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Significant performance improvement in terms of reduced cathode flooding in polymer electrolyte fuel cell using a stainless-steel microcoil gas flow field

Shiro Tanaka [a,b,*], Toshio Shudo [a]

[a] Department of Mechanical Engineering, Tokyo Metropolitan University, 1 Minamiosawa, Hachioji, Tokyo 192-0397, Japan
[b] HySA/Catalysis, Department of Chemical Engineering, University of Cape Town, Private Bag X3, Rondebosch, Cape Town 7701, South Africa

HIGHLIGHTS

- We achieve flood-free performance using aligned microcoils as a flow field.
- The GDL has no observable effect on flooding if air is uniformly distributed.
- The HFR is high in GDL-less design with less contact point of the microcoils.
- The membrane is dehydrated in high current density with big temperature raise.

ARTICLE INFO

ABSTRACT

Flooding at the cathode is the greatest barrier to increasing the power density of polymer electrolyte fuel cells (PEFCs) and using them at high current densities. Previous studies have shown that flooding is caused by water accumulation in the gas diffusion layer, but only a few researchers have succeeded in overcoming this issue. In the present study, microcoils are used as the gas flow channel as well as the gas diffuser directly on the microporous layer (MPL), without using a conventional carbon-fiber gas diffusion layer (GDL), to enable flood-free performance. The current–voltage curves show flooding-free performance even under low air stoichiometry. However, the high-frequency resistance (HFR) in this case is slightly higher than that in grooved flow channels and GDLs. This is due to the differences in the electron conduction path, and the in-plane electron conductivity in the MPL is the key to enhancing the microcoil fuel cell performance.

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1. Introduction

Polymer electrolyte fuel cells (PEFCs), which convert the chemical energy of hydrogen directly into electrical energy, are regarded as a promising alternative clean power source for automotive, stationary, and portable applications [1–5]. The acceptance of fuel cells for automotive use requires reduction in their cost and size, as current automotive fuel cells are not yet profitable and are still too large, putting limitations on the layout of the fuel cell system [6].

Currently, most automotive fuel cells are operated at a maximum current density of 1.0 A cm^{-2} [7–9]. This low current density requires a large active area in order to produce a vehicle power output of 70–120 kW, which necessarily increases the number of polymer electrolyte membranes (e.g., Nafion, Acipex, and Dow membrane), catalyst layers with precious metals, and gas diffusion layers (GDLs). The expense of these materials means that enlarging the active area significantly increases the cost of fuel cell stack. In addition, the thickness of the GDL at 90–200 μm for each electrode increases the length in the stacking direction, as the total volume of the GDL occupies about half of the fuel cell stack. These considerations create limitations for conventional fuel cell structures with regard to cost and size.

Solving these commercialization issues for automotive fuel cells requires a novel fuel cell structure that enables high current density

* Corresponding author. HySA/Catalysis, Department of Chemical Engineering, University of Cape Town, Private Bag X3, Rondebosch, Cape Town 7701, South Africa. Tel.: +27 21 650 3852; fax: +27 21 650 5509.
E-mail address: shiro.tanaka@uct.ac.za (S. Tanaka).

operation without a GDL. Increasing the current density of conventional fuel cells causes them to undergo a sudden decrease in performance, known as cathode flooding. When current density increases, more water is generated in the electrochemical reaction $(O_2 + 4H^+ + 4e^- \rightarrow 2H_2O)$ at the cathode, and more water is transported from the anode to the cathode through the membrane via electro-osmotic drag [10–12]. The water at the cathode condenses and accumulates, blocking oxygen diffusion and causing a sudden decrease in the cell voltage.

Simultaneously, increasing the current density causes the membrane to dry out at the anode. The membrane is dehydrated by electro-osmotic drag from the anode to the cathode, decreasing the water concentration [13]. The increased temperature arising from the electrochemical reaction and the ohmic heat also dehydrate the membrane. The proton conductivity of the membrane heavily depends on its water content, meaning that dehydration of the membrane decreases proton conductivity and thus cell performance.

There are a few reports on efforts to overcome membrane flooding/dry-out. Shudo et al. replaced the conventional land/channel flow field structure with a porous stainless steel in direct methanol fuel cell (DMFC) operations [14]. Using electrochemical impedance spectroscopy (EIS), they found that their fuel cell with a porous flow field allowed for uniform distribution of the reactant gas onto the catalyst layer, thus enabling higher performance than that of conventional cells even at a high current density of 0.5 A cm^{-2}. Strickland et al. developed a unique flow field with in-situ polymerized wicks coated onto the cathode channel in a hydrogen fuel cell [15]. The performance of their cell design was flood-free even at 1.5 A cm^{-2}. They concluded that the cathode channel wick provided a hydraulic pathway from the reaction sites to the outside of the fuel cell that removed the product water and diffused oxygen to the catalyst layers. Turhan et al. explored the effect of changing the width ratio of the land and channel (L/C) on water accumulation using an in-situ neutron imaging technique [16]. They concluded that liquid water tended to accumulate under the lands rather than under the channels, and that accumulation depended strongly on the flow-field geometry. As L/C decreased, less liquid water accumulated in the cell, permitting higher cell performance with less flooding.

