## X-RAY TOMOGRAPHY FOR INVESTIGATING DEGRADATION AND WATER TRANSPORT IN POLYMER ELECTROLYTE FUEL CELLS

<u>Pratiti Mandal</u>\*1, Iryna V. Zenyuk<sup>†2</sup>, Shawn Litster<sup>‡3</sup>, Adam Z. Weber<sup>1</sup>, Harold S. Barnard<sup>1</sup>, Dilworth Y. Parkinson<sup>1</sup> & Alastair A. MacDowell<sup>1</sup>

<sup>1</sup>Lawrence Berkeley National Laboratory, Berkeley, CA, USA
<sup>2</sup>Tufts University, Medford, MA, USA
<sup>3</sup>Carnegie Mellon University, Pittsburgh, PA, USA

**Keywords:** PEM fuel cell, in-operando, water-transport, morphology

**Summary:** We have used micro- and nano-scale x-ray tomography for *ex-situ*, *in-situ* and *in-operando* study of various components of hydrogen fuel cells. The structure and morphology of the electrodes, gas diffusion medium and the interfaces provide insight about the performance of the fuel cell system and degradation mechanisms. We also investigate water-distribution and defect-formation/evolution for devising cells with high performance and durability.

## 1. INTRODUCTION

Polymer electrolyte fuel cells (PEFCs) use hydrogen gas as a fuel that electrochemically reacts with air to produce electrical energy and water as by product. Fuel cell electric vehicles are zero tail pipe emission systems that offer high efficiency and power density for medium-heavy duty and long-range transportation. However, the widespread commercialization of PEFC technology is impeded due to the high cost, limited durability and lack of hydrogen infrastructure, especially in the United States [1].

At the most basic level, PEFCs are comprised of a cathode, anode, and an ion conducting membrane. The anode converts hydrogen to protons (H<sup>+</sup>) and electrons (e<sup>-</sup>). The electrons go through an external circuit driving the electric load, while the membrane allows protons to move from the anode to the cathode. At the cathode, the electrons and protons meet with oxygen to produce water. A conventional PEFC electrode is a porous, microstructured film consisting of carbon supported platinum (Pt) catalyst nanoparticles bound in polymer electrolyte (ionomer) films. On either side of the electrodes, a gas diffusion layer (GDL) is present, which provides mechanical support, aids in uniform gas distribution and water removal from the electrodes. Performance or power output of a PEFC depends on rate of gas, water, ion and electron transport within the components of the cell, which in turn depends on the connectivity and properties of the conducting networks.

The high cost per kWh of energy produced by a PEFC can be reduced by increasing the performance of a fuel cell, by achieving better mass transport characteristics, reducing the amount of expensive platinum catalyst, increasing the Pt utilization in the electrodes or replacing the Pt catalyst with less-expensive platinum-group-metal-free (PGM-free) electrodes. Investigating the porous structure of PEFC electrode has provided insight into the relation of cell performance with the morphology of the electrode, resulting from different fabrication protocols [2]. Recent studies have shown how high resolution x-ray tomography can help understand the behaviour in the newly developed PGM-free electrodes, caused by inhomogeneous distribution of ionomer and pore network [3]. An efficient water-management system is also imperative towards high performing fuel cells. Hydration is required in the system for maintaining high ionic conductivity; however, water accumulating in the pores of electrodes and GDL, increases mass transport losses in the system. Zenyuk, et al. have studied water transport characteristics in different GDL structures, under fuel cell operating conditions using in-situ micro-tomography [4].

Here, we present some of our in-situ, in-operando and ex-situ studies to investigate transport properties and

<sup>\*</sup> e-mail: <u>pmandal@lbl.gov</u>

<sup>†</sup> e-mail: <a href="mailto:iryna.zenyuk@tufts.edu">iryna.zenyuk@tufts.edu</a> ‡ e-mail: <a href="mailto:litster@andrew.cmu.edu">litster@andrew.cmu.edu</a>

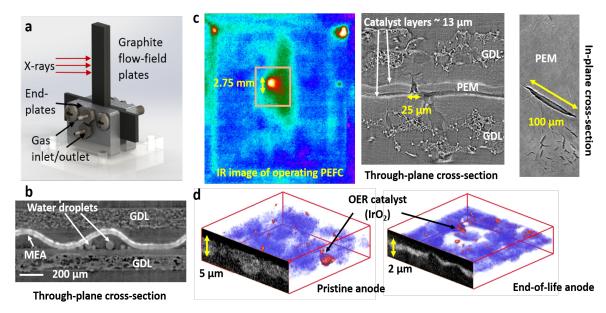
degradation of the various components of a PEFC affecting the performance and durability of PEFCs using nanoand micro-tomography using lab-based x-ray and synchrotron x-ray sources, respectively.

## 2. RESULTS AND DISCUSSIONS

We have used x-ray micro-tomography (Beamline 8.3.2, Advanced Light Source, Berkeley, CA) to study water transport in operating fuel cells with different electrode architectures, to correlate performance of different electrodes with water management in the system. We used specialized fuel cell hardware, which can be adapted for various in-operando fuel cell studies (Fig. 1(a)). Fig. 1(b) shows a virtual-cross-section through the fuel cell assembly showing water accumulated at interfaces and in gas channels. We are also developing fuel cell testing hardware for in-situ study of the evolution of membrane pinholes during continuous operation. Fig. 1(c) shows a damaged membrane in a post-mortem image of a degraded cell, exhibiting local hot spots during operation. We are expanding on this study by designing a cell-testing set-up, which will allow IR imaging to locate local hot spots and correlate them with the degradation observed within the cell components using tomography. This can be performed in-operando, in order to emulate the different stress conditions that a PEFC undergoes during automotive operative conditions. We also investigated another degradation phenomena, which is caused by cell reversal (caused by hydrogen starvation) and loss of catalytic activity of oxygen-evolution reaction (OER) catalyst used in reversal tolerant anodes (RTA), using ex-situ nano-tomography (UltraXRM L200, Carl Zeiss X-ray Microscopy, formerly Xradia, Inc., Pleasanton, CA). The study elucidated the agglomeration of the OER catalyst and eluded that the deactivation of the catalyst resulted in the eventual failure of the RTAs, as well as confirming the loss of carbon due to severe carbon-corrosion during cell reversal. Fig. 1(d) shows side-by-side views of a pristine and end-of-life anode with OER catalyst.

## References

- [1] DOE Annual Merit Review, Fuel Cell Program, 2016.
- [2] P. Mandal & S. Litster. Morphological analysis of polymer electrolyte fuel cell electrode using high resolution X-ray computed tomography and subsequent performance analysis. *ECS Trans.* 58, 481-488, 2013.
- [3] S. K. Babu, H. T. Chung, P. Zelenay & S.Litster. Resolving Electrode Morphology's Impact on Platinum Group MetalFree Cathode Performance Using Nano-CT of 3D Hierarchical Pore and Ionomer Distribution. *ACS Appl. Mater. Interfaces* 8, 32764–32777.
- [4] I.V. Zenyuk, D.Y. Parkinson, G. Hwang & A.Z. Weber. Probing Water Distribution in Compressed Fuel-Cell Gas-Diffusion Layers Using X-ray Computed Tomography. *Electrochem. Commun.* 53, 24-28, 2015.



**Figure 1:** (a) *In-operando* fuel cell test hardware (b) through-plane cross-section showing water droplets at MEA-GDL interface (c) IR image showing hot spots (left) and tomographs showing pinhole in the membrane (d) Volume rendering of anodes with OER catalyst in pristine (left) and end-of-life (right) states.