Development of low temperature fuel cell holders for Operando x-ray micro and nano computed tomography to visualize water distribution

Devashish Kulkarni1,2,7, Stanley J Normile3,7, Liam G Connolly4 and Iryna V Zenyuk1,2,3,5,6

1 Department of Materials Science and Engineering, University of California Irvine, United States of America
2 National Fuel Cell Research Center, University of California Irvine, United States of America
3 Department of Mechanical Engineering, Tufts University, United States of America
4 Department of Mechanical Engineering, The University of Texas at Austin, United States of America
5 Department of Chemical and Biomolecular Engineering, University of California Irvine, United States of America
6 E-mail: Iryna.Zenyuk@uci.edu

Keywords: x-ray computed tomography, fuel cells, water management, cell design

Abstract

Synchrotron x-ray imaging techniques, like x-ray computed tomography (CT) and radiography have proven instrumental in expanding the communities knowledge of complex transport and reaction kinetics in electrochemical devices such as fuel cells and electrolyzers. This work presents the development of novel x-ray CT imaging techniques for operando visualization of water within low temperature fuel cells at spatial resolutions spanning the micro and nano scales. The design of operando sample holders, for both micro x-ray CT and nano CT experiments is described in depth, and prototypes of these sample holders were evaluated across a set of requirements, the most important of which are x-ray transmissibility, electrical conductivity and mechanical stability. Water segmentation from micro x-ray CT data was enabled by an image subtraction method, where the image without water is subtracted from the one with water. Through iterative experimentations, the operando nano CT cell was developed to optimize mechanical compression, electric conductivity and gas flow. While three-dimensional fuel cell reconstructions were shown possible, there remain challenges to overcome at typical lower energies (8 keV) due to beam damage, whereas it is not as significant for higher energies (>17.5 keV).

1. Introduction

Recent developments in transmission x-ray microscopy (TXM) techniques such as x-ray radiography and computed tomography (CT) have enabled the acquisition of high-resolution images at both the micro and nano scales with relatively short scan times [1–8]. These advances in TXM, have enabled imaging of processes within electrochemical energy devices such as batteries, fuel cells and electrolyzers. Recently, operando studies have shown new insights into morphological changes of the various components of electrochemical devices [9–13]. However, these studies generally require custom test apparatuses, which have to be adapted for a use at each specific beamline, limiting broader applicability. Fuel cells, electrochemical energy conversion devices that use hydrogen as a fuel at the anode and oxygen at the cathode to produce electricity, with water as the by-product [14, 15], require effective water management to have high power densities and reliability. For polymer electrolyte membrane (PEM) fuel cells, water is a by-product on the cathode side, where the oxygen reduction reaction (ORR) takes place, as shown by figure S1 (https://stacks.iop.org/JPENERGY/2/044005/mmedia) of the supplementary material (SM). For alkaline exchange membrane (AEM) fuel cells, water management is even a larger issue, as water is formed on the...
anode side and is a reactant in the ORR on the cathode side [16]. The key focus of study revolves around the stack of membrane, the catalyst layers and often the gas diffusion layers (GDLs) typically termed the membrane electrode assembly (MEA). Water is needed to hydrate the membrane, as ion conductivity depends on water content in the membrane [17], however, too much water can be detrimental to fuel cell performance, as it can flood the catalyst layers and block oxygen flow, effectively lowering electrochemically accessible surface area in the MEA. It is critical to understand water distribution during fuel cell operation and to observe its formation in the catalyst layer, so future MEA designs can be optimized to reduce the power density impact of unwanted flooding. While in previous work MEA components morphology was extracted with ex-situ and in-situ imaging and used as input to drive computational simulations to predict water distribution [18], operando studies are needed to further validate and parametrize models [19].

Current state of the art operando x-ray techniques are capable of visualizing water at the microscale, however microscale resolution is not sufficient to resolve the porous structure of the catalyst layer and the water contained therein. This paper presents a first of its kind operando fuel cell that is capable of nanoscale resolution, thus enabling the visualization of water in the porous catalyst layer.

Significant effort was dedicated to micro x-ray CT and radiography cell hardware development for visualizing water transport in PEM fuel cells. Manke et al pioneered some of the first fuel cell for x-ray study by drilling an 8 mm hole into the metallic plate of a conventional fuel cell and imaging through it using x-ray radiography [20]. This apparatus offered a significant improvement in resolution over neutron imaging (1.5 µm vs 75 µm), however the field of view (FOV) was limited to sub 10 mm × 10 mm and in radiography no three-dimensional information can be obtained. Schneider et al built upon these results and developed the first operando cell compatible with x-ray CT [21]. Eller et al improved on Schneider’s design by changing the orientation of the flow channels to be perpendicular with the incident x-rays. This also lengthened the flow channels so that the flow of gasses would be fully developed once the flow reached the imaging FOV. Eller et al reported that their cell could reach a current density of 1 A cm⁻² [22] and maximum active area of 30 mm². Initially this cell design relied on rubber O-rings for cell compression, limiting control of cell compression important factor in fuel cell performance [23]. Although no additional details are provided, the images in Eller et al’s more recent work show metal bolts were added to improve the mechanical compression [24].

