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Enhanced low-humidity performance in a proton exchange membrane fuel cell by the insertion of microcrystalline cellulose between the gas diffusion layer and the anode catalyst layer

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ABSTRACT

A high-performance self-humidifying membrane electrode assembly (MEA) was prepared by inserting hygroscopic microcrystalline cellulose (MCC) between the gas diffusion layer (GDL) and the catalyst layer (CL). At low humidity, the MEA exhibited good self-humidification, including high output and good stability. With our optimal MEA, in which the MCC loading was ca. 0.5 mg cm⁻², the current density at 0.6 V reached 1100 mA cm⁻² and the maximum power density was 751 mW cm⁻², at a relative humidity (RH) of 30% for both anode and cathode gases and a cell temperature of 70 °C; the performance was comparable to that of a MEA prepared without added MCC and operated at 100% RH. Further, after 24 h of operation at low humidity and 0.6 V, the MEA's current density decreased by only 9.1%, compared with the 60% decline experienced by the MEA without MCC after 3 h under the same conditions, demonstrating the former's good self-humidification stability. When we attempted to insert the MCC layer elsewhere, including between the membrane and the anode CL, we found that inserting it between the GDL and the anode CL yielded the best performance. The high self-humidification performance of this MEA is attributable to the strong wettability and water-retention capacity of MCC. The MCC layer between the GDL and the anode CL ensured the latter would remain sufficiently wet and accelerated hydrogen activation and proton transfer, resulting in the MEA having high self-humidification under conditions of low humidity and high cell temperature.

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Introduction

Proton exchange membrane fuel cells (PEMFCs) are widely acknowledged as efficient power sources for portable devices, electric vehicles, and residential cogeneration systems [1–3]. However, their commercialization is presently hindered by several problems, a key one being water management, which affects both the performance and the life span of a PEMFC [4–6]. For the cell to perform effectively, the Nafion membrane and the Nafion material in the catalyst layers (CLs) must be well hydrated to achieve good proton conductivity. Low hydration leads to poor proton conductivity and higher cell resistance, ultimately causing rapid decay in cell performance [7,8]. To maintain enough hydration, water is normally introduced into the cell externally by passing the hydrogen and air feeds through humidifiers prior to the gases entering the cell. However, these additional humidifying devices complicate the system and increase costs. To respond to this challenge, many researchers have developed high-performance membrane electrode assemblies (MEAs) capable of self-humidification. This is an attractive prospect, as these MEAs offer the possibility of simplifying the auxiliary systems of PEMFCs—specifically, eliminating the whole water management apparatus—which would decrease the overall cost.

Recent years have seen various efforts to prepare self-humidifying MEAs. One approach is to modify the membrane with a hydrophilic material to render the membrane self-humidifying [9–17]. Watanabe et al. [9–11] created a self-humidifying electrolyte membrane by adding Pt nanoparticles and a nanosized hygroscopic material such as TiO_2 or SiO_2 to the membrane. The Pt nanoparticles combined H_2 and O_2 into water, and the hygroscopic oxides absorbed the water, thereby maintaining the membrane's water content. Yang et al. [14] designed a thin double-layer composite membrane consisting of one layer of plain Nafion and another of Nafion–silica composite placed on the anode side; this thin layer transported protons from active sites in the CL to the membrane and retained water to wet the membrane and the CL. Han et al. [15] also reported a self-humidifying MEA achieved by adding a Nafion–silica suspension in the immediate vicinity of the catalyst sites.

The second method is to fabricate self-humidifying MEAs by adding hygroscopic particles into the CL or fabricating a self-humidifying composite catalyst [17–29]. Researchers who added SiO_2 [20,24] or Al_2O_3 [29] particles into the anode CL confirmed that these improved wettability and thereby enhanced MEA performance under low-humidity conditions. Jung et al. [24] prepared a self-humidifying MEA by directly dispersing hydrophilic SiO_2 particles into the anode CL. With 0% relative humidity (RH) at the anode and 80% RH at the cathode, their MEA containing 40 wt.% SiO_2 exhibited better performance than a blank MEA at a cell temperature of 70 °C: the current density at 0.6 V was up to 840 mA cm⁻². Su et al. [26] prepared a Pt– SiO_2/C self-humidifying catalyst via an organic colloid method. A MEA with this Pt– SiO_2/C as the anode catalyst showed good long-term performance under low-humidity conditions; at a cell temperature of 50 °C and with 28% RH at both anode and cathode, the current density at 0.6 V remained at 650 mA cm⁻², undegraded after 120 h. Su

et al. suggested that the silica on the carbon support served as a mini water reservoir, absorbing and retaining back-diffused water from the cathode and thereby improving the PEMFC's low-humidity performance. CLs containing SiO_2 nanoparticles were also prepared by Inoue et al. [21,27] to improve the low-humidity performance of a PEMFC. They found that using SiO_2 CLs increased the water content and enhanced the electrochemically active surface area of the Pt catalyst.

The third approach is to fabricate self-humidifying MEAs by adding hydrophilic materials at the MEA's interfaces [30–38]. Kitahara et al. [31] developed a new gas diffusion layer (GDL) coated with a hydrophilic and hydrophobic double microporous layer (MPL) to enhance the performance of a PEMFC under both low- and high-humidity conditions. The hydrophilic layer coated on the hydrophobic MPL was effective for preserving the MEA's hydration under low-humidity conditions, while the hydrophobic layer between the carbon paper substrate and the hydrophilic layer prevented the removal of water from the hydrophilic layer. Kannan et al. [33,38] prepared self-humidifying MEAs by adding inorganic oxides (such as SiO_2 , TiO_2 , Al_2O_3 , and polyaniline (PANI)) to the GDL. The MEAs with a thin hydrophilic layer showed high performance even at low humidity. Kannan and colleagues also investigated how placing the PANI in different places in the anode gas diffusion electrodes (GDEs) affected PEMFC performance [34]. These various studies have indicated that a GDL modified with a thin hydrophilic layer can yield better low-humidity performance in a PEMFC than a pristine GDL with only a hydrophobic MPL.

Although the last decade has seen great progress achieved in developing self-humidifying MEAs, the low working temperature of the cell still must be addressed. Thus, exploring new materials that have high hygroscopicity and high stability at high temperatures is a meaningful line of inquiry. After screening a large number of possibilities, we determined that microcrystalline cellulose (MCC) might be a perfect candidate, superior to general inorganic and organic materials.

Microcrystalline cellulose (MCC) is a purified natural polymer material, with excellent hygroscopicity, high chemical and temperature stability, and insolubility in water. It is widely used in the cosmetic, pharmaceutical, and food industries [39]. In this work, we attempted to use MCC in the preparation of self-humidifying MEAs by adding it to the CL or inserting a MCC layer at various interfaces in the MEA. Ultimately, we found that inserting a MCC layer between the anode CL and the GDL achieved the best self-humidification performance, even at a cell temperature of up to 70 °C and with 30% RH at both anode and cathode.

Experimental

GDL preparation

The GDL used for this study was prepared as follows. Carbon paper (TGP-H-060, Toray, Japan) was washed with acetone to remove possible surface contaminants and then impregnated with a PTFE emulsion (60%, Aldrich). This was followed by drying and calcining at 400 °C for 30 min to uniformly disperse the PTFE in the carbon paper; the loading of the dry PTFE was

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