The interactive effect of heat and mass transport on water condensation in the gas diffusion layer of a proton exchange membrane fuel cell

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A B S T R A C T

Despite recent advancement in fuel cell technology, significant challenges remain in achieving high power density operation to meet the stringent targets of performance, durability and cost. This is due to the lack of fundamental understanding in interactive transports of oxygen, protons, heat, and water. In this study, we employed both experimental and analytical methods to study water onset condensation using Toray and Freudenberg diffusion media, which have different thermal and diffusion properties. Toray performs better under dry conditions, while Freudenberg has improved performance under wet conditions. The dry and wet effective diffusivities obtained using the in situ limiting current support the performance results. Neutron images show that liquid water exists throughout the layer of diffusion media for Toray material, but only under the land for Freudenberg keeping the area under the channel open for oxygen transport. To further understand this fundamental mechanism, we developed a 1-D model to simulate fuel cell performance. In addition, we identify the water condensation behavior is controlled by the product of thermal conductivity and the ratio of tortuosity to porosity. The findings provide new insights into improving material design and boosting energy conversion efficiency under a wide range of fuel cell operation conditions.

1. Introduction

The world is facing a severe energy crisis. The need for petroleum fuel is increasing and global warming is reaching new magnitudes every day. In 2018, 69% of US total petroleum use went toward transportation, which comprised 92% of total transportation energy use [1]. Therefore, identifying a reliable and sustainable energy source for automotive application is crucial. Though electric battery is a potential solution, it is not suitable for continuous driving in high power mode due to its low energy density and long charging time [2]. These aforementioned problems make hydrogen and fuel cell technology the most viable solution to our energy issue [3]. However, there are still multiple technical challenges related to fuel cell systems. One of the key challenges is due to severe water condensation when operating at high current density [4]. In this work, we have provided direct qualitative evidence of the water condensation features at high current density, along with a quantitative analysis of critical material parameters. The results of this research will provide guidance in speeding up the material development and stack design to enhance performance and reduce cost.

A proton exchange membrane fuel cell (PEMFC) is a low-temperature (<95 °C) electro-chemical energy conversion device that combines hydrogen and oxygen to generate electricity with byproducts of water and heat. A PEMFC consists of five major components: membrane, catalyst layer (CL), microporous layer (MPL), gas diffusion layer (GDL), and flow field (FF). To achieve high power density performance, however, it is imperative to optimize the transport of gas, proton, electron, heat, liquid water, and compression load. When a PEMFC is operated at high current density, the amount of generated water often exceeds the limit of vapor transport. Therefore, liquid water starts to condense inside the GDL, blocking the pores for gas transport and increasing the resistance to oxygen.

Water condensation has been one of the most challenging issues in increasing the operating power density of a PEM fuel cell for more than 20 years [5,6]. Both experimental techniques and numerical models have been used to study water condensation and transport behavior in the GDL. The limiting current method has been used by several researchers to study the oxygen transport resistance in a GDL under dry operating conditions [7–9]. Baker et al. [10,11] extended the limiting
pores in the GDL facilitate water removal capability. However, none
of a different pore size, and the simulation results indicated that macro-
channel in a GDL. Park et al. [38] simulated two GDL samples with
water saturation is higher under the land compared to that under the
achieved by increasing cell compression. Li et al. [37] showed that GDL
overall water saturation level. Hang et al. [36] studied the effect of
anisotropy can increase the local temperature and therefore reduce the
distribution inside the GDL. Cao et al. [35] observed that the GDL
addition, several experimental efforts have been made to improve water
management in the GDL [19–23]. A number of 1-D and 3-D numer-
ical models [24–34] have also been used to obtain water saturation
studies all focused primarily on water capillary pressure and saturation,
and condensation on the oxygen transport resistance (\(\eta_2\)) of the diffusion
medium (DM). From these experiments, it is proven that the limiting
current methods can effectively study the oxygen transport resistance in
GDL under both dry and wet conditions. Bazylak [12,13], Litster et al. [14] used fluorescence microscopy, Zenyuk et al. [15–17] used X-ray computed tomography, Yang et al. [18] used an optical PEMFCs
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a different pore size, and the simulation results indicated that macro-
pores in the GDL facilitate water removal capability. However, none
of the studies directly investigated the effect of water condensation on