That said, the curved design of Eller et al’s cell makes it difficult to machine, and thus inhibits rapid testing of the effects varying flow-channel geometry has on cell performance. White et al developed an operando cell for a lab-scale x-ray CT machine, leveraging a 3D printed holder which allowed for rapid prototyping [25]. The downside of this approach is that the material that the cell was made from is not electrically conductive. While a copper current collector was added to one end of the cell, they were only able to achieve a current density of 0.1 A cm⁻² due to Ohmic limitations. It should be noted that the reported current densities of Eller et al and White et al were recorded using different catalyst layer materials and under different conditions. Nevertheless, the order of magnitude difference in current density underscores the effect of cell architecture on electrochemical performance. The cell designs discussed thus far were for micro x-ray CT or radiography experiments and designed to fit into a FOV of 3–5 mm in width and varied heights.

Many in-situ and operando techniques have been used to image the fuel cells [20, 21, 26–29], but the quality and utility of information produced is dependent on their spatial resolution and the FOVs. Spatial resolution describes how well small details can be imaged or small features can be located with respect to a point. It can be quantified in several ways, for example one way quantification is in terms of the smallest separation at which two points can be perceived as discrete entities [30]. Spatial resolution in the context of TXM depends upon not only the pixel spacing of the detector employed but also the ability to collimate the source, since source divergence leads to image blur [31]. Hence, as a general rule, the FOV is reduced when spatial resolution is increased to ensure each pixel, or volumetric pixel (voxel), is measured with a greater level of statistical significance.

Water is formed inside the nano-scale pores of the catalyst layer, at the surface of the nano-scale catalyst but then is removed into micro-scale GDL, (see figure S1), and then into mm-scale gas flow-channel. Since it is infeasible to capture all these water transport processes by a single imaging technique, various techniques having different resolutions and FOVs levels are used to bridge the scales allowing a complete understanding of the underlying transport phenomena. Figure 1 shows the relationship between the spatial resolution and FOV for different imaging techniques in the style of an Ashby chart.

The most powerful transmission electron microscopes (TEMs) have a spatial resolution of 0.5 Angstrom at an electron beam energy of > 1 MeV [32]. The FOV of TEMs lies between 20 nm and 1 µm and measurement is limited to in-situ studies and does not allow observation of water formation in fuel cell catalyst layers in operando. FIB-SEMs has a slightly higher FOV between 5 µm and 50 µm providing a spatial resolution of 5 nm–30 nm [33]. However, FIB-SEM is a destructive imaging technique hence, it is not possible to do anything beyond ex-situ with this technique. Nano x-ray CT allows for 3D imaging with a
Figure 1. Resolution as a function of FOV for different techniques relevant for imaging electrochemical devices. (1) TEM resolution limits: 0.5 Å–1 nm, FOV: 20 nm–1 µm. (2) FIB SEM resolution limits: 5 nm–30 nm, FOV: 5 µm–50 µm. (3) Nano x-ray CT resolution limits: 10 nm–50 nm, FOV: 20 µm–1 mm. (4) Micro x-ray CT resolution limits: 1 µm–50 µm, FOV: 0.1 cm–2 cm. (5) Neutron imaging resolution limits: 5 µm–200 µm, FOV: 5 mm–400 mm.

In this work, we present the details of micro and nano x-ray CT cell designs aimed at furthering imaging capabilities water within the catalyst layers and other fuel cell components. We will focus only on full-field imaging and will not discuss the techniques that are based on raster scans. The examples presented here are imaged exclusively with synchrotron radiation, using monochromatic beams and absorption imaging mode for micro CT and phase-contrast mode for nano CT. We will present examples of the results generated with several versions of our operando cells, as well as discuss the challenges of operando x-ray imaging, including x-ray beam damage, synchrotron stages limitations, materials selection limitations and others. Lastly, we will present ideas for future cell improvements.

2. Methods: experimental/hardwaredesign

2.1. Micro-CT operando cell design

2.1.1. Design constraints.

The most important quality of the operando cell is that it must be sufficiently x-ray transparent. As x-rays pass through a medium, they are attenuated as described by the Beer–Lambert law:

\[ \frac{I}{I_0} = \exp(-\mu l) \]

where \( I_0 \) and \( I \) are the incident and resultant intensity of the x-rays, \( \mu \) is the attenuation coefficient, and \( l \) the thickness of the medium. In absorption contrast imaging, the intensity of the light is lowered by some of the photons being absorbed by material. X-ray attenuation is a degree of sum of all interaction between photons and matter, including scattering and absorption by a material. Phase contrast imaging relies on the shift of the x-ray wave that travels through the sample, with respect to the wave that is not perturbed by the sample. A detailed derivation of intensity of the light incident onto a detector in absorption and phase modes may be found in our previous work [26].

The attenuation coefficient is dependent on the photon energy of the incident x-rays. Figure 2(a) shows the x-ray transmission \( (I/I_0) \), as a function of energy for various materials that were considered for the cell construction across typical micro x-ray CT range (20 keV–30 keV) and nano
Figure 2. X-ray transmission for various materials across (a) micro and (b) nano x-ray CT beamlines energy ranges. The material thicknesses specified represent the total thickness of material on both sides of the cell (i.e. 1/4” (6.35 mm) Graphite refers to two 1/8” (3.18 mm) plates, one at the anode and one at the cathode). The 1/8” graphite represents the thickness of the half-graphite half-Kapton cell which features a graphite plate on the cathode and Kapton film on the anode.

x-ray CT energy range (4 keV–23 keV). In addition, these materials must be electrically conductive or have a layer of electric conductor (such as gold plating). The higher the transmission, the lower the signal-to-noise ratio that we will observe in the resulting images. At least 30% of x-ray transmission is required in single radiograph to achieve reconstructed tomography data with minimal artefacts, using current state-of-art detectors installed at the synchrotron facilities in the U.S. For low transmission samples unwanted artefacts, such as rings on the images will appear. For example, in figure 2(a) we observe that 1/2” graphite plate (which most of the operando cells for fuel cell imaging use) is above the 30% threshold for all energies above 20 keV. On the other hand, gold-sputtered polyether ether ketone (PEEK), another possible material for operando cells, only becomes a viable option at energies above 25 keV.

