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Understanding the gas diffusion layer in proton exchange membrane fuel cells. I. How its structural characteristics affect diffusion and performance

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HIGHLIGHTS

- We provide a comprehensive discussion of GDL & MPL structural characteristics & how they are measured experimentally.
- We further examine the impact of these characteristics on transport & performance in proton exchange membrane fuel cells.
- Correlation between the effective diffusivity & the limiting current density is provided.
- The impact of the MPL structure on cell performance under wet as well as dry conditions is discussed.

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ABSTRACT

The proton exchange membrane fuel cell (PEMFC) has a significant potential in transportation, backup, and portable power applications, although there still are remaining technical and cost challenges. A key current goal is improving the performance while reducing the cost of the gas diffusion layer (GDL). Designing a commercial GDL, however, is far more complex than simply making a porous, sturdy, conductive layer, because of the trade-offs among performance, manufacturability, and cost. An improved understanding of its multifarious functions in the fuel cell can help attain this goal. Here, we identify 11 key characteristic parameters of the GDL and their significance to its performance. We begin a discussion of some of these parameters in this paper, specifically those related to the structure of the GDL substrate and the microporous layer (MPL), how these are measured experimentally *ex-situ*, how they influence fuel cell performance, and how they can be altered via the manufacturing process. In particular, we investigate the correlation between *ex-situ* measured effective diffusivity of water vapor and *in-situ* performance and limiting current density in a PEM fuel cell. Further, we examine the effect of adding multiple MPLs, MPL loading, and MPL particle size on cell performance under both wet and dry operating conditions.

1. Introduction

Polymer electrolyte membrane (PEM) fuel cells are poised for transportation, backup power, and portable power applications [1]. They have many advantages including low operating temperature, quick starting time, high efficiency, low weight, and simple design. Despite these advantages, however, modern PEMFCs face many challenges before fully realizing their commercial potential. These include high cost, inadequate durability, hydrogen storage and distribution issues, and water management difficulties.

The cost breakdown of PEMFCs has been analyzed both by the Department of Energy (DOE) [2] and in the literature [3]. It has been concluded that there needs to be a significant reduction in both the capital (high-volume, low-cost production capability) and operational (improved stack efficiency) costs before PEMFCs can be fully commercialized. The life of a PEM fuel cell is limited by: 1) membrane durability, 2) catalyst durability, and 3) that of the balance of plant (BOP) components. Many investigators have examined the issue of membrane durability, e.g., the impact of side chain degradation by hydroxyl radicals [4], performance loss due to chemical and electrochemical degradation [5–7], and with the mechanisms of membrane degradation [8–10]. Others have focused on catalyst durability, specifically Pt migration [11–13], carbon support corrosion [14,15] and catalyst poisoning [16–18]. Much work has also been

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done to investigate the BOP components for PEMFCs, namely, air blowers [19], heat exchangers [20,21], humidifiers [22–24] and power converters [25,26]. Our effort is focused on the often-neglected gas diffusion layer (GDL), its experimental and theoretical characterization, and issues related to its design, cost, and manufacturability.

The GDL is comprised of a highly porous woven carbon fabric, or a carbon fiber paper, that is treated with PTFE (Teflon) to make it hydrophobic, and then coated with a microporous layer (MPL) to form a 2-layer graded porous structure, as shown schematically in Fig. 1. The GDL is characterized by the following five key features [27], each of which requires specific, sometimes competing, GDL properties for desired performance:

1) *Reactant permeability*: This should be as high as possible to ensure an adequate supply of reactants (O_2/H_2), especially oxygen from air, to the catalyst layer so that the performance is limited by electrode kinetics, not by reactant transport. This requires the GDL to be macroporous and largely free of liquid water.
2) *Product permeability*: This should be high to remove water to the flow field effectively and prevent it from building up at the cathode catalyst layer (CCL), thereby blocking the reactant access to the catalyst, and. On the other hand, it should not be so high as to lead to membrane drying, which would reduce the proton conductivity of the membrane [28].
3) *Electrical conductivity*: This should be as high as possible to minimize Ohmic losses by effectively conducting electrons between the catalyst layer and the current collectors via the bipolar plate. However, it is reduced by increased porosity and PTFE content of the GDL, necessary for high reactant and product permeabilities.
4) *Thermal conductivity*: This should be high to effectively remove the heat from the membrane electrode assembly (MEA), where it is produced, to the bipolar plates, where cooling is available. The heat effects are mainly associated the cathode reaction, the Ohmic losses in the membrane, and water evaporation/condensation.
5) *Mechanical support*: The GDL must provide robust mechanical support to protect the membrane and catalyst layers from damage during assembly, while maintaining good interfacial contact among the MEA layers, to prevent damage from any pressure differentials across the MEA, and to avoid its deflection into the flow field channels during cell compression.

