

LA-UR-

08-5051

Approved for public release;
distribution is unlimited.

Title: High Resolution Neutron Imaging of Water in PEM Fuel Cells

Author(s):
Rangachary Mukundan,
Rodney L. Borup
John R. Davey
Jacob Spendelow
Tommy Rockward
Muhammad Arif
David Jacobson

Intended for:
2008 Fuel Cell Seminar and Exposition
Phoenix
October 27-30, 2008



Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.

Optimal water management in Polymer Electrolyte Membrane (PEM) fuel cells is critical to improving the performance and durability of fuel cell systems especially during transient, start-up and shut-down operations. For example, while a high water content is desirable for improved membrane and catalyst ionomer conductivity, high water content can also block gas access to the triple-phase boundary resulting in lowered performance due to catalyst and gas diffusion layer (GDL) flooding. Visualizing liquid water by neutron imaging has been used over the past decade to study the water distribution inside operating fuel cells.

In this paper, the results from our imaging at NIST using their recently installed higher resolution (≈ 25 mm) Microchannel Plate (MCP) detector with a pixel pitch of 14.7 mm are presented. This detector is capable of quantitatively imaging the water inside the MEA(Membrane electrode assembly)/GDL(Gas diffusion layer) of working fuel cells and can provide the water profiles within these various components in addition to the channel water^{1,2}. Specially designed fuel cells (active area = 2.25cm^2) have been used in order to take advantage of the full detector resolution. The cell design is illustrated in Figure 1 where one of the current collector/end plates is shown. The serpentine pattern was machined into a block of aluminum and plated with nickel and then gold to form the flow field. The measurements were performed using beam #1 and aperture #2 with a fluence rate of $1.9 \times 10^6 \text{ neutrons cm}^{-2} \text{ sec}^{-1}$. The cells were assembled with GoreTM Primea[®] MEAs and SGL Sigracet[®] 24 series GDLs (PRIMEA, GORE-SELECT and GORE are trademarks of W. L. Gore & Associates, Inc). All the cells were tested at 80°C with 1.2 stoichiometry H_2 and 2.0 stoichiometry air flows.

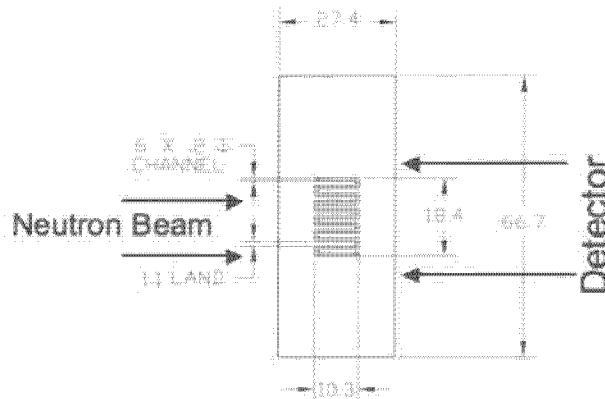


Figure 1. Cell hardware used for the high-resolution imaging. All dimensions are in mm.

The water profiles provide valuable qualitative information about the relative water content in the various fuel cell components. We have operated cells with different GDL materials under a variety of operating conditions to better understand the effect of electro-osmotic drag and water diffusion on the water distribution in the fuel cell components. The measured water content correlates well with performance data. Higher MEA water content results in lower high frequency resistance (HFR) and higher cathode GDL water results in lowered performance due to oxygen mass transport limitations.

Our previous results have shown more water accumulation in the GDLs under the land than GDLs under the channels illustrating the importance of water diffusion in the GDL to keep the pores open for gas diffusion.³ The flow field water is also concentrated more towards the edges of the flow

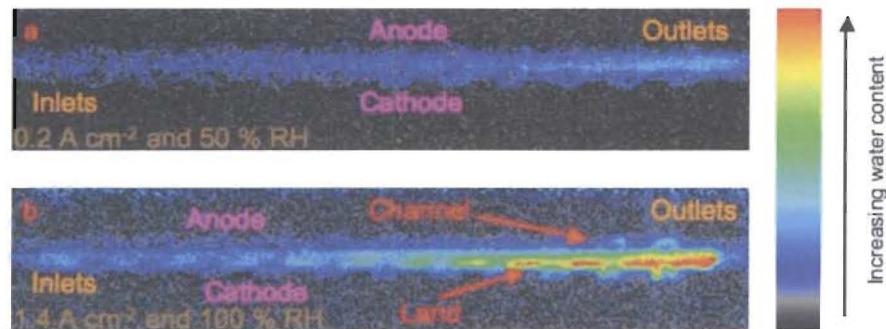


Figure 2. Typical water density images of 2.25 cm^2 cells operated under two different conditions: a) Current Density = 0.2 A cm^{-2} , cathode inlet RH = 50%, and b) Current Density = 1 A cm^{-2} , cathode inlet RH = 100%.

field away from the middle, indicating liquid water drainage into the flow fields from the GDL above the lands. This can be seen in Figure 2 where a typical high-resolution neutron image is presented. The higher current density operation (Fig. 2b.) shows the accumulation of more water

