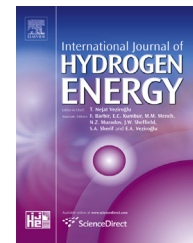


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Investigation of the effect of microporous layers on water management in a proton exchange membrane fuel cell using novel diagnostic methods

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ARTICLE INFO

Article history:

Received 2 June 2014

Received in revised form

25 July 2014

Accepted 27 July 2014

Available online 23 August 2014

Keywords:

Water management

Proton exchange membrane fuel cell

Microporous layer

Gas diffusion layer

Mass transport losses

Fuel cell diagnostics

ABSTRACT

Water management remains a significant challenge for the Proton Exchange Membrane Fuel Cell (PEMFC) with respect to performance, lifetime and operational flexibility. In recent years, microporous layers (MPL) have been widely used on the cathode side of the PEMFC in order to improve fuel cell performance and water management capabilities. Many modeling and experimental studies have with limited success attempted to analyze the underlying mechanisms that are responsible for the performance improvement due to the MPL. In this study, porous inserts along with various in-situ experimental techniques are used to investigate the MPLs. It was observed that the anode pressure drop increased when a cathode MPL was present, indicating water cross-over from the cathode towards the anode side. Further testing identified that the MPL improved cell performance due to the reduction of water saturation in the cathode catalyst layer, which resulted in enhanced oxygen diffusion. The influence of the MPL on the anode side was also studied with the aid of porous inserts and other techniques, and it was observed that the anode MPL improves cell voltage stability and reduces water accumulation in the anode catalyst layer. The present investigation provides further important information on the critical role of the MPL in the PEMFC.

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Introduction

Proton exchange membrane fuel cells (PEMFC) are generally considered to be the fuel cell technology closest to wide scale commercialization and are being considered for a number of

applications including automotive, back-up power, portable devices, small power generation, and micro applications [1,2]. However, these fuel cells face unique difficulties with each application, such as cost, efficiency, durability, degradation, harsh operating conditions, and water management. Thus, a lot of emphasis has been given to study, understand, and

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<http://dx.doi.org/10.1016/j.ijhydene.2014.07.147>

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Nomenclature

CCM	catalyst coated membrane
CFP	carbon fiber paper
CL	catalyst layer
FF	flow field
GDL	gas diffusion layer
HFR	high frequency resistance
MEA	membrane electrode assembly
MPL	micro-porous layer
OA	open area
PEMFC	proton exchange membrane fuel cell
PS	perforated sheet
C_{H_2O}	water concentration, mol m^{-3}
D_{H_2O}	water diffusion coefficient in the membrane, $\text{m}^2 \text{s}^{-1}$
k_p	membrane hydraulic permeability, m^2
$N_{H_2O,BD}$	molar flux of water vapor due to back diffusion, $\text{mol s}^{-1} \text{m}^{-2}$
$N_{H_2O,hyd}$	molar flux of water vapor due to hydraulic permeation, $\text{mol s}^{-1} \text{m}^{-2}$
P_c	pressure cathode, kPa abs
P_a	pressure anode, kPa abs
T_{dp}	dew point temperature, °C
T_{cell}	cell temperature, °C
T_{gas}	gas temperature, °C
λ	membrane hydration
μ	fluid viscosity, $\text{kg m}^{-1} \text{s}^{-1}$
τ	membrane thickness, m

alleviate some of these issues. Water management in PEM fuel cells is a critical parameter that determines the performance and durability of a fuel cell. At humidified conditions the water will accumulate over time in the catalyst layers, porous gas diffusion layers (GDL), and flow field channels. This accumulation of water causes flooding, which in turn, decreases the performance of the system and can result in critical failure modes [1]. In addition, water management also requires maintaining the right water balance inside the cell in order to keep the membrane humidified at low to no gas humidification conditions. In order to improve water management and overall fuel cell performance, a microporous layer (MPL) can be located between the GDL and catalyst layer (CL). The MPL also reduces the contact resistance between the GDLs and the catalyst layer, gives mechanical support to the catalyst layer, and prevents catalyst particle loss and clogging of the GDL pores with catalyst particles [3–8].

The MPL, which is usually made of carbon black and a hydrophobic agent (e.g., PTFE), is normally deposited on top of one of the GDL surfaces (forming a diffusion double-layer). This layer is dense and with small pores. In fact, the MPL forms smaller pores than the GDL (20–200 nm pores for MPLs [9] and 0.05–100 μm pores for typical carbon fiber paper (CFP) GDLs [10,11]). The MPL can also be made out of a carbon based polymer porous sheet that is simply placed between the CL and GDL when assembling the fuel cell [9,12–14].

Microporous layers are now commonly used in order to improve the overall performance of a fuel cell at both fully

humidified and low humidity conditions [15,16]. It is believed that they play a key role in the overall water management within the fuel cell. However, it is still unclear exactly how the MPL affects the water transport mechanism inside the GDL and the MEA and there are many contradictory studies and observations. The following is a brief summary of some of the studies that investigated MPLs and their effect on PEM fuel cells.

Passalacqua et al. [17] were able to show that with an MPL the performance of the cell improves substantially. They concluded that the MPL reduced the size of the water droplets, thus enhancing the oxygen diffusion. This layer also prevented the catalyst particles from penetrating into the GDL. Park et al. [18] also concluded that with the addition of an MPL both the water management and electrical conductivity improved. Similar observations were also presented by Song et al. [19] and Holmstrom et al. [20], especially when investigating the fuel cell's performance at high current densities. GDLs without an MPL on the cathode side lead to major mass transport losses at high humidity conditions due to water flooding. The MPL has also been shown to effectively improve the start-up performance of a PEMFC by suppressing water accumulation at the electrode, which is important especially when dealing with sub-zero conditions [21–23].

After testing a number of GDLs with and without MPLs, Lin et al. [15] postulated that the MPL appears to push more liquid water back to the anode through the membrane. Basically, the small hydrophobic pores in the MPL result in low liquid water permeability and reduce the water transport from the catalyst layer towards the GDL. Therefore, more water accumulated in the CL is forced towards the anode (back-diffusion). This reduces the amount of water that is removed through the cathode GDL, decreases the number of blocked pores within the cathode GDL, and improves the overall gas transport in the GDL towards the active zones.

One contradictory point regarding how the MPL works is related to the water saturation in the catalyst layer of the cathode side. Nam et al. [24] stated that by using a microporous layer near the CL, the water condensed in the GDL cannot enter the CL, thus reducing the overall saturation of the active catalyst zones. This idea was also presented by Pasaogullari et al. [25] in which they concluded that in the presence of an MPL the liquid saturation in the catalyst layer is reduced substantially. Kitahara et al. [26] demonstrated that the MPL reduced water content in the catalyst layer and increased water accumulation inside the GDL area closest to the flow field channels. Recently, the study presented by Deevanhxay et al. [27], in which soft X-ray radiography was used to observe water transport inside an MEA, confirmed that the MPL reduced the water accumulation in the CL/MPL interface. These concepts contradict those presented earlier since it is not clear whether the liquid saturation does in fact increase in the cathode catalyst layer or not. This may depend directly on the rate at which the water goes back (or is forced) to the anode.

Through neutron radiography imaging, Owejan et al. [28] were able to observe that MEAs that had cathode GDLs with MPLs had better distribution of water over the active area at high current densities. GDLs without MPLs tended to have more water accumulated in one location of the active area (closer to the outlet). One issue with this work was that the

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