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Effects of fabrication processes and material parameters of GDL on cell performance of PEM fuel cell

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Abstract

In this work, the main theme is to study the effects of electrode fabrication processes and material parameters on cell performance of PEM fuel cell. Adding a micro porous layer to traditional gas diffusion layer (GDL) can enhance the ability of water management, and therefore achieve better cell performance and higher limiting current density. For the effects of fluorinated ethylene propylene (FEP), either too high or too low content will deteriorate the performance of fuel cells. If the FEP content in the GDL is too high, indicating the GDL with a quite small pore size, the fuel gas will be difficult to diffuse in the GDL. If the content is too low, the water generated in the cell cannot be effectively removed. It is disclosed by the present experiments that when using air as cathode oxidant at an operating voltage above 0.6 V, the best performance can be achieved by using a GDL with 10% FEP content in the carbon paper and 20% content in the micro porous layer. The Vulcan XC-72R carbon loading of 1 mg/cm^2 in the micro porous layer is sufficiently high to obtain the maximum performance.

Keywords: Gas diffusion layer; FEP contents; PEMFC; Micro porous layer; Fabrication process

1. Introduction

Proton exchange membrane fuel cell (PEMFC) consists of seven layers, namely, anode flow field, anode gas diffusion layer, anode catalyst layer, proton exchange membrane, cathode catalyst layer, cathode gas diffusion layer (GDL) and cathode flow field. The GDL is an important part of membrane electrode. It should be hydrophobic and provide passages for gas transport from flow fields to catalyst layers and water removal from the catalyst layers to the flow fields. It is found that improvement of the gas distribution and water management in PEMFC can avoid water flooding. Conventional GDL usually used carbon substrates as diffusion media such as carbon paper or carbon cloth. Prasanna et al. [1] used carbon paper as GDL. They found that cells with different thicknesses of GDL have almost the same performance at low current density. But at high current density, too thin GDL would perform poorly, as it causes high mass transfer loss, high contact resistance and water flooding. Too thick GDL would lead to high activation potential, high ohmic resistance and high mass transfer resistance. Therefore, there should be an optimal GDL thickness. Their experiments also found that the cells with 20% PTFE content have the best performance.

Novel GDL is composed by carbon substrates and micro porous layer. Passalacqua et al. [2], in their experimental measurement, found that cells perform better with a gas diffusion electrode made of mixture of carbon black with PTFE in carbon paper, especially at high current density. When no mixture of carbon black with PTFE is contained in gas diffusion electrode and oxygen was used as cathode fuel gas, liquid water would block the pore, resulting in serious water flooding, and catalyst layer would enter carbon paper easily, deteriorating cell performance. If a carbon micro porous layer is added, the catalyst

