Woven gas diffusion layers for polymer electrolyte membrane fuel cells: Liquid water transport and conductivity trade-offs

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HIGHLIGHTS

- Effect of different FEP loading on water breakthrough is investigated for woven GDLs.
- Water transport of woven GDLs was visualized employing fluorescent microscopy.
- The optimum FEP coating for thermal conductivity is 30 wt% among samples (0 and 55).
- Water propagate in treated GDLs and the breakthrough happens in compressed regions.

GRAPHICAL ABSTRACT
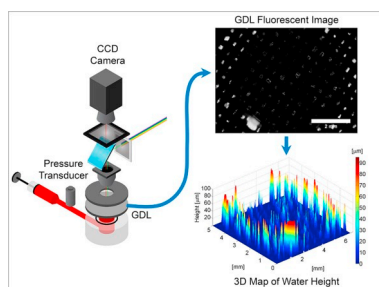
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ABSTRACT

Gas diffusion layers (GDLs) provide pathways for water removal in a polymer electrolyte membrane (PEM) Fuel Cell. Woven GDLs, have shown higher capability to retain water and improve performance under humid conditions compared to non-woven GDLs. In this work, we investigate water transport, distribution and location of breakthrough in woven GDLs using fluorescent microscopy. GDLs with no coating, 30, and 55 wt% fluorinate ethylene propylene (FEP) were investigated. FEP increases hydrophobicity and affects thermal and electrical conductivities. The results show that the FEP-treated GDLs have higher breakthrough pressures and water contact angles than non-treated GDLs. For untreated samples, water breakthrough occurs in non-compressed regions; whereas, for FEP-treated samples emergence occurs in the compressed regions. Furthermore, water was observed to first cover visible pores inside the GDLs prior to breakthrough. Increasing FEP loading promotes the propagation of water inside the GDLs. Thermal conductivity is found to improve with FEP coating and attains a maximum at 30 wt% FEP loading, whereas electrical conductivity decreases with increasing FEP loading. This analysis shows more pores are engaged in water transport with higher FEP loading. Implementation of woven GDLs in fuel cell design requires a balancing of the water and heat transport benefits with the reduced electrical conductivity.

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1. Introduction

The gas diffusion layer (GDL) plays a central role in the performance of polymer electrolyte membrane fuel cells (PEMFCs) [1]. The GDL facilitates transport of reactants from the flow channels to the catalyst layer and contributes to the transport of electrons and heat from the membrane electrode assembly (MEA), where the electrochemical reactions occur. Additionally, the GDL helps control the level of moisture in a fuel cell. Proper water management ensures that by-product water is removed from the catalyst layer to prevent flooding while maintaining the catalyst layer and MEA hydrated at the same time [2].

The GDL is a porous structure that is fabricated either by weaving carbon fibers into a carbon cloth or randomly distributing carbon fibers to form a non-woven carbon paper. Carbon fibers are mostly made of polyacrylonitrile (PAN) using a solvent spinning process [1]. Spun PAN yarns, used to make carbon cloth, are produced through the Worsted process, where yarns are generated and wrapped around a bobbin for weaving. The woven carbon fiber is then carbonized at a minimum temperature of 1600 °C (often under vacuum) [1]. This manufacturing process leads to a more flexible GDL structure. Non-woven GDLs are manufactured using the papermaking technology followed by sintering [1]. This process leads to a different microstructure for the non-woven GDLs than their woven counterparts. The pore sizes in woven GDLs vary in a wide range from 2 to 100 μm, whereas pore sizes in non-woven GDLs range from 10 to 30 μm. The wide range of pore size distribution in the woven GDLs is due to the multiscale microstructure of the constituent yarns that are formed from packed fine fibrils with large pores located between the yarns [3,4]. Moreover, woven GDLs have lower porosity and less tortuous structure compared to non-woven GDLs [3]. In addition, the in-plane porosity distribution in woven GDLs has a sinusoidal shape, which varies between 80 and 90%, whereas non-woven GDLs porosity distribution is more random. GDLs are commonly treated with hydrophobic polymers such as polytetrafluoroethylene (PTFE) or fluorinated ethylene propylene (FEP) [5] to improve the hydrophobicity of the GDLs using dipping, spraying, or brushing methods. Another process that improves the performance of the GDL is applying a thin microporous layer (MPL), to the side which is in contact with the catalyst layer (CL), to facilitate the wicking of liquid water from the CL to the GDL. MPLs have a pore size distribution much smaller than GDLs, from ~100 to 500 nm [6].

