INVESTIGATION OF GAS DIFFUSION MEDIA USING CFD MODELING

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ABSTRACT

The performance of a gas diffusion layer comprised of a macro porous and micro porous layer has been studied both experimentally and by numerical simulation. Experimental data at different humidification conditions has been compared to full cell, three dimensional computational fluid dynamics calculations to validate the physical model of the cell. Local distributions of current density, membrane conductivity, temperature, and gas composition are discussed in detail. Model calculations with and without the micro porous layer show that this layer has a small effect on the overall performance. However, the local distributions show significant differences.

INTRODUCTION

The main purpose of Gas Diffusion Media (GDM) in a Polymer Electrolyte Membrane Fuel Cell (PEMFC) is to distribute the reactants along the active surface of the electrodes. In addition, the GDM has to ensure proper transport of product water, electrons, and heat of the reaction. Porous carbon materials are most often used to accomplish this complex task. A micro porous layer or diffusion layer (1,2) is sometimes added between the main macro porous GDM layer and the Membrane Electrode Assembly (MEA) as illustrated in Fig. 1. The purpose of this micro layer is to aid in the distribution of the reacting gas flows to the MEA surface, mechanical compatibility between the layers, the contact resistance, local current density distribution, and water management. The micro layer is usually comprised of carbon for electrical conductivity and PTFE for hydrophobicity. The objective of this study is to understand the role of the micro porous layer as well as the interactions with the macro layer and the flow field configuration.

Computational Fluid Dynamics (CFD) is gaining more interest as a tool to understand fuel cell performance (3-6). The model calculations provide insight into the fuel cell on a local level and describe distributions of current density, temperature, reacting gases, and water (vapor and liquid). Thus, modeling will help in gaining an understanding of the physics inside the PEMFC, aid in data analysis, and identify limiting parameters. A
previously published model (6-8) has been used to estimate the performance of a PEMFC using a layered GDM structure. In this study, experimental performance data are compared to numerical results to validate the CFD model and to study the local distributions. Furthermore, the effect of the micro porous layer is studied by comparison of numerical results to calculations without the micro layer.

EXPERIMENTAL

Experimental data was obtained with 40% hydrogen in nitrogen on the anode and air on the cathode using a 5620 PRIMEA® membrane electrode assembly from W.L. Gore & Associates, Inc. (Elkton, MD). It was assembled in a 25 cm² test cell (Fuel Cell Technologies, Inc., Albuquerque, NM) using SGL hydrophobized macro gas diffusion media (SGL Carbon AG, Meitingen, Germany) and CARBEL™ MP micro gas diffusion media (W.L. Gore & Associates, Inc.). The average compression in the cell was about 150 psi. The cell contained a triple serpentine flow pattern and was operated in the co-flow configuration. All tests were performed with a standard gas delivery unit (Globe Tech, Inc., Bryan, TX) with a Scribner 890b electronic load with PC control (Scribner Associates, Inc., Southern Pines, NC). All mass flow controllers and sparger bottles were calibrated for flow rate and dew point, respectively. All polarization curve data was collected using automated test protocols (Fuel Cell™ software by Scribner Associates, Inc.) by holding 10 minutes at each set point and averaging the last minute of collected data. The operating conditions were 65°C cell temperature with 0 psig back pressure and an anode and cathode stoichiometry of 1.2 and 2.0, respectively. The gas humidification was varied during the experiments for a comparison of model calculations and experimental data over a range of operating conditions. Four conditions were tested: fully humidified gas streams (condition #1: 65°C/65°C dew point on anode and cathode, respectively), highly oversaturated streams (condition #2: 85°C/85°C), dry anode (condition #3: dry/65°C), and dry cathode (condition #4: 65°C/dry).