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layer would not contact the carbon paper directly. Giorgi et al. [3] reported that cell performance decreases with PTFE content in GDL. When PTFE content increases, the porosity decreased. At high current density, the diffusion layer controlled electrode characteristics. Jordan et al. [4] found experimentally that cells with thinner carbon paper in GDL perform better, because the thinner GDL has shorter gas diffusion path. But the thinner GDL also has a larger electric resistance. Thicker GDL made cell performance worse due to its longer gas diffusion path. So, there should be an optimal thickness of GDL. They also found that the cell using Acetylene Black (AB) carbon powder performs better than that using Vulcan carbon powder. They found that the AB carbon is denser and has less micro pores than XC-72R carbon [5]. In their experiments, cells can achieve the best performance with the AB carbon loading of 1.9 mg/cm^2 when oxygen is used as the cathode fuel gas. As air is used, the optimal AB carbon loading is 1.25 mg/cm^2. Their results also demonstrated that the cell performance increases by 15% when using AB carbon instead of XC-72R carbon. Song et al. [6] employed AC impedance method to study the effects of carbon loading in catalyst-supporting layer on cell performance. Their results revealed that the catalyst active area decreases with a decrease in carbon loading. The catalyst active area reached the maximum value with the carbon loading of 3.5 mg/cm^2. Also, the thickness of catalyst-supporting layer is related to the maximum active area of Pt in catalyst layer. They also obtained the optimal cell performance with 30% PTFE content. Qi and Kaufman [7] added a sublayer onto GDL to improve mechanical stability and cell performance. Their experimental results proved that the performance of a cell with 24% PTEE contents in GDL slightly increases with increased carbon loading. With the GDL of 35% PTEE content, the cell performs better. As the GDL contains 45% PTEE, the cell performance becomes worse. Antolini et al. [8] coated two types of carbon, Vulcan XC-72R and Shawinigan AB, onto carbon cloth or carbon paper to manufacture GDL. They found that coating Shawinigan AB onto the both sides of carbon paper/cloth results in a better performancewhen oxygen partial pressure was 1 atm; while coating Vulcan XC-72R onto catalyst side is better when oxygen partial pressure was 3 atm. Thus, they suggested that it is better to coat Shawinigan AB onto channel side and Vulcan XC-72R onto catalyst side at high pressures. Park et al. [9] investigated the effect of hydrophobic treatment on GDL. They found that the porosity and pore radius of GDL material is related to PTEE content. The porosity decreases with PTEE content. But PTEE content has no influence on the thickness of GDL. They had used micro porous layer in their investigation to improve cell performance. The micro porous layer plays an important role on water management and electrical conductivity. Also, the micro porous layer is able to improve cell performance effectively. Lim and Wang's [10] investigation indicated that the cell with 10 wt% FEP performs better than that with 30 wt% FEP. Since the cell with 30 wt% FEP has smaller pores and may lead to much more saturation water on the surfaces of flow passages and GDL, and too much FEP would hamper mass transfer and water removal. Chen et al. [11] reported that, at high current density, cell performance can be improved by adding a water management layer between traditional GDL and catalyst layer. The water management layer is able to avoid PEM drying out and cathode water flooding, especially at low temperature humidified gas. Wang et al. [12] had investigated the effects of carbon black in the PEMFC on the cell performance. A GDL with AB carbon is found to have a high permeability. But, the water management is not good. Aa GDL with Black Pearls 2000 carbon exhibited less permeable to cause mass transport limitations. The results indicated that an optimal cell performance can be obtained by 10 wt% Black Pearls 2000 carbon and 90 wt% AB carbon in composite carbon black.

Lee et al. [13] found that the best cell performance can be achieved with the torque of 125 in-lb/bolt using ELAT carbon cloth or 100 in-lb/bolt using TORAY carbon paper. Because the TORAY carbon paper is brittle, high torque could crush carbon paper, making electric conductivity decrease. As the CARBEL is used as the GDL material, the optimal torque is 125 in-lb/bolt. Kong et al. [14] explored the effects of pore distribution in GDL on cell performance of PEM fuel cell. They found experimentally that the summation of pore number has an optimal value. At high current density, mass transfer loss would restrict cell performance, resulting in large excessive potential loss due to water flooding. When water flooding occurs, large pore would still provide a gas transport path, improve water management effectively and decrease mass transfer limit. As a result, large pore could drain water at high current density. But too large pore would decrease electric conductivity. Therefore, there must be an appropriate ratio of the large pore to the small pore and the pore distribution is important rather than the number of pores. Lee et al. [15] dealt with the influences of GDL fabrication processes on GDL characteristics. They found that the GDL coated by spraying or printing method is suitable for reaction because pores in carbon powder are large enough to separate water vapor and liquid water. Also, the GDL prepared in this way has rough surface, which is suitable for coating catalyst layer. The critical properties of five commercial and one in-house GDLs for PEMFC was characterized and compared by Williams et al. [16]. Recently, a hybrid technique for fabricating PEMFCs low platinum loading electrodes was proposed by Abaoud et al. [17]. It is concluded that the results of electrodes by hybrid method are superior to those with spraying and screen-printing techniques.