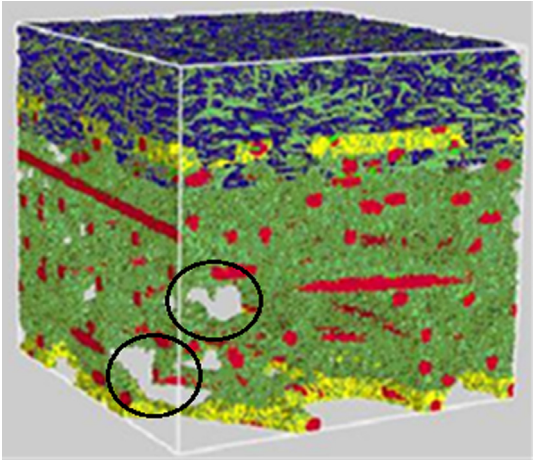

Fig. 1. 3-D rendering of GDL structure with carbon fiber in red, fill matrix in green, PTFE loading in yellow, MPL structure in blue, and some large pores circled. Made with GeoDict® software, and provided by Ballard Power Systems. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The reactant permeability of the GDL, typically characterized by the effective oxygen diffusivity of the cathode GDL, usually determines the limiting current density, and is dependent on the pore structure and the thickness of the GDL as well as the saturation level, or water loading. The effective diffusivity is a function of the porosity, tortuosity, and the mean pore radius of the GDL [29–31], as discussed further below. More recently, efforts have also been made to relate it to other microstructural properties, such as pore shape, orientation, and connectivity [32,33]. Similarly, the product permeability of the GDL is controlled by its pores structure, thickness, as well as the PTFE content.

In addition, the transport properties of a GDL are greatly influenced by the MPL design (Fig. 1). Although, the GDL is often treated as a homogenous material, in reality the GDL substrate and MPL are separate, distinct layers. For clarity, thus, here we will refer to the carbon paper base layer without any MPL coating as simply the "substrate", while "GDL" implies the composite of the substrate plus the MPL. Further, although the substrate can be woven carbon cloth or a carbon fiber paper, the focus in this work is exclusively on the carbon fiber paper, since the woven fabrics are significantly more expensive than their paper counterparts, due to the higher amounts of expensive carbon fiber. This cost difference can be as much as ten-fold at high volume production, which is prohibitive to fuel cell commercialization.

Significant efforts have been made to examine the influence of the presence of an MPL [34–37], the optimization of the MPL thickness and composition [38–40], and the effect of critical MPL properties including pore size distribution, porosity, and hydrophobicity [41–43]. Some recent publications [44,45] provide an excellent review of experimental characterization of the *ex-situ* electrical, thermal, and mechanical properties of the GDL, so these are not the focus in this paper.

A key characteristic of the transport performance of a GDL is measuring *in-situ* the limiting current density of a fuel cell, at which the cell voltage drops to zero owing to mass transportation limitations, typically in the cathode GDL. Mathematical models have been developed for the limiting current density in terms of transport properties of the GDL [46–49]. The limiting current density, of course, affects the performance at lower currents as well and is, thus, critical. The development of GDLs with higher wet limiting current densities could allow for smaller, more energy efficient fuel cells with lower overall costs.

Despite significant progress in recent years, the further advancement of PEM fuel cells is limited in part by the lack of a good understanding of how the performance of a GDL is correlated to its structure and design. While much effort has been spent on optimizing GDL properties for specific applications, there has been little effort devoted to understanding how the demands on the GDL change with application, and how those demands can be met by altering the GDL design. Here, we focus on the structural properties of the GDL and MPL, describing how they are measured experimentally, and exploring how they influence fuel cell performance.

A total of 11 *ex-situ* GDL characteristics were identified that influence GDL performance, and are listed in Table 1 in order of their perceived importance. Some of these, i.e., those related to the GDL and MPL structure and design are discussed below in detail including why they are important, how they can be measured experimentally, and how they can be altered in the GDL manufacture. Other characteristics listed in Table 1 are left for discussion in future publications. All the GDLs used in this work were fabricated using a commercial low-cost, high-volume production process at AvCarb® Material Solutions in Lowell, MA, USA.