Sources of Current Density Distribution in the Land-Channel Direction of a PEMFC
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Proton exchange membrane fuel cell (PEMFC) is an electrochemical reactor with three geometrical directions: flow channel (length scale 1 to 100 cm), land-channel (length scale 100 to 1000 μm), and through-plane (length scale 10 to 100 μm) directions. Because of the geometry of a PEMFC the distribution of reactants and products in these directions are highly inhomogeneous, as a result making current generation non-uniform. In order to study the local non-uniformities in the flow-direction, segmentation of a PEMFC to many differential cells is widely used. The differential cell means a fuel cell with small enough active area so that various conditions within the cell can be considered uniform along the flow channel direction. On the other hand, because of its relatively smaller length scale, variations in the land-channel direction are often neglected in most of such studies. However, due to the non-uniform transport length distribution from channel to catalyst layer in the land-channel direction, the differential cells aligned in the flow direction give only the averaged values over each differential cell, and therefore the local information in the land-channel direction cannot be captured.

In a wet condition, for example, due to the land-channel geometry liquid water tends to distribute non-uniformly in gas diffusion layer (GDL). Many modeling studies present in literature have predicted liquid water distribution in land-channel direction. One example of such study is numerical investigation of liquid water saturation distribution in the land-channel direction of a flow field with 1 mm wide land and channel presented by Wang et al. According to their findings, liquid water saturation at 80 °C, 100% RH, and 2.5 A/cm² was five times higher under the land than in the channel area. Recently, in-situ imaging techniques have been widely used to explore liquid water distribution in through-plane and in land-channel direction of an operating PEMFC. Eller et al. applied X-Ray Tomography imaging technique to visualize water in the land-channel direction of a PEMFC operating in a wet condition and found heavy liquid water accumulation under the land. Other studies available in literature have confirmed that the current density distribution in the land-channel direction cannot be captured.

In dry conditions, on the other hand, ohmic resistance is a major contributor in voltage loss at a given current density. Ohmic resistance comprises of two components – electronic and protonic resistances. In a dry condition, the amount of liquid water in the open pores can be considered negligibly small, and therefore the oxygen transport resistance is not affected by the presence of liquid water. However, the proton transport resistance through the electrolyte membrane and the ionomer film in the catalyst layer is a strong function of the local water content. Hence local current generation is heavily influenced by the variation in membrane water content along the land-channel direction. The distribution of protonic resistance in the land-channel direction is not well documented in literature. However, a few experimental and modeling studies have shown a major variation in electrical resistance distribution induced by the land-channel geometry. An electrical resistance distribution study conducted by Butsch et al. on four types of GDLs has demonstrated a significantly uneven pressure distribution which translates to non-uniform electrical resistance distribution. Their results have shown that for one of the GDL samples the electrical resistance in channel areas can be as high as 160 mΩ-cm² whereas in land areas below 20 mΩ-cm².

Also, non-uniform compression and resulting thermal transport length distribution lead to uneven distribution of thermal resistances. Konduru et al. incorporated solid phase distribution in their pore network model to simulate the temperature distribution at 70 °C (fuel cell temperature) in the through-plane and land-channel directions for TGP-H-90 Toray diffusion medium. They estimated that the temperature of the GDL-catalyst layer interface varied from 73 °C at the center of channel to 70 °C at the extremity of land. This difference in local temperature is high enough to alter local water saturation as the electrochemical kinetics and the saturation vapor pressure are strong function of the temperature.

In aforementioned operations, cumulative effect of irregularities in liquid water saturation, membrane water content, and electrical and thermal resistances brought by the land-channel geometry can have appreciable effect on the current density distribution, and vice versa. An ample amount of studies based on theoretical and computational models present in literature to study the impact of land-channel geometry on current density distribution. However, there are very few experimental studies reported on current density distribution in the land-channel direction. Owing to small length scale of land-channel direction (typically 1 mm or less for a land and a channel each), segmentation becomes a very challenging task as explained in our previous work. Schneider et al. and our previous work have confirmed that the current density distribution in the land-channel direction can be immensely influenced by the humidity conditions. However sources of such inhomogeneities are yet to be quantified.

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This work describes a method to discern the sources of current density distribution in land-channel direction. First an ex-situ electrical resistance distribution experiment was carried out to measure electrical resistance distribution in the land-channel direction. Then, a customized in-house catalyst coated membrane (CCM) was prepared, and a cell with segmented anode was used to map current density and high frequency resistance (HFR) distributions in the land-channel direction (1 mm-wide land and 1 mm-wide channel) of cathode flow field at 350 μm resolution in various conditions. This configuration was also applied to measure distribution in electrochemical surface area (ECSA). Methodology to identify the sources of current density distribution in wet condition (90% RH) explained in our previous work27 is extended to dry (0% RH) and moderate (60% RH) conditions to estimate oxygen concentration, local transport resistance, electrode resistance, and membrane water content distribution in the land-channel direction.