Recently, a fundamental understanding of liquid water transport in hydrophobic GDL and the effect of flooding on the PFMFC performance was examined by Pasaogullari and Wang [18]. It is found that flooding diminishes the cell performance as a result of decreased oxygen transport and surface coverage of active catalyst by liquid water. Yan et al. [19] numerically studied the effects of flow passage distribution and excessive electric potential on the local characteristics in the cell. The simulation results indicated that the local current distribution inside cells becomes homogeneous and leads to a better cell performance as the number of the flow passages increases. They also found that the cells achieve got better performance when the porosity increases from 0.3 to 0.6. Both the pressing pressure during electrode assembly and the water generated in cathode GDL could change pore distribution and influence

diffusion coefficient. Roshandel et al. [20] employed a numerical model to explore the influences of pore distribution in GDL on the cell performance. At low current density, the pore distribution in GDL has no influence on the cell performance; while at a high current density, the influence becomes noticeable. More recently, Zhan et al. [21] numerically studied the liquid water flux distributions with different GDL designs. When a micro porous layer is added between catalyst and GDL, the liquid saturation is redistribution across MPL and GDL to improve the water management within the PEM fuel cell. The liquid water flux is increased in the GDL with those caused by increasing MPL porosity and decreasing MPL thickness.

In the above literature survey, some literature is available on the effects of GDL fabrication processes and material parameters on cell performance, little is known about the effect of FEP content in the GDL and micro porous layer on the performance of PEMFC. This motivates the present study.

2. Experimental methods

2.1. Fabrication principle and steps of GDL

The fabrication process of GDL has been presented by Mathias et al. [22]. In [22], they presented the carbon substrates as diffusion media, and coated with PTFE. Carbon substrates, especially carbon paper or carbon cloth, are the most prevalent gas diffusion medium used in PEMFC for its high electrical conductivity and suitable porosity. The GDL fabrication processes involved in literature can be classified into two different types, one-stage and two-stage. Conventional GDL is usually produced with a one-stage fabrication process. The procedure for coating the hydrophobic material onto carbon paper or carbon cloth is shown in Fig. 1(a). Since carbon paper has low electric resistance and is hydrophobic in nature, it is chosen as the basic gas diffusion medium in this work. To further enhance the hydrophobicity of carbon paper, fluorinated ethylene propylene (FEP) solution is used for carbon paper coating. Novel GDL consists of two components: carbon paper and micro porous layer through a two-stage fabrication process is shown in Fig. 1(b). At stage one, the hydrophobic material is coated onto a carbon paper or carbon cloth. While at stage two, blending with hydrophobic material and carbon powder to form a micro porous layer is coated onto the treated carbon paper or carbon cloth. Because the micro porous layer is formed by carbon powder aggregation, it has much smaller pores than that using carbon paper as the backing material, the main purpose of which is to improve water management capability, provide effective mass transfer of liquid water, and carry liquid water in catalyst layer to GDL. According to the above introduction, the GDL produced by the two-stage fabrication process consists of two parts. One is called gas diffusion medium, that is, carbon paper; the other part is a micro porous layer between the carbon paper and the catalyst layer. Adding a micro porous layer onto catalyst layer can hinder saturation water from entering catalyst layer immediately to decrease the concentration of saturation water in the catalyst layer. Besides, coating carbon slurry onto the carbon paper can make its surface smooth to decrease electric resistance effectively since the roughness of the carbon paper surface has influences on the catalyst layer.

2.2. PEMFC materials and specifications

The membrane electrode assembly (MEA) used in the present experiments consists of three layers, i.e., the anode catalyst layer, the cathode catalyst layer and the PEM. It is manufactured by GORE®, named PRIMEA 5621, with the