NUMERICAL

The numerical work is based on a steady state, multi-phase phenomena, and three-dimensional mass transfer model including heat transfer aspects of a full-cell PEMFC. The typical single cell PEMFC used for testing is a sandwich of two graphite flow channel blocks laced with channels with designed flow-field pattern, separated by GDM and a MEA. The MEA consists of a membrane and two electrodes that have dispersed platinum catalysts. The flow channel geometries used in this work is shown in Fig. 2. It has triple pass channel with 10 serpentine fashions. The flow channel has dimension of 0.09 (height) x 0.07 (width) cm² cross-section flow area in each channel with the length of 5.00 cm. Each diffusion layer (macro and micro) has dimensions of 0.033 (height) x 5.00 (width) x 5.00 (length) cm³. Therefore, the active area of the MEA is 25 cm². A total of 68 x 272 x 41 cells (elements) were used in the flow channels and gas diffusion layers to model the fuel cell. The MEA is placed in between anode and cathode diffusion layers, which are containing macro and micro structures.
Model equations

The equations used in this work (conservation of mass, Navier-Stokes equations, species transport equations, energy equation, and water phase change model, with the source terms) were developed in previous studies (7,8). In addition, the water phase change model is extended in this work to include a liquid water film presented on the membrane surface for both anode and cathode (9). When the local activity of water exceeds unity, liquid water is present in the control volume and a liquid water film forms. The thickness of this liquid film depends on the rate of condensation/evaporation and the production of water by electrochemical reaction. The solubility of H₂ and O₂ is included in this model using Henry's Law to account for the diffusion of reacting gases to the MEA surface through the liquid water film. Thus, the local current is decreased as water condenses. Further, the overpotential equation shown in references (7,8) has been modified by simplifying the general form of the Butler-Volmer Equation to include the effect of concentration for both anode and cathode as shown in Eq. [1]:

\[
\eta(x,y) = \frac{RT}{4F} \ln \left( \frac{P_{O_{2}in}}{P_{O_{2}}(x,y)} \right) - \frac{RT}{2F} \ln \left( \frac{P_{H_{2}}(x,y)}{P_{H_{2}in}} \right) + \frac{RT}{0.84F} \ln \left( \frac{i(x,y)P_{O_{2}in}}{i_{o,02}P_{O_{2}}(x,y)} \right) + \frac{RT}{2F} \left( \frac{i(x,y)P_{H_{2}in}}{i_{o,H_{2}}P_{H_{2}}(x,y)} \right)
\]  

[1]

A control volume technique based on a commercial flow solver, FLUENT (version 4.56), is used to solve the coupled governing equations. This software requires specification of the source terms for species transport equations, two-phase equations for water, and heat generation equations created by electrical losses as shown in references (7,8). The new subroutines with the inclusion of the solubility effect were written to calculate the electrochemical overpotential and permeability for this simulation. Also, FLUENT requires a subroutine to account for the flux of protons and water across the membrane. The operating conditions and parameters used in the numerical model are shown in Table 1. All material properties were determined from ex-situ observations, except the exchange current density for the hydrogen oxidation reaction, \( i_{o,H_{2}} \), which was adjusted to obtain a better fit to the experimental data at condition #1. The results will be presented in the form of distributions of current density, membrane conductivity, temperature, liquid water fraction, and alpha. The net water transfer coefficient per proton, alpha, is a measure of the water management in the cell. A positive value indicates that electro-osmotic drag dominates over back diffusion and a net transport of water from anode to cathode. A negative value for alpha shows a net transport from cathode to anode. A well-balanced fuel cell will result in an alpha value close to zero.

RESULTS AND DISCUSSION

Figure 3 shows four polarization curves taken from experimental data compared to numerical results. The four data points at a cell potential of 0.65 V were chosen to compare to the model calculations. The numerical results agree very well with experimental data for all cases. Both experiment and numerical calculations show that condition #1 (65°C anode dew point and 65°C cathode dew point) gives the highest...
performance with a current density of 644 mA/cm², and condition #4 (65°C anode dew point and dry cathode) gives the lowest performance with a current density of 298 mA/cm². Further, condition #2 (85°C anode dew point and 85°C cathode dew point) gives lower performance (569 mA/cm²) than condition #1 due to the water flooding. Finally, condition #3 (dry anode and 65°C cathode dew point) provides a current density of 462 mA/cm² at 0.65 V.

