



Non-isothermal two-phase transport in a polymer electrolyte membrane fuel cell with crack-free microporous layers



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ABSTRACT

Liquid water breakthrough events were observed in crack-free microporous layers (MPLs) of operating polymer electrolyte membrane (PEM) fuel cells through in situ synchrotron X-ray radiography. The measured water thicknesses were used as inputs into a one-dimensional (1D) heat and mass transport model. This 1D model was developed to describe the coupled relationship between liquid and vapour transport through the cathode GDL and the temperature distributions in the fuel cell that accompany the in operando measurements. The current density was increased from 1.4 to 2.4 A cm⁻², during which MPL breakthrough was observed. Immediately following MPL breakthrough events, the fraction of vapour-phase transport of the total water flux in the MPL increased by up to 5%. Post MPL breakthrough, further increases in current density resulted in increased thermal and water vapour concentration gradients in the MPL, and vapour transport was further enhanced. A temperature gradient at the cathode catalyst layer (CL)–MPL interface resulted in decreased threshold capillary pressures, and as a result higher liquid water saturations were observed near this interface. These findings suggest that the temperature gradient should be considered in two-phase flow modeling and pore network modeling due to its impact on liquid water distributions.

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1. Introduction

Reducing the cost of the polymer electrolyte membrane (PEM) fuel cell is vital for enabling its widespread commercialization [1]. Effective catalyst utilization through high current operation is an attractive strategy for overcoming this cost challenge in transportation applications [2]. However, high current density operation leads to a greater rate of water generation, which tends to result in the accumulation of liquid water in the cathode gas diffusion layer (GDL) and the hindrance of reactant oxygen gas transport. As the saturations in the catalyst layer and GDL reach prohibitive levels, the cathode and cathode GDL become flooded [3], and the performance of the fuel cell becomes dominated by mass transfer polarization. Hence, fuel cell components must be designed for

effective water management, particularly, at high current density operation.

It has been observed that the implementation of a microporous layer (MPL), which typically consists of carbon black particles and polytetrafluoroethylene (PTFE), placed between the carbon fiber substrate and the catalyst-coated membrane (CCM) (as shown in Fig. 1) mitigates cathode flooding and leads to the improvement of the overall performance of PEM fuel cells at high current densities [4–11]. Weber and Newman [8] suggested that the MPL mitigates the liquid water content at the cathode catalyst layer (CL) by forcing water across the membrane to the anode. Through electrochemical impedance spectroscopy (EIS) and synchrotron X-ray radiography, Antonacci et al. [12] observed that thicker MPLs led to decreased oxygen transport resistance due to the decreased water content in the cathode GDL. Thomas et al. [13] measured the water flux in the cathode and anode at various temperature settings. They concluded that the presence of the MPL led to the reduction of liquid water accumulation in the cathode CL. Multiple studies [6,14,15] have shown that the MPL constrains the number

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Nomenclature

c	vapour concentration	λ	water uptake
D^{eff}	effective diffusion coefficient	ϕ	heat flux
D_{vap}	diffusivity for water vapour	κ^{eff}	effective ionic conductivity
E	measured fuel cell voltage	ε_{cl}	volume fraction of ionomer in the catalyst layer
E°	theoretical open circuit voltage	μ_w	X-ray attenuation coefficient for liquid water
F	Faraday constant		
i	current density of the fuel cell	Superscript	
I	pixel intensity in collected X-ray radiograph	a	anode GDL
k	thermal conductivity of the dry fuel cell material	c	cathode GDL
k^{eff}	effective thermal conductivity of the fuel cell material with the presence of liquid water		
l	length of the active area	Subscript	
L	through-plane length	ch	flow field
L_f	specific latent heat for water condensation	cl	catalyst layer
N	molar flux of water	$diff$	diffusive flux
n_d	electro-osmotic drag coefficient	dry	dry-state image
P_c	capillary pressure	gdl	gas diffusion layer
P	pressure	j	GDL material: MPL or substrate
r_{pc}	volumetric phase change rate	m	membrane
R	gas constant	mpl	the bulk microporous layer
s	water saturation	pc	phase change
\bar{s}	mean water saturation	$pixel$	value at a certain pixel
SV	number of the solid voxels at a through-plane position	$PTFE$	polytetrafluoroethylene
T	temperature	ref	reference
VF	voxel fraction	sat	saturated water vapour
VV	number of the void voxels at a through-plane position	sub	the bulk carbon fiber substrate
x	through-plane position in the cathode GDL	tot	total
X	material thickness	tra	transition GDL region between the bulk MPL and substrate regions
		V	void voxel
Greek		w	water
ε	porosity	wet	wet-state image

