UNITED STATES DEPARTMENT OF COMMERCE
National Institute of Standards and Technology
Gaithersburg, Maryland 20899-8461

Neutron Imaging Study of the Water Transport in Operating Fuel Cells

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IA016

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Overview

This project started in 2001 as a partnership between NIST, DOE and General Motors to develop methods and infrastructure to non-destructively image water transport in operating fuel cells. Funding from the project has been provided through both DOC and DOE with major contributions to the fuel cell infrastructure from our main testing partners at General Motors.

The role of the NIST imaging facility has been to develop state of the art neutron imaging, maintain a state of the art fuel cell testing infrastructure and to provide these capabilities and services to the public through our user program.

Barriers

(A) Durability

(C) Performance

(D) Water Transport within the Stack

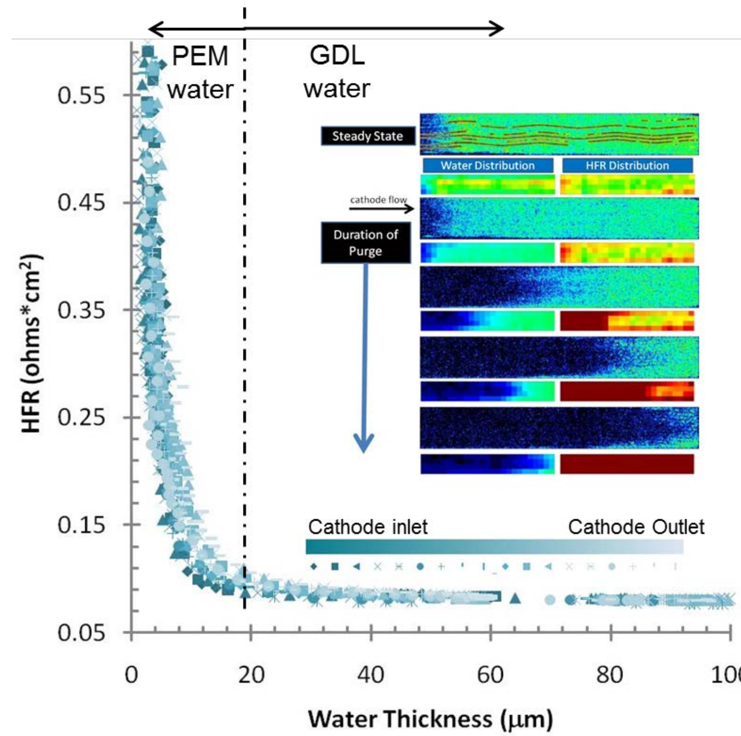
Partners/Users/Collaborators

Project Lead: National Institute of Standards and Technology

- General Motors
- Honda
- Hydrogenics Corp.
- HySA Infrastructure
- NASA, MSFC
- Lawrence Berkeley National Laboratory
- Los Alamos National Laboratory
- NECSA
- Oak Ridge National Laboratory
- Sensor Sciences
- Toyota
- Colorado School of Mines
- Massachusetts Institute of Technology
- Pusan National University
- Rochester Institute of Technology
- University of California, Merced
- University of Connecticut
- University of Hawaii
- University of Michigan
- University of South Carolina
- University of Tennessee
- University of Toronto