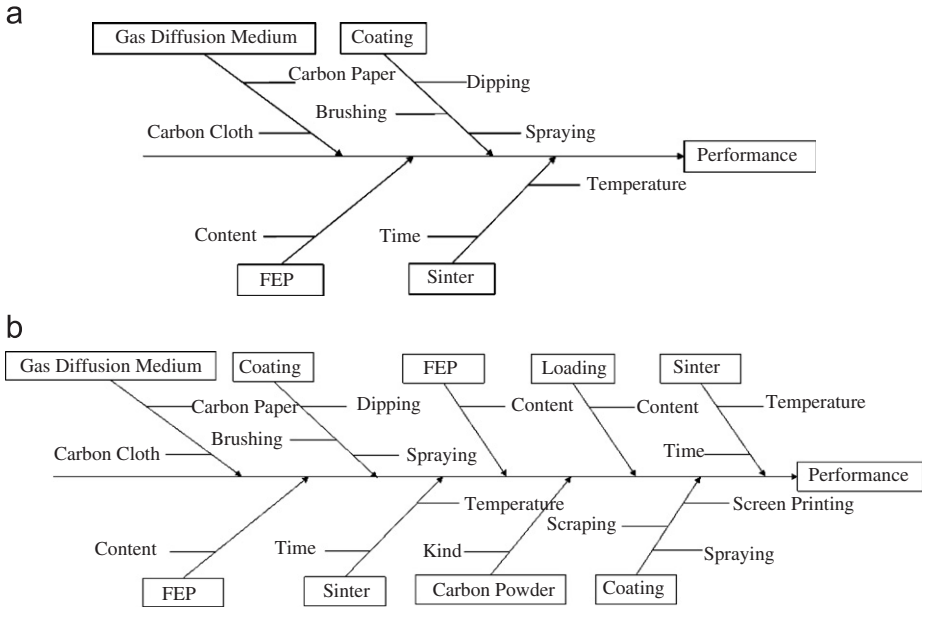

Fig. 1. Flow charts of the fabrication processes of gas diffusion layer (GDL): (a) one-stage process; (b) two-stage process.

reactive area of $5 \times 5\,cm^2$, the Pt loading of $0.45\,mg/cm^2$ at anode and $0.6\,mg/cm^2$ at cathode and the PEM thickness of 35 μm. The test GDL is made by us. The backing material is TGP-H-090 carbon paper produced by Toray Corp., with a thickness of 270 μm and size of $5 \times 5\,cm^2$. The hydrophobic material and carbon powder used in GDL are FEP solution produced by Dupont and Vulcan XC-72R, respectively. The bipolar plates used in the experiments are AXF-5QCF high density carbon plate produced by POCO, which has the material of pure graphite plate, the size of $10 \times 10\,cm^2$, the thickness of 10 mm. In the central section, with the size of $5 \times 5\,cm^2$, the serpentine flow passages are machined with the cross section of $1 \times 1\,mm^2$. The collector plates with the size of $10 \times 10\,cm^2$ and a thickness of 2 mm are made of copper and gold-plated surface to increase its surface conductivity and reduce contact resistance in connection of the collector plate with bipolar plates. The end plate surfaces are made of stainless steel with the size of $10 \times 10\,cm^2$ and the thickness of 10 mm.

2.3. Test conditions

In this paper, flow is controlled with stoichiometric mode with values of 1.5 at anode and 2.0 at cathode. In the experiments, the cell temperature is set as 65 °C, and the inlet humidifying temperatures at cathode and anode are 70 and 80 °C, respectively. The cell pressure is the same as the normal atmosphere.

3. Results and discussion

Adding micro porous layer on GDL of PEMFC has influences on water management, limiting current density and cell overall performance. In this paper, the influences of one-stage and two-stage fabrication processes on cell performance will be discussed as follows. To study the micro porous layer effects, three types of carbon paper with 10%, 20%, 30% FEP contents are used in GDL, and both the conditions with and without micro porous layer (30% FEP + 70% XC-72R, carbon loading $2\,mg/cm^2$) are considered. The unit fuel cell test results are shown in Fig. 2. When the inlet fuel gas at the cathode is air, which comprises $\frac{1}{5}$ oxygen and $\frac{4}{5}$ nitrogen, the limiting current density phenomenon occurs at a high current density. So, it is more important to improve the diffusion of air. When adding micro porous layers into the carbon paper, not only the cell performance improves significantly but also the limiting current density is delayed. It is because the micro porous layer is beneficial to the mechanical support, water management, and gas diffusion.

