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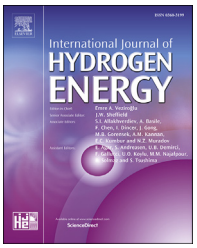

Simultaneous characterization of oxygen transport resistance and spatially resolved liquid water saturation at high-current density of polymer electrolyte membrane fuel cells with varied cathode relative humidity

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ABSTRACT

In this study, the impacts of relative humidity on liquid water accumulation and mass transport resistance at high current densities were investigated for polymer electrolyte membrane (PEM) fuel cell cathode gas diffusion layers (GDLs). Through-plane liquid water saturation distributions were measured *in situ* using synchrotron X-ray radiography while simultaneously performing limiting current-based characterizations of oxygen transport resistance. In experiments with inlet cathode reactant relative humidity ranging from 0% to 100%, high local saturations (>0.6) were consistently observed in the GDL region adjacent to the flow field lands when the fuel cell reached limiting current. High land-region saturation is a possible indication of lower local temperatures. In the carbon fiber substrate portion of the GDL (i.e. excluding the MPL), the liquid water volume in the regions adjacent to the flow field channels was consistently small relative to the water volume adjacent to the land regions. We observed a trend of increasing liquid water saturation of the GDL adjacent to the channels with increasing levels of cathode inlet relative humidity, along with corresponding small increases in total oxygen transport resistance.

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Nomenclature

A	Fuel cell active area [cm^2]
c	Molar concentration [$mol \cdot m^{-3}$]
D_{eff}	Effective diffusion coefficient [$m^2 \cdot s^{-1}$]
E	Measured fuel cell voltage [V]
F	Faraday's constant, 96485 [$C \cdot mol^{-1}$]
i	Current density of the fuel cell [$A \cdot cm^{-2}$]
i_L	Limiting current density [$A \cdot cm^{-2}$]
I_{dry}	Pixel intensity in the dry reference image
I_{wet}	Pixel intensity during fuel cell operation
L_z	Length of the active area and gas diffusion layers in the direction parallel to the X-ray beam path [cm]
$\dot{N}$	Molar consumption rate of a reactant [$mol \cdot s^{-1}$]
p	Absolute pressure [kPa]
R_T	Transport resistance [$s \cdot cm^{-1}$]
R	Gas constant, 8.314 [$J \cdot K^{-1} \cdot mol^{-1}$]
T	Temperature [°C]
$x_{O_2}^{dry-in}$	Oxygen mole fraction before humidification
t_w	Measured water thickness [cm]
Greek	
μ	Attenuation coefficient [cm^{-1}]

Introduction

An overall cost reduction is necessary to make polymer electrolyte membrane fuel cells (PEMFCs) competitive with conventional internal combustion engines in the transportation market. As a means of reducing cost, stack sizes can be reduced for a given power requirement if fuel cell performance can be improved at high current densities [1]. Management of mass transport is critical for maintaining high performance during operation at high current densities, at which large fluxes of reactants and products must be accommodated through the porous gas diffusion layer (GDL) [2]. Additionally, the accumulation of liquid water within the pore space of the GDL restricts transport pathways for reactant gases.

State-of-the-art GDLs employed in PEMFCs have a bi-layered structure composed of a carbon fiber substrate and a microporous layer (MPL) [3,4]. Oxygen gas must pass through the GDL from the gas channels to the reaction sites in the catalyst layer (CL). Concurrently, water produced by the oxygen reduction reaction at the cathode CL must flow in the opposite direction for removal through the gas channels. If liquid water accumulates in the GDL, portions of the void pore space will be occupied, thereby reducing the effective diffusivity of oxygen within the GDL. With this knowledge, a concerted research effort should be made to measure the liquid water saturation distribution in a GDL in operando, to study the parameters that affect this distribution, and to determine the performance implications.

The liquid water saturation distribution in the GDL is a function of water and heat transport behaviour in the PEMFC [5–12]. Water transport in the GDL is typically characterized via two mechanisms, liquid percolation and vapour diffusion. Liquid percolation is a capillary-force dominated process [13–16], wherein liquid water advances into a pore space via a connecting throat when the difference between the invading liquid pressure and the defending gas phase pressure exceeds the threshold capillary pressure of the throat. The GDL saturation gradually increases until a breakthrough event occurs, defined as the moment when a connected water pathway through the thickness of the GDL is formed [17]. Heat is also produced at the catalyst coated membrane (CCM) from the electrochemical reaction and due to the ohmic resistance of the ionomer [18]. The result is a temperature gradient between the CCM and the gas flow field. Since the saturation water vapour pressure is temperature-dependent, this in turn creates a gradient in water vapour partial pressure, which drives vapour-phase diffusion of water [19–21].

In addition to the percolation of liquid water, liquid water may also accumulate in the GDL due to condensation at locations where the local temperature is below the dew point of the gas phase [21,22]. Therefore, the distribution of liquid water in the GDL is sensitive to the gradient of water vapour partial pressure between the CL-GDL interface and the flow-field channels. We can impose some control over this gradient by changing the relative humidity (RH) of the inlet gases. Pore-network modeling studies by Hinebaugh et al. [23], Straubhaar et al. [24], and Gostick et al. [13] have demonstrated that condensation rates can have a significant impact on GDL water saturation. Boillat et al. [25] used neutron radiography to observe water accumulation in a PEMFC and reported that liquid water accumulation increased when the relative humidity of reactant gases increased. Chevalier et al. [26] measured water accumulation using synchrotron X-ray radiography and reported the significant sensitivity of liquid water saturation to relative humidity with an increase in peak local saturation from <5% to 86% when the cathode relative humidity increased from 25% to 100% at an operating current density of 1.5 A/cm^2. In addition to the direct studies of GDL water mentioned above, numerous works have investigated PEM fuel cell liquid water accumulation in the channels via numerical modeling, direct visualization, or measurement of pressure drop [27–39]. In studies by Banerjee et al. [28], Hussaini and Wang [29] and Spernjak et al. [32], increased relative humidity of the inlet reactant gases was observed to increase the presence of liquid water in the channels.

Due to the blockage of transport pathways, liquid water saturation in the cathode GDL has a strong impact on oxygen transport resistance [13,26,40–44]. The limiting current technique is an established methodology for directly measuring oxygen transport resistance and has been used in recent published works to assess the parameters that affect water and oxygen transport [40,45–48]. For example, Baker et al. [45] determined the relative contributions of the channels, GDL substrate, MPL, and catalyst layer to total transport resistance by manipulating GDL and MPL thicknesses and the operating pressure. Simon et al. [48] observed that the transport resistance at current densities >2.0 A/cm^2 decreased when cathode GDL compression was increased in the range from 0 to 20%, postulating that poor thermal contact at low compression resulted in low local temperatures in the GDL, encouraging condensation. Caulk and Baker [40] studied the impacts of fuel cell operating pressure and temperature on the oxygen

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