

Short communication

Porosity-graded micro-porous layers for polymer electrolyte membrane fuel cells

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Abstract

In this paper, the effect of porosity-graded micro-porous layer (GMPL) on the performance of polymer electrolyte membrane fuel cells (PEMFCs) was studied in detail. The GMPL was prepared by printing micro-porous layers (MPL) with different content of NH_4Cl pore-former and the porosity of the GMPL decreased from the inner layer of the MPLs at the membrane/MPL interface to the outer layer of the MPLs at the gas diffusion electrode/MPL interface. The morphology and porosity of the GMPLs were characterized and the performance of the cell with GMPLs was compared with those having conventional homogeneous MPLs. The result demonstrates that the fuel cells consisting of GMPL have better performance than those consisting of conventional homogeneous MPLs, especially at high current densities. Micro-porous layer with graded porosity is beneficial for the electrode process of fuel cell reaction probably by facilitating the liquid water transportation through large pores and gas diffusion via small pores in the GMPLs.

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1. Introduction

Polymer electrolyte membrane fuel cells (PEMFCs) have been attracting enormous research interest as promising power sources for vehicles, stationary power generations and portable electronics devices [1–4]. The performance of a PEMFC is critically dependent on the electrocatalytic activity of the precious metal catalyst such as platinum and platinum alloy and performance of various components including the gas diffusion electrode (GDE) [5–7]. The gas diffusion electrode (GDE) is one of the key components of PEMFCs where the reactions of the diffusion of reactant gas and the discharge of the liquid water formed occur. As a result, the microstructure of the GDE has significant influence on the performance of the PEMFCs [6,8–10].

The performance of a GDE can be improved by attaching a micro-porous layer (MPL) consisting of carbon black and hydrophobic polytetrafluoroethylene (PTFE) [11–15]. Qi et al.

experimentally demonstrated the advantage of placing a MPL between the gas diffusing layer (GDL) and the catalyst layer [16]. Chang et al. [17] studied the effects of the pore size on gas transport and cell performance, and suggested that porosity distribution of GDL should be bimodal, facilitating the water discharge through large pores and gas diffusion through small pores. Wilkinson et al. [18] simulated the structure of the GDE, and concluded that both the operation parameter and the MEA structure should be graded. Based on the calculation of the capillary pressure, Nam et al. [19] indicated that a pore and water saturation “jump” in GDL would improve the water and gas transport. Chu et al. [20] investigated the effect of average porosity on the oxygen transport and suggested that a porosity-graded GDL should be beneficial to the fuel cell performance. The results of Roshandel et al. also revealed the important effects of porosity distribution on PEM fuel cell performance [21]. Recently, based on a two-phase flow in fuel cell electrode, Hotinen et al. [22] suggested a GDE with a graded porosity is more favorable for the liquid water removal from the catalyst layer. This conclusion was also supported by the computer simulation results [23].

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However, as shown above there is lack of experimental studies on the effect of the porosity-graded MPL on the electrocatalytic activities of catalyst layer and performance of PEMFCs. In this paper, MPL with graded porosity (GMPL) was prepared and the effect of graded porosity in the MPL on the cell performance was studied in detail. The results indicate that GMPL enhances the performance of PEM fuel cells.

2. Experimental

2.1. Preparation of GMPL

The wet-proofed carbon paper was prepared by immersing a carbon paper (Toray TGPH-060) in a polytetrafluoroethylene (PTFE) dispersion (ZhongHao, 60 wt%, China) for 5 min, followed by drying in air for 15 min and heat-treating in oven at 350 °C for 30 min. The PTFE loading was about 30 wt%.

To prepare the GMPL, carbon powder (Vulcan XC-72, Cabot Co.) was mixed with an appropriate amount of distilled water and PTFE dispersion in a supersonic mixer for 60 min. NH_4Cl was added as pore-former to the suspension and stirred for another 60 min to form MPL slurry.