In order to investigate the influences of FEP content in the first stage of the two-stage fabrication process on cell performance, the GDLs made with carbon papers of 10%, 20%, 30% FEP contents and coated by micro porous layers (30% FEP + 70% XC-72R, carbon loading $2\,mg/cm^2$) are used. The test results of the unit cells using above GDLs are shown in Fig. 3, in which it is found that cell performance and limiting current density increase with decreasing FEP content in carbon paper for the cases with air as the fuel gas. This indicates

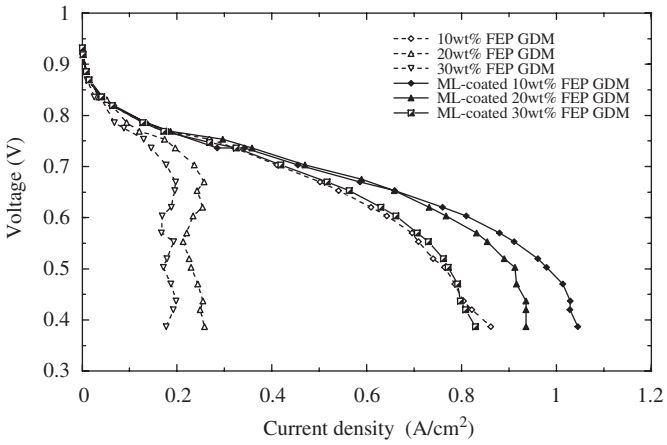

Fig. 2. Effects of FEP contents in GDLs with and without coating micro porous layer.

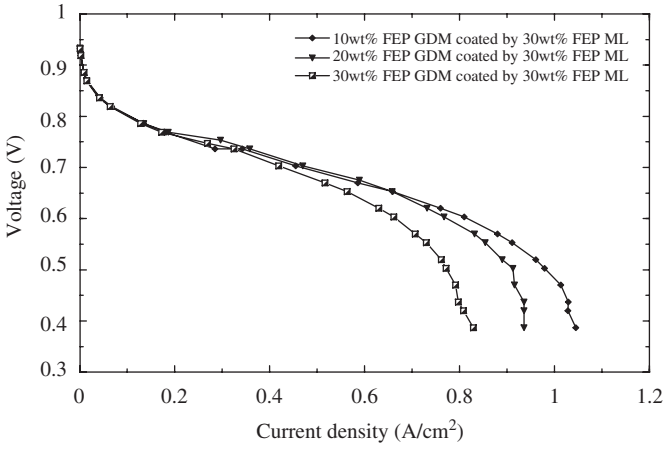

Fig. 3. Effects of FEP content in GDL for the first stage of two-stage fabrication process on cell performance.

that low hydrophobicity is good for air transport. Moreover, the limiting current density reduces with a decrease in the FEP content, because oxygen content in air is only $\frac{1}{5}$, and the rest are nitrogen and other gas components, and therefore nitrogen can accumulate on catalyst layer surfaces and hinder air transport. Hence, less hydrophobic material is needed for air to avoid pore decreasing. As known from the above discussion, in the first stage of two-stage fabrication processes, the cell performs better with 10% FEP in carbon paper.

Fig. 4 illustrates the influences of FEP content in micro porous layer on cell performance to understand the effects of FEP content in the second stage of the two-stage fabrication process. In the experiments, GDLs are prepared with 10% FEP in carbon paper, and micro porous layer coating with three different FEP contents, namely, 10%, 20%, 30%, and the carbon loading of $2\,mg/cm^2$. With these GDLs unit cell tests are performed. The results showed that when air is the oxidant, the operation voltage is above 0.45 V and the micro porous layer contains 20% FEP, the cells perform the best because of the optimal usage of catalyst. It is also found that less FEP