**Experimental**

**Ex-situ electrical resistance distribution measurement.**—A schematic of test fixture to measure electrical resistance distribution in the land-channel direction is illustrated in Figure 1 where segments 1–3 and 7–9 are under the land, and segments 4–6 are under channel. A piece of carbon paper (TGP-H-90, Toray Inc.) of dimension 5 mm × 5 mm was sandwiched between a cathode graphite bipolar plate with 1 mm-wide channel and a printed circuit board (PCB) with 200 μm-wide copper segments with 150 μm gaps. First, an HFR meter with 1 kHz applied AC (Hioki Battery Hi-tester model 3561-01, Hioki, Japan) was connected between individual segments and graphite bipolar plate to measure the contribution of cathode side assembly in total electrical resistance distribution. Similarly, in a separate experiment, a copper foil of 50 μm thickness coated both sides with carbon layer containing ionomer (to imitate the catalyst layer) was sandwiched between TGP-H-90 carbon paper and PCB to measure the contribution of segmented anode side assembly to the total electrical resistance. In the latter case the electrical resistance was determined between copper foil and each individual segment with the abovementioned instrument. The total of anode and cathode local electrical resistances at each segment was calculated from the measured data.

**Preparation of catalyst coated membrane (CCM).**—Pt catalyst on carbon (40% Pt/Vulcan C, Fuel Cell Earth) and Nafion ionomer solution (D2020, Ionpower) were used to prepare catalyst ink. Ionomer/carbon weight ratio of 0.8 was selected for ink preparation.28 To obtain catalyst ink slurry isopropyl alcohol and de-ionized water were added to ionomer solution and catalyst mixture to maintain 0.18 solid to liquid ratio. This catalyst ink was stirred using magnetic stirrer for two days and ultrasonicated for 30 minutes before coating. Then catalyst ink was coated using knife coater on a flat ethylene tetrafluoroethylene (ETFE) sheet and dried for a day to obtain an 18 μm-thick catalyst layer with 0.4 mg/cm² Pt loading. Method explained by Zhao et al. was applied to prepare CCM.29 Windows of 3 mm × 3 mm and 4 mm × 4 mm were created on cathode and anode Kapton sub-gaskets, respectively, so that transferred active area of 9 mm² on cathode side and 16 mm² on anode side can be obtained. A 50 μm thick Nafion membrane (NRE-212) was sandwiched between Kapton sub-gaskets to achieve sub-assembly and catalyst was transferred by hot pressing catalyst coated ETFE decals to sub-assembly at 150 °C and 2.7 MPa for three minutes.

**Segmented cell assembly.**—An image of segmented fuel cell assembly is displayed in Figure 2. This study uses the same experimental set-up that was described in our previous work.30 On cathode side, a 1 mm-wide channel was milled on a graphite plate to depict 1 mm wide land-channel configuration. A carbon paper (10% wet proof TGP-H-90, Toray Inc.) with 5 mm × 5 mm area is used as GDL, and 225 μm-thick VPTFE gaskets were chosen as compensating layers to obtain 20% compression of GDL. On anode side, nine 200 μm-wide current collecting segments and 150 μm-wide flow channels for gas flow were milled on a PCB (copper clad on FR-4), and then gold was electroplated on entire PCB to obtain corrosion resistance surfaces. The fabricated PCB acts as a hydrogen distributor and segmented current collector. The custom CCM was sandwiched between cathode assembly and segmented anode assembly by applying a compression pressure of 2 MPa. Then segments from anode current collector were attached to a shunt resistor module that consists of nine shunt resistance of 200 mΩ for local current measurement.

Mass flow rate, temperature, pressure, and humidity of gases were controlled by fuel cell test station (Fuel Cell Technologies). An electrochemical instrument (Vertex, Ivium Technologies) was used to perform cyclic voltammetry, and to control cell overall potential in fuel cell mode. A digital multimeter (Keithley Instruments, DMMA-2700, 100 nV resolution) was employed to measure AC and DC voltage drop across the shunt resistors. In order to measure local and overall high frequency resistances (HFRs) a custom current source was developed so that the amplitude and the frequency of AC current applied to the entire cell can be controlled. All the test conditions are available in Tables I and II.
Analysis of Segment-to-Segment Current Density Distribution

As explained in earlier section, the non-uniform transport distance from channel to catalyst layer can significantly impact the species distribution in the land-channel direction. Therefore it is important to analyze the effect of non-uniform transport distance on each species separately.

The local cell potential at each segment is expressed in terms of individual losses by the equation given below

\[ E_{cell}^n = E_{eq}^n - \eta_{ORR}^n - \eta_{HOR}^n - \eta_{ohm}^n - \eta_{crossover}^n \]  

where superscript \( n \) specifies \( n^{th} \) segment, \( E_{cell} \) is cell potential, \( E_{eq}^n \) is thermodynamic equilibrium potential, \( \eta_{ORR} \) is oxygen reduction reaction overpotential, \( \eta_{HOR} \) is hydrogen oxidation reaction overpotential, \( \eta_{ohm} \) is ohmic overpotential, and \( \eta_{crossover} \) is potential due to hydrogen crossover. Here the concentration overpotential is included in \( \eta_{ORR} \) for cathode and in \( \eta_{HOR} \) for anode, since the decreased reactant concentration ultimately affects the activation overpotential. Assuming the hydrogen oxidation reaction overpotential is negligible, Equation 1 can be reduced to following equation.