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current operating conditions to high relative humidity (RH ≥ 80%) and \(O_2\) mole fractions to quantitatively study the effect of liquid water condensation on the oxygen transport resistance (\(R_{O_2}\)) of the diffusion media (DM). From these experiments, it is proven that the limiting current methods can effectively study the oxygen transport resistance in GDL under both dry and wet conditions. Bazylak [12,13], Litster et al. [14] used fluorescence microscopy, Zenyuk et al. [15–17] used X-ray computed tomography, Yang et al. [18] used an optical PEMFCs to study the water transport mechanism in a GDL. However, these studies all focused primarily on water capillary pressure and saturation, which is not directly correlated with oxygen transport resistance. In addition, several experimental efforts have been made to improve water management in the GDL [19–23]. A number of 1-D and 3-D numerical models [24–34] have also been used to obtain water saturation distribution inside the GDL. Cao et al. [35] observed that the GDL anisotropy can increase the local temperature and therefore reduce the overall water saturation level. Hang et al. [36] studied the effect of compression on the temperature distribution in a GDL. They observed that a more uniform temperature distribution inside the GDL can be achieved by increasing cell compression. Li et al. [37] showed that GDL water saturation is higher under the land compared to that under the channel in a GDL. Park et al. [38] simulated two GDL samples with a different pore size, and the simulation results indicated that macro-pores in the GDL facilitate water removal capability. However, none of the studies directly investigated the effect of water condensation on overall oxygen transport resistance, which requires an in situ water visualization tool.

Neutron radiography has been used to investigate liquid water transport in a PEMFC [39–45]. Researchers have applied neutron imaging to study liquid water accumulation in the flow channel [46–50], GDL [51–55], micro-porous layer [56] and membrane [57]. Due to the high contrast between liquid water and fuel cell components, the results from neutron imaging have been able to directly identify liquid water in a PEMFC. In this study, we applied neutron imaging combined with the limiting current technique to study the through-plane water profile in a PEMFC under varying degrees of water saturation for the first time. The aims of this work are to fundamentally understand the physics governing the onset of water condensation in the GDL. In addition, we develop and apply a 1-D steady state non-isothermal PEMFC model [58] to validate our experimental results. The experimental and numerical results provide new insights for future PEMFC material design and system operation to enable commercialization of fuel cell technology.

2. Experiments

2.1. Materials

Two wet-proofed and MPL coated diffusion media, Toray TGP-H-060 and Freudenberg H23C8, are investigated in this study. The uncompressed thickness of Toray TGP-H-060 and Freudenberg H23C8 is 210 μm and 220 μm, respectively. The microscopic structure of the two GDL materials is presented in Fig. 1. It can be observed from the SEM images that, the GDL micro-structure such as fiber orientation and pore structure, is highly anisotropic. Freudenberg has smaller pore size and the pores are more randomly distributed compared to Toray [59]. In addition, the thermal transport properties of these two diffusion media show significant differences due to material and processing differences [60]. The reported values of through-plane thermal conductivity of Toray and Freudenberg are 1.24 W m\(^{-1}\) K\(^{-1}\) and 0.11 W m\(^{-1}\) K\(^{-1}\), respectively [61–63].

