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Triple microporous layer coated gas diffusion layer for performance enhancement of polymer electrolyte fuel cells under both low and high humidity conditions



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HIGHLIGHTS

• Novel triple MPL coated GDL was developed to enhance the PEFC performance.

• The thin hydrophilic layer coated on the hydrophobic double MPL is effective at conserving MEA hydration under low humidity.

• The triple MPL with an appropriate hydrophobicity gradient was effective at reducing flooding under high humidity.

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ABSTRACT

Enhancement of the performance of polymer electrolyte fuel cells (PEFCs) requires an appropriate water balance between the conservation of membrane humidity and the discharge of excess water produced in the cell. In the present study, a novel triple microporous layer (MPL) coated gas diffusion layer (GDL), in which a hydrophilic layer was coated on a hydrophobic double MPL, was developed to enhance the PEFC performance under both low and high humidity. The thin hydrophilic layer in the triple MPL is effective at conserving the humidity of the membrane electrode assembly (MEA) under low humidity, while the hydrophobic double MPL between the hydrophilic layer and the carbon paper substrate prevents removal of water from the hydrophilic layer. This results in a significant enhancement of the ability of the GDL to prevent dehydration of the MEA. The triple MPL coated GDL, where the polytetrafluoroethylene (PTFE) content in the hydrophobic MPL in contact with the hydrophilic layer is set to 30 mass% and that in contact with the substrate is set to 10 mass%, is effective at expelling excess water from the catalyst layer, which results in much higher PEFC performance under high humidity than that for a conventional hydrophobic MPL coated GDL.

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1. Introduction

Polymer electrolyte fuel cells (PEFCs) generally have external humidifiers to supply humidified fuel and oxidant gases, which prevents dehydration of the membrane electrode assembly (MEA). However, such additional humidifiers make the PEFC system complex and increase the production cost. The humidification requirements of the anode and cathode are different. At the anode, it is possible to introduce humidified hydrogen gas using water

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generated during the reforming process in fuel production. Furthermore, humidified pure hydrogen gas that is not used for the electrochemical reaction can be recirculated in the PEFC system. It is therefore possible to remove external humidification at the anode. At the cathode, air, from which oxygen is consumed in the electrochemical reaction, is usually exhausted without recirculation. Therefore, the humidified air is generally supplied using external humidification. If a PEFC could be operated without cathode humidification, then external humidifiers could be removed, which would result in a very simplified PEFC system with increased total efficiency and reduced cost. Therefore, one of the most important issues to advance the commercial viability of PEFCs is the development of high performance PEFCs that can operate without cathode humidification [1–5].







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The design parameters for the gas diffusion layer (GDL) in the PEFC, such as its pore size, thickness and hydrophobic and hydrophilic properties, influence the water management characteristics during PEFC operation. Several investigations have demonstrated that a hydrophobic microporous layer (MPL) coated on a GDL substrate can effectively improve the water management characteristics and thereby enhance the PEFC performance [6–13]. An appropriate MPL coated GDL prevents dehydration of the MEA under low humidity conditions and reduces flooding under high humidity conditions. However, the appropriate design parameters for the MPL coated GDL are different under low and high humidity conditions [14]. An MPL coated GDL designed to prevent dehydration of the MEA under low humidity conditions is generally inferior at reducing flooding under high humidity conditions. Even when a PEFC is operated under low humidity conditions, it is essential to prevent flooding due to water produced in the cell at high current densities. Therefore, it is important to develop a robust MPL coated GDL that can enhance the PEFC performance under both low and high humidity conditions.

Although the hydrophobic MPL coated GDLs have been commonly used to improve the water management properties of PEFCs, some studies have reported that either the hydrophilic MPL [15,16] or hydrophilic treatment of the GDL substrate [17] are effective to improve the water management properties of PEFCs, thereby enhancing the PEFC performance. The authors have also reported that the hydrophilic MPL coated GDL using polyvinyl alcohol (PVA) was effective to enhance the PEFC performance under low humidity conditions [18]. Although the ability of the hydrophilic MPL coated GDL to conserve the humidity of the MEA was enhanced, it was relatively easy for water in the hydrophilic MPL to be expelled to dry air in the substrate. Therefore, enhancement in the PEFC performance was not significant when using the hydrophilic MPL coated GDL. Thus the authors have developed a new hydrophilic and hydrophobic double MPL coated GDL [18]. A thin hydrophilic layer using either PVA or titanium dioxide (TiO₂) coated on the hydrophobic MPL was effective to conserve the humidity of the MEA under low humidity conditions, while the hydrophobic intermediate MPL between the hydrophilic layer and the carbon paper substrate prevented the removal of water from the hydrophilic layer. This resulted in a significant enhancement of the PEFC performance under low humidity conditions, compared with that