\[ \eta_{ORR}^n = E_{eq}^n - \eta_{crossover}^n - E_{cell}^n - \eta_{ohm}^n \]  

where \( E_{cell}^n \) is open circuit voltage including overpotential due to hydrogen crossover. Ohmic losses to electronic and protonic resistances can be expressed as follows

\[ \eta_{ohm}^n = i^n \left( R_{el}^n + R_{H_2,mem}^n + R_{eff,ca}^n \right) \]  

where \( R_{el} \) is electronic resistance including bipolar plate, GDL, catalyst layer, and segmented anode, \( R_{H_2,mem} \) is protonic resistance through membrane, and \( R_{eff,ca} \) is effective cathode electrode protonic resistance that is the ratio of ohmic voltage loss across the catalyst layer to the current density. Local electronic resistance was measured by employing method explained in Ex-situ electrical resistance distribution measurement section whereas local HFR was measured using technique described in our previous work.\(^{30}\)

Oxygen concentration distribution in wet and moderate conditions.—To evaluate oxygen concentration distribution, ohmic losses in membrane and in catalyst layer, and charge transfer resistance in catalyst need to be estimated because these resistances have significant effect on the overall cell overpotential. Neyerlin et al.\(^ {32} \) derived an expression (see Equation 4) to compute local effective protonic resistance \( R_{eff,ca}^n \) of cathode catalyst layer

\[ R_{eff,ca}^n = \frac{R_{H_2,ca}^n}{\theta^n + \xi^n} \]  

where catalyst layer proton resistance \( R_{H_2,ca}^n \) can be calculated from equation below

\[ R_{H_2,ca}^n = \frac{t_l}{t_{mem} (\eta_{ohm}^n)} \]  

where \( t_l \) (\( = 18 \) \( \mu m \)) is catalyst layer thickness, \( t_{mem} \) (\( = 50 \) \( \mu m \)) is membrane thickness, \( \xi (\sim 0.113 \) from the design of electrode) is ionomer volume fraction in catalyst layer and \( \eta_{ohm} \) is tortuosity. The catalyst layer proton transport resistance \( R_{H_2,ca} \) is the through-plane protonic resistance in CL all the way from membrane-CL interface to CL-GDL interface, while the effective protonic resistance \( R_{eff,ca} \) accounts for the through-plane location of reaction area in CL that is a function of operating conditions. Following set of equations is used to calculate correction factor \( \xi\).\(^ {32} \)

\[ \xi^n = \frac{2\Theta^n \tan (\Theta^n)}{\ln (\frac{\Theta^n + \tan (\Theta^n)}{\tan (\Theta^n)})} \]  

where \( \Theta \) is a dimensionless number and described by following equation

\[ \Theta^n = \tan (\Theta^n) = \ln (10) \left( \frac{i^n R_{H_2,ca}^n}{b} \right) \]  

where \( b \) is Tafel slope (\( = 78 \) mV/decade at \( \alpha = 0.8 \))

Neyerlin et al.\(^ {32} \) used 60% and 100% RH humidity conditions to validate these equations. However one needs to be careful in using Equation 5 because this equation includes an assumption that proton conductivity of ionomer in catalyst layer and membrane is the same that may not be always the case. At 60 °C and relative humidity below 95% RH, Paul et al.\(^ {31} \) measured conductivity of ionomer film of 4 nm thickness (typical ionomer film thickness in catalyst layer) and found that the conductivity much less than bulk membrane. In our work the cell is operated in the similar conditions as that of Neyerlin et al.\(^ {32} \) so that the equations are useful to determine local oxygen reduction overpotential. It is further related to oxygen concentration \( C_{CA,Hel} \) available at catalyst layer at a given current density through Tafel equation described as follows

\[ i^n = u^n C^n_{CA,Hel} \frac{C_O2^{max}}{C_O2} \exp \left( \frac{\eta_{hor}}{\eta_b} \right) \]  

where \( i_b \) (A/cm²) is the exchange current density, and in this work the current density generated at 0.9 V is used as exchange current density. In Equation 8 another parameter used is the catalyst utilization factor \( u \) which accounts for the loss in the active area of catalyst layer due to the proton and the charge transport resistance and is defined in terms of \( \Theta \) in equation below.\(^ {32} \)

\[ u^n = \frac{\tan (\Theta^n)}{\Theta^n (1 + \tan^2 (\Theta^n))} \]  

Moreover, computed oxygen concentration distribution can be utilized to evaluate the local oxygen transport resistance in the land-channel direction at a given overall current density which is a useful parameter in determining the effect of liquid water distribution in the land-channel direction. Equation 10 below describes the local oxygen transport resistance \( R_{O2} \)

\[ R_{O2} = \frac{4F(C^n_{O2} - C^n_{O2,el})}{i^n} \]  

where \( C^n_{O2} \) is the oxygen concentration in flow channel.
Electrode resistance distribution in dry condition in H2/O2 cell.—

In extremely dry condition, water vapor is rapidly carried away from the catalyst layer and this causes severe dehydration of ionomer in catalyst layer. Therefore the estimation of cathode electrode effective protonic resistance is essential to analyze the performance loss.