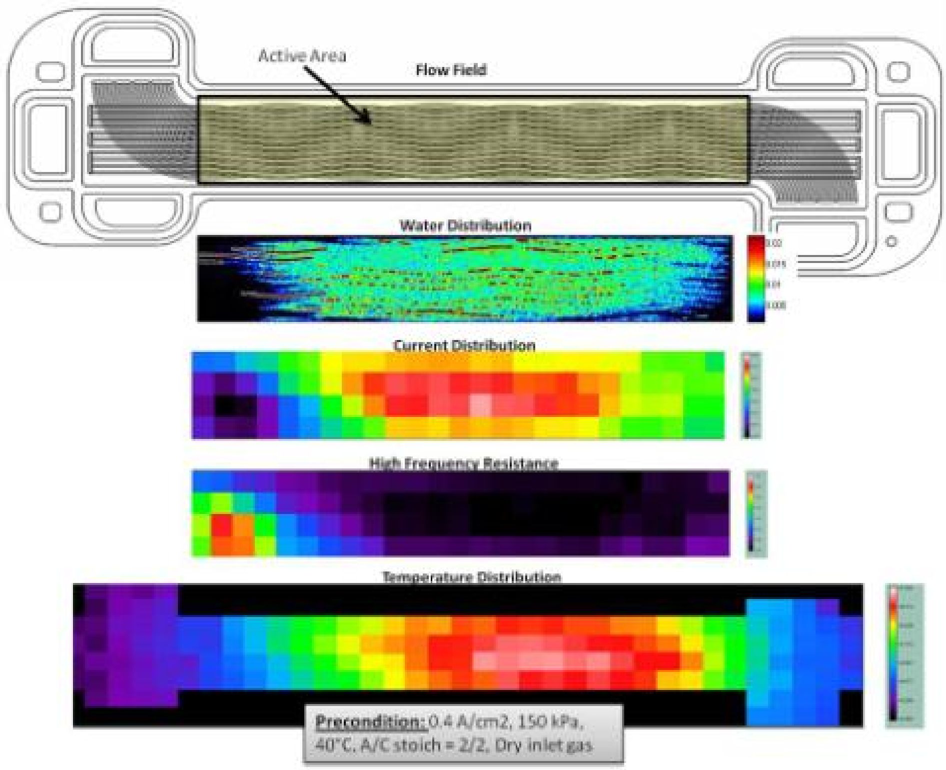
Relevance

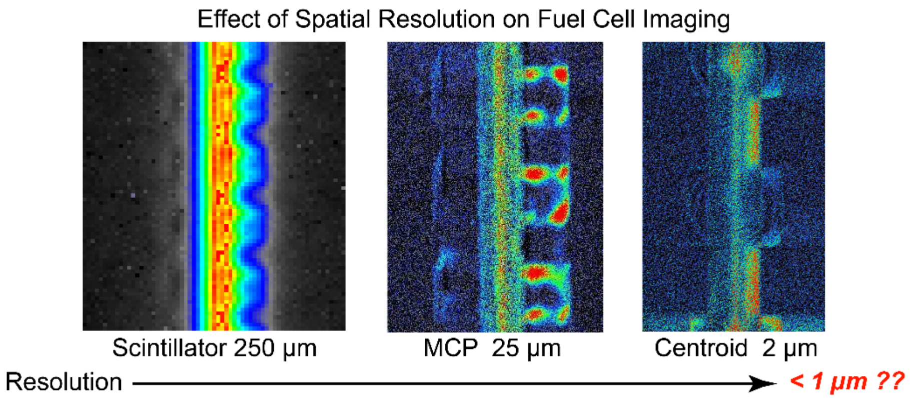
- Neutron imaging is the most powerful and sensitive method to *non-destructively* image water in the fuel cell *in operando* as neutrons readily penetrate common fuel cell hardware yet accurately measure small volumes of liquid water
- This enables one to develop a complete picture of the heat and mass transport in a fuel cell, namely:
  - Dynamic water transport in the flow fields and manifolds
  - Liquid water distribution anode versus cathode
  - Cold start and freeze-thaw effects
  - Catalyst degradation induced by liquid water
  - Catalyst layer liquid saturation level
- Objectives of the project include:
  - Study water transport in single cells and stacks
  - Enable fuel cell community to utilize state of the art neutron imaging capabilities to study water transport phenomena
  - Tailor neutron imaging to needs of the fuel cell community
  - Improve the spatial resolution to provide more detail of the water content in commercial MEAs



Approach

As an example of the method, the data shown below include water content, current and temperature distribution, and HFR measured simultaneously by General Motors at NIST.

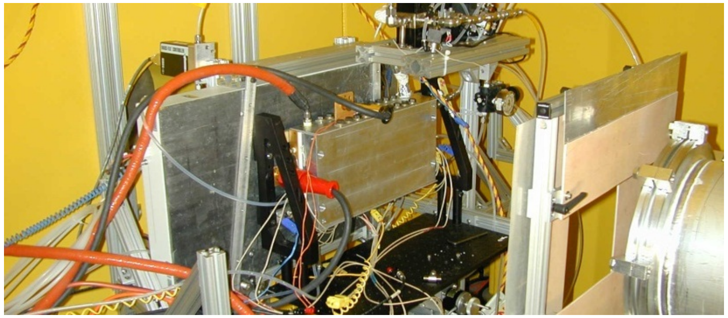




- In order to extend this capability to the catalyst layer, we are engaged in a continuous effort to enhance the image spatial resolution.
- We are constantly improving the image analysis to correct systematic effects and ensure accurate water content measurements.
- We provide to the fuel cell research community open access to state-of-the-art detectors, methods, and analysis techniques.

Approach

- Maintain a national user facility for neutron imaging of fuel cells
  - Develop and maintain state-of-the-art fuel cell testing infrastructure
  - Pursue facility improvements through collaboration and feedback with testing partners at General Motors and the fuel cell community
- Free access for open research
  - Experiments are proposed by users and selected through a peer review process managed by NIST
  - We collaborate as needed, data must be published
  - ***"Mail-in" service for high resolution imaging***
- Fee based access for proprietary research
  - Contact NIST for details
  - Stack developer owns data outright
  - Proprietary users trained to take and analyze image data
- User friendly operation
  - Ample area on beamline for complex setups
  - Can image automotive cells with 26 cm dia. beam
  - Photos show both 50 cm^2 and full size automotive cell
  - Test stands fully integrated with GUI and scripting
  - Image analysis software is tailored to fuel cell user needs

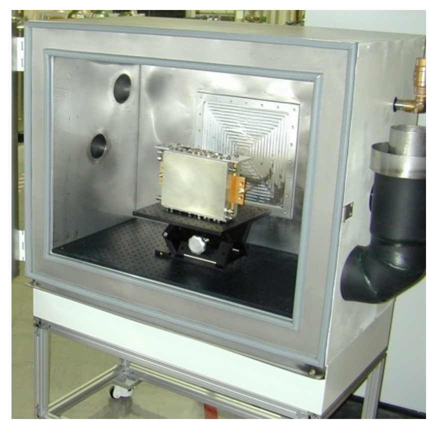


Approach

Fluids:
H_2 (18.8 slpm), D_2 (1.2 slpm), N_2, Air, O_2, He,
DI (18 MΩ/cm)
New H_2 Generator FY14