The GMPL was prepared by three steps. First, a wet-proofed carbon paper was screen printed with a MPL slurry with 50 wt% NH_4Cl pore-former, labeled as L-1. This was followed by a second coating with a MPL slurry with 30 wt% NH_4Cl pore-former (referred to L-2) and a third coating with a MPL slurry with 10 wt% NH_4Cl pore-former (referred to L-3). As-prepared GMPL was sintered in oven at 350 °C for 30 min. The carbon loading of each layer in the GMPL was about 0.8 mg cm^{-2} and total carbon loading was about 2.4 mg cm^{-2} . The PTFE content was about 30 wt%.

For the purpose of comparison, conventional homogeneous MPL samples were also fabricated by the screen-printing of a MPL slurry with NH_4Cl pore former content of 10 and 50 wt% on the wet-proof carbon paper.

Catalyst coated membrane (CCM) with Nafion 112 membrane and Pt loading of 0.4 mg cm^{-2} for both anode and cathode side was used to investigate the single cell performance of the GDE samples. The CCM was fabricated as follows. First, 1 g Pt/C catalysts (40 wt% Pt/C, Johnson Mathey) were mixed with 10 mL deionized water under vigorous stirring. Then 24 mL

Nafion DE 520 solutions (5 wt%, EW 1000, Du pont) was added to the mixture, followed by ultrasonic treatment for 30 min and high-speed homogenizer (20,000 rpm) for 1 h to form catalyst slurry. The catalyst slurry was sprayed to a PTFE sheet with Pt loading of 0.4 mg cm^{-2} . Then the catalyst layer was transferred to Nafion 112 membrane to form CCM. Fig. 1 shows schematically the preparation procedures of the membrane-electrode-assembly based on the GMPL.

2.2. Characterization

The surface and the cross-section of GMPL were observed using scanning electron microscopy (SEM, JSM-5600). The sample for the cross section analysis was prepared by freezing the sample in liquid nitrogen and cutting it with a blade.

The porosity distribution of GMPL and MPL was measured by a mercury-intrusion porosimetry (PM-33, Quantachrome Instruments). During the test, the sample was immersed in hydrargyrum, filling both hydrophilic and hydrophobic pores in the GDL and MPL.

The performance of the single cell was measured with H_2 as fuel gas and air as oxidant at 60 °C without back pressure in a fuel cell test station (ElectroChem. Co.). The external humidification temperature of both H_2 and air was kept constant at 60 °C. H_2 and air flow rates were 300 and 2000 sccm, respectively. Prior to record the performance curve, cells were activated by polarization at a constant current till stable performance was obtained.

3. Results and discussion

3.1. Microstructure of MPL

The surface micrographs of micro-porous layers, L-1, L-2, and L-3 are shown in Fig. 2. Fig. 3 shows the cross-section of a GMPL sample with three micro-porous layers of L-1, L-2, and L-3. The surface of MPL becomes coarse and more porous from L-3 to L-1. It appears that both pore size and porosity decrease from L-1, L-2 to L-3. As shown in Fig. 3, the contact between various micro-porous layers is good. From L-1 to L-3, MPL become denser, and size of pores also decreases. This indicates that a graded porous structure is formed in the MPL.

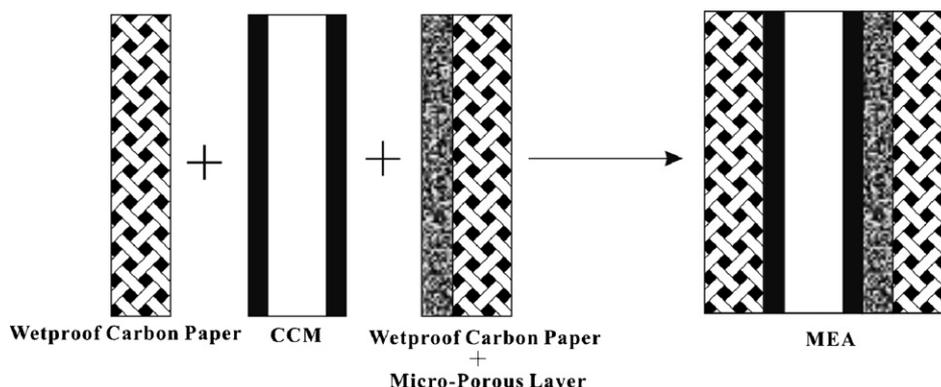


Fig. 1. Preparation procedure of membrane-electrode-assembly using porosity-graded micro-porous layer.

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