When a fuel cell is operated with H2/O2 and at 1 A/cm² overall current density, the effect of non-uniform distribution and ORR over-potential due to mass transport can be neglected. This is because (1) the fuel cell is not operated in mass transport limited region, (2) high concentration of oxygen available at channel, and (3) the diffusivity of oxygen in the land-channel direction is approximately two times greater than in through-plane direction. Equations 4 to 7 were used at this condition to estimate cathode electrode protonic and effective resistance.

Water distribution in membrane.—In a later subsection, it is shown that the membrane resistance is non-uniform in dry and moderately wet conditions. The local membrane water content is an indicator of humidity, and therefore the distribution of water content in membrane is important to understand the sources of current density distribution and water transport from catalyst layer to the channel.

As explained in a section earlier, HFR across each segment is measured to resolve local ohmic resistance. Electronic resistance of each segment is subtracted from local HFR to assess the local protonic resistance, assuming the protonic resistance in the electrode is not counted in HFR.

$$R_{H^+ \text{mem}}^n = HFR^n - R_e^n$$

$$\sigma_{H^+ \text{mem}}^n = \frac{R_{H^+ \text{mem}}^n}{\text{mem}}$$

where $\sigma_{H^+ \text{mem}}$ denotes conductivity of membrane that is a result of average local water content $\lambda$ and temperature $T$ averaged over the membrane thickness. An expression derived by Springer et al. is rearranged to calculate local membrane water content.

$$\lambda = \frac{\sigma_{H^+ \text{mem}} \exp[-1268(\frac{303}{T} - \frac{1}{T}}] + 0.00326}{0.00514}$$

Correlation between the membrane water content and the water vapor activity $a$ surrounding the membrane at equilibrium is also taken from Springer et al.

$$\lambda = 0.043 + 17.81 a - 39.85 a^2 - 36.0 a^3$$

Results and Discussion

Electrical resistance distribution.—Figure 3 displays the electrical resistance distribution measured using the ex-situ experimental setup explained in Ex-situ electrical resistance distribution measurement section. The electrical resistance under the land is relatively uniform and varies between 8 mΩ·cm² to 10 mΩ·cm² whereas under the channel electrical resistance is non-uniform and ranges from 12 mΩ·cm² near the edges of channel to 16 mΩ·cm² at the center of the channel. The distribution of electrical resistance is a clear indicator of non-uniform compression pressure distribution where pressure is high under the land and low under the channel. Result presented in this work is qualitatively similar to the one presented by Butsch et al. Such electrical resistance can cause non-uniform ohmic overpotential distribution. For example at uniform current generation of 2 A/cm² the ohmic loss can vary from 16 mV at the edge of land to 32 mV at the center of channel.

Electrochemical surface area distribution.—Overall electrochemical surface area (ECSA) was measured at 40°C and 76% RH. The mass flow rates of hydrogen and nitrogen were 100 scm, and scan rate of 200 mV/sec was applied. The overall ECSA is computed to be 40 m²/gpt with roughness factor of 158 cm²pt/cm²geometrical-area.

Figure 4 exhibits the distribution of roughness factor in the land-channel direction which was calculated for the each segment and found within ±15% of overall roughness factor. In the channel areas roughness factor is lower than the land areas and study presented by Schneider et al. corroborates this phenomenon. High gas flow rates can swiftly remove the evolved molecular hydrogen at cathode during the cyclic voltammetry and influence the shape of hydrogen desorption/adsorption (HAD) curve. Hence for segments 4–6 the calculated HAD area may be lower than the actual HAD area for these segments.

Current density and HFR distribution.—Operation with H2 and air.—Figure 5 represents the overall performance of the cell at three different gas RH conditions as described in Table 1. In ohmic region, at the voltages roughly from 0.8 V to 0.5 V, performance of the cell declines with the decreasing humidity (Figure 5a) due to the higher ohmic resistance of the cell as displayed in Figure 5b. At 0% RH (dry), HFR reduces from 400 mΩ·cm² at overall current density lower than 0.1 A/cm² to 160 mΩ·cm² at 1 A/cm² while at 60% RH (moderate), HFR varies from 200 mΩ·cm² at overall current density below 0.2 A/cm² to 88 mΩ·cm² at limiting current of 1.76 A/cm². However in 90% RH (wet) the overall HFR is lower than the both conditions and variation is very minute compared to other conditions.
In both dry and wet conditions limiting current density is approximately 1 A/cm² whereas in moderate condition limiting current density is 1.76 A/cm². In dry condition (0%RH), proton transport resistances in membrane and in catalyst layer are the limiting factor, and both membrane and catalyst layer dehydration causes elevation in protonic resistance. The cell has to rely on water generation due to electrochemical reaction to hydrate the membrane. In wet condition (90%RH), at 1 A/cm² severe flooding within porous media started to occur and inhibited oxygen transport to the catalyst, and concentration overpotential increases significantly.