Large scale test stand: 800 W,
6-1000 A @ 0.2 V
0 V – 50 V,
Liquid coolant
H_2/Air: 11/27 slpm
Contact humidifier (dew pt. 35-85 °C)
First User Data 03/15

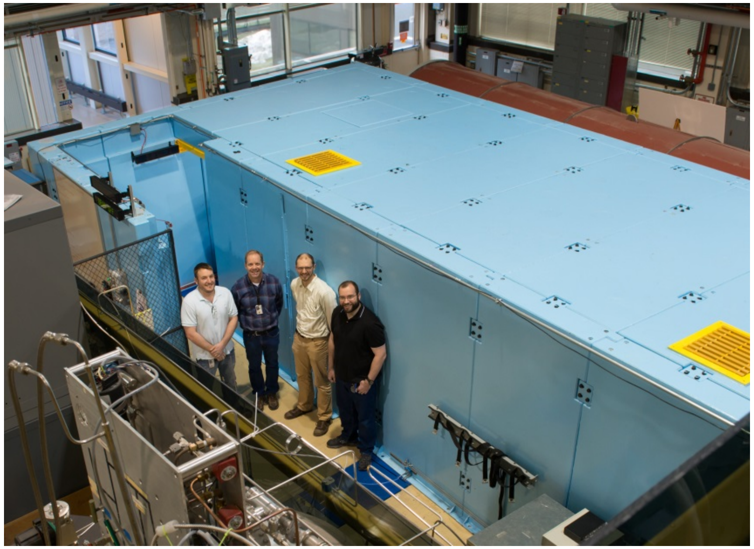
Small scale test stand:
Cell area ≤50 cm^2, dual & liquid temperature control, absolute outlet pressure transducers
2018 coming upgrade:
Full integration of EIS acquisition into scripting



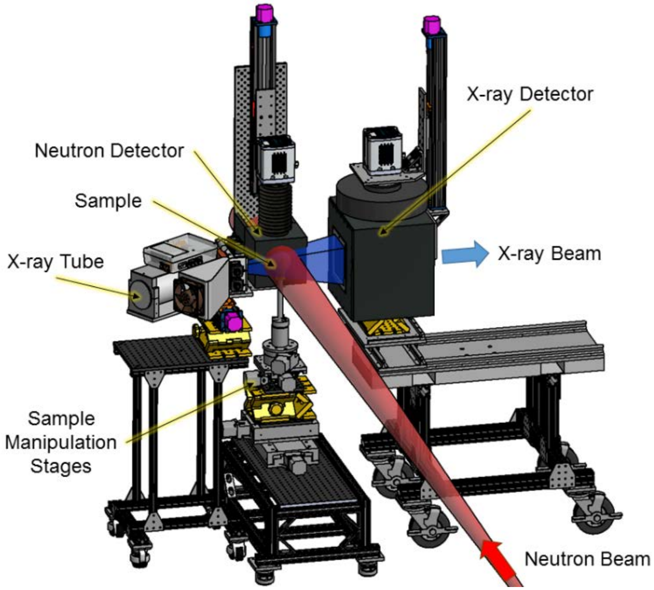
Environmental Chamber:
-40 °C – 50 °C
RH 20-90% above 20 °C
1 kW air cooling at -40 °C
Also available, liquid cooling to -45 °C

Recent Facility Advancements/Milestones

- Cold Imaging Facility Developments
  - Tested Engineering unit for 1:1 Neutron Microscope Lens
  - Installed Hydrogen Gas supply
- Methods to improve image spatial resolution – Ongoing
  - Scintillation light centroiding with 1.5 μm resolution, 2 h image exposure time
  - Neutron microscope project is receiving development support from NIST; delayed due to unexpected production delays at NASA, expect:
    - 20 μm spatial resolution, 10 s time resolution available 2022
- Complementary x-ray imaging system
  - Acquired first simultaneous dual x-ray / neutron tomogram of operating fuel cell
  - New improved design of fuel cell for x-ray neutron tomography has been tested
- User program – Ongoing
  - Procurement of new scriptable EIS
  - 6 new fuel cell proposals from last call for proposals
  - 20 % of open beamtime allocated to Fuel Cell and hydrogen storage experiments
  - Centroid imaging investigation with RIT and GM
  - Results from 2017 AEFC work and electrolysis have been recently published and new work planned



NIST Cold Neutron Imaging Instrument.

