases have same current density in same conditions like stoichiometry, reactant mass flux and product mass flux. The cell voltage is only experiment data. The transient variation of the voltage decreases by decreasing the air flow rate and the performance of the unit cell becomes low and unstable. Fig. 3(a) shows the transient variation of the voltage in the case of the 1.5 mm current collector thickness. Fig. 3(b) shows the transient variation of the voltage in the case of the 1.7 mm current collector thickness. Fig. 3(b) has a higher voltage than Fig. 3(a). However, the amplitude of the voltage fluctuation augments. It is considered that flooding issue in the case of thick current collector. Although, both of cases were controlled same reactant and product mass flux by constant current operation and flow rate, the difference of compression ratio affects microstructure of GDL and area size of flow channel between GLD and separator. These changes of structure affect to stability because of increasing mass transport loss on the cathode



(b) Current collector thickness, 1.7mm

Fig. 3 Transient variation of the voltage with respect to stoichiometry in constant current operation (100 mA/cm²)

side. Fig. 3(b) also shows that is difficult to operate behind a 0.27 LPM air flow rate. Conversely, Fig. 3(a) shows that it can operate in all regions of the air flow rate, although the performance is lower than Fig. 3(b). This reflects the effect of the mass transport, which directly depends on the compression ratio. The performance increases with a decreasing ohmic loss when the central area of the MEA has a higher compression ratio. On the other hand, the stabilities of the performance decreases with increasing compression ratio because the porosity of GDL decreases. As mentioned above, increasing the compression ratio is an effective way for decreasing ohmic loss. However, it is ineffective for decreasing mass transport loss. Decreasing porosity of the GDL makes it difficult to transfer the fuel on the catalyst layer from the bulk side.

Fig. 3 shows decreasing porosity directly affects mass transport loss and produces a voltage fluctuation. There is difficulty in the transportation of the oxidizer on the catalyst layer from the bulk side. This voltage fluctuation provides useful information about mass transport loss on the performance of the unit cell. To verify the quantitative effect of the mass transport loss, the results from fast Fourier transform (FFT) analysis are performed. Then, the fluctuation of the voltage is normalized by the mean value of the voltage. Finally, the results from the FFT analysis of the voltage variation at a 100 mA/cm² current density are integrated with each frequency, which shows the fluctuation amplitude of the voltage.³³

Fig. 4 shows the results of the amplitude of the voltage fluctuation for each air flow rate. This reflects that the voltage fluctuation increases with a decreasing air flow rate, which means that the mass transport loss increases with a decreasing air flow rate. This analysis shows the



Fig. 4 Amplitude of the voltage fluctuation with respect to air flow rate



Fig. 5 Unit cell performance with respect to thickness of the GDL

possibility for a quantitative amount of the mass transport loss and reflects that the unit cell can have a large difference on the amplitude of the voltage fluctuation with the variation of the current collector thickness. In particular, the amplitude of the voltage fluctuation in the case of the 1.7 mm current collector thickness, the current collector increases exponentially more than in the case of the 1.5 mm current collector thickness for all regions of air flow rate. The results show that the effect of the thickness of the current collector directly affects the mass transport loss, and that the compression ratio of the GDL can change the porosity of the current collector. The characteristic of the GDL is changed by these effects, and subsequently, the performance is varied.

As shown in Fig. 3 and 4, the thick current collector has better performance because of a decreasing ohmic loss with more compression ratio. However, the mass transport loss increases because the porosity of the GDL decreases. This makes the voltage fluctuation increase, which means that the stability of the unit cell decreases with an increasing compression ratio.

3.2 Control the Compression Ratio by PTFE Gasket

There are some ways to change the compression ratio of the GDL, which compresses more of the central area of the unit cell while using a current collector and changing the PTFE gasket thickness.

The PTFE gasket is usually used for sealing and spacing and thus prevents the over-compression of the GDL. The change of the PTFE gasket thickness can be another way to control the unit cell performance with the compression ratio of the GDL. Fig. 5 shows the unit cell performance for the PTFE gasket thickness of the unit cell.



Fig. 6 Amplitude of the voltage fluctuation with respect to the PTFE gasket thickness

The stoichiometric ratio of the air and fuel is fixed at 10 for decoupling the stoichiometric effect related with the effect of the fuel and air. The operating temperature is about 60°C and the relative humidity is 60%. The OCVs and activation loss in all cases look similar. However, the unit cell has different performance because of the mixing of the ohmic loss and mass transport loss. The performance of the unit cell increases in the case of the 150 μ m to 250 μ m current collector thicknesses. However, performance decreases in the case of the 250 μ m to 300 μ m current collector thicknesses. The unit cell using the 250 μ m PTFE gasket thickness has a maximum power of 72 mW/cm². This gives rise to vary the porosity of the GDL and compression ratio. Although there is an optimal performance, it is not easy to know what the major effects in losses are.