Experimental studies to characterize GDLs have been conducted (1) to understand transport properties, such as permeability, diffusivity, breakthrough pressure, electrical conductivity and thermal conductivity; and (2) to analyze the microstructure, including bulk porosity, pore size distribution, and porosity distribution. One of the key properties of GDLs is water breakthrough. Breakthrough analysis [7–10] provides information about the required pressure to overcome the capillary force and also the location of the water breakthrough in GDLs. Benziger et al. [7] investigated water breakthrough pressure for woven and non-woven GDLs without an MPL and showed that woven GDLs have a lower breakthrough pressure (~2 kPa) compared to non-woven GDLs (Toray samples ~7 kPa). The lower breakthrough pressure of woven samples is due to larger pores located between yarns of the woven GDLs. Furthermore, increasing the PTFE loading of the GDL slightly increases the breakthrough pressure. Lu et al. [9] investigated non-woven GDLs with and without MPL (SGL 25BA and SGL 25BC). SGL samples are more porous compared to Toray samples and have larger pores and porosity values [11]. A breakthrough pressure of 1.7 kPa was reported for GDLs without MPL and 6.7 kPa for GDLs with MPL [9]. This is due to the smaller pores of the MPL, which are expected to increase the breakthrough pressure.

Visualization of water breakthrough in GDLs has been studied with different imaging techniques. Two main techniques are X-ray micro-computed tomography (X-μCT) and fluorescent microscopy [12]. Flückiger et al. [10] performed X-μCT imaging of water breakthrough on non-woven GDLs to observe their water content. The scan time was as low as 5 min with a sample size of 2.5 mm in diameter. This study showed the saturation curve for different water intrusion pressures. More recently, Weber et al. [13] designed a new test setup to replicate the land and channel in the flow field and observe the water saturation in GDLs. The scan time was about 8 min with a sample size of 3.2 mm in diameter. Although X-μCT provides high spatial resolution and allows characterization of GDLs and interfaces in MEAs, the small sample size and the low temporal resolution limit the breakthrough analysis [11,14]. However, in the recent 3D X-μCT study of Eller et al. [15] resolution was improved with scan times of 3.2 s and 1% false water detection. Nonetheless, fluorescent microscopy temporal and spatial resolutions currently allow better tracking of the emergence of water. The challenges with optical fluorescent microscopy are the depth of field, which does not allow observation of the whole structure of GDLs, and the need for modified sample holders to provide access of light to the GDLs structure [16]. Litster et al. [16] visualized water transport through the thickness of non-woven GDLs and found the location of the breakthrough on the surface. Bazylak et al. [2] investigated the effect of compression on the location of the breakthrough in non-woven GDLs and showed that compression damages the PTFE and fiber structure and creates preferential pathways for water removal in the compressed areas.

Previous studies mainly characterized non-woven GDLs. However, a comprehensive study to understand water transport in woven GDLs is warranted since these GDLs have a higher capacity to retain water compared to non-woven GDLs (cf. [17]). The present study aims to investigate water transport in woven GDLs at the microstructure level. For this purpose, carbon cloth GDLs with three different FEP loadings (0, 30 and 55 wt%) were used to visualize water transport. The study reveals why woven GDLs have a higher capacity to keep water inside compared to non-woven GDLs, and documents the associated changes in thermal and electrical conductivity.

2. Experimental

2.1. GDL

In this study, a woven GDL, Avcarb 1071 HCB (from Fuelcell Earth), was used. This woven GDL has a thickness of 350 μm and porosity of 65 %. FEP solution (Teflon FEPD 121 Fluoroploymer Dispersion) was used to treat the AvCarb GDLs with 30 and 55 wt% FEP loadings. GDLs were dipped into the solution for 1 min and then placed on needle-point holders. The holders were kept in the vacuum oven at room temperature for an hour; the temperature was then increased to 100 °C for one additional hour. This process allowed evaporation of water and other solvents from the GDLs. To evaporate the surfactant, the GDLs were kept in a muffle oven for 50 min while the oven temperature was ramping up to reach 260 °C, and then for an additional 10 min at a constant 260 °C. To sinter the polymer, the temperature was ramped up to 280 °C over 20 min and kept at 280 °C for 20 min (also see Ref. [5]).

2.2. Visualization and breakthrough pressure

The apparatus to measure the breakthrough pressure and perform fluorescence microscopy to visualize water transport is described below. A dilute water solution of 1 mM rhodamine B (excitation: emission 540 nm: 625 nm) was prepared to trace water transport in the plane of the GDLs. Since this solution was dilute, dying the water had negligible effect on the properties of water (compared to pure water).

2.2.1. Apparatus

Samples were placed in an assembly and water was injected with a syringe pump at a rate of 0.02 ml min^{-1}. The clamping device has a top plate made from Plexiglas to visualize water, and it has a small hole (with diameter of 5 mm) for water removal. The O-ring diameter is 9 mm and the area between O-ring and open hole is under pressure. The