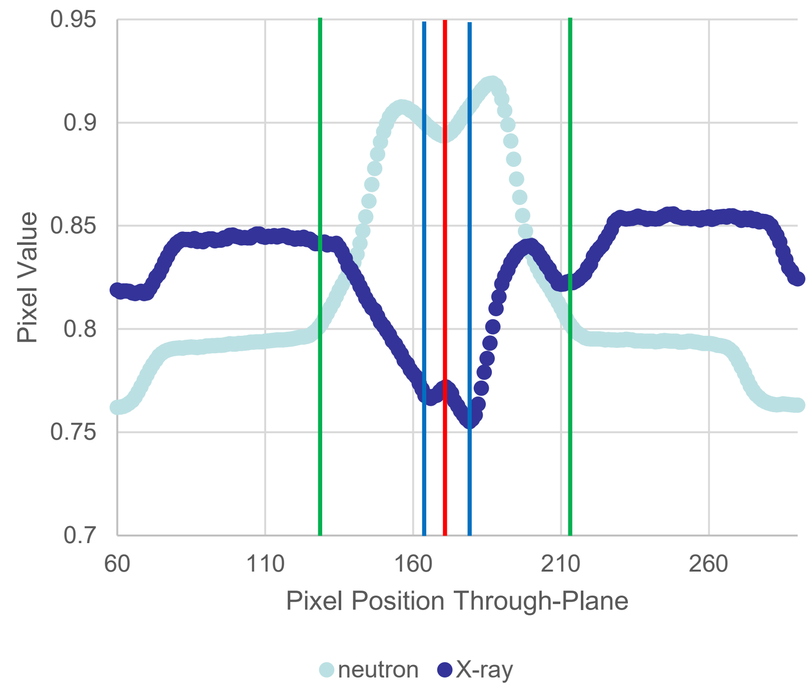


Dual x-ray/neutron tomography

Improving interface detection with addition of X-ray imaging

- Current high resolution neutron imaging provides high sensitivity for water detection
- It is often difficult to know where layer boundaries exist in the neutron images
  - Complicated by the non-planar catalyst layers and membrane
  - Critical to know where boundaries are to know correct porosity values for conversion from water thickness to saturation

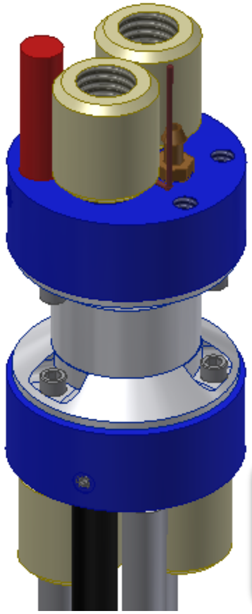
Adding X-ray image facilitates identification of catalyst layer locations.

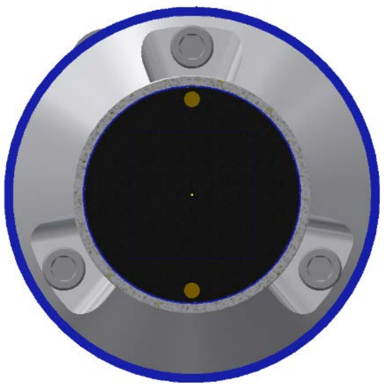


Mass Attenuation Coefficients		
	90 keV x-ray	Thermal neutrons
	[cm²/g]	
H_2O	0.07	5.62
C	0.16	0.37
Al	0.19	0.04
Pt	6.86	0.07

Improving cell design to reduce artifacts

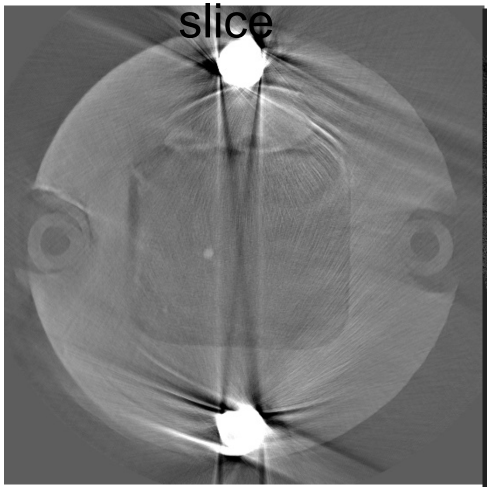
- New design has:
  - Full temperature control
    - Differential control possible
  - Stiffer mounting
    - Reduces blurring
  - Removable flow fields
    - Selectable flow patterns
  - Compact fluid and electrical connections
  - No screws in field-of-view
    - Removes CT artifact
  - Fiducials to track an/ca and inlet/outlet in reconstructions



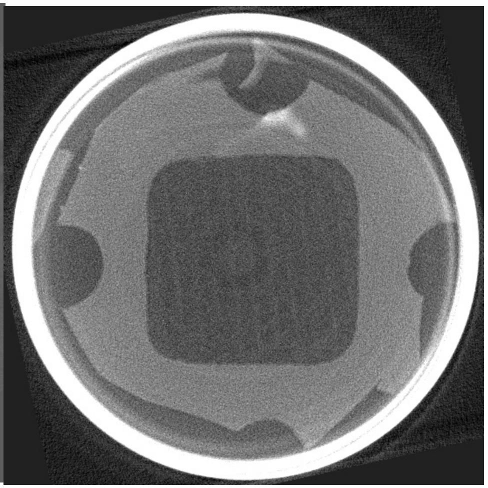


Cell design improvements to remove sources of artifacts and increase testing flexibility