In moderate condition (60%RH), both proton and oxygen transport resistances are important to voltage loss at 1 A/cm². With increase in current density the hydration of membrane and of catalyst layer ionomer increases that alleviates the ohmic resistance. At the lower voltages, the higher current generation and water production rate decrease the concentration of oxygen at the catalyst layer. Due to the better water balance (explained later in this section) in moderate condition than in dry or wet condition, the limiting current is much higher.

Figures 6 and 7 exhibit the current density and HFR distribution in the land-channel direction, respectively, corresponding to the overall performance shown in Figure 5. The profile of current density distribution produced in this work is similar to the ones demonstrated in Reference 27 at similar conditions. In wet condition (Figure 6a) current generation in channel area is much higher than in land area at cell voltages lower than 0.6 V, corresponding to overall current density higher than the 0.8 A/cm². At 1 A/cm² and 0.5 V the local current density peaks around 1.4 A/cm² at the center of the channel and at the extremity of the land the local current generation is approximately as low as 0.7 A/cm². However at voltages lower than 0.5 V the current generation under the channel drops and becomes smaller than current generated at 0.5 V, indicating severe flooding in channel area. Since the local current generated in channel area is a major contributor to the overall current density, flooding in channel area degrades the overall performance of the cell (Figure 5a, 90% RH). At all the cell voltages and overall current density the HFR distribution is within 5 mΩ·cm² variation from overall HFR (Figure 7a). HFR distribution demonstrates that the membrane is well hydrated, and the key reason of current density distribution is the non-uniform liquid water saturation in the land-channel direction as revealed by the imaging studies.10–13

In dry condition (see Figure 6b), at all the cell potentials the current density generated in channel area is much lower than the land area. At overall current density of 1.1 A/cm² and voltage of 0.1 V the current density at the center of channel is 0.6 A/cm² and at the extremity of land is 1.4 A/cm². At the same cell voltage, the current density distribution trend in dry condition is opposite to the trend observed in wet condition i.e. current density is lower in channel areas than in land areas. In dry condition the electrolyte dehydration in membrane and catalyst layer dominates the voltage loss thus the local current generation becomes function of the local ohmic resistance which in turn is affected by the presence of water. It is clear from Figures 6b and 7b that a strong inverse relation exists between local HFR and current density. For example, at an overall current density of 0.8 A/cm² and cell voltage 0.2 V, the current density and HFR at segment 5 (at center of the channel) is 0.5 A/cm² and 300 mΩ·cm² whereas for segment 1 (at extremity of the land) the current density and HFR is 1.4 A/cm² and 100 mΩ·cm². Also, local HFR of segments in channel region notably affect the overall HFR. As overall and local current density increases water content in membrane increases as well, and local HFR in channel region and overall HFR decreases. Moreover, with increasing current density, HFR distribution becomes less non-uniform and overall HFR does not vary much between 0.2 V and 0.1 V. Along with the dehydration of membrane the ionomer dehydration in catalyst layer also accounts for the significant ohmic voltage loss because proton conductivity of few nanometer thick ionomer film is much lower than the conductivity of bulk membrane, especially in dry condition.33

In Figure 6c, at 60% RH the local current generation is uniform for all the segments at cell voltages above 0.4 V, corresponding to overall current density lower than 1.3 A/cm². However there is variation in HFR to a certain extent. For example at overall current density of 1 A/cm² and 0.5 V (condition same as wet condition), current generation is uniform within 10% difference from overall current generation, while HFR at center of channel is 110 mΩ·cm² and at the edge of land is 87 mΩ·cm² (see Figure 7c). The distributions of current density and HFR indicate that the both oxygen and proton transport are playing critical role in cell voltage loss. As cell voltage decreases below 0.5 V the overall current density increases and the local current density tends to distribute non-uniformly, and at limiting current density of 1.76 A/cm² the current density distribution profile is similar to the wet condition. The local current density produced by segments in and near the channel region (segments 3–7) is approximately 2.2 A/cm² whereas segments close to and the extremity of land (segments 1, 2, 8, and 9) is approximately 1.5 A/cm². However, unlike wet condition, HFR distribution is also non-uniform along with the non-uniform current density. From the channel area to the center of the land HFR is approximately 93 mΩ·cm² and at the extremity HFR is 70 mΩ·cm². The fact that the non-uniform distribution of current density and

Figure 5. Overall cell a) polarization curves and b) HFR at 60°C with dry, wet and moderate conditions.
lower HFR at each segment suggests that the condition in the cell has transitioned from moderate to wet condition. This transition is mainly associated with the production of liquid water at local level due to the high local current densities.

Operation with H₂ and O₂.—The performance of the cell with pure oxygen was tested in conditions shown in Table II. In both conditions at all the cell voltages, performance of cell was superior to H₂/air in same conditions (see Figure 8). On the contrary, trend of overall HFR with respect to current density showed behavior similar to the ones observed for H₂/air cell in same conditions. Overall HFR in wet condition remained almost constant while in dry condition overall HFR continuously decreases with increasing current density and eventually stabilized at 140 mΩ-cm².