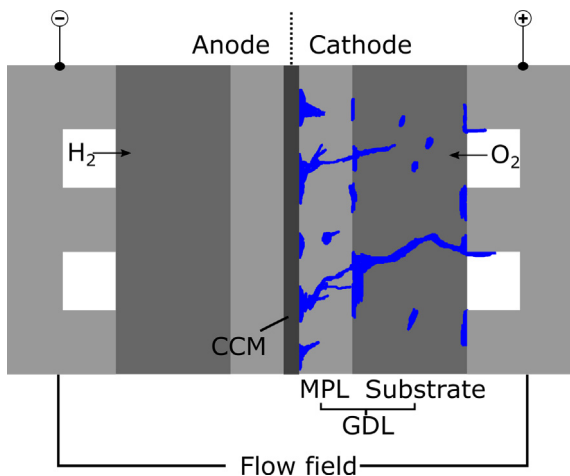


Fig. 1. Schematic of a PEM fuel cell with an example liquid water distribution.

of liquid water injection points to the substrate, which leads to the reduction of substrate water saturation.

The pursuit of a comprehensive understanding of the water transport mechanisms in the MPL can be traced the literature, which includes extensive studies focused on the dominance of the MPL in liquid-phase and vapour-phase water transport. In one proposed mechanism, liquid water travels across the MPL in continuous pathways as the liquid-phase pressure increases [4,16,17]. In this mechanism, high levels of liquid water saturation occur near the CL–MPL interface [4,18], as shown in Fig. 2(a). When

the resulting liquid-phase pressure overcomes the threshold capillary pressure of an adjacent pore, liquid water penetrates and saturates the pore. This process is termed *invasion percolation*. At the moment of *breakthrough*, liquid water reaches the opposite side of the MPL to form a liquid pathway through the MPL. After the breakthrough event, all subsequent liquid water introduced at the CL travels via the established pathway, whereby the saturation level in the MPL is no longer influenced by the production of liquid water [19,20]. This saturation trend in Fig. 2(a) was experimentally confirmed through the synchrotron visualization results reported by Lee et al. [4].

In general, liquid water preferentially occupies larger and more hydrophilic pores [21]; hence, MPL cracks provide primary pathways for liquid transport. Using visualization tools, several research groups [6,22,23] observed that MPL cracks facilitate liquid water transport at high current densities. However, cracks lead to poor mechanical durability of the MPL, so crack-free MPLs have been suggested [24]. Furthermore, the temperature has an impact on the capillary pressure [25–28]; however, there is a lack of information about how the temperature distribution within the GDL affects the nature of liquid transport in the MPL.

In the other proposed mechanism, vapour-phase diffusion across the MPL is driven by the water vapour concentration gradient (Fig. 2(b)). This concentration gradient is caused by the temperature gradient from the cathode CL to the flow field [29,30], due to the heat generated from the electrochemical reaction at the CL, Joule heating, and water phase change (condensation). Several studies [31,32] have reported that the addition of the MPL to the GDL leads to a decrease in thermal conductivity, increased temperatures at the CCM, and enhanced vapour transport. Yet,

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