



Experimental study on the membrane electrode assembly of a proton exchange membrane fuel cell: effects of microporous layer, membrane thickness and gas diffusion layer hydrophobic treatment



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ABSTRACT

In this study, electrochemical impedance spectroscopy (EIS) is employed to analyze the influence of microporous layer (MPL), membrane thickness and gas diffusion layer (GDL) hydrophobic treatment in the performance of a proton exchange membrane (PEM) fuel cell. Results show that adding a MPL increases cell performance at low to medium current densities. Because lower ohmic losses are observed when applying a MPL, such improvement is attributed to a better hydration state of the membrane. The MPL creates a pressure barrier for water produced at the cathode, forcing it to travel to the anode side, therefore increasing the water content in the membrane. However, at high currents, this same phenomenon seems to have intensified liquid water flooding in the anode gas channels, increasing mass transfer losses and reducing the cell performance. Decreasing membrane thickness results into considerably higher performances, due to a decrease in ohmic resistance. Moreover, at low air humidity operation, a rapid recovery from dehydration is observed when a thinner membrane is employed. The GDL hydrophobic treatment significantly improves the cell performance. Untreated GDLs appear to act as water-traps that not only hamper reactants transport to the reactive sites but also impede the proper humidification of the cell. From the different designs tested, the highest maximum power density is obtained from that containing a MPL, a thinner membrane and treated GDLs.

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1. Introduction

A proton exchange membrane (PEM) fuel cell is an electrochemical device that combines hydrogen with oxygen to produce electrical energy. Owing to their high efficiency, quick/cold startup, easy scale-up and zero emissions, PEM fuel cells appear as a viable power generation technology for the future.

In the heart of a PEM fuel cell lies the membrane electrode assembly (MEA): a 5-layer structure with a proton exchange membrane in the center, two catalyst layers (CLs) for the anode and cathode and two gas diffusion layers (GDLs). The membrane acts as electrolyte, allowing protons to move from the anode to cathode while forcing electrons to travel through an external circuit. Nafion is to the present date the best known electrolyte for PEM fuel cells. Electrochemical reactions take place in the CLs, typically catalyzed by platinum supported on carbon particles. GDLs, generally

consisting of carbon fiber-based porous materials (papers, cloths and nonwovens), have several important functions including: to allow for an uniform distribution of reactants gases to the CLs; to provide pathways for product water and heat to be removed from the CLs to the gas channels of the bipolar plates; to electrically connect the CLs to the bipolar plates; and to provide mechanical support for the MEA [1].

In order to maintain proper proton conductivity of Nafion and its mechanical integrity, humidified gases are commonly used. However, excess water can lead to electrodes flooding, hindering the reactants access to the CLs active sites. To facilitate water removal, GDLs are usually subjected to hydrophobic treatment, using agents such polytetrafluoroethylene (PTFE or Teflon) [2–5] or fluorinated ethylene propylene (FEP) [6]. Moreover, a microporous layer (MPL) has been placed between the CL and GDL. As the name suggests, the MPL has smaller average pore size (1–10 μm) than that of the GDL (10–100 μm), and usually consists of carbon black Teflon as a hydrophobic binder and pore-forming agent [7]. Generally, the addition of a MPL to the GDL has been shown to improve water management and the performance of a PEM fuel

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cell. Although the role of the MPL is still not completely understood [8,9], it has been shown that the MPL seems to create a capillary barrier between the CL and the GDL substrate, causing water to move from the cathode to the anode side, improving the hydration state of the membrane [2,10,11]. At the same time, less water remains in the cathode porous media and liquid water saturation is reduced. The MPL is also referred to suppress water accumulation in the interfacial gap between the CL and the GDL by allowing the passage of water in vapor form, further improving the cell tolerance to flooding [9]. Moreover, cracks in the MPL are found to limit the number of water entries into the GDL, stabilizing the water paths and therefore decreasing water saturation [12]. In addition, MPL is referred to increase electrical conductivity and to prevent GDL fiber intrusion into the CL [7].

As seen above, in the MEA components many processes take place, very often with conflicting requirements, so their properties and design must be carefully selected. Electrochemical impedance spectroscopy (EIS) is a non-invasive diagnostic tool capable of measuring the fuel cell impedance over a wide range of frequencies. This powerful technique can determine various sources of polarization loss in a short time, being very useful to the optimization of the MEA design.

In this work, EIS is employed to evaluate the performance of four commercially available MEAs. The effects of MPL, membrane thickness and GDL hydrophobic treatment are investigated at different air stoichiometries and humidities.