Nano x-ray CT synchrotron beamlines operate at lower energy ranges, as nano x-ray imaging requires the beam to be focused onto the sample. This is typically done with a Fresnel zone plate [37, 38], which is a diffractive optical element (figure 4(a)). From figure 2(b) it is evident that 15 μm Kapton film is x-ray transparent, ¼” graphite plate can be used only at energies above 15 keV, and ¼” PEEK plate can be only be used at energies close to 20 keV. Another option is to use graphite on the cathode side of the cell and Kapton on the anode. This half-graphite half-Kapton cell uses a total thickness of 1/8” of graphite and represents a compromise between the transparency of Kapton and the electrochemical advantages of graphite. The nano CT beamlines that these experiments were performed at operate at energies of 8 keV, 15 keV or 17.5 keV. From this analysis, we can conclude that nano x-ray CT cell material selection is limited to Kapton film for 8 keV energy, however at the higher energies, graphite and PEEK become feasible provided that their thickness is minimized.

Accurately controlling the temperature of the cell is critical to its performance, as water transport is dependent on both the temperature of the cell and the local temperature gradients within the cell. Weber et al noted that water which is evaporated in the cathode catalyst layer tends to move down the temperature gradient and condense in areas of lower temperature [39, 40]. This temperature gradient exists through the thickness of the cell (thru-plane). The in-plane temperature gradient should be minimized, as a large variation in temperature along the length or width of the cell could result in uneven current density and water pooling in the cooler areas. This can be avoided by minimizing the spreading resistance of the bipolar plate material. Thermal spreading resistance describes the resistance of heat transfer from a small source (in this case, the cartridge heater) to a larger area (in this case, the graphite plate) [41]. Conductive thermal resistance in direction i is given by

$$\frac{R_i}{k_iA_i}$$

where R is the thermal resistance, l is the length along which the heat is travelling, k is the thermal conductivity and A is the cross-sectional area. If the thermal conductivity is isotropic, the thermal resistance in the two in-plane directions will be higher than that of the through plane, due to the longer length scales and smaller cross-sectional areas. The plate would therefore be defined as having a high spreading resistance, resulting in an uneven temperature distribution along the length of the channel. On the other hand, if the
in-plane thermal conductivity is much higher than the thru-plane thermal conductivity, as is the case for graphite, the thermal resistances in the three coordinate directions will be much closer to equal, allowing the heat to spread evenly in all directions.

More detailed material selection for the micro x-ray CT cell design are summarized in SM tables S1 and S2. We consider the same set of materials shown in figure 2(a): two ¼” PEEK plates with 300 nm gold sputtering, two 1/4” graphite plates, as well as two ¼” aluminum plates. The physical properties that we consider relevant are x-ray transmission, electrical resistance, thermal conductivity, maximum displacement, durability and machinability. These metrics are provided for all three materials under consideration.

2.1.2. Cell geometry development

Figure 3 shows the generational development of the micro x-ray CT cell. All of the cells featured two gas channels. The generation (Gen) 1 cell was designed based on Eller et al’s cell [22] and the main cylindrical components of this cell were 3D printed by resin stereolithography for rapid prototyping. However, this cell lacked adequate compression, thermal stability, electrical conductivity and was only able to reach a steady-state current density of 0.1 A cm\(^{-2}\). The Gen 2 cell was made by sputtering 300 nm gold on a machined ¼” PEEK flow field and using aluminum bolts for compression. While this improved the overall performance of the cell, it had some shortcomings. Sputtered gold has low electrical in-plane conductivity due to inter-grain or grain boundary contact resistances and moreover, poor external electrical connections further increased Ohmic resistance. Increased resistance in turn results in higher joule heating:

\[ W = i^2 R \]  

where \( W \) is the heat produced in Watts, \( i \) is current flowing through the cell, and \( R \) is resistance.

Furthermore, as the cell was used repeatedly, the sputtered gold flaked off in places. This resulted in an uneven electrical resistance and caused localized temperature gradients with hot spots in areas of higher resistance. If the temperature gradient along the length of the gold sputtered plate is uneven, pockets of condensed water may form in the areas of lower temperature. The next generations of cells addressed this thermal gradient issue by using fuel cell grade graphite (properties shown in table S2, SM) for the bipolar plates with variations in the flow field design and compression techniques. The Gen 3 cell had a long area with a serpentine channel that could be used for imaging and was compressed with an external compression fixture, which introduced a pressure gradient and was not repeatable. To solve this issue, Gen 4 and 5 cells
were made with integrated Al compression plates, which provided even compression, heat distribution, and a capacity to hold a cartridge heater close to the active area.