While there are a few published studies on flow-field alterations for extending current densities, there is little in the literature suggesting novel structures or materials to overcome flooding and dry-out with smaller cells and lower pressure drop designs. In this report, we describe a unique fuel cell design that utilizes stainless steel microcoils to generate the flow field and gas diffusion layer. We compared the fuel cell performance of microcoils with and without a GDL and conventional flow fields with a GDL under various conditions, and discussed how the microcoils in the fuel cell reduced flooding and dry-out. In addition, these new structures and materials were expected to perform differently compared with conventional structures and materials, and so we subsequently characterized the electrical and thermal properties of several elements of the fuel cells.

2. Experimental

2.1. Fuel cell preparation

The bipolar plates for our fuel cell had 5 cm^2 active areas (1 cm × 5 cm) made of graphite (Mechanical Carbon Industry, Kanagawa, Japan). The end-plates for these bipolar plates were made with stainless steel, with eight holes for fastening the fuel cell with a compression pressure of 1 MPa. The end-plates had sheet heaters on the outside of both the anode and cathode to control the cell temperature. A stainless steel current-collector with 1-μm-thick gold-plate was inserted between the bipolar plate and the end-plate on both sides. 0.6 mm-thick Ethylene−propylene diene monomer rubber (EPDM) gaskets (NOK Corp, Tokyo, Japan) were used to surround the active area and seal the reactant gases. Bolt torques of 0.3 N m were used to achieve the compression pressure of 1.0 MPa for the fuel cell assembly.

25-μm-thick Nafion (NRE211, du Pont de Nemours, Delaware, USA) was chosen as a polymer electrolyte membrane, and carbon-supported platinum (Pt/C, 50 wt% Pt, TEC10E50E, Tanaka K.K., Tokyo, Japan) was selected as a catalyst. The catalyst-coated membrane (CCM) was prepared as follows: Pt/C powder was mixed with a Nafion dispersion (DE2020 du Pont de Nemours, Delaware, USA) using a zirconia bead mill for 8 h on a rotation table. The diameter of the beads was 1 mm, and the speed of rotation of the table was 400 rpm. The solid contents of the catalyst were prepared using a 1:1 mixture of carbon support:Nafion-ionomer. The catalyst was coated on both sides of the membrane with a spray coater (Mic Lab, Kanagawa, Japan) several times until the coating thickness was about 30 μm. The platinum loadings were 0.4 mg cm^{-2} for both the anode and the cathode. The coated membrane was dried at 80 °C for 60 min and annealed at 130 °C for 10 min to obtain the CCM.

Microporous layers (MPLs) were used in the anode and cathode catalyst layers to control the water management of the fuel cell [17]. MPLs were prepared as follows: powdered carbon black (CB; Denka Black, Denki Kagaku Kogyo K.K., Tokyo, Japan), polytetrafluoroethylene (PTFE) aqueous dispersions (D-1, Daikin Industries Ltd., Osaka, Japan), nonionic surfactant (Triton X-100, Sigma−Aldrich), and DI water were mixed with 1 mm zirconia beads in a bead mill for 3 h at 400 rpm using 5:1:20:20 CB:PTFE:surfactant:water. This coating was cast onto an expanded PTFE porous sheet (30 μm thick, Poreflon Membrane, Sumitomo Electric Industries Ltd., Osaka, Japan) and dried at 350 °C for 60 min to obtain MPLs 40 μm and 60 μm thick.

The CCM was hot-pressed with MPLs on both sides for the anode and cathode at 130 °C and 3 MPa g for 10 min, using 200 μm-thick Teflon backing sheets on both sides to avoid MPL attachment to the plates of the hand-press.

The cathode was chosen for these experiments because the cathode reaction has been found to be the limiting step in PEFCs, in contrast to the fast kinetics of the anode reaction as demonstrated by Springer et al. in their EIS study [18].

We evaluated four designs of cathode flow fields in this study. The first design was a conventional flow field, with a cathode land 50 mm long and 1 mm wide, and cathode channels 50 mm long, 1 mm wide, and 1 mm deep. The GDL was laid between the flow field and MPL at the cathode (Design 1; conventional flow field with GDL). The second design utilized a microcoil with a 0.6 mm outer diameter and a GDL (280 μm thick, 5 wt% PTFE, TGP-H-090, Toray, Tokyo, Japan) between the microcoil and the MPL on the cathode side (Design 2; 0.6 mm microcoil with GDL). The third and fourth designs both featured microcoil flow fields tightly arranged parallel to the cathode air flow on pool-shaped graphite bipolar plates. Both microcoils were purchased from Micro Spring K.K. (Nagano, Japan) and were made of 50-μm stainless steel fiber with 0.1-μm-thick gold plating. The pitch of the coil was 90 μm, and the width of the opening was 40 μm; this configuration prevented the fibers from getting entangled with one other and facilitated the tight arrangement of the microcoils. The outer diameter of the microcoil for the third design was 0.6 mm (Design 3; 0.6 mm microcoil), and that for the fourth design was 0.2 mm (Design 4; 0.2 mm microcoil). The microcoils for both designs were directly laid on the cathode MPLs without a GDL.