The membrane electrode assembly (MEA) used in this study consists of 0.30 mg cm\(^{-2}\) Pt loading on both anode and cathode sides with an NRE 211 membrane purchased from Ion Power Inc. An in-house designed 2 cm\(^2\) low pressure-drop straight parallel channel flow field is used for both performance and limiting current experiments [64]. PTFE gaskets with different thickness are used to control the compression strain of Toray TGP-H-060 and Freudenberg H23C8 to be 22 % ± 1.5% and 17 % ± 1.5%, respectively, with the target compression load to be 2 MPa under the land. Therefore, the target compressed thickness for Toray TGP-H-060 and Freudenberg H23C8 is 165 μm and 180 μm, respectively.

2.2. Fuel cell testing

Single cell 2 cm\(^2\) tests were performed on an automated G20 fuel cell test station manufactured by Greenlight Innovation [65]. High frequency resistances were measured using a Gamry Reference 3000 with 30k booster [66]. Polarization curves at 70 °C under both wet (100% RH and 300 kPa) and dry (60% RH and 100 kPa) conditions were tested with constant flow rates of 0.4 slpm of pure hydrogen and 2.0 slpm of air to evaluate fuel cell performance. In addition, limiting current experiments under different pressure and oxygen mole fraction were conducted to study dry and wet oxygen transport properties in the GDL. Detailed operating conditions of break-in, polarization, and limiting current are listed in Table 1.

The limiting current method is an in situ experimental technique to quantify oxygen-transport resistance in a fuel cell. Following the protocol developed by Baker et al. [10], we conducted limiting current tests under both dry and wet conditions to study the oxygen transport properties in both Toray and Freudenberg GDL materials. To ensure
The total oxygen transport resistance in a fuel cell is composed of the channel, the catalyst layer, the MPL, and thin film diffusion, transport resistances resulted from both Knudsen diffusion and intermolecular diffusion [58]. In the MPL and CL, the Knudsen diffusion may become an important transport method in the small size pores (<50 μm). In addition, oxygen gas can also diffuse through liquid water and ionomer films in the CL. Unlike inter-molecular diffusion, transport resistances resulted from both Knudsen and thin film diffusion are independent of pressure. Based on the transport mechanism, we can separate oxygen transport resistance into pressure dependent, \( R_p \), and pressure independent, \( R_{NP} \), as:

\[
R_{O_2}^{Total} = R_p + R_{NP}
\]

In limiting current tests, we performed the experiments at four different pressure conditions to quantify pressure-dependent and pressure-independent resistances under dry conditions [10].

2.3. Neutron radiography

In our study, the liquid water in an operating cell was quantified using Neutron Radiography at the National Institute of Standard and Technology (NIST). A detailed layout and camera setup of the neutron imaging facility can be found in the literature [67,68]. The experiment used an Andor Neo neutron camera, which is a scientific complementary metal-oxide (sCMOS) camera with a pixel pitch of 6.5 μm [69]. A set of specially designed fuel cell hardware was used to study the through-plane water distribution. The straight parallel channel design features 15 channels with a depth of 890 μm and a width of 635 μm. The hardware was fabricated using 6061 aluminum alloy to minimize neutron attenuation and plated with gold to enhance electrical conductivity and avoid corrosion. A custom-built fuel cell test station manufactured by the Micro Instrument Corporation was used for cell testing at the NIST [44]. Neutron images were obtained under steady-state operating conditions of 50 °C, 77% RH, 300 kPa, and 50 % OCV, for the tests at NIST.
0.30 V at 2%, 4%, 8%, and 16% oxygen concentrations. To increase the spatial resolution and reduce noise, the final neutron image was filtered and averaged for 12 min. Detailed image processing steps can be found in the literature [51,67,68]. To quantify the through-plane distribution, the water thickness is calculated based on the Beer–Lambert law [55]. Lastly, the final images are false-colored for better contrast and visualization.

