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Effects of local gas diffusion layer gas permeability variations on spatial proton exchange membrane fuel cells performance

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HIGHLIGHTS

- The effects of a local GDL permeability defect on PEMFC performance were studied with a segmented cell.
- The defective GDLs through-plane gas permeabilities differed by a factor of 2.6.
- The defects were intentionally created at dimensionless flow field lengths equal to 0.4 and 0.9.
- The defect caused a change in the local performance that was more intense at high current densities.
- The higher permeability GDL improved local performance whereas the lower permeability GDL had the reverse effect.

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ABSTRACT

The effects of local gas diffusion layer (GDL) gas permeability variation and its location on spatial and overall proton exchange membrane fuel cell (PEMFC) performance were studied using a segmented cell approach. Variations in the physical and chemical parameters of the main membrane electrode assembly's (MEA) components (e.g., the membrane, electrode, and GDL) are considered defects and might negatively affect fuel cell performance. An artificial GDL defect was introduced by exchanging a standard (or intact) cathode GDL at one segment (segment 4 or 9) with a defective GDL. The standard and defective cathode GDLs had different through-plane gas permeabilities, while values were similar for in-plane permeability and some other structural parameters. The effects from a defective GDL were observed at a high current. Introducing a highly permeable GDL as a defect increased local performance due to a decrease in mass-transfer overpotential. For a defective GDL with lower permeability than the standard GDL, a local performance decrease was observed because mass-transfer losses increased. Simultaneously, downstream segment performance improved, which might be due to changes in water management. Defect localization at the cell outlet resulted in the detection of the defect at a lower current density compared with localization at the cell inlet. Spatial polarization curves (VI) and electrochemical impedance spectroscopy (EIS) facilitated detection and localization of GDL defects. Thus it was demonstrated that the local GDL anomalies are detectable by the segmented cell system.

1. Introduction

The gas diffusion layer (GDL) is a membrane electrode assembly (MEA) component. It ensures electronic/heat conductivity and mechanical strength to the MEA. However, the primary GDL function is to provide water and reagent transport in proton exchange membrane fuel cells (PEMFC). The GDL must simultaneously control water transport in and out of the electrodes and set conditions for high rates of gas transport under a wide range of operating conditions. For example, the GDL must maintain an appropriate level of membrane hydration in hot and dry environments at low currents when water production is low. On the other hand the GDL must also be adaptable to high water production rates at a high current, which facilitates efficient water removal and simultaneously directs water flow from a cathode to an anode to prevent membrane dehydration.

GDL permeability is a major parameter that influences gas and liquid transport in a PEMFC. Permeability is determined by the internal GDL structure and texture. Traditionally, a GDL comprises an electronically conductive macroporous substrate, such as carbon paper or carbon cloth, which is impregnated by a polytetrafluoroethylene (PTFE) solution to modify the hydrophobic/hydrophilic

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properties. In addition, a thin microporous layer (MPL) of carbon material and PTFE is applied on the substrate. This modification provides a layered structure for the GDL, and the layers have different textural properties and pore size distributions [1,2]. Because the average MPL pore size is similar to the catalyst layers, the MPL ensures enhanced electrical contact, mechanical compatibility, as well as improved water management between a GDL and an electrode. As a result of its multi-layered structure, the GDL is highly anisotropic, and its permeability is typically represented through two values: "in-plane" (x–y direction) and "through-plane" (z direction). The viscous permeability coefficient (dimensions L^2) reflects the viscosity loss between the fluid and pore walls. The inertial permeability coefficient (dimensions L) describes losses due to changes in flow direction at the microscopic level (tortuosity) and has been combined with the viscosity loss equation of Darcy by Forchheimer (see for example [3–5]).

Most papers on GDL permeability present methods for permeability determination and its connection with GDL textural and morphological properties [3,6–9]; some studies have demonstrated the impact of gas permeability on fuel cell performance [10–15]. However, most publications that characterize GDL permeability report modeling results [16–24]. It was shown that the in-plane and through-plane permeabilities have different values, and most GDLs have higher in-plane than through-plane permeabilities because of the layered structure [3,7,10,11,15,20]. Typically, in-plane permeability does not significantly depend on an MPL; however, permeability in two perpendicular in-plane directions may produce significant anisotropy due to the direction of the aligned fibers [7]. MPL strongly impacts through-plane permeability [3,7,8,12,15,20]. The effect of an MPL on through-plane permeability confirms that textural GDL properties, such as porosity, pore volume, and pore size, determine this parameter. For example, a decrease of the GDL porosity and pore size leads to a decrease of the material permeability [10,11,13,15,17]. Based on the same reasons, GDL compression decreases through-plane permeability, as previously reported [6].