Original design slice

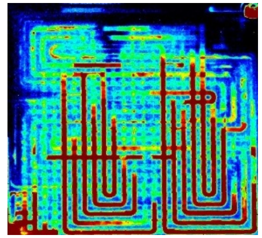


New design slice

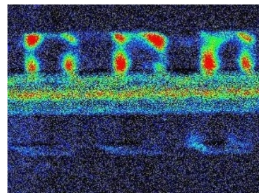


Spatial Resolution Development Timeline

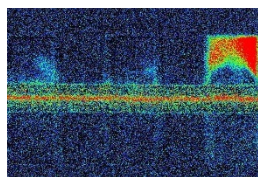
2001: 250 µm



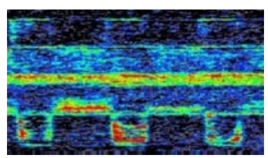
2006: 25 µm



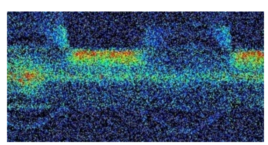
2009: 10 µm



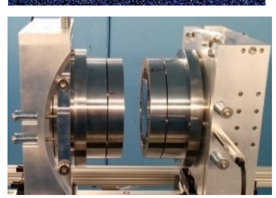
2016: 4 µm w/ slits



2017: 1.5 µm w/ centroiding

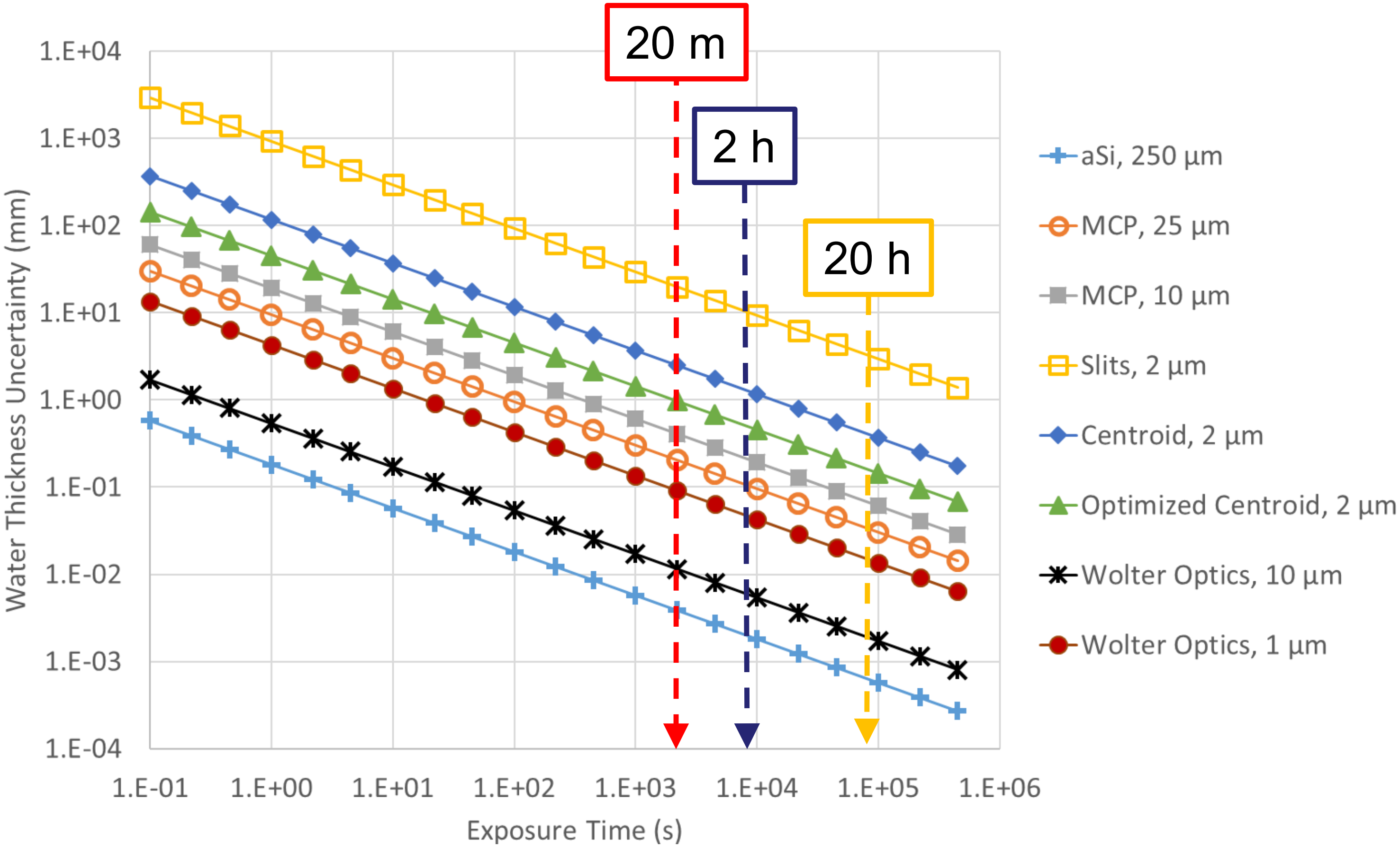


2022: 20 µm w/ Wolter Optics



- 250 µm **(1-30 Hz frame rate)**: In-plane studies of total water content and manifold.
- 25 µm (**20 minut**e): Through plane water distribution to begin GDL transport studies.
- 10 µm (**20 minut**e): More accurate measurement of diffusion media as well as temperature driven phase change flow and thermal osmosis, studies of PGM-free catalyst layers.
- ***To resolve water in state of the art MEAs, need resolution of 1 µm ... at the very least***
- 1.5 µm (**2 hours**): Recent work on centroiding improves resolution but requires long exposure times.
- **Goal 1 µm (20 min):** A neutron lens (Wolter optics) is under development which will improve ***both*** spatial and time resolution. Currently optics are expected to approach 20 µm by 2022.

