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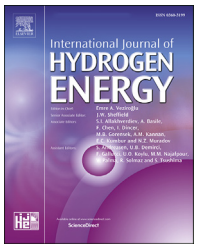

Transient, spatially resolved desaturation of gas diffusion layers measured via synchrotron visualization

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ABSTRACT

The transient 3-D visualization of the desaturation process of flooded gas diffusion layers (GDLs) is presented for the first time. Desaturation rates and pathways are reported for two commercial GDL samples, one with a PTFE treatment and the other without. On-the-fly Synchrotron X-ray CT is performed while the GDL sample is subjected to a humidified or dry air flow stream to visualize the transient desaturation. The two humidification conditions assist in separating the convective and evaporative components of the desaturation process, showing a slight contribution from the convective effect, while the majority of the desaturation is due to evaporative removal. The convective removal is found to be insufficient to fully desaturate either GDL, with water remaining trapped underneath the channel rib with the more hydrophobic GDL and within the pore space in the more hydrophilic GDL. Stop-and-shoot Synchrotron X-ray computed tomography (CT) is then used in conjunction with program-assisted segmentation to determine initial saturation water volumes. These are then combined with the desaturation times found from the on-the-fly experiments to determine desaturation rates for both GDLs. The desaturation rate using dry air flow for the more hydrophobic GDL is found to be nearly four times faster than that of the more hydrophilic GDL. Results demonstrate that an evaporative contribution is necessary for either GDL sample to reach full desaturation.

Introduction

Polymer Electrolyte Membrane (PEM) fuel cells are a scalable energy conversion technology with the potential for zero local emissions, high power density, and rapid start-up, making them an attractive option for both mobile and stationary applications [1]. One of the main challenges to successful long-term, efficient fuel cell operation is water management, where water produced by the reaction and brought into the cell via a humidified air stream must be controlled [2]. Insufficient water management can lead to poor performance or total cell failure and reduce long term durability and efficiency [3,4]. Proper water management includes maintaining enough water to hydrate the electrolyte membrane for proton conductivity while not allowing for excess water accumulation,

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which blocks effective gas transport to the catalyst layer [5,6]. Thus, water management occurs over a range of length and time scales, from the catalyst layer and membrane to the flow field. Central to this challenge is the transient nature of fuel cell operation, previously reviewed by Banerjee and Kandlikar [7]. Any changes in power production result in changing water production rates, further complicating the balance of the overall system.

A key component to the overall water management of the fuel cell is maintaining appropriate water transport within the porous Gas Diffusion Layer (GDL), which is utilized to connect the flow field channels to the active reaction sites. At the cathode, it must allow for efficient oxygen transport to the catalyst layer while also facilitating the removal of water produced by the reaction. Multiple studies have been performed to better understand the transport phenomena occurring within this thin porous layer. These studies have included investigations of various properties related to water transport, as well as investigating structural modifications of the entire membrane electrode assembly (MEA). Such research includes the exploration of introducing perforations to specific locations of a GDL [8], modifying microporous layer (MPL) composition based on the gas flow conditions [9], and changing the surface properties of the different layers on the cathode side [10]. Investigations of transport properties of GDLs have been performed in both an in-situ and ex-situ manner. Owejan et al. [11] measured vapor and liquid transport rates through different GDLs ex-situ, and then investigated how these properties affected active cell performance. From this work, they proposed that one of the main functions of the MPL was to help prevent the formation of a water film on the surface of the catalyst layer (CL). Morgan et al. [12] measured the vapor diffusivity and permeability of various GDLs using a specially-designed diffusivity cell and identified how different GDL design parameters can enhance performance in high or low current density operation. Yau et al. [13] measured overall MEA water crossover rates between the anode and cathode without a current load through infrared sensors and outlet gas processing.

Signal analysis has also been investigated as a way to infer water movement or accumulation within a fuel cell. This enables a responsive control scheme to better avoid suboptimal performance by controlling the transient saturation of the GDLs. Hussaini et al. [14] proposed a dynamic external intermittent humidification scheme for multiple flow fields based on a feedback loop with the cell voltage. Song et al. [15] proposed a hydrogen purge protocol based on the analysis of the hydrogen pressure drop to indicate cathode flooding. Damour et al. [16] designed a model to control air flow to regulate the overall water content of the membrane and optimize performance. Battrell et al. [17] analyzed the anode pressure drop of an initially dry anode stream to calculate the amount of water being removed from a flooded fuel cell during the transient desaturation process. In each of these methods, a macroscopic parameter, such as the voltage or a pressure drop, is measured to infer a piece of the overall water behavior. These macro level studies have provided important information about the system, but understanding water management at the pore scale is necessary to inform the intelligent design and control of fuel cell systems.

Visualization has been a key tool to provide additional understanding of the various water management characteristics within the fuel cell at the micro level, and a review of the strengths and weaknesses of various visualization methods was provided by Bazylak [18]. One of the challenges related to visualization is that the natural architecture of a PEM fuel cell dictates that either specialized instrumentation or major modification to the overall fuel cell is required. Hussaini et al. [19] proposed a channel flow regime map of various two-phase flow patterns based on direct visualization experiments performed using an optically transparent fuel cell. Deevanhxay et al. [20] used soft X-ray tomography to visualize and confirm the increased flooding that occurs between the GDL and catalyst layer (CL) if an MPL is not applied to the GDL, which had previously been proposed by Owejan [11]. Iranzo et al. [21] utilized neutron beam imaging to visualize the transient water build-up during start-up and identified that water initially builds up at the cathode exit, but that the location was dependent on the specific flow field design. Another transient investigation by Banerjee et al. [22] looked at water distributions in the GDL using synchrotron radiography with increasing current density and found that the saturation increased to a certain threshold and would then stay steady as the current density was further raised. Lamibrac et al. [23] investigated water transport through the GDL with neutron radiography by varying the capillary pressure to understand the pathways that the water takes through the GDL and found that consecutive injections followed similar pathways, but that small water volumes would accumulate in unconnected pores. Such visualization experiments have been key in confirming previously inferred conclusions.

Another visualization technique used to investigate PEM fuel cells is synchrotron radiography, due to the available phase contrast along with high temporal and spatial resolution. Synchrotron radiography was used by Kruger et al. [24] to demonstrate the increase in cathode GDL saturation that occurs when a fuel cell is operated at a high current density. Lee et al. [25] used synchrotron radiography to visualize and quantify water within the GDL to investigate the effects of MPL thickness on GDL water content. Arlt et al. [26] investigated how artificially aged GDLs changed water management capabilities using synchrotron radiography and found that aging had a complex impact on water management, and not a simple increase in water. Chevalier et al. [27] used synchrotron radiography to characterize liquid water transport capabilities of various electrospun GDLs. All of the above studies demonstrate the important and conflicting roles that different layer properties and fuel cell designs can have on fuel cell operation. For example, in the study of novel electrospun GDLs [27], it was found that the GDLs could reduce water accumulation underneath the channel ribs but were also facilitating significant membrane dry-out. In many of the discussed studies, the focus was on the behavior of water ingress into the GDL or steady-state conditions. A key area which has been overlooked is an exploration of the mechanisms by which water is removed. For instance, as previously discussed, water management strategies that utilize a dry gas stream [14,15,17] rely on evaporation to provide GDL desaturation. However, this dynamic process has not been resolved in both a spatial and temporal manner in previous