The final design sought to combine the most advantageous properties of graphite and aluminum by adhering graphite plates to an aluminum backing along the entire length of the cell except for a region that is left open to allow x-rays to pass through as shown in figure 3 (Gen 3–5). The length of the x-ray window varies between 0.5 cm and 1 cm. A smaller length provides higher compression of the MEA, but limits FOV. The active area of the cell can be as large as 1 cm². This design allows the aluminum plates to provide all the mechanical stiffness without cracking. The temperature fluctuations are modeled to be within 1 degree along the 3 cm of graphite modeled that include the flow channel (figure S2). Typically, Teflon gaskets are used as a hard-mechanical stop to control compression of the porous materials within the fuel cell. The thickness of the gaskets is chosen in such a way that the overall porous media components within the fuel cell are compressed by 15%–20%. The local pressure distribution was measured using a Fuji Prescale Low Film that registers a range of 2.5 MPa–10 MPa and while the x-ray window was only compressed by 3 MPa of clamping pressure, the rest of the cell area measured 10 MPa of clamping pressure. An image of the compression pattern is shown in figure S3. The design of Gen 5 cell also allows it to be assembled and disassembled quickly, an especially advantageous feature when the cell is used at synchrotron facilities where beamtime is limited. The use of acetal alignment pins ensures that the components are roughly aligned during reassembly.

The Gen 4 and 5 cells can also be used as either fuel cells or electrolyzers. The cells shown in figure 3 have parallel channels, however they can be easily swapped for serpentine channel plates to understand water transport near the serpentine bends [42]. The Gen 5 cell has also been used as an electrolyzer in our previous work with parallel channels [43, 44]. An example of different flow channel geometries that can be employed in this setup can be found in the SM, figure S4.

The mechanical compression of the cell coupled with the high electrical conductivity of the graphite led to measured electrical resistance of just 0.7 Ω when assembled without any MEA or gaskets. In order to reduce the resistance of the junction between the graphite plate and the copper wire used to connect the plate to the potentiostat, the contact region of the plate is electroplated with copper. The copper wire can then be soldered directly to the copper contact pad, creating a low-resistance bond. Graphite is an excellent heat conductor due to its very high in-plane (275 W mK⁻¹) and high thru-plane thermal conductivity (5.0 W mK⁻¹), as shown in table S1.

Using graphite as the gas flow-field and a hole in the aluminum plate to hold a cartridge heater enabled closed loop heating control of the cell, using a single 0.75 W cartridge heater on each side of the cell. We created a thermal model of the cell and found that the temperature varied by only 1.1 °C along the length of the channel, as shown by figure S2. The complete thermal analysis is provided in the SM. Figure S5 shows the heater response to a step temperature change. The cell's electrochemical performance for fuel cells and electrolyzers reached 500 mA cm⁻² for Pt-group metal free (PGM-free) catalyst layers [42–46]. Comparing Gen 4 and Gen 5 micro CT cell polarization curves, as shown in figure S6, they both show very similar performance. However, Gen 5 has shorter channel design, and is more mechanically robust with better thermal management and thus it is recommended to use Gen 5 cell.

2.2. Nano x-ray CT operando cell design

2.2.1. Material selection.

The nano x-ray CT cell poses more stringent design requirements than its micro x-ray CT counterpart. First and foremost, it must be x-ray transparent at photon energies as low as 8 keV. The first-generation nano x-ray CT cell was used at beamline 32-ID-C at the Advanced Photon Source (APS) at Argonne National Lab (ANL). This beamline cannot vary the energy for nano-scale x-ray imaging using phase contrast imaging mode and had energy set at 8 keV. Later iterations of the first-generation nano x-ray CT cell were imaged at beamline 6–2 at the Stanford Synchrotron Light Source (SSRL) at SLAC National Accelerator Laboratory (also at 8 keV) and beamline ID16B at the European Synchrotron Radiation Facility (ESRF) in Grenoble, France (17.5 keV). The second-generation (Gen 2) nano x-ray CT cell was used at beamline 18-ID at NSLS II at Brookhaven National Laboratory (15 keV). As was the case for the design of the micro CT cell, the Beer–Lambert law can be used to determine which materials will be sufficiently transparent for use in the nano x-ray CT cell (figure 2(b)). A thin Kapton window is the only viable option at 8 keV. At 17.5 keV, 1/4" of graphite may be used. Table 1 summarizes the possible materials at the two extremes of available energies across the three selected beamlines.

