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Effect of diffusion layers fabricated with different fiber diameters on the performance of low temperature proton exchange membrane fuel cells

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HIGHLIGHTS

► A new material for GDL precursor is introduced.
► FEP and phenolic resin distributions for PAN fiber felt.
► Diameter of PAN fiber, concentration of phenolic resin and FEP are related to permeability.
► Fuel cell operation has been influenced by the permeability.

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ABSTRACT

This study discusses the relationship between performance and carbon fiber diameter (400 nm–1 μm) in fuel cells employing carbon fiber paper produced from PAN fiber felt, and also examines the effect of carbon fiber paper thickness, air permeability, porosity, and surface resistivity on performance. The researchers fabricate gas diffusion layers (GDLs) with a small carbon fiber diameter from PAN fiber employing the two processes of stabilization and carbonization, and investigate the relationship between fiber diameter and air permeability in the gas diffusion layer material. Carbon fiber paper made in this study is left as is or impregnated with 10 wt% phenolic resin or FEP. When the tested area is 25 cm^2, the test temperature 40 °C, and the carbon fiber paper impregnated with 10 wt% phenolic resin, the paper has a fiber diameter of 1 μm and an air permeability is 29 $cm^3\ cm^{-2}\ s^{-1}$, and a test fuel cell yields 997 mA cm^{-2} at a load of 0.5 V. Carbon fiber paper impregnated with 10 wt% FEP has a smaller carbon fiber diameter of 400 nm and an air permeability of only 1 $cm^3\ cm^{-2}\ s^{-1}$; a test fuel cell made with this material yields 683 mA cm^{-2} at a load of 0.5 V.

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1. Introduction

Proton exchange membrane fuel cells (PEMFCs) are considered to be superior to other fuel cell systems in vehicle applications due to their higher efficiency and power density and lower operating temperature and noise [1–4]. Gas diffusion layers (GDLs), which provide a channel for the transport of fuel and the transmission of current in PEMFCs, are typically made of carbon fiber paper or carbon fabric, and the composition of a GDL can significantly affect PEMFC performance [5].

Carbon has the advantages of high conductivity and resistance to corrosion, and is well suited to the environment inside a PEMFC. Graphitization of carbon fiber at a temperature of up to 1800 °C increases the conductivity of the fiber while reducing the hydrophobicity of surface functional groups on the fibers [6,7]. When carbon fiber is used to produce a GDL, generally employing a heat treatment temperature in excess of 1400 °C, the different carbon structures obtained employing different carbonization methods can affect the performance of the resulting PEMFC [8]. The production of carbon fiber with good electrical conductivity from polyacrylonitrile (PAN) fiber requires three steps: stabilization, with a temperature range of 200–300 °C, carbonization, at a temperature of approximately 1000 °C in an inert gas such as nitrogen, and graphitization, which requires heating to 1500–3000 °C under controlled conditions [11–18]. While carbon fiber

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fabric is prone to warping and shrinking in fuel cells, carbon fiber paper offers an excellent stability. Moreover, the addition of phenolic resin to the carbon fiber paper can improve PEMFC performance [9]. In addition, although the addition of fluorinated ethylene propylene (FEP) will reduce PEMFC performance, it can increase hydrophobicity and therefore reduce flooding [10].

Due to the beneficial effect on reaction kinetics, catalyst tolerance, heat rejection, and water management, the operation of a PEMFC at a high temperature (>80 °C) is considered to be an effective way to improve fuel cell performance [21–32]. While there are several compelling reasons for operating at a higher temperature, several researchers have studied PEMFC operation at room temperature [33,34], and we decided to operate PEMFCs at 40 °C in this experiment since we plan to apply fuel cells in products that require a low relative humidity. Gasket thickness will also affect performance, and PEMFC performance is optimal when GDL thickness is slightly greater than that of the gasket [20]. We specifically chose a gasket thickness of 0.06 mm in this experiment.

Past research [11–18] indicates PAN fibers need much time and processing before they can be can be transformed into graphite fiber. Graphite fiber has the best structure, followed by carbon fiber, and finally by PAN fiber with the worst structure. In addition, graphite fiber is the most expensive, carbon fiber is second, and PAN fiber is the least expensive. In our previous research [8], the better the structure of the fiber, the greater the conductivity. Graphite fiber has the highest conductivity, followed by carbon fiber and PAN fiber. When commercial carbon fiber paper (such as SGL, TORAY) is used to produce fuel cell electrodes, the first step is to carbonize the carbon fiber at a temperature of 1800 °C or higher to obtain graphite fibers. In our previous study [19,20], we used oxidized carbon fiber felt made from oxidized fibers, which had been carbonized at a temperature of 1000 °C–2500 °C. In this study, we used selectively oxidized fibers of the precursor material, PAN fiber felt, to investigate the relationship between fuel cell performance and fiber diameter.