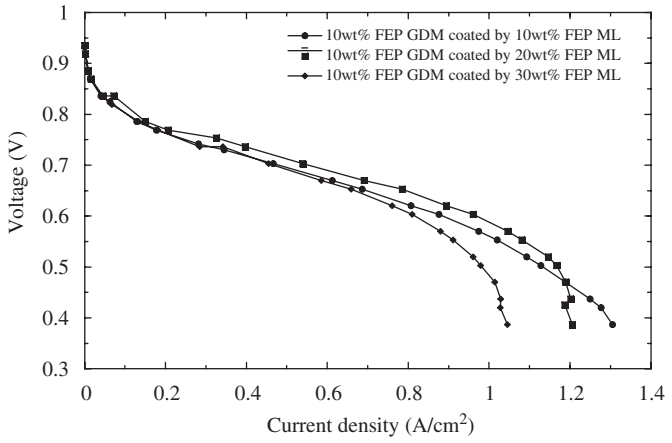

Fig. 4. Effects of FEP content in micro porous layer for the second stage of two-stage fabrication process on cell performance.

content leads to more delay of limiting current density. In the past literature, too large or too small pores in micro porous layer could affect the distribution of liquid water content and then the cell performance. Discovered in our measurement, the optimal FEP content in micro porous layer is 20% when the operating voltage is above 0.45 V.

The comparison of performance curves is shown in Fig. 5 to understand the influences of FEP content on GDL. In this figure, all GDLs with different FEP contents and different fabrication processes of carbon paper, including untreated, one-stage, and two-stage processes are compared. For the cases of oxygen as the oxidant and without coating micro porous layer, a higher FEP content such as 20% or 30% at low voltage region ($V < 0.6$ V) may lead to liquid water blocking pores and hindering oxygen transfer in GDL, see Fig. 5(a). But if the FEP content is too low, such as 0% or 5%, the cell performance decreases. While with a micro porous layer coating on the GDL, the cell performance is obviously better than that without coating micro porous layer. In Fig. 5(a), it is found that the cells with a micro porous layer on GDL perform even better than that with commercial carbon cloth. Furthermore, it is also found that the cells achieve the best performance with 10% hydrophobic material in carbon paper and 20% in micro porous layer. When air is used as oxidant and no micro porous layer is coated on carbon paper, cell performance at low voltage region ($V < 0.6$ V) is quite poor obviously. This is because of serious water flooding appearing in GDL. For GDL with coating micro porous layer, cell performance is improved significantly due to proper water management. When the operating voltage is above 0.6 V, the cell with GDL of 20% FEP in micro porous layer performs best. For limiting operation condition, the cells with two-stage fabrication process perform better in extending limiting current density than that with one-stage fabrication process. But carbon paper of 30% FEP with micro porous layer of 30% FEP and carbon paper of 10% FEP without micro porous layer have the same limiting current density, because too much FEP content in carbon paper and micro porous layer lead to small pores and then large mass transfer loss. However, in two-stage