2.2.2. First generation nano x-ray CT cell.

Figure S7 in SM shows the first generation of nano x-ray CT cell design. The cell dimensions are 25 mm × 25 mm at the base and 15 mm height. It consists of a PEEK base that acts as a stable block to avoid vibrations due to introduction of gases and it also has internal channels for gas delivery. Gas lines are
connected to the PEEK base via Swagelok connectors and 1/16\textquotedbl| flexible tubing is used for the gas delivery. Figures S7(b) and S8 shows the paths for gas inlet and outlet. The tubing material must be adherent to the beamline hydrogen protocols—as it must supply hydrogen to the anode—while simultaneously being flexible so as to not introduce any external torque to the sample stage during the cell rotation. The cell is designed to be in a vertical configuration, where Au-sputtered PEEK frames are used as structural supports for the MEA and current collectors. The frames slide onto the top of the base, and two silicon O-rings at the bottom enable sealing between the base and the frames, as shown by figure S8. At the bottom of the frames, two small magnets are used to lock the frames to each other. This was mainly a precaution, as the sliding mechanism enables good contact to the MEAs at the bottom of the frames. Small tabs are protruded from the frame, and these are the locations where copper wire is soldered to, to enable connection to the potentiostat. The PEEK frames are covered by a Kapton window, and are designed to have a space within to enable gas transport to the MEA. The gas flow within this design is not directed by gas channels but rather is accomplished by a constant flow of gas into a small chamber that is in contact with MEA. The GDLs, with cut out windows, are shown as well by figure S7. The GDLs provide structural stability and electrical conductivity. Because the cell is to be used at two different energies, 8 keV and 17.5 keV it was designed to support both Kapton windows and graphite plates (figure S7(b)). The advantage of the graphite plate with a gas flow-channel is that it enables introduction of gas in a more controlled fashion, as well as the fact that graphite provides structural stability and electric conductivity. This was accomplished using a standard base, to which either the Kapton frame or graphite could be mounted. The exploded view of the cell is shown by figure S7c, where the view also shows how the cell connects to the magnetic base that is used at the APS 32-ID-C. Lastly, the assembled cell with gas and electric connectors is shown at 32-ID-C beamline at APS.

Typical nano x-ray CT beamlines have a FOV of 80 \( \mu \)m, therefore it is preferable to have a small electrochemically active area so that the current is concentrated within the FOV. The MEA is cut in a T-shape and surrounded by Kapton to form a seal. The active area is approximately 6 mm \( \times \) 1 mm. After operating this fuel cell for several experiments at APS, ESRF and SSRL the following design issues were identified: (1) poor mechanical compression on the top of the MEA, (2) poor electric conductivity of sputtered Au and delamination of Au on PEEK, and (3) difficulty of assembly due to tightness of the o-ring preloaded slip-fit. Issues 1 and 2 were present, but much less severe, for the graphite plate configuration of the cell. The electric conductivity issue was partially resolved after we introduced several Au sputtered bars across the frame, and surrounded the MEA and current collectors. The frames slide onto the top of the base, and two silicon O-rings at the bottom enable sealing between the base and the frames, as shown by figure S8. At the bottom of the frames, two small magnets are used to lock the frames to each other. This was mainly a precaution, as the sliding mechanism enables good contact to the MEAs at the bottom of the frames. Small tabs are protruded from the frame, and these are the locations where copper wire is soldered to, to enable connection to the potentiostat. The PEEK frames are covered by a Kapton window, and are designed to have a space within to enable gas transport to the MEA. The gas flow within this design is not directed by gas channels but rather is accomplished by a constant flow of gas into a small chamber that is in contact with MEA. The GDLs, with cut out windows, are shown as well by figure S7. The GDLs provide structural stability and electrical conductivity. Because the cell is to be used at two different energies, 8 keV and 17.5 keV it was designed to support both Kapton windows and graphite plates (figure S7(b)). The advantage of the graphite plate with a gas flow-channel is that it enables introduction of gas in a more controlled fashion, as well as the fact that graphite provides structural stability and electric conductivity. This was accomplished using a standard base, to which either the Kapton frame or graphite could be mounted. The exploded view of the cell is shown by figure S7c, where the view also shows how the cell connects to the magnetic base that is used at the APS 32-ID-C. Lastly, the assembled cell with gas and electric connectors is shown at 32-ID-C beamline at APS.

2.2.3. Second generation nano x-ray CT cell.

The second-generation cell had significant advantages over its predecessor in terms of manufacturability, modularity of design, electrical conductivity, and mechanical compression control. Additionally, it is less expensive to manufacture due to both its geometry and lack of need for expensive treatments such as gold sputtering. Like the Gen 1 cell, the Gen 2 cell is also a vertical cell. The base has been redesigned to reduce its size and to divide it into two portions for anode and cathode. Graphite with two flow-channels is incorporated into the base to conduct electrons and deliver gases, as shown in figure 4(d). The graphite flow distributors are in contact with the electrically conductive copper sheet current collectors. The adhesion between the graphite flow channel and the copper electrode is achieved using electrically conductive epoxy. The copper current collectors run out of the cell body to establish electrical connections with the potentiostat. Gases are introduced in the cell through the openings within the base as shown in figure 4(c). The last component of the cell was the PEEK frame with a 1 mm \( \times \) 4 mm window for x-ray transmission. Out of the total MEA active area of 0.84 cm\(^2\), 0.067 cm\(^2\) was exposed to x-rays through the window. The MEA compression was achieved using anodized compression bolts. The cell was designed to compress the MEA as evenly as possible, even though the Kapton window does not mechanically support the MEA. This compression prevents the MEA from moving because of gas flow, which was an issue with the Gen 1 cell. Figure 4(e) shows the readings of the pressure paper, where we still observe four-fold reduction in compression under the Kapton windows, from 200 psi to 50 psi (1378.9 kPa to 344.7 kPa). The cell was
mounted on the NSLS 18-ID beamline stage with a 3D printed fixture. Figure 4(b) shows the cell mounted on the rotation stage and a zoom-in shows the cell during rotation. All the gas and electric lines were managed in the same bundle.

