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Preparation of microporous layer for proton exchange membrane fuel cell by using polyvinylpyrrolidone aqueous solution

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ABSTRACT

A new method of preparing microporous layer (MPL) for proton exchange membrane fuel cell (PEMFC) was presented in this paper. Considering the bad dispersion of PTFE aqueous suspension in the carbon slurry based on ethanol, polyvinylpyrrolidone (PVP) aqueous solution was used to prepare carbon slurry for microporous layer. The prepared gas diffusion layers (GDLs) were characterized by scanning electron microscopy, contact angle system and pore size distribution analyzer. It was found that the GDL prepared with PVP aqueous solution had higher gas permeability, as well as more homogeneous hydrophobicity. Moreover, the prepared GDLs were used in the cathode of fuel cell and evaluated with fuel cell performance and EIS analysis, and the GDL prepared with PVP aqueous solution indicated better fuel cell performance and lower ohmic resistance and mass transfer resistance.

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Introduction

As a promising alternative power sources for stationary and mobile applications, the development of proton exchange membrane fuel cells (PEMFCs) has drawn considerable interest over the past few years [1–3]. Situated between the catalyst layer and bipolar plate, the GDL should be a conductive porous material in which reactant gas can diffuse from the flow field to the active catalyst surface where the electrochemical reaction occurs and simultaneously produced water can be transported from the catalyst layer to the flow field [4].

Effective water management is considered to be of great importance in achieving high performance and improving the durability of PEMFC [5]. However, achieving an optimal water management requires a good balance between the timely removal of produced water from catalyst layer to avoid water flooding and the necessary hydration level inside the polymer electrolyte membrane to increase the hydrogen proton flux from the anode to the cathode [6,7]. Without good water management, the reactant convection and diffusion can be hindered and the fuel cell performance will degrade accordingly [8,9]. GDL is considered to be the component most

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responsible for the optimal water management [10,11]. Typically, the GDL is a dual-layer structure with a substrate and a microporous layer (MPL). The non-woven carbon paper or woven carbon cloth with good electric conductivity and high porosity is always adopted as the substrate material [12]. The MPL composed of the mixture of carbon black particles and polytetrafluoroethylene (PTFE) is coated on one side of the substrate facing the catalyst layer. The MPL works well in minimizing the contact resistance and providing a better mechanical compatibility with the catalyst layer [13]. Besides, the addition of MPL can improve the total water management capability of GDL, and therefore achieving higher limiting current density and better fuel cell performance [14,15]. Thus, the MPL deserves much attention in improving the performance of PEMFC. Studies on the optimization of the content of PTFE [12,16,17] and carbon loading [18–20] in MPL and modification of pore structure of MPL [21–25] have been carried out. Besides, the preparation of MPL should be a key parameter to be optimized and designed [26]. In general, the carbon slurry for MPL is obtained by dispersing carbon black and PTFE aqueous suspension in ethanol based dispersants. But in fact, the PTFE aqueous suspension with low surface energy cannot be homogeneously dispersed in the ethanol based dispersants, which would cause a great effect on the characteristics of MPL. Up to now, very few literature have paid attention to optimization of the preparation of MPL and studied the effect of carbon slurry adopting other dispersants instead of ethanol based dispersants on the characteristics of MPL.

The objective of the present work is to develop a new method for fabrication of GDL, especially the MPL part. The carbon slurry for MPL adopts polyvinylpyrrolidone (PVP) aqueous solution instead of ethanol as the dispersant. The carbon paper substrate is painted PTFE aqueous suspension on the side close to bipolar plate rather than totally dipped into the PTFE aqueous suspension. Besides, the characteristics of prepared GDL and its effect on fuel cell performance are carried out.

Experimental

Fabrication of GDLs

The GDL was fabricated with carbon paper (Toray, TGP-H-060) as the substrate and the hydrophobic characteristic of the MPL was provided by PTFE. The carbon slurry for MPL with PVP aqueous solution as the dispersant was prepared by blending acetylene black carbon powder with PVP aqueous solution using ultrasonication and mechanical stirring and then 5 wt% PTFE aqueous suspension was added. Herein, the molecular weight of PVP was 58,000 and the supplier was Aladdin Chemistry Co. Ltd. For comparison purposes, a carbon slurry for MPL with ethanol as the dispersant was also prepared. The carbon slurry was coated onto one side of the carbon fiber substrate, followed by drying at 80 °C and then sprayed with some PTFE aqueous suspension on the other side of the substrate. After coating MPL on one side and spraying PTFE on the other side, the GDL was dried at 240 °C and sintered at 340 °C for 30 min, respectively. For the GDLs with different dispersant, the carbon loading on the substrate was 0.5 mg cm⁻² and the mass ratio of PTFE and carbon in the MPL was 0.4:1.

Characterization of GDLs

The surface morphology of GDLs was examined with a field emission scanning electron microscopy (FESEM; Hitachi, S-4800), the surface contact angles of GDL were measured by a contact angle system (Drop Shape Analyzer 100, Kruss, Germany). The pore size distribution of GDLs was characterized using mercury intrusion porosimetry (Quantachrome PoromasterGT 60). The gas permeability of GDLs was characterized by pore size distribution analyzer (PSDA-20) and calculated by equation (1).

$$\mu = \eta \times \frac{L}{A} \times \frac{Q}{\Delta P} \quad (1)$$

Where η is gas viscosity, L is the thickness of the sample and A is the cross-sectional area through which the gas passes, Q is the volumetric flow rate and ΔP is the pressure difference across the sample.

Dispersibility and stability of PTFE in different dispersants

500 mg PTFE aqueous suspension (5 wt%) was added into 5 ml deionized water, ethanol and PVP aqueous solution (5 ml deionized water : 4 mg PVP), respectively. After the above dispersions were well stirred, the dispersibility and stability of PTFE was examined.

Single cell test conditions

A single cell test was carried out using a 5 cm² active area test cell and the commercial catalyst coated membrane (CCM). For the CCM, the membrane material was Nafion 212 membrane and the catalyst loading of anode and cathode was 0.2 and 0.4 mg cm⁻², respectively. Two pieces of GDLs with an effective area of 5 cm² were hot-pressed onto two sides of one piece of CCM to fabricate a MEA. The hot pressing was performed at 140 °C under a pressure of 0.1 MPa for 2 min. The MEA was sandwiched into a single cell consisted of Ag-plated stainless end plates and two graphite flow fields. Hydrogen (99.99%) and oxygen (99.5%) were used as the anode fuel and cathode reactants, respectively. The fuel cells with different cathode GDLs were operated at 65 °C with H₂/O₂ at the gauge pressures of 0.05 MPa and flow rates were 50 ml min⁻¹ and 100 ml min⁻¹, respectively. The relative humidity of the reactant gases were maintained at 100% by controlling the humidity bottle temperatures. The output performances of the cells were tested with an electronic load PLZ70UA (Kikusui, Japan). Besides, the in-situ electrochemical impedance spectroscopy (EIS) was performed at the current density of 200, 300 and 2000 mA cm⁻² under the same operation conditions of the evaluation of single cells to investigate the effect of MPL with different preparation. The measurements were carried out in the frequency range from 0.1 Hz to 10⁶ Hz.

Results and discussion

Dispersibility and stability of PTFE

In order to investigate the disperse characteristic of PTFE aqueous suspension in different dispersants, 500 mg PTFE

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