

# Probing Water Distribution in Compressed Fuel-Cell Gas-Diffusion Layers Using X-ray Computed Tomography

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X-ray computed tomography was used to investigate geometrical land and channel effects on spatial liquid-water distribution in gas-diffusion layers (GDLs) of polymer-electrolyte fuel cells under different levels of compression. At low compression, a uniform liquid-water front was observed due to water redistribution and uniform porosity; however, at high compression, the water predominantly advanced at locations under the channel for higher liquid pressures. At low compression, no apparent correlation between the spatial liquid water and porosity distributions were observed, whereas at high compression, a strong correlation was shown, indicating a potential for smart GDL architecture design with modulated porosity.

## 1. Introduction

Effective liquid-water management is essential for commercializing polymer-electrolyte fuel cells (PEFCs). At lower operating temperatures, there is a need to remove liquid water from the cathode electrode to ensure reactant (oxygen) transport . To achieve maximum water permeation, it is necessary to optimize the gas-diffusion layers (GDLs), which are porous backing layers between the catalyst layer and flowfields. Although water transport in GDLs has been investigated with modeling and experiments , there are still unresolved questions, especially under *in-operando* conditions wherein the GDLs are selectively compressed under flowfield lands but remain uncompressed under flowfield channels. Several studies have used X-ray computed tomography (XCT), which is a high-resolution, non-intrusive characterization technique that can visualize water distribution in GDLs and PEFCs . Previous *in-situ* studies of water permeation through GDLs predominantly concentrated on uncompressed samples. Recently, the morphology of dry GDLs with land and gas-channel effects was characterized using XCT , but those studies did not investigate the water redistribution under different GDL compressions. Currently, there is a gap in understanding how flowfield compression effects liquid-water permeation and distribution inside the GDLs under the flowfield lands and channels. Herein, we report spatially-resolved liquid-water distributions in GDLs at different levels of compression with land and channel effects using an *in-situ* and *ex-operando* experimental set-up and XCT. Liquid-water pathways for different compressions are correlated to local porosity distributions and the three-dimensional morphology of water clusters is quantified.

## 2. Experimental

Fig. 1a) and b) show a schematic of the experimental setup and a photograph of the sample holder mounted on the stage. The experimental setup consists of an aluminum stage with a water column inside, a thin-walled, Kapton tube with cap, and a grooved 1 mm punch to represent the PEFC land/channel geometry. Ultra-fine pitch thread was used to fine-tune the compression of the GDL sample. SGL® 10 BA carbon paper was used with an average thickness of 400  $\mu\text{m}$ , a reported uncompressed porosity of 0.88, and containing 5% PTFE treatment. Capillary pressure and water saturation were controlled hydrostatically using liquid-water column height.

The collection of tomographic data was performed at Beamline 8.3.2 at the Advanced Light Source (ALS). The source energy was 14 keV, the number of back-projections 1025, an exposure time of 500 ms, 0.5 mm LuAG scintillator, 5x lenses, and sCMOS (PCO.edge) camera with 3.3x3.3 mm field of view produced an image with a resolution of 1.33  $\mu\text{m}$ . Image reconstruction was done with commercial software Fiji and Octopus 8.5. The imaged data was of a high quality and minimal filtering was necessary for accurate segmentation.

## 3. Results and Discussion

### 3.1 Tomography of Compressed GDLs with Water Intrusion

Figs. 2a), b) and c) shows spatially-resolved liquid-water distribution for a 2.4 mm field of view under locations of land and channel for GDL compressed thicknesses of 340, 260, and 210  $\mu\text{m}$ , herein referred to as A, B, and C, respectively. The stress and compression of the samples are reported in Table 1. Imaging was done for increasing capillary pressure (liquid-column height) until water breakthrough was observed. The local porosity for each sample, the reconstructed and segmented tomographs at a centerline cross-section are shown as well, where reduced porosity under the lands is observed due to compression. Porosity and saturation were calculated as cross-

section averages parallel to the channel and land directions (*i.e.*, in-plane direction). Liquid-saturation levels remain low as breakthrough is reached at low values, in agreement with *ex-situ* measured water-retention curves . Higher saturation levels are observed for higher liquid-water pressures. The liquid-water saturation front is nonuniform, where variations in local liquid saturations can be as high as 0.45.