3. Synchrotron beamlines

The supplemental material shows an overview of the capabilities of the various micro and nano CT beamlines that were used in this study. It also shows the imaging setups that were used to achieve the results shown in the next section. The micro-CT beamlines included Beamline 8.3.2 at ALS, LBNL and 2-BM at APS, ANL. The nano x-ray CT beamlines where we used operando cell included 32-ID at APS, ANL, 18-ID, NSLS II at BNL, 6–2 at SSRL and ID16B at ESRF. The optics specifications, resolution, FOV and various environments that were used to generate results for this work are described in-detail in SM.

4. Results

4.1. Micro x-ray CT studies

Figure 5 shows micro x-ray CT cross-section tomographs of the Gen 2, 4 and 5 operando fuel cells fully assembled all imaged with resolution of 1.3 µm using 5× lenses. The details of the materials used for fabrication these cells are provided in SM. PGM-based Pt/C or Pd/C MEAs are used in these cells. The catalyst layers are bright in all three cross-sections and are clearly visible. The membrane used in the Gen 2 cell was twice as thick as those used in the Gen 4 and Gen 5 cells. The microporous layer (MPL) next to the catalyst layer is seen in Gen 2 and 4, but was not used in Gen 5, as it was an AEM fuel cell. The GDL fiber cross-sections are seen in all three tomographs. In the case of the Gen 2 cell, the highly absorbing gold sputtering on the PEEK frame distorts the contrast on the nearby catalyst coated membrane (CCM) and
Figure 5. Micro CT operando x-ray CT images of a fuel cell (a) Gen 2 cell cross section (b) Gen 4 cell cross section (c) Gen 5 cell cross section. 1.3 µm resolution. Gen 2 data collected at 2-BM APS, Gen 4 and 5 collected at 8.3.2 at ALS.

GDL. Median filter of radius 2 was used to smooth the greyscale image shown for Gen 2 cell, while the Gen 4 and Gen 5 images were not filtered. The Gen 4 and Gen 5 cell images have an even contrast throughout the cross section and the graphite bipolar plates are clearly visible. Although there is little visible difference between the Gen 4 and Gen 5 cross sections, the Gen 5 cell has better compression and heat distribution. The Gen 3 cell results have been previously published in Normile et al [42]. The Gen 5 cell results with AEM have been published by Peng et al [47].

Since water attenuates x-rays weakly, studies that aim to show water content in carbon materials should theoretically be imaged in a phase contrast mode. Figure S9 shows that the linear attenuation coefficients of water and carbon are similar across a wide energy range. That said, phase contrast can introduce artefacts from the sample holder, so the images were therefore taken in absorption contrast mode. Image subtraction with a dry, non-active CT data set is used to visualize water location. This method of water separation was first developed by Eller et al [22]. As shown by figure 6(a), the first scan is taken with the cell at open circuit voltage (OCV) conditions, which will not produce any water. Since the gases are humidified to 100% RH before flowing into the cell, they will cause the MEA to swell. This is necessary for the base subtraction data set so that it will be possible to align the dry and wet images. The second scan is taken with the cell running at a specified current density. The reason the cell is maintained at constant current, and not constant voltage, is that the amount of water produced is proportional to the current density by Faraday’s law of electrolysis:

\[
N_{H_2O} = \frac{A}{2F} \int i dt
\]

where \(N_{H_2O}\) represents the number of moles of water produced, \(A\) is the active area of the cell, \(F\) is Faraday’s constant, and \(i\) is the current density. After 2–3 min, the cell reaches steady-state, where the rate of water production and removal will be equal. It is at this time when the scan is taken to identify the steady-state water content in the GDLs, channels and ideally, in the catalyst layer. The polarization curves for this cell have been previously published in Normile et al [42], and are reproduced in SM, figure S10. Figure 6(b) shows a cross-section tomograph for the cell operating at a low 40 mA cm\(^{-2}\) current density, where water can be observed. The reason for water observed at this low current density is that the cell temperature was 30 °C and inlet gas was fully humidified and thus water was formed as liquid, as phase-change-induced flow and evaporation is minimal at this low temperature. Cell operation was stable at this current density for close to 2 h (figure S10). Constant current holds are often repeated for several different current densities in order to establish a trend in water content. The dry image is then subtracted from the wet image in order to threshold the water. This process can be done in absorption contrast mode, as the minute changes in attenuation from the water will be visible once the images are subtracted. The result of this subtraction is shown in figure 6(c). Notice that dry and wet scans cannot be aligned perfectly, for example due to
Figure 6. Cross-section tomographs of the assembled fuel cell having a PGM-free cathode at 30 °C under (a) OCV (dry), (b) with a current density of 40 mAcm⁻² drawn from the cell (wet), (c) thresholded water clusters and (d) overlay of thresholded water and image from plot b. (e) A histogram of greyscale dry and wet images, with the region belonging to water, (f) the greyscale values along the dashed line shown in a and b, where the window for greyscale values for water is shown too. Gen 3 cell. 1.3 µm resolution. Data is collected at 8.3.2 beamline at ALS.

membrane and ionomer swelling. Thus, some noise and error is expected in thresholded image, as can be seen in figure 6(c), where smaller specs of single voxel are most likely noise. The thresholded water domain is then overlaid with the grey-scale image with water and is shown by figure 6(d). Through the 8-bit greyscale histogram we observe the region that corresponds to water, as shown by figure 6(e). This region also overlaps with some of the carbon material. Finally, figure 6(f) shows the grey-scale values distribution along the dashed line drawn in figures 6(a) and (b). In future work, machine learning algorithms will make it possible to automatically segment water without need for a background dry scan.