Liquid water uncertainty for various methods/detectors



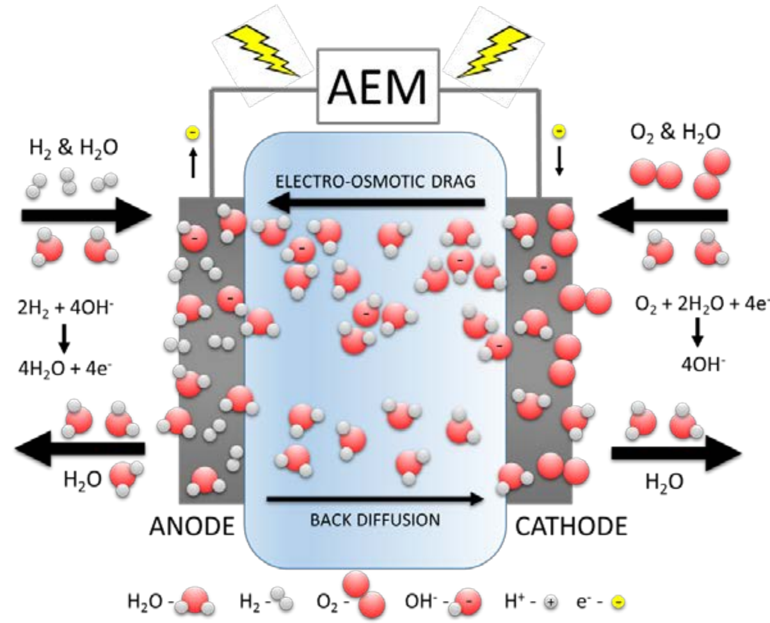
- The exposure time includes dead time, for instance the time to scan a slit
- To reduce exposure time, one can also average over many pixels in the in-plane direction to reduce the water thickness uncertainty in the through-plane direction

Water Accumulation and flooding in AEMFCs

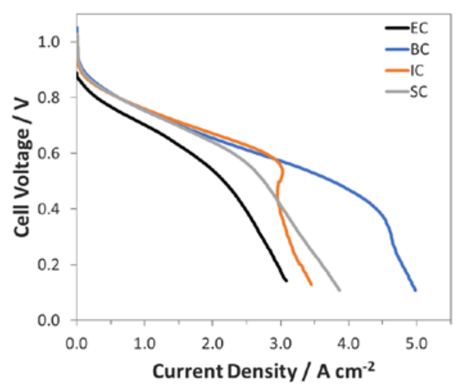
Travis J. Omasta, Yufeng Zhang, Xiong Peng, William E. Mustain - University of South Carolina and UCONN
Andrew M. Park, Bryan S. Pivovar - NREL
Lianqin Wang, John R. Varcoe - University of Surrey

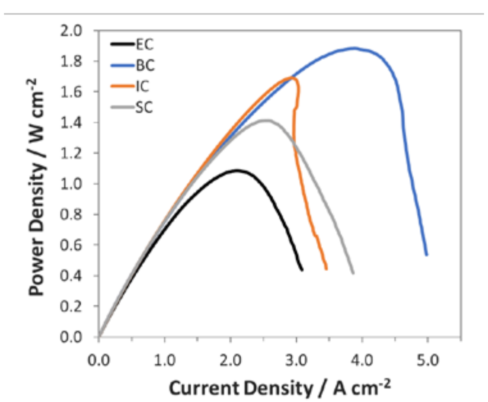
* Omasta, Varcoe, Mustain *et al*., *Energy Environ. Sci.* DOI: 10.1039/C8EE00122G.

- Anion exchange membrane fuel cells (AEMFCs)
- Previously low peak current and power densities believed due to poor water balance
- Record results achieved guided by neutron imaging:
  - Peak Power Density – 1.9 W cm-2
  - Limiting Current Density – 5 A cm-2
- Changes studied:
  - Electrode:
    - Increase water capacity increasing C
    - SC-Standard Carbon, IC-Increased Carbon, BC-Balanced Carbon, EC-Extra Carbon
  - Membrane:
    - ETFE
    - PFAEM



Anode Type:	IC	BC	EC
Pt Loading, mg cm^{-2}	0.47	0.47	0. 50
C Loading, mg cm^{-2}	0.75	1.07	1.76
Carbon weight %	41.4 %	48.0 %	56.0 %
AEI weight %	17.2 %	**20.0 %**	**20.0 %**
AEI:C ratio	***0.417**	***0.417**	0.357