This liquid-water distribution heterogeneity is normalized by water retention curves, commonly used to characterize liquid-water saturation as a function of liquid pressure in PEFC modeling community . As Fig. 3a) shows, for all measured liquid pressures, the volume-averaged liquid-water saturation remains below 0.27. Under the channels the average saturation is higher for all three samples. This increase is particularly pronounced at higher capillary pressures. Such local heterogeneities in saturation imply that expressions which use the volume-averaged values (e.g., effective diffusivities) will predict incorrect values, as they do not capture land/channel effects as well as local water distributions, which has recently been discussed .

The average liquid-front position normalized by the GDL's compressed thickness is shown in Fig. 3b). The liquid front advances approximately linearly with liquid pressure, and the front position under the channel is further than for the whole sample, thus indicating preferential water pathways. The average front position is much smaller at breakthrough than the GDL thickness, showing that water fingering is occurring. For PEFCs, the average liquid-front position represents the wetted length of GDL referenced from a catalyst layer. For the 340  $\mu\text{m}$  GDL, only the first 80  $\mu\text{m}$  (23% of thickness) are wetted on average at a capillary pressure of 1.8 kPa, thereby suggesting that in the cathode GDL oxygen diffuses through 77% dry GDL and then is obstructed by large water islands in the last 23% of the GDL closest to the catalyst layer. For the 210  $\mu\text{m}$  compressed sample at a similar capillary pressure, the liquid-front position is 60  $\mu\text{m}$ , representing 28% of the thickness.

Fig. 3c) plots the Pearson product-moment correlation coefficient,  $R$ , between spatial porosity and liquid saturation as a function of liquid pressure for the three samples. The coefficient examines whether the spatial porosity and saturation, as reported in Fig. 2, are correlated, where a correlation value of 1 and  $-1$  indicates that saturation is a function of porosity and a value of 0 states that there is no apparent correlation between them. For the sample with the lowest compression (sample A),  $R < 0.2$ , indicating no significant relationship between porosity and saturation. For sample B and  $p_L > 0.6$  kPa,  $R > 0.6$  indicating a strong positive correlation, and for the sample with the highest compression (sample C) and  $p_L > 1.4$  kPa, the porosity and saturation are highly correlated. From Fig. 2c, it is apparent that for the highest compression, the liquid front advances under the channel where the porosity is higher by 0.1 compared to that under the lands. For  $p_L < 0.6$  kPa, there is a linear increase in  $R$  and relatively weak correlation between porosity and saturation. For compressed GDLs, liquid water takes the higher-porosity transport pathways, thus implying that GDL architectures with modulated regions of high and low porosity could be used to control water removal, where the porosity difference has to be at least 0.1. The findings are consistent with recent studies of morphologically modified GDLs where milled or laser-perforated holes and lines serve as water-transport pathways, although these modifications were  $\sim 100$   $\mu\text{m}$ , much larger than they needed to be based on the current study. However, evaporation/condensation due to thermal gradients and convection along the channel are not captured here but can have a significant impact on water distribution as *in-operando* studies for uncompressed GDLs have shown.

### **3.2. Comparison of water profiles at capillary pressure of 1.4 kPa**

To examine the different compression cases in more detail, the liquid-water distributions are compared at a capillary pressure of 1.4 kPa. Fig. 4a) shows the total in-plane average saturation