**Condition #1: 65°C dew point on anode and 65°C dew point on cathode**

Figures 4 a-d presents the local current density, membrane conductivity, temperature, and alpha distributions on the membrane surface for condition #1 (65/65°C dew point). Fig. 4a shows the current density distribution with an average of 644 mA/cm² at 0.650 V. The distribution is very non-uniform with the highest current density of ~800 mA/cm² from the entrance region toward the outlet with the lowest value of ~400 mA/cm². This is caused by the concentration reduction of the reacting gases, even though the membrane conductivity increases from inlet toward outlet as shown in Fig. 4b. The membrane conductivity increases from inlet to outlet due to an increase in water activity. Even though the gases enter the fuel cell fully humidified, the activity inside the cell is less than unity due to the increased temperature of up to 72°C caused by the heat of reaction. Further, in the region of the channel bends, the current density is slightly lower at inner channel of the triple serpentine compared to the outer channel. This is because at the inner bend, the velocity magnitude is lower than the outer bend. Therefore, the heat transfer coefficient of the inner bend is lower than that of the outer bend. Thus, the temperature of the inner bend is higher than the outer area as shown in Fig. 4c and this creates the lower membrane conductivity at the inner bend that outer bend shown in Fig. 4b. The distribution of alpha as shown in Fig. 4d, the net water transfer coefficient, is very uniform and has an average value close to zero indicating a good balance of water transport between anode and cathode.

**Condition #2: 85°C dew point on anode and 85°C dew point on cathode**

Figures 5 a-d present the local current density, membrane conductivity, temperature, and liquid water distributions on the membrane surface for condition #2 (85/85°C dew point). Fig. 5a shows the current density distribution with an average of 569 mA/cm² at 0.660 V. The current distribution shows more non-uniformity than the distribution of 65/65°C dew point. The highest value is ~900 mA/cm² at the entrance region and the lowest value is ~200 mA/cm² at the exit region. This is because the higher inlet humidity applied in this condition leads to a well-hydrated membrane even in the entrance region. Fig. 5b shows that the membrane conductivity is very uniform with the high value. This is because the inlet humidity condition of both anode and cathode are higher than the actual cell temperature. The cell temperature is high in the inlet region (>75°C) as Fig. 5c shows caused by the high current density and water condensation. However, the temperature drops towards the outlet, which results in the accumulation of liquid water resulting in flooding. Fig. 5d shows that the liquid water fraction increases above 10% especially in the bends of the channels. This will significantly affect the concentration of oxygen at the MEA by the liquid water film resistance.
Condition #3: dry anode and 65° C dew point on cathode

Figures 6 a-d present the local current density, membrane conductivity, temperature, and alpha distributions on the membrane surface for condition #3 (dry/65°C dew point). Fig. 6a shows the current density distribution with an average of 462 mA/cm² at 0.645 V. This condition results in the opposite distribution compared with Figs. 4a and 5a. The lowest current density is shown at the entrance region with the value of -200 mA/cm² and the current density increases toward the exit region with the value of ~520 mA/cm². This is because the dry anode inlet results in low membrane hydration at the entrance region, giving the lowest local performance. Further down the cell, water from the cathode humidity and electrochemical reaction is transported across the membrane by back diffusion to hydrate the MEA. This results in increasing membrane conductivity from inlet toward outlet as shown in Fig. 6b. The temperature distribution illustrated in Fig. 6c is very uniform with the minimum value of 68°C at the entrance region and the maximum of 72°C at the exit region. The distribution of alpha in Fig. 6d shows a large negative value in the inlet region, indicating a strong water back transport from cathode to anode. Towards the outlet of the cell alpha approaches zero, which shows a balance between electro osmotic drag and back diffusion.