Figure S11 shows the cross-section tomographs for a conventional Pt/C catalyst fuel cell with the Gen 5 hardware. Water was observed both within the cracks of the MPL and the flow channel. For this data set the OCV scan was of marginal quality, and therefore proper thresholding was not possible. As can be seen, the image quality and grey scale of water is very similar to that reported by figure 6. From these micro x-ray CT
images, it is also evident that for conventional Pt/C catalyst layer less than $10 \, \mu m$ thick, it is not possible to see water within the catalyst layer due to the resolution limitations of micro x-ray CT of about $1 \, \mu m$.

### 4.2. Nano x-ray CT studies

First attempts to image the first-generation nano CT cell at 32-ID-C at APS (8 keV) yielded poor quality images due to missing angles imposed by the PEEK frame, as well as due to beam damage. The missing angles were due to close to 0 transmission when the PEEK frame is parallel to the x-ray beam. Reducing the frame thickness from 1 mm to 0.75 mm helped reduce the number of missing angles by 2 degrees. Figure S12 shows a schematic of x-rays transmission and frame geometry with respect to incident x-rays. At 8 keV, the attenuation coefficients of all the relevant materials is higher than at 17.5 keV. This increases the absorbed radiation dose and makes beam damage more likely. Figure 7 shows beam damage to the CCM imaged in the Gen 1 nano CT cell at 32-ID at APS. Figures 7(a)–(c) shows the in-plane radiography images at the same degree of rotation, and the inset the reconstructed tomographs. In addition, 3D rendered MEAs are shown in figure S13. It is evident from both the tomographs and radiographs that 60 min of beam exposure reduced the CCM thickness by 66% (figure 7(e)) since material degradation is proportional to the absorbed radiation dose \[ R = \frac{NTE_\mu}{\rho} \] (5).

Where $R$ is the radiation dose in keV g$^{-1}$ (often converted to rad), $N$ is the flux density in photons/cm$^2$/s, $T$ is the exposure time in s, $E$ is the photon energy in keV, $\mu$ is the linear attenuation coefficient in cm$^{-1}$, and $\rho$ is the material density in g cm$^{-3}$. Although the resolution was 30 nm, the noise introduced into the tomograph images by both the missing angles and the fact that the sample moved during the scan due to beam damage yielded an image quality that was not high enough to visualize water in the nano-scale pores of the catalyst layer. Figure 7(d) also shows that visible cracks were formed in the CCM, which is possibly due to beam heating. Since membrane is a low-density material and is made of a polymer material, this is the component that degraded the most, as its thickness reduced from about $25 \, \mu m$ at the beginning to less than $10 \, \mu m$ after 60 min of operation (figure 7(e)). The thickness measurements were performed on reconstructed images, where thickness of membrane within the 60 $\mu m$ FOV was evaluated and averaged across 70 tomographs (roughly every 1/10th image). The observed membrane thinness was the most...
pronounced in the center of the FOV (where beam intensity is the largest) and at the edges of the FOV the membrane was thicker. This is indicative that membrane thinning is mostly due to beam damage and will be localized to the area exposed to the beam.

There are generally three strategies to reduce x-ray beam damage: (1) reduce time of imaging (by reducing exposure time), (2) use materials that are more radiation resistant (avoid polymers) and (3) use higher energies, where x-ray absorption is lower. Collecting data in radiography mode is another possibility. Radiography is a 2D imaging technique that exposes the sample to the beam for $1/N$ the amount of time that tomography does as it only requires a single exposure. Here $N$ is number of projections used in equivalent tomography scan, in this work at APS it was 720. To better understand the degradation dynamics, the operando cell was imaged in through-plane radiography mode at beamline 6–2 SSRL. Imaging in through-plane mode allows us to observe the surface of the CCM and any associated degradation due to beam damage (formation of cracks, formation of bubbles etc. since x-rays pass in a perpendicular direction to it. In order to expand the FOV, five scans were taken of each sample and stitched together to form one large image called a mosaic. It is possible to separate five images stitched together in mosaic by eye due to non-uniform intensity of the beam. Figure 8 shows the images that were taken, as well as the electrochemical performance of the cell. The first scan (figure 8(a)) was taken with dry air flowing over the CCM in order to provide a dry baseline image. The cell was then fed humidified hydrogen and air. The second scan was under OCV conditions. The cell was then imaged under three different current conditions by applying a specified voltage across the cell. The current response to these constant voltages is plotted in figure 8(b). For all three applied potentials, currents started high and then decayed with time. As the current density is higher at 0.3 V than at 0.1 V, it can be inferred that the cell may have a limit to its maximum current.

The increased current density caused the formation of cracks that become more pronounced with increasing current and reducing potential. It also caused the overall x-ray attenuation of the sample to decrease. This can be seen in figure 8(a) since the reconstruction algorithm inverts the color contrast so highly attenuating objects appear more brightly. The very bright spots in figure 8(a) are gold fiducials placed on the sample to register the sample within the coordinate frame. The grayscale value of each pixel in the resultant image is proportional to the attenuation of the sample at that point, however the relationship between the intensity calculated using the Beer–lambert law and the grayscale value is nonlinear due to the

![Figure 8](image-url)
camera lens, the reconstruction algorithm, and the image processing software. The increase in grayscale value is plotted in figure 8(c). This trend of decreased attenuation with increased potential was found to be due to cracks formation. It is difficult to say if water was observed in the CCM as the evolution of cracks was the most pronounced morphological change. We observe that even during radiography imaging at 8 keV with a scan time of roughly 1 min, the beam damage is significant. The cracks could also form due to localized heating due to the x-rays burning the membrane.