3. Results and discussion

3.1. Limiting current measurement

Limiting current experiments were performed under both dry and wet operating conditions. The results from the dry conditions were conducted at 80 °C and 64% RH and are mainly used to analyze the diffusion properties of the materials without liquid water. As shown in Fig. 2(a) and (b), limiting currents were measured at four oxygen concentrations and four pressure conditions. The results from both Freudenberg H23C8 and Toray TGP-H-060 GDLs show linear trends in all conditions, which indicates constant and reliable transport resistances were obtained without liquid water formation in the cell. Based on Eq. (3), the total oxygen transport resistance can be calculated as a function of pressure. As shown in Fig. 2(c), Toray TGP-H-060 has a significantly higher oxygen transport resistance than Freudenberg H23C8. Following the analyses developed by Baker et al. and Rahman et al. [10,11,58], the ratio of tortuosity to porosity is calculated to be 5.0 and 3.5 for Toray TGP-H-060 and Freudenberg H23C8, respectively. The dry limiting current results demonstrate that, despite being thinner, the structure of TGP-H-060 has a higher gas transport impedance than the Freudenberg H23C8.

In contrast to the dry limiting current measurement, the wet limiting current tests are probing the oxygen transport resistance from the onset of liquid water condensation to reaching maximum liquid water saturation in the GDL by increasing oxygen concentration from 1% to 21%. Fig. 3 shows the total transport resistances of both GDLs operating at 70 °C, 300 kPa, and three relative humidity conditions. Toray TGP-H-060 exhibits a clear transition of the transport resistance from no liquid water (≈ 2 s cm⁻¹) to the maximum saturation (≈ 4 s cm⁻¹). In addition, the onset water condensation of Toray TGP-H-060 has an immediate effect on total transport resistance, which shows a rapid increase as in Fig. 3(a). This explains why the limiting current density associated with onset water condensation increases with increasing channel RH. Similar to the observations from Caulk et al. [70], the transport resistance of Toray TGP-H-060 reaches a wet plateau, which indicates that the maximum saturation in the GDL is achieved. In contrast, the transport resistances of the Freudenberg H23C8 only increased gradually with increasing limiting current densities, regardless of the channel RH, as shown in Fig. 3(b). This behavior differs significantly from the conventional trend shown in Fig. 3(a) and can be attributed to the following two potential reasons: (1) there is much less liquid water condensation occurring in the Freudenberg H23C8 and/or (2) the liquid water condensation does not significantly impede the oxygen transport.
To investigate the liquid water distribution in both GDLs to directly study this phenomenon.

3.2. Fuel cell performance

To evaluate fuel cell performance with both GDLs, polarization curves under both dry and wet operating conditions as listed in Table 1 are performed. As shown in Fig. 4(a), the Toray 060 shows similar performance to the Freudenberg H23C8 at a low to intermediate current density region (≤1.25 A cm⁻²) and then diverges slightly at a high current density region (≥1.5 A cm⁻²) under dry operating conditions. The performance difference between these two materials can be mostly attributed to the high frequency resistances (HFR), as indicated in Fig. 4(a). The HFR measured at 1000 Hz represents ohmic resistances, which include both electronic and ionic resistances in the fuel cell [71]. The electronic resistance is primarily a function of cell compression and remains relatively constant during operation. On the other hand, the ionic resistance of the conducting proton is strongly dependent on the hydration of the membrane and ionomer in the CL, which varies with material properties and operating conditions [72]. Since the Freudenberg GDL has a higher thermal resistance, the cell would have higher membrane temperature and lower hydration than the Toray GDL under similar operating conditions [58]. Additionally, the heat generated from the ohmic heating can contribute to a further temperature increase in the adjacent CL [73]. In addition, the Freudenberg GDL has a lower gas transport resistance than the Toray GDL under dry conditions as shown in Fig. 2(c). Therefore, the generated water in the cell with Freudenberg GDL can be diffused to the channel more easily, which also results in reduced membrane hydration. In conclusion, high thermal resistance and low diffusion resistance are responsible for a higher HFR and thus a lower performance at a high current density region (≤1.5 A cm⁻²) for the Freudenberg H23C8 under dry operating conditions.