To show the effect of the mass transport loss for varying the PTFE gasket thickness, the unit cell is tested with varying the air flow rate. Fig. 6 shows the results the amplitude of the voltage fluctuation in the case of 150 μ m, 250 μ m and 300 μ m PTFE gasket thickness. The unit cell is controlled with a fixed current density of 100 mA/cm² and by air flow rate. Fig. 6(a) shows the amplitude of voltage fluctuation with varying air flow rate. The 300 μ m PTFE gasket thickness has smaller amplitude of voltage fluctuation than the other PTFE gasket thicknesses with a decreasing air flow rate from 0.41 lpm to 0.27 lpm. The amplitude of voltage fluctuation, which has more than 10 mV of voltage fluctuation compared to the other PTFE gasket thicknesses. Conversely, the amount of the voltage fluctuations looks the same below 250 μ m PTFE gasket thickness, which means that the mass transport loss in the case of above 300 μ m gasket thickness is relatively low and the voltage



Fig. 7 Peak power density and pressure drop with respect to the PTFE gasket thickness

fluctuation while increasing air flow rate decreases. On the contrary, the ohmic loss in the case of using the 300im PTFE gasket thickness is larger than the other PTFE gasket thicknesses. Fig. 6(b) shows the amplitude of voltage fluctuation with varying PTFE gasket thickness. The amplitude of voltage fluctuation increases with decreasing PTFE gasket thickness. However, it has the same amplitude below a 250 μ m PTFE gasket thickness at an air flow rate of 0.41 lpm. Fig. 6(b) shows that the PTFE gasket thickness. However, a differential effect between the different PTFE gasket thicknesses looks small at the higher air flow rate region.

The unit cell performance depends on the channel pressure drop that is directly affected by the compression ratio because of the GDL bending rate inside the channel.18,32,33 The pressure drop starts to increase with an increasing compression ratio until the elastic region. To illustrate the compression ratio effect in the channel, Fig. 5 shows the peak power density and pressure drop in the unit cell for the various PTFE gasket thicknesses. Fig. 5 and 7 show the unit cell with the 250im PTFE gasket thickness has the best performance. The peak power density increases while increasing the PTFE gasket thickness from 300 μ m to 250 μ m, and dramatically decreases while increasing the gasket thickness. The unit cell with a 250 μ m PTFE gasket thickness has the highest pressure drop. This is a very similar pressure drop pattern that increases while increasing the PTFE gasket thickness until 250 µm, and then decreases while increasing the PTFE gasket thickness. However, the pressure drop has a gentle slope between the PTFE gasket thickness of 150 μm and 250 µm compared to the drastic slope region between 250 µm and 300 µm. All things considered from Fig. 6 and 7, the PTFE gasket thickness has an elastic region limitation at 250 μ m. The pressure drop and the amplitude of voltage fluctuation drastically decrease with an increasing gasket thickness above 250 μ m. A 250 μ m gasket thickness is almost the compression ratio limitation for this unit cell.

4. Conclusions

We verified in this study that the effect of the compression ratio on the cathode side directly influences fuel cell performance. The gas diffusion layer is a key element of the fuel cell. The characteristics of the gas diffusion layer directly affect fuel cell performance. This paper suggests a way to show the amount of mass transport loss by amplitude of voltage fluctuation. We also show that there is a relationship between the pressure drop and peak power of the unit cell. Studies have indicated an effective way to control the compression ratio of the gas diffusion layer, which are the design of the stack structure such as the control of the current collector thickness without any change of compression force and control of the gasket thickness. Experimental results on varying the thickness of current collectors reflect a very efficient way to decrease the ohmic loss by concentration of the compression force at the central area of the unit cell. However, an over-compressed central area of the stack can affect an increasing mass transport loss. The transient voltage variation is more fluctuated with a decreasing air flow rate because of the over-compression ratio in this research.

The controlled PTFE gasket thickness is another effective way to control the compression ratio of the gas diffusion layer. The results show the unit cell of the direct methanol fuel cell has an optimal point using proper PTFE gasket thickness of $250 \,\mu m$. The performance increases with a decreasing gasket thickness up to $250 \,\mu m$ because the gas diffusion layer is well-compressed down. The performance decreases with decreasing gasket thickness under $250 \,\mu m$ because the gas diffusion layer is damaged from an over-compression ratio. This gives rise to the ohmic loss decreasing up to $250 \,\mu m$ however decreasing the porosity of the gas diffusion layer, which affects the mass transport loss of the air. As a result, the unit cell can be optimized by control of the current collectors and PTFE gasket thickness. The membrane electrode assembly condition by the amplitude of voltage fluctuation and pressure drop can also be identified.

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