In wet condition, the current density distribution (shown in Figure 9a) was found uniform at ~1 A/cm² and 0.6 V and a very slight variation in HFR (80 ± 5 mΩ-cm²) was observed (see Figure 10a). At similar overall current density the uniform current density distribution and higher voltage than air show that the abundant oxygen is present in the catalyst layer which is due to the higher concentration of oxygen at the channel, and that the transport distance is not a limiting factor in this case. However, as overall current density increases, local current density distribution becomes non-uniform at overall current density generated beyond 1 A/cm². This is probably because the liquid water starts to fill the pores of GDL and play a major role in appreciating the effect of transport distance on non-uniform oxygen concentration distribution. Even at higher overall current generation the variation in local current density is much less than that observed for current distribution in H₂/air cell.

In dry condition, as seen in Figure 9b, similar to H₂/air, current density generated in channel region is lower than the land region till overall current density of 1 A/cm² at 0.5 V. Again such distribution is related to the HFR distribution displayed in Figure 10b. Contrary to H₂/air cell, along with further reduction in voltage and increase in overall current density, HFR and current density distribution started to become more uniform, and eventually current generation in channel areas surpassed the current generated in areas close to extremity of land. Again excess oxygen concentration near catalyst allowed higher local current generation, and as a result enough water is generated to hydrate the membrane and ionomer in catalyst layer which aided in keeping uniform current density distribution. Segments 3 and 7 close to the edge of channel have produced highest current in wet condition. This may be due to the fine balance of water transport in this region where high concentration of oxygen is available to sustain high reaction rates and membrane is relatively well hydrated.

Sources of Current Density Distribution

In all the conditions the effect of land-channel geometry is evident. Therefore it is imperative to analyze the effect of land-channel geometry on distributions of oxygen concentration and water content in membrane because distribution of these two species plays key role in current density distribution. Using the methodology described in Analysis of segment-to-segment current density distribution section the distribution of oxygen concentration at catalyst layer/GDL interface, oxygen transport resistance distribution, cathode electrode protonic resistance, and water content in the membrane are quantified.
Figure 7. HFR distribution in the land-channel direction at 60°C with a) 90% RH, b) 0% RH, and c) 60% RH conditions.

Wet and moderate conditions.— Membrane water content in wet and moderate conditions.— Membrane water content ($\lambda$) was calculated from the conductivity using Equations 11 to 13 in Water distribution in membrane section. Figure 11 shows membrane water content in moderate and wet conditions when air is supplied to cathode side.

In both moderate and wet conditions membrane water content does not vary much in the land-channel direction. In wet condition because product water is mostly in liquid phase even at low current densities, the water content in membrane is nearly uniform (see Figure 11a). In moderate condition (Figure 11b), average $\lambda$ is 8.5 (value corresponds to 85% RH) from center of channel to the center of land.

Figure 8. Overall cell a) polarization curves and b) HFR with H$_2$/O$_2$ at 60°C in dry and wet conditions.
Figure 9. Distribution of a) current density and b) HFR with H₂/O₂ at 60°C and 90% RH.

Figure 10. Distribution of a) current density and b) HFR with H₂/O₂ at 60°C and 0% RH.

Figure 11. Membrane water content distribution with H₂/air at 60°C with a) 90% RH, and b) 60% RH.
which is slightly lower than land region ($\lambda = 9.6$, value corresponds to 89%RH). This minute variation stems from high local current densities which keeps the membrane hydrated. At limiting current, at the extremity of the land average $\lambda$ is 12 (value corresponds to 95%RH) again higher than other regions where average $\lambda$ is 9 (value corresponds to 87%RH). This confirms that the longer transport length for water removal from the extreme edge of land to the channel causes water accumulation under the land.

Oxygen concentration and transport resistance distribution.—To compute oxygen concentration distribution in the land-channel direction Equations 1 to 9 were used, and then calculated oxygen concentration was applied to Equations 10 to 13 for assessment of the local oxygen transport resistance. Comparison of estimated oxygen concentrations and oxygen transport resistances between moderate and wet conditions are displayed at 1 A/cm$^2$ and limiting current density in Figures 12 and 13, respectively. In both wet and moderate cases at similar overall current density of 1 A/cm$^2$, oxygen concentration under the channel is higher than under the land, because diffusion distance is shorter from channel to the catalyst layer directly than channel to the catalyst layer under the land. Consequently oxygen concentration is higher in channel region.

As pointed out in our previous work, non-uniform distribution of liquid water saturation due to the transport length is vital to the oxygen concentration distribution at electrode. To understand the effect of land-channel geometry on oxygen transport and to define the wet or dry state, the calculated oxygen transport resistance is compared with the ones estimated by Baker et al. (see Table III) in dry and wet states. Baker et al. used 0.5 mm wide land-channel configuration to estimate oxygen transport resistance, and according to them 50% of mass transport losses originates from 160 $\mu$m thick GDL (TGP-H-60) and rest from the channel, MPL, and catalyst layer. In the present work oxygen transport resistance is only affected by catalyst layer and 270 $\mu$m thick GDL (TGP-H-90). Measured ECSA is high enough that the impact of transport resistance in CL can be safely neglected and the calculated oxygen transport resistance represents the transport resistance in GDL only. Considering all these factors oxygen transport resistance evaluated by Baker et al. is a valid benchmark to define the state in GDL.