The same procedure was repeated for a cell without a window cut in the GDL. The addition of a complete GDL vastly improved the electrochemical performance of the cell’s resilience in maintaining a constant current for a prolonged period of time, such as 10 min (figure S14). However, the GDLs completely blocked the view of the CCM behind it. This is because the GCM is 30 µm thick, whereas the GDL is 225 µm thick. This means that the CCM will absorb about 4% of the incident intensity, whereas the GDL will absorb 20%, by the Beer–Lambert law (equation (1)). From these two comparisons, we conclude that nano x-ray CT imaging should be conducted with GDLs, but because of poor x-ray transmission, they must be reduced in thickness, or the possibility of free-standing MPLs explored.

Experiments performed at beamline ID16B at the ESRF benefitted from imaging at a photon energy of 17.5 keV. This allowed the use of graphite plates as opposed to Kapton windows; however, the porous nature of the graphite produced phase artifacts that made the sample itself difficult to resolve. A compromise between the improved electrochemical performance afforded by the graphite and the visibility afforded by the Kapton windows (the PEEK supports around the window do not block x-rays at 17.5 keV) was to use one graphite plate and one Kapton window. This half-graphite half-Kapton cell maintained reasonable electrochemical performance while still feasible to image. Furthermore, as discussed above, we were able to include GDLs in this cell to improve its electric conductivity.

Beamline ID16B allows the cell to be imaged at several different resolutions by adjusting the distance between the stage and the beam-source. Figure 9 shows tomographic scans of a PEM fuel cell having conventional Pt/C catalyst layer taken at three different resolutions: (a) equivalent to micro x-ray CT resolution, (b) 100 nm resolution and (c) 50 nm resolution. The ability to image at multiple length scales is especially important for the development of models of the porous structure of the MEA, which must bridge the nano- and micro-scales to prove useful for future MEA design optimization. Furthermore, even after several scans the MEA remained intact with no observable thinning or degradation.

In spite of the improved resiliency to beam damage, the electrochemical performance of the cell still suffers when exposed to the beam, as can be seen by the order of magnitude difference between the current extracted from the cell when under constant irradiation (figure 9(d)), compared to at SSRL where it was irradiated in short bursts (figure 9(b)). This is in line with the theory of an onset dose for beam damage presented by Vaselabadi et al [49] and Lim et al [50]. In spite of the drawback in measured current, the ability to image a cell in an operando mode at nanometer resolution presents a unique opportunity to visualize water in the catalyst layer at the same scale as the finest morphological features found in commonly used catalyst layers. As already discussed, this cannot be done with micro CT as the pore sizes in most fuel cell catalyst layers lie between 20 nm and 60 nm [51].

5. Conclusion

In this paper, we presented the roadmap for the operando cells designed in our group for imaging water in fuel cells with x-ray CT and x-ray radiography. The operando cells presented here enable the visualization of water at both the micro and nano scales. The micro x-ray CT fuel cell underwent five generations of design
iterations to optimize electrical conductivity, mechanical stability, thermal management and compression. This cell has been used to image fuel cells with various catalyst layers with the aim to visualize water inside the fuel cells. Water segmentation was achieved by subtracting an OCV background scan from the data measured at constant current density to reduce number of image phases from three (water, carbon and pore) to just two (water and pore/carbon). Further, the development of a novel operando nano CT fuel cell is detailed including lessons learned and the challenges in x-ray imaging on this scale. The main challenge of beam damage is difficult to overcome due to the low energy <10 keV that many of the beamlines use. We showed that this can be overcome by using beamlines that rely on Kirkpatrick-Baez (KB) mirrors (at ESRF) instead of Fresnel zone plates (APS, NSLS II) for beam focusing, as KB x-ray optics can enable imaging at nanoscale at higher energies (such as 17.5 keV at ESRF). Despite the challenges, the nano- x-ray CT sample holder design enables multiscale 3D imaging.

Acknowledgments

This material is based upon work supported by the Department of Energy, Office of Energy Efficiency and Renewable Energy (EERE), under Award Number DE-EE0007270. This research used resources of the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357. We thank Dr. Vincent De Andrade for the beamtime support at beamline 32-ID at APS and Dr. Xianghui Xiao for the beamtime support at beamline 2-BM at APS. The Advanced Light Source is supported by the Director, Office of Science, Office of Basic Energy Sciences, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231. We thank Dr. Dula Parkinson for beamtime support at beamline 8.3.2 at ALS. Use of the Stanford Synchrotron Radiation Lightsource at SLAC National Accelerator Laboratory is supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences under Contract No. DE-AC02-76SF00515. We thank Dr. Johanna Nelson Weker for beamtime support at beamline 6-2 at SSRRL. The experiments were performed on beamline ID16b at the European Synchrotron Radiation Facility (ESRF), Grenoble, France. We are grateful to Dr. Julie Villanova at the ESRF for providing assistance in using beamline ID16b. We thank Mr. Osvalo Calzada with the help of manufacturing earlier generations of micro CT cell.

ORCID iD

Iryna V Zenyuk • https://orcid.org/0000-0002-1612-0475

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