and that under the channel as a function of normalized through-plane position. For sample A, a relatively high saturation is observed near the injection plate; however, it falls off steeply and beyond 30% of the sample's thickness only water fingering is observed. For samples B and C, the saturation is lower near the injection plate and falls off more smoothly. The inset in Fig. 4a) shows the spatial liquid-water distributions, where the width of the advancing liquid front decreases from 2,376  $\mu\text{m}$  for the least compressed sample to 1,370  $\mu\text{m}$  for the most compressed GDL. For a PEFC, the difference of  $\sim 1,000$   $\mu\text{m}$  is significant as it is on the same order of magnitude as the dimensions of the flow channels and 2 to 3 times larger than the GDL thickness. Fig. 4b) shows the cumulative volume of liquid water as a function of effective water diameter ( $d = \sqrt[3]{6V/\pi}$ , where  $V$  is the volume). The size of the water clusters decrease with increasing compression; this decrease amounts to 100  $\mu\text{m}$  from sample A to C. Larger effective diameters indicate a larger amount of interconnected water clusters. As Fig. 4c) shows, for the least compressed GDL, larger water volumes form near the interface with the injection plate and spread laterally, whereas, for the GDL with the highest compression, the water clusters lose their connectivity and form smaller islands, predominantly under the channel.

#### **4. Conclusion**

The liquid-water distribution is visualized with X-ray computed tomography for the *in-situ* water intruded gas-diffusion layer (GDL) with 0.4 and 0.8 mm lands and 1 mm channel for three levels of compression and for increasing capillary pressures. No noticeable trends between GDL samples at different compressions were observed when using a volume-averaged saturation – a standard metric used in the fuel-cell community. It has been shown that local liquid-water saturation differs significantly in the GDLs under locations of land and channel and from the average, resulting in a correlation between porosity and saturation for compressed samples. This

finding provides insights into improving fuel-cell modeling and designing optimal GDL architectures with modulated porosity, where liquid-water removal can be directed through controlled porosity levels for desired water redistribution.

### **Acknowledgments**

This work was funded by Assistant Secretary for EERE, Fuel Cell Technologies Office, U.S. DOE and made use of facilities at the ALS, supported by the Office of Science, BES under contract number DE-AC02-05CH11231.



## Tables

Table 1. Tested samples' parameters (uncompressed GDL thickness: 400  $\mu\text{m}$ ).

Sample	Thickness, t [ $\mu\text{m}$ ]	Compression [%]	Stress [MPa]
A	340	15	0.35
B	260	35	1.8
C	210	47	2.8 (extrapolated)

## Figure Captions

Fig. 1. Experimental set-up including a) the schematic of a sample holder and b) a photograph of the sample holder mounted stage.

Fig. 2. Average liquid-water saturation and porosity as a function of position under lands and channel for GDLs under three levels of compression (see Table 1) with pressure ranges of a) 0.1 to 1.8 kPa, b) 0.3 to 2.45 kPa and c) 0.1 to 1.8 kPa. The reconstructed and segmented tomographs located on top; the region of interest (ROI) used to compute the distributions is marked.

Fig. 3a) Volume-averaged liquid-water saturation as a function of liquid pressure for the three samples. b) Position of the averaged liquid-water front normalized by the sample thickness. c) Correlation coefficient between the porosity and liquid saturation.

Fig. 4a) Liquid saturation as a function of normalized GDL thickness. The inset shows saturation as a function of land-channel position. b) Cumulative sum of normalized volume of water clusters,  $V$ , as a function of clusters' effective diameter,  $d = \sqrt[3]{6V/\pi}$ . c) Isosurfaces of liquid water superimposed on the same plot.