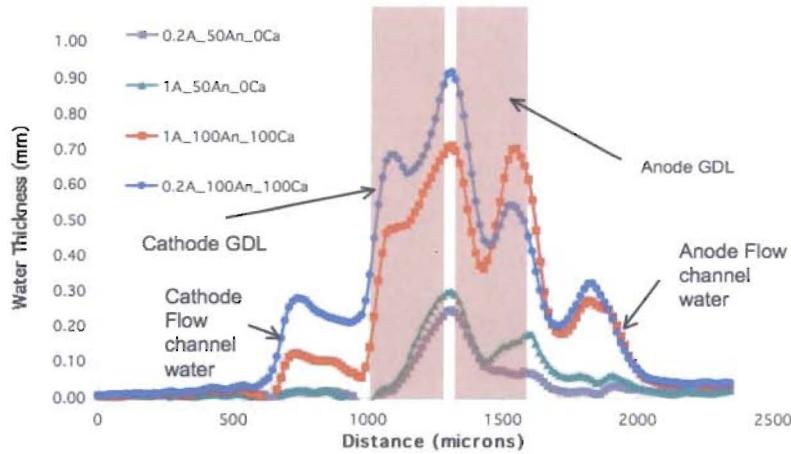


Figure 3. Typical water profiles across the various cell components at different current densities and inlet RHs

especially near the outlet (right side) under co-flow operations. This outlet water is concentrated under the lands and on the sides of the flow channels. Typical water profiles across the various cell components under different operating conditions are shown in Figure 3. The unit of water thickness is in mm for an entire active length of $\approx 12\text{ mm}$. Here, it is seen that the higher current density (1 A

cm^{-2}) results in more anode GDL water indicating that some of the generated water actually back diffuses through the thin (18 μm thick) Gore-select[®] membrane and is removed through the anode GDL. These results also indicate an accumulation of water on the anode GDL substrate close to the flow fields especially near the outlet. This illustrates how low gas velocities (1.1 stoich at the anode) can lead to poor water removal (by droplet shear mechanism) from the GDL. The presence of a liquid water peak in the anode GDL near the flow field can also be explained by a heat pipe effect due to a temperature gradient where water is present primarily in the vapor phase in the MPL and starts to condense to liquid at the GDL substrate closer to the flow channels. There is overall much more water in all the components when the inlet gases are saturated and this water content drops significantly under the drier operating condition (50% anode inlet RH and 0% cathode inlet RH). The drier condition also shows an increased HFR and lower fuel cell performance indicating membrane dryout.

The effect of gravity on these small cells with very little pressure drop was also studied. This is illustrated in Figure 4 where the water content of the MEA and cathode GDL along the cell length

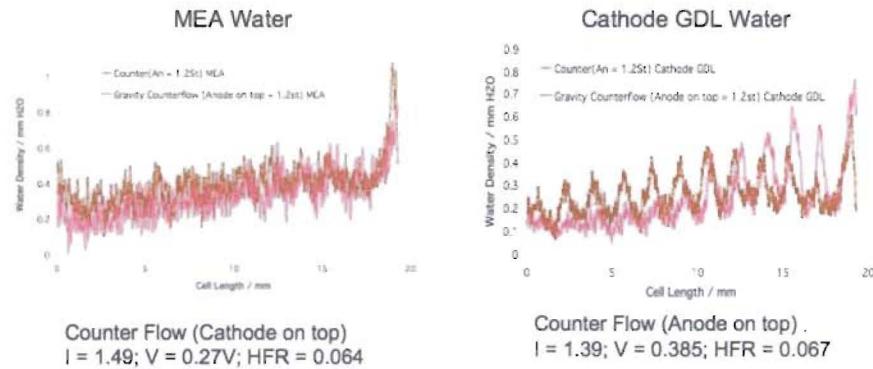


Figure 4. Water density profiles along the cell length (counterflow with cathode inlets at the right of the cell) for 2 different configurations: cathode on top (thick lines with square markers) and anode on top (thin line with no markers).

are presented for two different operating conditions. A counter-flow configuration was used in these cells with the cathode inlets at the right of the cell and the anode inlets at the left of the cell. In one configuration the cathode was on top (counter) and in the other the anode was on top (gravity counter). The HFR of both these cells were very similar, consistent with the almost identical membrane water content observed along the length of the cell. The cell with the anode on top showed significantly better performance at high current densities, which correlated well with the lower cathode GDL water content (especially near the outlets). The cathode GDL water was lower on the cell with the cathode on the bottom indicating the gravity can assist in the water removal from the cathode into the flow channels when the cathode GDL is at the bottom of the cell. The peaks in the cathode GDL water correspond to the lands and the valleys correspond to the channels again showing the difficulty in getting water from the land to the channel. The higher cathode water near the inlets of the cell with the anode on top is due to the back diffusion of significant liquid water near the anode outlet due to the low stoichiometry (1.2) operation.

The quantitative determination of the water in the membrane revealed the presence of < 10 volume% (< 1.2 mm of water in an active length of 12 mm of membrane) water even under fully hydrated conditions. Moreover, this water content was found to decrease as the thickness of the MEA decreased (not shown). These results indicate that the detector spread function plays a role in the quantitative determination of water from these neutron images, especially when the water content is varying greatly over short distances. This issue needs to be resolved before quantitative water content determinations can be made using this technique.

REFERENCES

1. D. S. Hussey, D. L. Jacobson, M. Arif, J. P. Owejan, J. J. Gagliardo, T. A. Trabold, "Neutron images of the through-plane water distribution of an operating PEM fuel cell", J. Power Sources, Vol. 172, pp. 225-228, 2007.
2. M. A. Hickner, N. P. Siegel, K. S. Chen, D. S. Hussey, D. L. Jacobson, M. Arif, "In Situ High-Resolution Neutron Radiography of Cross-Sectional Liquid Water Profiles in Proton Exchange Membrane Fuel Cells", J. Electrochem. Soc., Vol. 155, No. 4, pp. B427-B434, 2008.
3. R. Mukundan, J. R. Davey, T. Rockward, J. S. Spendelow, B. Pivoar, D. Hussey, D. Jacobson, M. Arif, R. Borup, "Imaging of Water profiles in PEM Fuel Cells Using Neutron Radiography: Effect of Operating Conditions and GDL Composition", ECS Transaction, Vol. 11, No. 1, pp. 411-422, 2007.