Under wet operating conditions, the HFRs shown in Fig. 4(b) from both cells are flat and close to each other, indicating that both GDLs have similar electrical resistance and the membrane is fully hydrated at its minimum resistance. Therefore, the performance difference when current density is greater than 1.0 A cm⁻² between the two GDLs can be attributed to oxygen transport loss due to water condensation. The GDL performance under wet operating conditions agrees very well with the wet oxygen transport resistance measurements by limiting current density is greater than 1.A cm⁻² at its minimum resistance. Therefore, the performance difference when both cells are flat and close to each other, indicating that both GDLs have similar electrical resistance and the membrane is fully hydrated at its minimum resistance. Therefore, the performance difference when current density is greater than 1.0 A cm⁻² between the two GDLs can be attributed to oxygen transport loss due to water condensation. The GDL performance under wet operating conditions agrees very well with the wet oxygen transport resistance measurements by limiting current density.

The Toray TGP-H-060 shows significant performance loss due to high oxygen transport loss as soon as there is water condensation in the Freudenberg H23C8. To date, the exact cause for this behavior is unknown. In this study, we employed neutron radiography to investigate the liquid water distribution in both GDLs to directly study this phenomenon.

3.3. Neutron radiography

To develop a fundamental understanding of how liquid water affects the oxygen transport, we designed a specialized cell and performed in situ neutron radiography experiments to study the through-plane liquid water distribution in the GDL. A photo of the test cell sitting in front of the neutron detector is shown in Fig. 5(a). To obtain reliable liquid water content in the cell, a baseline neutron image at the operating temperature under perfectly dry conditions is required. Therefore, the baseline neutron images shown in Fig. 5(b, c) were obtained right after the cell was built and before flowing any humidified gas through the cell. In addition, the illustration in Fig. 5 shows the cross-section of the corresponding location of a neutron image in a fuel cell. Besides being used in image processing, the analyzed pixel intensity of the baseline dry neutron images is also used to identify the physical pixel location of the MEA, diffusion media, and channel. As shown in Fig. 5, the Freudenberg GDL is slightly thicker than the Toray GDL, which agrees with the material properties discussed in Section 2.1.

Steady-state neutron images were taken at four oxygen concentrations (2%, 4%, 8%, and 16%) of a fuel cell operating at 0.3 V to capture the onset and transition of water condensation. The wet neutron images were processed with the baseline dry images to obtain liquid water data. The final false-colored liquid water distribution images together with the quantified saturation for the Toray TGP-H-060 and the Freudenberg H23C8 are shown in Figs. 6 and 7, respectively. The water saturation level is calculated based on the measured liquid water thickness and the porosity of 63% and 47% for the Toray TGP-H-060 and the Freudenberg H23C8, respectively [75]. There are a total of 15 parallel channels in the cell and the land/channel water saturation results were obtained by averaging the liquid water in the GDL under the middle 3 lands and channels to avoid any edge effect. Since the channels are coated with gold, which is hydrophilic, liquid water can be observed in the channel. Nevertheless, the liquid water in the channel
was observed to have minimal effect on water distribution in the GDL due to high channel flow rate and hydrophobic GDL.