## References

- [1] A.Z. Weber, J. Newman, Modeling transport in polymer-electrolyte fuel cells, *Chem. Rev.*, 104 (2004) 4679-4726.
- [2] A.Z. Weber, R.M. Darling, J. Newman, Modeling two-phase behavior in PEFCs, *J. Electrochem. Soc.*, 151 (2004) A1715-A1727.
- [3] U. Pasaogullari, C.Y. Wang, Two-phase modeling and flooding prediction of polymer electrolyte fuel cells, *J. Electrochem. Soc.*, 152 (2005) A380-A390.
- [4] A.Z. Weber, R.L. Borup, R.M. Darling, P.K. Das, T.J. Dursch, W. Gu, D. Harvey, A. Kusoglu, S. Litster, M.M. Mench, R. Mukundan, J.P. Owejan, J.G. Pharoah, M. Secanell, I.V. Zenyuk, A critical review of modeling transport phenomena in polymer-electrolyte fuel cells, *J. Electrochem. Soc.*, 161 (2014) F1254-F1299.
- [5] R. Alink, D. Gerteisen, Coupling of a continuum fuel cell model with a discrete liquid water percolation model, *Int. J. Hydrogen Energy*, 39 (2014) 8457-8473.
- [6] E.F. Médici, J.S. Allen, Evaporation, two phase flow, and thermal transport in porous media with application to low-temperature fuel cells, *Int. J. Heat Mass Transfer*, 65 (2013) 779-788.
- [7] J.T. Gostick, Random pore network modeling of fibrous PEMFC gas diffusion media using Voronoi and Delaunay tessellations, *J. Electrochem. Soc.*, 160 (2013) F731-F743.

- [8] S.G. Kandlikar, E.J. See, M. Koz, P. Gopalan, R. Banerjee, Two-phase flow in GDL and reactant channels of a proton exchange membrane fuel cell, *Int. J. Hydrogen Energy*, 39 (2014) 6620-6636.
- [9] Z. Lu, M.M. Daino, C. Rath, S.G. Kandlikar, Water management studies in PEM fuel cells, part III: Dynamic breakthrough and intermittent drainage characteristics from GDLs with and without MPLs, *Int. J. Hydrogen Energy*, 35 (2010) 4222-4233.
- [10] R. Flückiger, F. Marone, M. Stampanoni, A. Wokaun, F.N. Büchi, Investigation of liquid water in gas diffusion layers of polymer electrolyte fuel cells using X-ray tomographic microscopy, *Electrochim. Acta*, 56 (2011) 2254-2262.
- [11] J.T. Gostick, H. Gunterman, B. Kienitz, J. Newman, A. MacDowell, A. Weber, Tomographic imaging of water injection and withdrawal in PEMFC gas diffusion layers, *ECS Trans.*, 33 (2010) 1407-1412.
- [12] F.N. Buechi, R. Flückiger, D. Tehlar, F. Marone, M. Stampanoni, Determination of liquid water distribution in porous transport layers, *ECS Trans.*, 16 (2008) 587-592.
- [13] J. Eller, T. Rosén, F. Marone, M. Stampanoni, A. Wokaun, F.N. Büchi, Progress in in situ X-Ray tomographic microscopy of liquid water in gas diffusion layers of PEFC, *J. Electrochem. Soc.*, 158 (2011) B963.
- [14] J. Lee, J. Hinebaugh, A. Bazylak, Synchrotron X-ray radiographic investigations of liquid water transport behavior in a PEMFC with MPL-coated GDLs, *J. Power Sources*, 227 (2013) 123-130.

- [15] P.A. Garcia-Salaberri, Hwang G., Vera, M., Weber A.Z., Gostick J.T. , Estimation of universal effective diffusivity in partially saturated fibrous media. Part 1: Effect of through-plane saturation distribution, *J. Heat and Mass Transfer*, Submitted (2014).
- [16] P. Deevanhxay, T. Sasabe, S. Tsushima, S. Hirai, Effect of liquid water distribution in gas diffusion media with and without microporous layer on PEM fuel cell performance, *Electrochem. Commun.*, 34 (2013) 239-241.
- [17] C. Tötzke, G. Gaiselmann, M. Osenberg, J. Bohner, T. Arlt, H. Markötter, A. Hilger, F. Wieder, A. Kupsch, B.R. Müller, M.P. Hentschel, J. Banhart, V. Schmidt, W. Lehnert, I. Manke, Three-dimensional study of compressed gas diffusion layers using synchrotron X-ray imaging, *J. Power Sources*, 253 (2014) 123-131.
- [18] N. Khajeh-Hosseini-Dalasm, T. Sasabe, T. Tokumasu, U. Pasaogullari, Effects of polytetrafluoroethylene treatment and compression on gas diffusion layer microstructure using high-resolution X-ray computed tomography, *J. Power Sources*, 266 (2014) 213-221.
- [19] A. Groso, R. Abela, M. Stampanoni, Implementation of a fast method for high resolution phase contrast tomography, *Opt. Express*, 14 (2006) 8103-8110.
- [20] J.T. Gostick, M.W. Fowler, M.A. Ioannidis, M.D. Pritzker, Y.M. Volkovich, A. Sakars, Capillary pressure and hydrophilic porosity in gas diffusion layers for polymer electrolyte fuel cells, *J. Power Sources*, 156 (2006) 375-387.
- [21] G.S. Hwang, A.Z. Weber, Effective-diffusivity measurement of partially-saturated fuel-cell gas-diffusion layers, *J. Electrochem. Soc.*, 159 (2012) F683-F692.