2. Experimental

2.1. Setup

A scheme of the PEM fuel cell test facility employed in the present study is shown in Fig. 1. Hydrogen and air flow rates are regulated by rotameters (Omega FLDH3301C for hydrogen and Omega FLDA3213ST for air). Both gases are humidified by bubbling them through heated water. The relative humidity (RH) of each gas is read by humidity probes (Vaisala HMT337). Stainless steel tubing and glass vessels from water humidification to the fuel cell inlet, as well as the fuel cell, are surrounded by flexible heaters (50 W m^{-1}) and covered by glass wool for temperature control. At the vessels

for RH reading and at the fuel cell, temperature is adjusted using readings from the corresponding type-K thermocouples and switching on/off the flexible heaters, both connected to a data acquisition system controlled by the LabVIEW software (National Instruments). Temperature at other points is controlled by stand-alone temperature controllers (Osaka Ok31). Electrochemical measurements are made using a Zahner Zennium electrochemical workstation coupled with an EL300 electronic load.

2.2. Fuel cell hardware and MEAs

A cell with an active area of 25 cm^2 ($5 \text{ cm} \times 5 \text{ cm}$) is used. End-plates are made of 10.0 mm thick stainless steel plates. Gold coated copper plates with thickness of 0.5 mm are employed as current collectors. Single-serpentine flow fields are machined into 3.5 mm thick graphite plates, having cross-section height and width of 1.0 mm and 1.4 mm, respectively. Brand new commercially available MEAs from QuinTech e.K. are used, each one containing 0.3 and $0.6 \text{ mg Pt cm}^{-2}$ in the anode and cathode, respectively. All MEAs are utilized as received. Table 1 displays the features of the MEAs tested in the present work, related with the membrane and GDLs employed. MEA 1 is used as the base case design. It can be seen that MEA 1 is equal to MEA 2 except the former has a MPL. Therefore, the effect of the MPL is analyzed by comparing MEA 1 and MEA 2 performances. The GDL thickness of MEA 2 is slightly lower than that of the others MEAs because it does not contain MPL. The effect of the membrane thickness is studied by comparing the results obtained with MEA 1 and MEA 3, which only differ in the Nafion membrane thickness employed. Results obtained with MEA 4 are compared with those of MEA 1 to investigate the effect of the GDLs hydrophobic treatment. Apart from a slight deviation in the GDLs thickness, the only difference between these MEAs is the PTFE treatment applied to the GDLs of MEA 1.

2.3. I-V curves and EIS measurements

Tests are conducted at 40°C cell initial temperature (measured placing a thermocouple at the cathode outlet in contact with the MEA surface) and atmospheric pressure. H_2 stoichiometry (λ_{H_2}) of

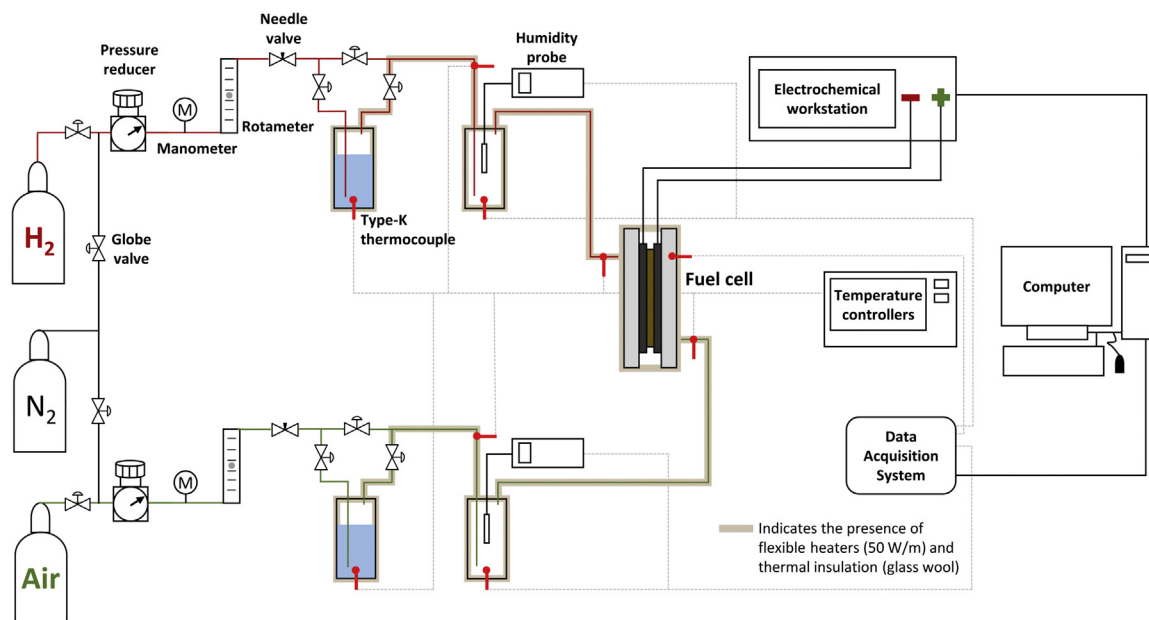


Fig. 1. Scheme of the PEM fuel cell test facility employed in the present study.

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