From Fig. 6, we can clearly observe the progression of increasing water condensation in the cathode DM from 2%, to 16% of oxygen dry mole fractions for the Toray TGP-H-060. At a 2% oxygen concentration, the cathode GDL is entirely dry and only some water molecules bonded to the sulfuric acid groups in the membrane can be observed. Please note that the lines located at the GDL and channel/land interface of the false-colored image are caused by the slight misalignment during the image processing steps and are considered artifacts. When increasing the oxygen concentration to 4%, significant water condensation can be observed in the cathode GDL both under the channels and the lands, which signifies onset condensation. Because the channel is the source of the oxygen flow with 77% RH, the GDL saturation directly under the channel is lower than that under the land. Throughout the GDL thickness, high liquid water saturation can be observed near the MEA and decreases toward the channel/land. Since the oxygen reduction reaction happens at the cathode catalyst layer in the MEA, the liquid water in the cathode GDL directly impedes all oxygen transport paths from the channel to the catalyst layer. This observation explains the sharp increase in oxygen transport resistance after the onset of water condensation of the Toray GDL in Fig. 3(a). When increasing the oxygen concentration to 8%, similar liquid water distribution in cathode GDL can be observed and the saturation level in the GDL reaches 40%.
This liquid water profile in the cathode GDL remains the same when the oxygen concentration is further increased to 16%, indicating that the wet plateau of oxygen transport resistance shown in Fig. 3(a) is reached. The liquid water distribution and saturation in the Toray GDL agree well with the in-situ measurement of oxygen transport resistance.

Similar to Toray, the progression of increasing liquid water condensation in the cathode DM from 2% to 16% of oxygen dry mole fractions can also be observed for the Freudenberg H23C8 as shown in Fig. 7. The same artifact between the GDL and channel/land interface can be clearly observed. At 2% oxygen concentration, the cathode GDL is also completely dry. When increasing the oxygen concentration to 4%, liquid water can be observed under the land with reducing saturation toward the MEA. On the other hand, the GDL under the channel is almost completely dry and, therefore, remains open for oxygen diffusion. When further increasing the oxygen concentration to 8%, the gap of the liquid water distribution between the land and the channel is further widened. The GDL under the land reaches a maximum saturation of 45%, while the GDL under the channel is still relatively dry. When the oxygen concentration is further increased to 16% the water starts to condense in the GDL directly under the channel. Nevertheless, the GDL under the channel still remains dry, which allows for oxygen transport. The observations from water condensation agree with the gradual increase of wet transport resistance after the onset condensation observed in Fig. 3(b).

### 3.4. Coupled diffusion and thermal transfer analyses

To further investigate the water condensation behavior in the GDL, a 1-D, steady state, non-isothermal fuel cell model has been developed to simulate PEMFC performance [58]. The model includes reaction kinetics, proton transport loss in electrodes, gas diffusion in porous media, ohmic loss, water transport across membrane, and coupled heat and mass transport. 1-D steady-state heat transfer and mass conservation equations are solved to obtain the temperature, \( T \) (K), and water vapor partial pressure, \( P_{H_2O} \) (Pa), distribution inside the GDL. At high current density operation, a thermal gradient of more than 5 °C can be observed in PEMFCs and the thermal conductivity of the GDL is the critical parameter which governs the temperature distribution inside the cell [58]. The model results provide a better understanding of how thermal and diffusion properties impact the relative humidity distribution in the GDL. The governing equations for heat and water vapor transport are:

\[
\frac{\partial}{\partial x} \left( \varepsilon \frac{\partial T}{\partial x} \right) = 0 \tag{6}
\]

\[
\frac{\partial}{\partial x} \left( \frac{D_{eff}}{\tau} \frac{\partial P_{H_2O}}{\partial x} \right) = 0 \tag{7}
\]

where \( x \) (cm) is in the through-plane direction, \( k \) (W cm\(^{-1}\) K\(^{-1}\)) is the through-plane thermal conductivity of the GDL, \( D_{eff} \) (cm\(^2\) s\(^{-1}\)) is the through-plane effective diffusivity of water vapor, and \( R \) is the universal gas constant. The through-plane water vapor effective diffusivity can be obtained by:

\[
D_{eff} = \frac{D_{H_2O}}{(\varepsilon/\tau)} \tag{8}
\]

where \( D_{H_2O} \) (cm\(^2\) s\(^{-1}\)) is the diffusivity of water vapor in open media, \( \varepsilon \) is the porosity and \( \tau \) is the tortuosity of the media. Alternatively, the ratio of porosity to tortuosity, \( \varepsilon/\tau \), can also be obtained experimentally by the limiting current tests [10]. To solve the heat and water transport equations, the following boundary conditions are applied at the CL-GDL interface:

\[
k \frac{\partial T}{\partial x} = Q' \tag{9}
\]

\[
\frac{\partial F P_{H_2O}}{R \partial x} = \frac{1 - f_{w}}{\varepsilon} \frac{j}{2F} \tag{10}
\]

where \( (1 - f_{w}) \) represents the fraction of generated water transported from the catalyst layer toward the cathode channel and \( Q' \) (W cm\(^{-1}\)) is the heat conducted from the catalyst layer toward the cathode land. Both parameters are solved based on the membrane water balance equation and thermal resistance network in the 1-D steady-state model and the detail solution algorithm can be found in our previous work [58]. To study the water condensation behavior, the 1-D steady-state simulation with the Toray and Freudenberg material properties is performed at a current density of 1.50 A cm\(^{-2}\) under 300 kPa, 70 °C and 90% RH. The ratios of the tortuosity over porosity are obtained from the limiting current measurement discussed in Section 3.1. The through-plane thermal conductivity of the two materials can be found in the literature [61,62]. Both the thermal conductivity and gas diffusivity are material properties as listed in Table 2.

To further investigate the water condensation behavior, the 1-D steady-state simulation with the Toray TGP-H-060 and Freudenberg H23C8. The critical transport parameter is presented in Fig. 3(a). Table 2 shows the critical transport parameter for both the Toray TGP-H-060 and the Freudenberg H23C8. When the value of \( k \left( \frac{j}{Q'} \right) \) is high, such as for the Toray material, liquid condensation first occurs in the GDL near the MPL interface, like the Toray material, or the channel/land interface, like the Freudenberg material, as shown in Fig. 8(b). Even though the 1-D steady-state model only deals with vapor transport, the RH distribution in the GDL provides key insights into the onset water condensation location. The simulation results agree well with the liquid water distribution observed from the neutron images in Figs. 6 and 7.

In summary, the location of onset condensation in the cathode GDL is governed by the complex interaction of the coupled heat and gas transport. The identified critical transport parameter is \( k \left( \frac{j}{Q'} \right) \). Table 2 shows the critical transport parameter for both the Toray TGP-H-060 and the Freudenberg H23C8. When the value of \( k \left( \frac{j}{Q'} \right) \) is high, such as for the Toray material, liquid condensation first occurs in the GDL near the MPL interface and directly blocks the oxygen paths. Therefore, liquid water in Toray materials causes a significant increase in oxygen transport resistance, as shown in Fig. 3(a). On the other hand, with a small \( k \left( \frac{j}{Q'} \right) \) value, such as observed for the Freudenberg material, liquid condensation first occurs in the GDL near the channel/land interface. Due to high convective flux and low heat transfer at the GDL and channel interface, water is less likely to condense. As a result, water condensation only occurs in the GDL under the land as shown in Fig. 7. This also explains why liquid water in the Freudenberg GDL only induces a gradual increase of oxygen transport resistance as shown in Fig. 3(b). Based on our simulation results, the transition of the onset condensation location exists when the critical transport parameter, \( k \left( \frac{j}{Q'} \right) \), is at around 0.81 W m\(^{-1}\) K\(^{-1}\) as shown in Fig. 8(b) at 70 °C and 300 kPa. Further, this transition of the critical transport
Fig. 7. Neutron radiograph profiles of water saturation and false colored neutron images for the Freudenberg H23C8 captured during wet limiting current experiments at 50 °C; 77% RH; 0.3 V; 300 kPa abs (a) 2%, (b) 4%, (c) 8%, and (d) 16% oxygen dry mole fractions.