- [22] J.H. Nam, M. Kaviany, Effective diffusivity and water-saturation distribution in single- and two-layer PEMFC diffusion medium, *Int. J. Heat Mass Transfer*, 46 (2003) 4595-4611.
- [23] A.L. Edwards, The Correlation Coefficient, in: *An Introduction to Linear Regression and Correlation*, W.H. Freeman, San Francisco, 1976, Ch. 4, pp. 33-46.
- [24] R. Alink, J. Haussmann, H. Markotter, M. Schwager, I. Manke, D. Gerteisen, The influence of porous transport layer modifications on the water management in polymer electrolyte membrane fuel cells, *J. Power Sources*, 233 (2013) 358-368.
- [25] H. Markotter, R. Alink, J. Haussmann, K. Dittmann, T. Arlt, F. Wieder, C. Totzke, M. Klages, C. Reiter, H. Riesemeier, J. Scholta, D. Gerteisen, J. Banhart, I. Manke, Visualization of the water distribution in perforated gas diffusion layers by means of synchrotron X-ray radiography, *Int. J. Hydrogen Energy*, 37 (2012) 7757-7761.
- [26] H. Markotter, J. Haussmann, R. Alink, C. Totzke, T. Arlt, M. Klages, H. Riesemeier, J. Scholta, D. Gerteisen, J. Banhart, I. Manke, Influence of cracks in the microporous layer on the water distribution in a PEM fuel cell investigated by synchrotron radiography, *Electrochem. Commun.*, 34 (2013) 22-24.
- [27] D. Gerteisen, T. Heilmann, C. Ziegler, Enhancing liquid water transport by laser perforation of a GDL in a PEM fuel cell, *J. Power Sources*, 177 (2008) 348-354.
- [28] D. Gerteisen, C. Sadeler, Stability and performance improvement of a polymer electrolyte membrane fuel cell stack by laser perforation of gas diffusion layers, *J. Power Sources*, 195 (2010) 5252-5257.

- [29] I.V. Zenyuk, R. Taspinar, A.R. Kalidindi, E.C. Kumbur, S. Litster, Computational and experimental analysis of water transport at component interfaces in polymer electrolyte fuel cells, *J. Electrochem. Soc.*, 161 (2014) F3091-F3103.
- [30] P. Krüger, H. Markötter, J. Haußmann, M. Klages, T. Arlt, J. Banhart, C. Hartnig, I. Manke, J. Scholta, Synchrotron X-ray tomography for investigations of water distribution in polymer electrolyte membrane fuel cells, *J. Power Sources*, 196 (2011) 5250-5255.
- [31] K. Seidenberger, F. Wilhelm, J. Haußmann, H. Markötter, I. Manke, J. Scholta, Grand canonical Monte Carlo study on water agglomerations within a polymer electrolyte membrane fuel cell gas diffusion layer, *J. Power Sources*, 239 (2013) 628-641.
- [32] V. Norouzifard, M. Bahrami, Deformation of PEM fuel cell gas diffusion layers under compressive loading: An analytical approach, *J. Power Sources*, 264 (2014) 92-99.
- [33] I. Nitta, Inhomogeneous compression of PEMFC gas diffusion layers, in, Helsinki University of Technology, 2008.