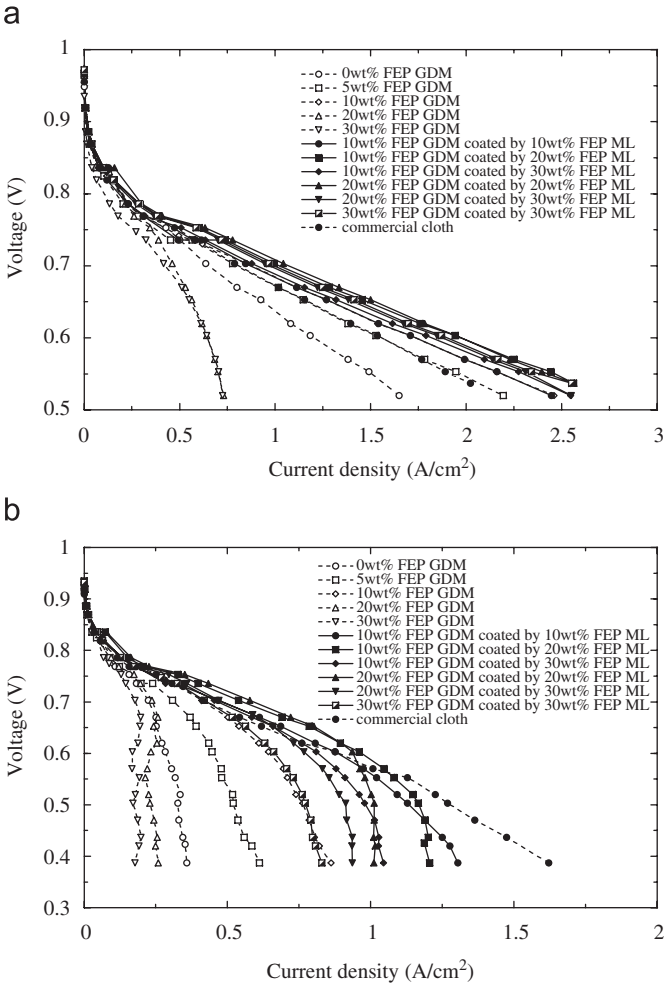

Fig. 5. Effects of FEP content in GDL with one stage or two-stage fabrication process on cell performance: (a) oxygen; (b) air.

fabrication processes, the GDL with low FEP content can delay the occurrence of limiting current density. Thus, GDL with 10% FEP in both carbon paper and micro porous layer delays the occurrence of limiting current density most effectively; GDL with 10% FEP in carbon paper and 20% FEP in micro porous layer will cause the optimal power curve of fuel cell.

Carbon loading also has influences on GDL performance. To investigate the influences, carbon paper of 10% FEP is used in the GDL fabrication processes and then micro porous layer of 10% FEP and 90% XC-72R is coated, therein carbon loadings of 1, 2, 3 mg/cm^2 are, respectively, used. Unit cell tests on those micro porous layers with different carbon loading are performed and the results are shown in Fig. 6. For the cases of air as the cathode fuel gas and the carbon loading of 3 mg/cm^2, the data implies that the micro porous layer with the carbon loading of 3 mg/cm^2 is confronted with gas diffusion and water management problems. Because the gas diffusion in too thick micro porous layer needs to undergo long distance and small pores in the mass transport, which leads to the early occurrence of limiting current density. When micro porous layer has the carbon loading of 1 mg/cm^2, the cell performs better and

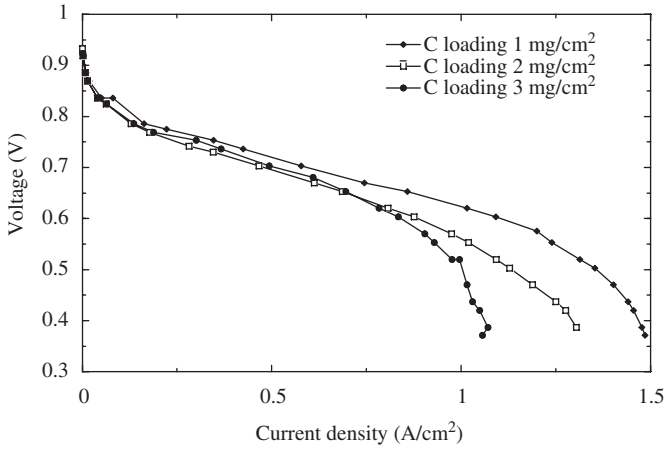

Fig. 6. Effects of carbon loading in micro porous layer on the cell performance.