Condition #4: 65°C dew point on anode and dry cathode

Figures 7 a-d present the local current density, membrane conductivity, temperature, and alpha distributions on the membrane surface for condition #4 (65°C/dry dew point). Fig. 7a shows the current density distribution with an average of 298 mA/cm² at 0.660 V. The distribution is similar to Figs. 4a and 5a, but the value is much lower. The current density shows the highest value at the inlet region with a value of -450 mA/cm². The current density decreases toward the outlet with the lowest value of -250 mA/cm². The performance is dominated by the low membrane conductivity illustrated in Fig. 7b, which gives a similar distribution as the current density contour. The low membrane conductivity is due to the reduction of anode water activity by electro-osmotic drag from anode to cathode. The distribution of alpha shown in Fig. 7d indicates that alpha is always positive, which indicates that the electro-osmotic drag dominates over back diffusion even though the current density is low. There is little water back diffusion because of dry gas on the cathode. The temperature distribution shown in Fig. 7c is very uniform with an average value of 70°C.

Influence of micro layer

Simulations were performed without the micro diffusion layer for conditions #1 and #3 to study the influence of the micro layer on the behavior of the fuel cell. Fig. 8 shows current density and membrane conductivity distributions for condition #1 (65°C dew point on anode and 65°C dew point on cathode). Comparison of Fig. 8a with Fig. 4a, which is the current density distribution including the micro layer reveals that the configuration including the micro layer gives slightly higher performance than the case of no micro layer (see Fig. 8a) by 18 mV at similar averaged current density. Furthermore, the distribution without the micro layer shows more non-uniformity than the case including the micro layer with a higher current density at the entrance region and lower value at the exit region. This could be because the micro layer aids in the distribution of the reacting gas flows to the MEA surface. The membrane conductivity including the

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micro layer (Fig. 4b) shows higher values throughout, but particularly in the exit region compared to the case without the micro layer (Fig. 8b). It appears that the micro layer has a beneficial effect on local water management at this condition.

Fig. 9 shows current density and membrane conductivity distributions for condition #3 (dry anode and 65° C dew point on cathode). The current density distribution without the micro layer shown in Fig. 9a increases from inlet to about ¾ of electrode width and then it decreases toward outlet. Fig. 6a shows that the current density including the micro layer has a more uniform distribution and is continuously increasing from inlet to outlet. Again, the performance including the micro GDM layer is slightly higher by about 15 mV. Interestingly, Fig. 9b shows that the membrane conductivity without the micro layer shows higher values throughout, but particularly in the exit region compared to the case including the micro layer (Fig. 6b). The reason for this behavior is the low current density in the exit region, which results in increased water transport towards the anode and subsequent membrane humidification.

In summary, the micro diffusion layer appears to have a beneficial effect. Even though the overall performance of the fuel cell is very similar, the local distributions may be very different depending on the operating condition.

CONCLUSIONS

Full-cell numerical calculations of PEM fuel cell performance agree very well with experimental data. The cell potentials at four different operating conditions with vastly different humidification conditions are predicted to be within 2% of the experimentally determined value. The simulation provides valuable insight into the local distribution of current density, membrane conductivity, temperature, and transport rates. Comparison of these distributions explains the performance under the different operating conditions. Super-saturation of the incoming gas streams results in a very high current density at the inlet, but also to the formation of liquid water (flooding), which lowers the performance due to the increased film resistance for diffusion. Dry gas streams on either anode or cathode cause a low membrane conductivity and low performance. The two cases show opposite current density distributions and reveal details of the water management inside the cell.

Simulations without the micro porous gas diffusion layer show slightly better performance when the micro layer is present. The local distributions, however, can be very different, and it appears that the micro layer assures more uniform local distributions.

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Table 1: Inlet conditions for 4 data points.