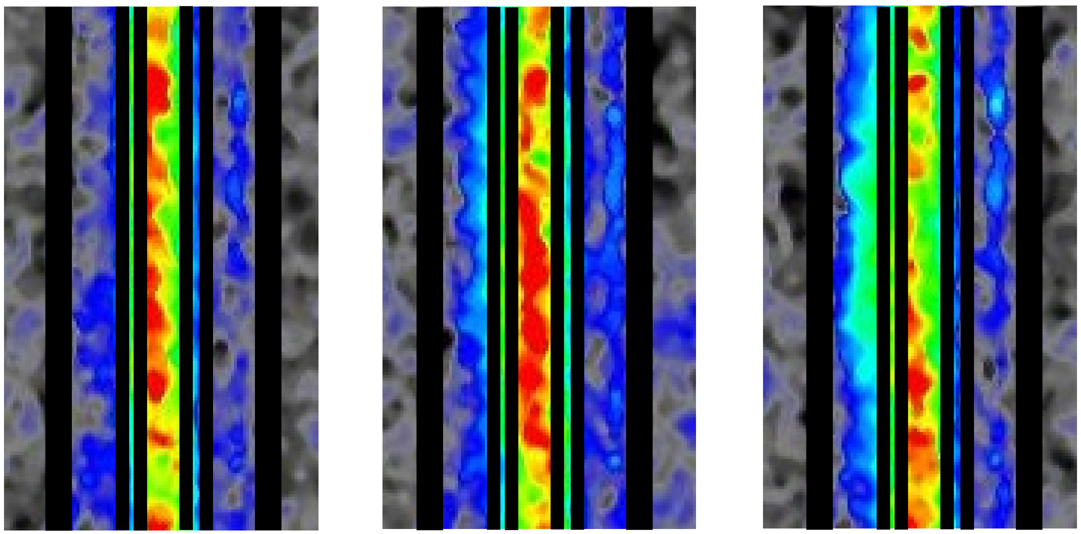


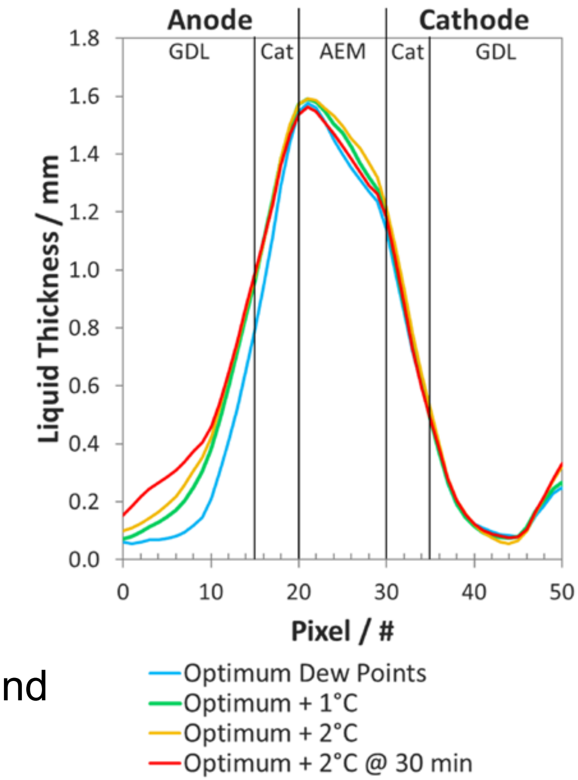


Neutron Imaging of AEMFC
University of Surrey – ETFE-BTMA membrane

Anode – Left; Cathode – Right, Membrane is ~70 μm Compressed GDE is ~150 μm

Optimal Dew Points $1.5\ A\ cm^{-2}$ | **Optimal + 2°C** $1.5\ A\ cm^{-2}$ | **Opt. + 2°C @ 30 min** $0.0\ A\ cm^{-2}$