Fig. 1. Hydrophobic MPL, double MPL and triple MPL coated GDLs used at the cathode.

for a hydrophilic MPL coated GDL. Under high humidity conditions, the double MPL coated GDL with the hydrophilic layer using PVA increased the water accumulation at the cathode catalyst layer, which increased the concentration overpotential and thereby lowered the PEFC performance. However, the double MPL coated GDL with the hydrophilic layer using TiO₂ was effective to reduce flooding under high humidity conditions, which resulted in higher PEFC performance than that for the conventional hydrophobic MPL coated GDL [19].

In the present study, a novel triple MPL coated GDL was developed, in which the hydrophilic layer was coated on a hydrophobic double MPL. The hydrophobic double MPL in the triple MPL coated GDL had a gradient of hydrophobicity due to variation of the polytetrafluoroethylene (PTFE) content, where the PTFE content in the hydrophobic MPL in contact with the hydrophilic layer was set to 30 mass% and that in contact with the substrate was set to either 10 or 40 mass%. The triple MPL was evaluated to determine the optimal parameters for promoting the discharge of excess water from the catalyst layer and thereby enhancing the PEFC performance under both low and high humidity conditions.

2. Experimental

2.1. Tested GDLs

The GDL used at the anode was a commercial carbon paper without an MPL (SGL SIGRACET[®] 24BA), which was loaded with 5 mass% PTFE to impart hydrophobicity [8]. The SGL24BA GDL has a thickness of 190 μ m, an areal weight of 54 g m⁻², and 84% porosity. Fig. 1 shows hydrophobic MPL, double MPL and triple MPL coated GDLs used at the cathode. The hydrophobic MPL coated GDL consisted of a carbon paper substrate (SGL24BA) coated with an MPL of 30 mass% PTFE and carbon black. The hydrophobic MPL was coated on the substrate as follows. A slurry containing a PTFE dispersion liquid (Daikin Polyflon D-210C), carbon black (Denki Kagaku Kogyo DENKA BLACK[®] HS-100), distilled water and a surface-active agent was mixed using an impeller blade-type mixer, and then spread on the substrate using a bar coating machine. The MPL was dried in an oven and then heated at 350 °C to remove any remaining solvent and surfactant, sintering PTFE and carbon black onto the substrate. For both the double MPL and the triple MPL coated GDLs, a hydrophilic layer of 25 mass% TiO₂, 5 mass% silicone, and carbon black was coated on the hydrophobic MPL coated GDL [19]. The PTFE content in the hydrophobic intermediate MPL of the double MPL coated GDL was varied between 10 and 40 mass%. For the triple MPL coated GDL, the PTFE content in the hydrophobic MPL in contact with the hydrophilic layer was set to 30 mass% and that in contact with the carbon paper substrate was set to either 10 or 40 mass%.

2.2. PEFC performance tests

A commercial MEA (GORE PRIMEA[®]5580, membrane thickness: 30 μ m, Pt loadings at the anode and cathode: 0.4 mg cm⁻²) was used in the PEFC performance tests under low and high humidity conditions. The active area of the MEA was 4.2 cm². A separator with a triple serpentine flow channel configuration was used, and the anode and cathode gases were supplied in a counter-flow orientation. The cell temperature was set at 75 °C. Hydrogen and air utilization was set to 70 and 60%, respectively. For PEFC performance tests under low humidity conditions, the relative humidity (RH) of the gas supplied to the cathode was set to 0%, while that supplied to the anode was maintained at 60% RH. For PEFC performance tests under high humidity conditions, the RH of the gases supplied to both the anode and cathode was set to 100%. The

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