In the following discussion the oxygen concentration and oxygen transport resistance distribution are compared in moderate and wet conditions at two cases. The first case is at overall 1 A/cm$^2$ where the rate of oxygen diffusion is defined (Figure 12), and the second case is at limiting current density where the oxygen concentration is considered zero in the CL (Figure 13).

At 1 A/cm$^2$, the available oxygen concentrations in channel area in wet and moderate conditions have similar magnitude (6.5 mol/m$^3$)

![Figure 12. Distribution of a) oxygen concentration in cathode CL, and b) oxygen transport resistance at 60°C and 1 A/cm² in wet and moderate conditions.](image)

![Figure 13. Distribution of oxygen transport resistance at 60°C and at limiting current density in wet and moderate conditions.](image)

| Table III. Range of transport resistance estimated for TGP-H-60 at temperatures from 50°C to 90°C and pressures from 110 kPa to 300 kPa signifying the state of GDL. |
| Oxygen Diffusion Transport Resistance $R_{O_2}$ (S/cm) | State of GDL in terms of presence of water |
|------------------------------------------------------|------------------------------------------|
| $R_{O_2} \leq 1$                                      | Dry Region                               |
| $1 < R_{O_2} \leq 2$                                 | Transition Region                        |
| $R_{O_2} > 2$                                        | Wet Region                               |

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in wet condition and 7.7 mol/m\(^3\) in moderate condition) but under the land oxygen concentration in wet condition is much lower than the moderate condition (Figure 12a). The difference in the oxygen concentration distribution between wet and moderate condition is related to the local oxygen transport resistance. In wet condition the oxygen transport resistance in channel area is \(\sim 1\) s/cm (see Figure 12b), and comparable to the estimated oxygen transport resistance in moderate condition. This shows that the region under channel is either in dry state or at very beginning of transition zone. But in land areas the oxygen transport resistance increases from the land/channel edge (segment 7, for example) to the land edge (segment 9), and the oxygen transport resistance distribution is asymmetric which may be related to local pore geometry and pore distribution or simply the experimental variation. At the extremity of the land oxygen transport resistance is 4.7 s/cm for segment 1 and 5.4 s/cm for segment 9. Both of these values are higher than oxygen transport resistance determined in moderate condition. The higher oxygen transport resistance in land region must attribute to both transport length of oxygen diffusion and liquid water accumulation. However, without determining the local oxygen transport resistance in dry condition it is difficult to differentiate the effect of local saturation and the effect of transport length. As elucidated by imaging techniques more water accumulation occurs under the land region. The local saturation is a likely reason of high oxygen transport resistance under the land. In moderate condition estimated oxygen concentration distribution is more uniform than that in the wet condition, and has higher magnitude in land areas. However the oxygen concentration computed in moderate condition is not symmetric. For example, concentration at segment 9 is 2.6 mol/m\(^3\) which is lower than the concentration at segment 1 (3.7 mol/m\(^3\)) and this might be related to either pore size distribution or presence of liquid water due to the local condensation. Oxygen transport resistance increases from the center of the channel (0.97 s/cm at segment 5) to extremities of lands (2.4 s/cm at segment 1 and 3.6 s/cm at segment 9) (Figure 12b). The oxygen transport resistance indicates the dry state of GDL in channel region and transition of the state from dry to liquid phase in land region.

At limiting current densities which are 1 A/cm\(^2\) for wet case and 1.76 A/cm\(^2\) for moderate case, oxygen concentration was in the order of \(10^{-3}\) mol/m\(^3\) at every segment in both cases, and oxygen transport resistance was calculated assuming zero concentration at catalyst layer/ GDL interface. Figure 13 displays estimated oxygen transport resistance distribution in the land-channel direction at limiting current density of 1 A/cm\(^2\). In both conditions the water content in membrane increases from the center of the channel region to the edge of land. In H\(_2\)/air case \(\lambda\) is 3 (value corresponds to 37\%RH) at the channel center and 8 (value corresponds to 83\%RH) at the edge of the land (see Figure 14a), and in H\(_2\)/O\(_2\) case \(\lambda\) is 3.5 at the center of channel and 6 (value corresponds to 74\%RH) at the edge of the land (see Figure 14b). Because of the transport distance difference, the vapor produced under the channel area can be removed at faster rate to the channel than the vapor under the land. In addition, low porosity of GDL under the land due to the compression may decrease the diffusion rate of water vapor by reducing the effective diffusivity. Therefore more water tends to accumulate under the land. In contrast to H\(_2\)/air condition, in H\(_2\)/O\(_2\) the water distribution in the membrane becomes more uniform at higher current (Figure 14b). At 0.3 V and 1.5 A/cm\(^2\) overall current density, \(\lambda\) in channel region is 5.5 and in land region is 6, and at 0.1 V and 2.4 A/cm\(^2\) the membrane water content is almost constant from segments 2–8 and slightly higher at segments 1 and 9. These results clearly suggest that the rate of water production above certain current density such as 1.5 A/cm\(^2\) is high.