The ultimate purpose of this study is to increase PEMFC performance and reduce cost by improving GDLs. In particular, we investigate the relationship between fuel cell performance and GDLs produced with different PAN fiber diameters, and the examine the effect of varying the thickness, air permeability, porosity, and surface resistivity of the carbon fiber paper on fuel cell performance. The results obtained are compared with relevant data for GDLs made from commercial carbon paper (Toray TGP-H 030).

2. Experimental parameters

Three different types of PAN fiber felt (supplied by the Industrial Technology Research Institute), denoted as E, H and S, and phenolic resin (supplied by Chang Chun Plastics Co., Ltd.) were used. Methanol solutions with and without 10 wt% phenolic resin was prepared. Each type of PAN felt had different characteristics, including diameter, yard weight, air permeability, and thickness. Type E was produced by electro-spinning and types H and S were both made by melt spinning. Table 1 shows the characteristics of each type of PAN felt.

One set of type E, H, and S PAN fiber felt pieces were immersed in a methanol solution containing 10 wt% phenolic resin, yielding felt types denoted as E-10R, H-10R, and S-10R, which were then baked in an oven at 70 °C for 15 min. Two sets of the same types of PAN fiber felt were also immersed in a methanol solution containing no phenolic resin. One of the sets contained felt types denoted as E–N, H–N, and S–N, and the another set, which was impregnated with FEP in a later step, contained felt types denoted as E-10F, H-10F, and S-10F. All PAN fiber felts were then stabilized at 280 °C in an air atmosphere, creating oxidized fiber felts, before being carbonized at 1000 °C in nitrogen to produce carbon fiber papers. A carbonization temperature of 1000 °C was chosen because we expected that a lower carbonization temperature would reduce the carbon fiber diameter, and thus achieve a better carbon structure. The E-10F, H-10F, and S-10F carbon fiber papers were then impregnated with 10 wt% FEP and sintered in nitrogen at 340 °C.

Table 1
Characteristics of PAN fiber felt and carbon fiber paper.

	Yard weight (g m^{-2})	Permeability (cm^3 cm^{-2} sq^{-1})	Thickness (mm)	Fiber volume fraction (kg m^{-3})
Type E	32	2	0.3	107
Type E-N(1000 °C)	37	0.9	0.07	529
Type E-10R	77	0	0.2	385
Type E-10F	40	1	0.07	571
Type S	50	30	0.3	167
Type S-N(1000 °C)	41	27	0.16	256
Type S-10R	46	21	0.18	256
Type S-10F	43	22	0.17	253
Type H	35	35	0.2	175
Type H-N(1000 °C)	21	36	0.11	191
Type H-10R	28	29	0.13	215
Type H-10F	22	29	0.11	200
Toray TGP-H 030	44	78	0.11	400

The thickness, air permeability, porosity, and surface resistivity of each piece of carbon fiber paper were measured. The thicknesses of the carbon fiber papers were measured using a Teclock SM-114 thickness tester, and consisted of the average of measurements taken at five random points. Air permeability measurements were performed in accordance with Gurley Model 4110 guidelines using a Gurley Model 4320 m. The porosity of the carbon fiber paper was measured in accordance with ASTM D-570. A Loresta GP MCP-T600 m was used to measure the surface resistivity of the carbon fiber paper in accordance with JIS K 7194. A cold field emission scanning electron microscope was used to observe the surface of the carbon fiber paper.

The carbon fiber paper and the commercial carbon paper (Toray TGP-H 030) were prepared as PEMFC GDLs. Each type of paper was cut into 5 × 5 cm pieces, which were used to make three-layer membrane electrode assemblies (MEAs) containing catalyst-coated membranes (CCM) consisting of Dupont NRE-211. The MEAs were then placed in a fuel cell testing module. Each MEA had an activated area of 25 cm^2, and the bipolar plates consisted of gate-type grooved graphite plates made of highly compacted graphite. The gas flows at the anode (H_2) and the cathode (O_2) were 500 cc min^{-1}, and the gases had a relative humidity of 95%. The temperatures of the anode and cathode were both 40 °C. All single cell operations were performed without external pressurization, and pure humidified hydrogen and pure oxygen were used. The gasket thickness was 0.06 mm.

3. Results and discussion

Fig. 1 shows thickness and surface resistivity curves for different types of carbon fiber paper produced from PAN fiber felt. The commercial carbon paper (Toray TGP-H 030) had a thickness of 0.11 mm. The thicknesses of the E–N, H–N, and S–N paper were 0.07 mm, 0.11 mm, and 0.16 mm, and those of the E-10F, H-10F, and S-10F paper were 0.07 mm, 0.11 mm, and 0.17 mm. Because the FEP formed an extremely thin film-like hydrophobic interface layer on the surface of the carbon fiber paper, impregnation with FEP did not significantly change the thickness of the paper. However, the carbon fiber papers impregnated with 10 wt% phenolic resin were relatively thick. The thicknesses of E-10R, H-10R, and S-10R paper

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