The impact of PTFE loading on a macroporous carbon substrate [8,12–14,25,26] and MPL [3,15] on GDL permeability was studied. A decrease of the GDL permeability with an increase of PTFE content was mainly reported [13–15,26]. In general, such observations are attributed to a decrease in GDL pore volume, porosity, and pore size due to infill of GDL pore volume by an excess of PTFE. However, some authors have found that an increase in PTFE loading enhances permeability [3,8,12]. The PTFE content influences GDL rigidity; materials with higher PTFE content are more rigid and therefore can crack under a compressive load. Crack formation on an MPL surface in a GDL, and hence, greater permeability was reported by M.S. Ismail et al. [8]. M. Uchida et al. [27] studied the effect of PTFE on porosity of a carbon powder–PTFE mixture. The mixture comprised 20–40 nm carbon grains, which formed larger agglomerates (200–300 nm). The pores can be classified as primary and secondary pores, respectively. The primary pores are located between the carbon grains and are 20–40 nm, whereas secondary pores are located between the carbon agglomerates, and their size ranges from 40 to 1000 nm. It was reported that the secondary pore volume increases with an increase in PTFE, whereas the primary pore volume remains unchanged [3,27]. The large PTFE particles cannot penetrate the primary pores but can penetrate the secondary pores, which increases total porosity and permeability.

The effect of GDL permeability on PEMFC performance has primarily been reported under a high current or low voltage in a limiting current region [9–15,18,24]. M.V. Williams et al. [10,11] showed a correlation between through-plane permeability and limiting current under three operating conditions, 80°C/75% relative humidity (RH) cathode inlet, 100 °C/70% RH cathode inlet, and 120 °C/35% RH cathode inlet. GDLs with higher permeability had a higher limiting current. Moreover, it was suggested that these GDLs had larger pores and a smaller fraction of pores associated with the range affected by capillary condensation; therefore, it could avoid oxygen transport limitations due to water flooding at near-saturation conditions. G. Lin and T.V. Nguyen [12] studied the effects of GDL thickness and content of a hydrophobic agent on electrode flooding level in a PEMFC. The through-plane permeability for all the samples in this study was also measured. Higher performance was observed for GDLs with an MPL that had higher through-plane permeability.

C.-J. Tseng and S.-K. Lo [13] investigated the impact of several GDL parameters on PEMFC performance. One of the parameters was MPL thickness, which varied at 38, 84, and 136 μm. The GDL through-plane permeabilities were estimated using the semi-empirical Kozeny–Carman equation [19] as follows:

$$K = \frac{\varepsilon^{n+1}}{C(1-\varepsilon)^n}$$

where ε is porosity, while the exponent n and constant C are Kozeny–Carman constants. The through-plane gas permeability decreased with an increase in MPL thickness. However, the sample with an 84 μm MPL and an intermediate permeability value performed best at a 60 °C cell temperature and 70 °C humidifier temperature.

The impact of MPL properties on PEMFC performance was also studied in detail by T. Kitahara et al. [15]. A 110 μm-thick MPL was coated on a 24BA GDL. The MPL mean pore diameter varied from 1 to 10 μm. Through-plane permeability increased with an increase in the MPL mean pore size. Fuel cell performance was investigated at a low (0% RH) and high (100% RH) cathode humidity. Anode gas humidification was 100% RH in both cases. At low humidity the fuel cell performance was the best where the GDL had the smallest MPL pore size and lowest through-plane permeability because the MPL prevented MEA dehydration. At high humidities, the best performance was found using GDLs with a mean MPL pore diameter of 3 μm. This GDL had an intermediate through-plane permeability value as it was also reported in [13]. This GDL facilitates the transport of reagents from a gas channel to a catalyst surface as well as water transport from an electrode to the gas phase. Recent numerical analyses for PEMFCs with varying GDL permeabilities in the through-plane direction confirmed the experimental observations on the impact of permeability on PEMFC [9,18]. Water and thermal management were good in a system with high permeability in at least one direction (in-plane or through-plane), while water and heat management were poor in a system with low permeability in both directions [18].

In general, GDL permeability is connected to many GDL parameters, such as thickness, PTFE loading, compression, and MPL. Thus, permeability variation within one GDL component should be considered a defect that may affect PEMFC performance at a high current density and result in premature MEA degradation or failure. Because few papers provide experimental data for the impact of GDL permeability on a fuel cell, it is important to study effects of this GDL parameter on a PEMFC. A segmented cell system is an appropriate and powerful tool for the in-situ study of current density and voltage distributions [28–43]. The Hawaii Natural Energy Institute's (HNEI) segmented cell system was partially based on a design developed at the Los Alamos National Laboratory that used closed-loop Hall sensors [41,42] and an improved data acquisition system. This data acquisition system allows the simultaneous, rather than sequential, measurement of spatial electrochemical impedance spectroscopy (EIS), cyclic voltammetry (CV), as well as linear sweep voltammetry (LSV) to determine the