**Dry condition.**—In dry condition, as seen in Figure 14, the membrane water content in H\(_2\)/air and H\(_2\)/O\(_2\) is highly non-uniform at overall current density of 1 A/cm\(^2\). In both conditions the water content in membrane increases from the center of the channel region to the edge of land. In H\(_2\)/air case \(\lambda\) is 3 (value corresponds to 37\%RH) at the channel center and 8 (value corresponds to 83\%RH) at the edge of the land (see Figure 14a), and in H\(_2\)/O\(_2\) case \(\lambda\) is 3.5 at the center of channel and 6 (value corresponds to 74\%RH) at the edge of the land (see Figure 14b). Because of the transport distance difference, the vapor produced under the channel area can be removed at faster rate to the channel than the vapor under the land. In addition, low porosity of GDL under the land due to the compression may decrease the diffusion rate of water vapor by reducing the effective diffusivity. Therefore more water tends to accumulate under the land. In contrast to H\(_2\)/air condition, in H\(_2\)/O\(_2\) the water distribution in the membrane becomes more uniform at higher current (Figure 14b). At 0.3 V and 1.5 A/cm\(^2\) overall current density, \(\lambda\) in channel region is 5.5 and in land region is 6, and at 0.1 V and 2.4 A/cm\(^2\) the membrane water content is almost constant from segments 2–8 and slightly higher at segments 1 and 9. These results clearly suggest that the rate of water production above certain current density such as 1.5 A/cm\(^2\) is high.

**Figure 14.** Distribution of membrane water content with a) H\(_2\)/air, and with b) H\(_2\)/O\(_2\) at 0.1 V and 60°C.
enough to overcome rapid removal of water from the channel region and allows membrane to hydrate uniformly in land-channel direction. In dry condition ohmic losses are the major contribution to the overall losses in the cell, and ohmic overpotential due to cathode electrode protonic resistance has a major role in total ohmic losses. In the following discussion, therefore the proton transport resistance in cathode catalyst layer is evaluated in the land-channel direction. In H2/O2 case since the cell is operated in dry condition and oxygen is supplied to the cathode electrode, oxygen transport losses can be neglected at overall current density of 1 A/cm². In this condition, local cathode electrode effective proton transport resistance is calculated with Equation 4 in the land-channel direction of cell. As shown in Figure 15, at 1 A/cm² overall current density, the electrode effective proton transport resistance varies from 410 mΩ·cm² at the edges of the land (segments 1 and 9) to 720 mΩ·cm² (segment 6) in the channel. The effective proton transport resistance in the catalyst layer for each segment is approximately three times greater than the membrane resistance. Furthermore, compared to cathode electrode protonic resistance (R_H+_{eff}) the effective cathode electrode proton transport resistance (R_{H^+_{eff}}) is ten times lower which indicates that the most of the reaction occurs near catalyst layer/membrane interface.

**Limitation of differential cell.**—A differential cell is often used to study the fundamental characteristics of the materials and transport properties. In the flow channel direction, the flow rates of the reactants are so large that the concentration change is negligible, and therefore the conditions within the cell are considered uniform. However, the results from this study clearly show that even within the differential cell the conditions are not uniform in land-channel direction. Thus, the results obtained with differential cell should be considered as the mean properties that vary in the land-channel direction. This implies that in order to evaluate the true materials and transport properties, the effect of land-channel direction needs to be properly taken into account.

**Conclusions**

A segmented PEMFC was designed to measure local ECSA, current density, and HFR distribution in the land-channel direction at 350 μm and a specially developed method was used to estimate oxygen concentration, membrane water content, and cathode electrode protonic resistance. An ex-situ experimental set-up was developed to measure electrical resistance distribution. In channel areas the estimated surface roughness was lower than in land areas and measured electrical resistance in channel areas were higher than in the land areas. Both of these distributions are supported by previous studies.

In wet cases, high RH resulted in uniform HFR distribution and non-uniform current density distribution in the land-channel direction, and such profiles are associated with the effect of land-channel geometry on liquid water distribution in GDL and concentration of the oxygen to overcome the differences in the length of diffusion. In moderate condition, at low overall current density, the current density distribution depends on both distribution of oxygen and HFR whereas at high overall current density the current density distribution is more influenced by the liquid water distribution. Moreover, at 1 A/cm² overall current density, in H2/O2, the concentration of oxygen was found higher in channel region than land region for all the cases of relative humidity, and effect of relative humidity on the uniformity of oxygen concentration distribution was significant. In moderate and wet conditions two cases of flooding were presented and discussed using oxygen transport resistance distribution in the land-channel direction. These two cases showed that the oxygen concentration distribution is dependent on liquid water distribution in the land-channel direction.

In dry condition current density distribution is highly influenced by the presence of water in the membrane and catalyst layer. Determination of cathode electrode protonic resistance revealed that the catalyst dehydration accounts for majority of losses in dry condition. It is demonstrated from the comparison of HFR and membrane water content distribution between H2/air and H2/O2 that at high local current density enough water can be generated to compensate the membrane dehydration due to the rapid water diffusion.

The results from this study imply that the uniform condition assumption used in a differential cell is not valid, and the effect of land-channel geometry needs to be properly taken into account.

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**Figure 15.** Distribution of cathode electrode protonic resistance with H2/O2 at 1 A/cm² overall current density at 60°C and 0% RH.
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