Fig. 8. Simulation results of (a) Water vapor and saturation pressure and (b) Relative humidity distribution in Toray TGP-H-060 and Freudenberg H23C8 at 1.50 A cm⁻², 300 kPa, 70 °C and 90% RH.
parameter increases with (1) cell operating temperature due to the increasing gradient of water saturation pressure and (2) decreasing total operating pressure because of the increase in diffusivity. Even though the transition of the critical transport parameter varies with the operating conditions, the same guiding principle applies for optimizing GDL material design to reach high power density operation.

4. Conclusions

A comprehensive experimental and analytical study was conducted to investigate the effect of diffusion media on the performance of a PEMFC, especially at high current density. Two of the most widely used commercial gas diffusion materials, the Toray TGP-H-060 and Freudenberg H23CB, were selected for this study due to their difference in thermal properties. The through-plane thermal conductivities of Toray material is one order of magnitude higher than that of the Freudenberg material. Limiting current experiments were conducted to determine oxygen transport resistance under both dry and wet operating conditions. The Toray material shows a sharp increase in oxygen transport resistance after the onset of water condensation, while Freudenberg material exhibits a gradual increase. The polarization curves under dry and wet conditions agreed with the effect of thermal and oxygen transport properties. Toray performs better under dry conditions, but worse under wet conditions due to high thermal conductivity and low gas diffusivity. To further study the effect of water condensation, in situ neutron radiography experiments were performed. Neutron images showed that a maximum of 40–45% water saturation can be achieved in both the Toray and Freudenberg GDLs. In addition, water first condenses in Toray GDL near MPL and then propagates throughout the GDL material design to reach high power density operation. A comprehensive experimental and analytical study was conducted to investigate the effect of diffusion media on the performance of a PEMFC, especially at high current density. Two of the most widely used commercial gas diffusion materials, the Toray TGP-H-060 and Freudenberg H23CB, were selected for this study due to their difference in thermal properties. The through-plane thermal conductivities of Toray material is one order of magnitude higher than that of the Freudenberg material. Limiting current experiments were conducted to determine oxygen transport resistance under both dry and wet operating conditions. The Toray material shows a sharp increase in oxygen transport resistance after the onset of water condensation, while Freudenberg material exhibits a gradual increase. The polarization curves under dry and wet conditions agreed with the effect of thermal and oxygen transport properties. Toray performs better under dry conditions, but worse under wet conditions due to high thermal conductivity and low gas diffusivity. To further study the effect of water condensation, in situ neutron radiography experiments were performed. Neutron images showed that a maximum of 40–45% water saturation can be achieved in both the Toray and Freudenberg GDLs. In addition, water first condenses in Toray GDL near MPL and then propagates throughout the thickness. In contrast, water in the Freudenberg GDL first condenses in the GDL under the land and extends toward the MPL and leaves the GDL under the channel open for oxygen transport. The observation of the liquid water accumulation in the GDL explains the distinct trends of wet oxygen transport properties. Lastly, a 1-D steady-state model is applied to develop a fundamental understanding of the physics governing the water condensation location. A critical controlling parameter, \( k \left( \frac{\tau}{\varepsilon} \right) \), is obtained from the coupled heat and water vapor transport equations. A gas diffusion material with high \( k \left( \frac{\tau}{\varepsilon} \right) \) performs well under hot and dry conditions, but suffers from drastic oxygen transport loss as soon as liquid water condensation occurs in the GDL. On the other hand, a gas diffusion material with low \( k \left( \frac{\tau}{\varepsilon} \right) \) is more suitable for cold and wet operating conditions. Therefore, there exists an optimal material design that yields an intermediate \( k \left( \frac{\tau}{\varepsilon} \right) \) value, which allows the material to have robust performance across a broad range of operating conditions. The findings of this study provide clear guidelines for designing PEMFC materials to significant boost fuel cell energy conversion efficiency for future application. Note: Certain trade names and company products are mentioned in the text or identified in an illustration in order to adequately specify the experimental procedure and equipment used. In no case does such identification imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the products are necessarily the best available for the purpose.

CRediT authorship contribution statement


Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

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