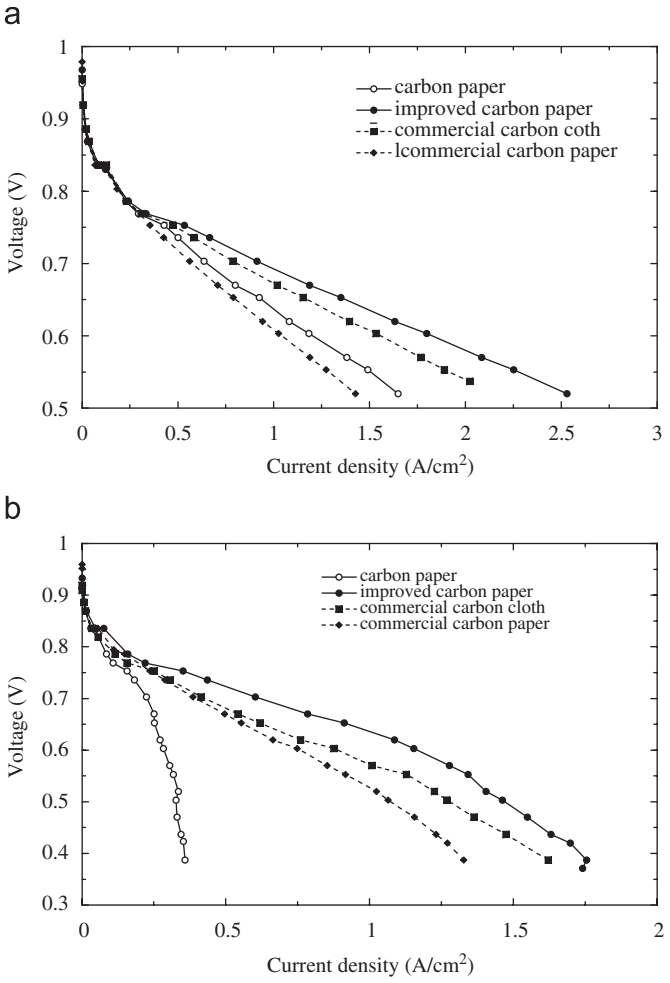

Fig. 7. Comparison of cell performance with different GDLs (untreated carbon paper, improved carbon paper, commercial carbon paper and commercial carbon cloth): (a) oxygen; (b) air.

the occurrence of the mass transport loss can be delayed. Because the thin micro porous layer and thus large pores provide short gas diffusion distance and can improve gas diffusion performance effectively. However, a thin micro porous layer has large electric resistance. Therefore, a too thin micro porous layer will decrease electrical and ion conductivity.

To find out the influences of various carbon papers on cell performance, untreated carbon paper, improved carbon paper, commercial carbon paper and commercial carbon cloth are used as GDL for test cells in this study. Unit cell tests with these GDLs under the same experimental conditions are performed and the results are compared in Fig. 7. For oxygen as the oxidant, as shown in Fig. 7(a), improved carbon paper has much better performance than untreated carbon paper and even better than commercial carbon paper and commercial carbon cloth. For the cases of air as the oxidant, as shown in Fig. 7(b), comparison between improved carbon paper and untreated carbon paper reveals that the current density increases from 0.3 to 1.15 mA and cell performance curve increases by about 70% under the operating voltage of 0.6 V. Besides, improved carbon paper recommended in this paper also has better performance than commercial carbon paper or commercial carbon cloth. In general, improved carbon paper proposed and manufactured in the present study can effectively improve gas transfer, ohmic resistance and water management in fuel cells.

4. Conclusion

In this paper, the effects of GDL fabrication process, FEP content, and carbon loading on cell performance of PEM fuel cells have been investigated experimentally. According to the present measurements, the following conclusions can be drawn.

1. The GDL fabrication process employing two-stage method, I i.e., coating GDL with a micro porous layer, can improve cell performance effectively and delay the occurrence of limiting current density, because adding micro porous layer onto GDL improves gas transport and reduces ohmic resistance inside GDL effectively, and also provides proper water management capability.
2. In the two-stage fabrication process of GDL, FEP contents in carbon paper and micro porous layer have obvious effects on cell performance. The GDL with the carbon paper of 10% FEP and micro porous layer of 20% FEP results in the best cell performance.
3. With air, the fuel cell achieves much better cell performance with the carbon loading of 1 mg/cm^2 in the micro porous layer due to the presence of the thin micro porous layer and large pores in GDL.

Acknowledgement

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