| Condition | Anode channel inlet | Cathode channel inlet | Operating conditions |
|-----------|---------------------|-----------------------|----------------------|
| # 1 | $I_{avg} = 644$ mA/cm$^2$ | $I_{avg} = 569$ mA/cm$^2$ | $I_{avg} = 462$ mA/cm$^2$ | $I_{avg} = 298$ mA/cm$^2$ |
| Velocity (m/s) per channel | 7.552 | 18.363 | 2.383 | 3.494 |
| Mole fraction of $H_2$ | 0.30 | 0.17 | 0.40 | 0.30 |
| Mole fraction of water vapor | 0.25 | 0.57 | 0.0 | 0.25 |
| Mole fraction of $N_2$ | 0.45 | 0.26 | 0.60 | 0.45 |
| $I_{H_2}$ (mA/cm$^2$) | 60,000 | 60,000 | 60,000 | 60,000 |
| Temperature (K) | 338 | 338 | 338 | 338 |
| Velocity (m/s) per channel | 11.99 | 29.15 | 8.60 | 2.44 |
| Mole fraction of $O_2$ | 0.152 | 0.086 | 0.152 | 0.20 |
| Mole fraction of $N_2$ | 0.608 | 0.344 | 0.608 | 0.80 |
| Mole fraction of water vapor | 0.24 | 0.57 | 0.24 | 0.0 |
| $I_{O_2}$ (mA/cm$^2$) | 60 | 60 | 60 | 60 |
| Temperature (K) | 338 | 338 | 338 | 338 |
| Condensation rate (l/s) | 1.0 | 1.0 | 1.0 | 1.0 |
| Membrane thickness ($\mu$m) + catalyzed layers ($\mu$m) | 50.0 | 50.0 | 50.0 | 50.0 |
| Graphite collector conductivity (W/m-K) | 5.7 | 5.7 | 5.7 | 5.7 |
| MEA conductivity (W/m-K) | 0.143 | 0.143 | 0.143 | 0.143 |
| Operating pressure (Psig) | 0 | 0 | 0 | 0 |

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Fig. 1: Schematic representation of GDM layers and the MEA.

Fig. 2: The geometrical model of the 25-cm² triple serpentine flowfield.
Fig. 3: Polarization curves of four operating conditions compared to four calculated points.
Fig. 4a: Current density distribution, $i_{avg} = 644 \text{ mA/cm}^2$.

Fig. 4b: Membrane conductivity distribution, $\sigma_{avg} = 7.5 \text{ S/m}$.

Fig. 4c: Temperature distribution, $T_{avg} = 342.2 \text{ K}$.

Fig. 4d: $\alpha$ distribution, $\alpha_{avg} = 0.129$

Fig. 4: Local distributions on MEA surface at 0.650 V for condition #1: 65°C anode and 65°C cathode dew point
Fig. 5a: Current density distribution, \( I_{avg} = 0.569 \, \text{A/cm}^2 \).

Fig. 5b: Membrane conductivity distribution, \( \sigma_{avg} = 12.0 \, \text{S/m} \).

Fig. 5c: Temperature distribution, \( T_{avg} = 344 \, \text{K} \).

Fig. 5d: Liquid water mass fraction at cathode, average = 0.07.

Fig. 5: Local distributions on MEA surface at 0.660 V for condition #2: 85°C anode and 85°C cathode dew point.
Fig. 6a: Current density distribution, $I_{\text{avg}} = 0.462 \text{ A/cm}^2$.

Fig. 6b: Membrane conductivity distribution, $\sigma_{\text{avg}} = 2.66 \text{ S/m}$.

Fig. 6c: Temperature distribution, $T_{\text{avg}} = 342.2 \text{ K}$

Fig. 6d: $\alpha$ distribution, $\alpha_{\text{avg}} = -0.70$

Fig. 6: Local distributions on MEA surface at 0.645 V for condition #3: dry anode and 65°C cathode dew point
Fig. 7a: Current density distribution, \( I_{\text{avg}} = 0.298 \text{ A/cm}^2 \).

Fig. 7b: Membrane conductivity distribution, \( \sigma_{\text{avg}} = 2.6 \text{ S/m} \).

Fig. 7c: Temperature distribution, \( T_{\text{avg}} = 342.2 \text{ K} \).

Fig. 7d: \( \alpha \) distribution, \( \alpha_{\text{avg}} = 0.54 \).

Fig. 7: Local distributions on MEA surface at 0.660 V for condition #4: 65°C anode dew point and dry cathode.
Fig. 8: Local current density (A/cm²) and membrane conductivity distribution at 0.632 V for condition 65/65°C dew point without micro layer.

Fig. 9: Local current density (A/cm²) and membrane conductivity distribution at 0.630 V for condition dry/65°C dew point w/out micro layer.