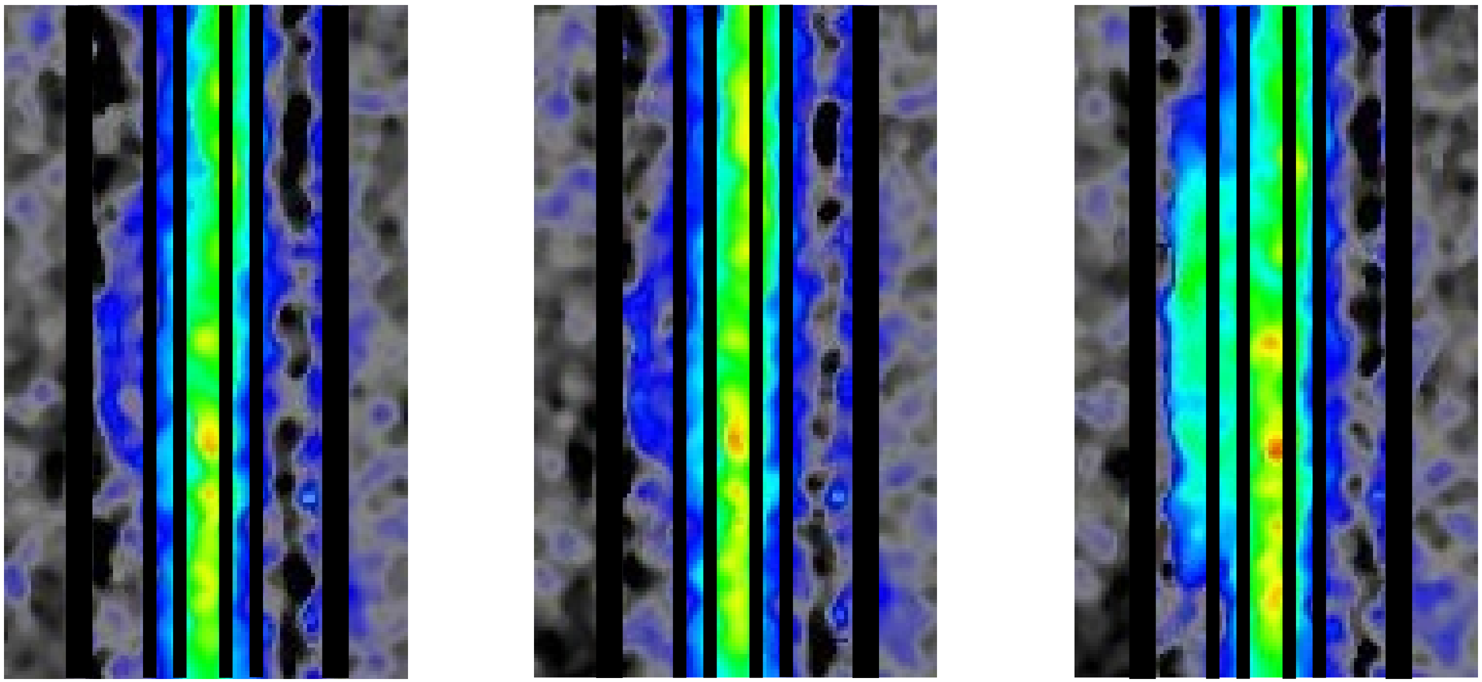




- Increase of gas dew point results in anode GDL flooding and voltage drop
- By 30 minutes of A+2 / C+2 the cell is unable to hold current due to anode GDL flooding
- AEMFC is still VERY sensitive to water
- Decreased membrane hydration observed at flooding location
- Decreased cathode water from reduced back diffusion

Neutron Imaging of AEMFC with NREL – PFAEM / TMA membrane

OA / OC 1.5 A cm^{-2} | A+1 / C+1 1.5 A cm^{-2} | A+2 / C+2 1.2 A cm^{-2}



- Behavior trends the same as the ETFE based membrane
- Overall less water present in the membrane
- More rapid cell crash than with ETFE membrane
- Performance recoverable by lowering current

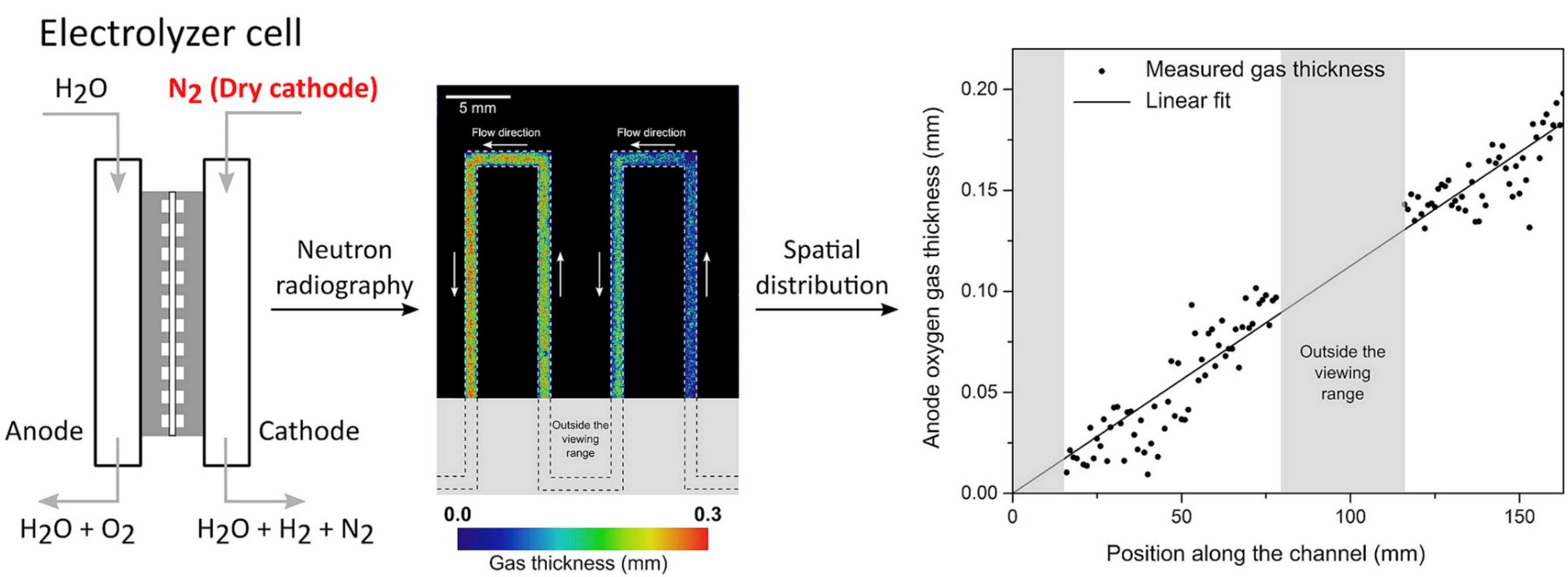
The effect of cathode nitrogen purging on cell performance and in operando neutron imaging of a polymer electrolyte membrane electrolyzer

- Neutrons used to differentiate between anode feed water and cathode water to resolve oxygen gas bubbles in the anode
- Excessive dry nitrogen purging of cathode increase ohmic resistance and decreases OCV
- Neutron visualization allows for optimum cathode nitrogen purge rates determined from visualization of anode oxygen gas distribution as a function of current density

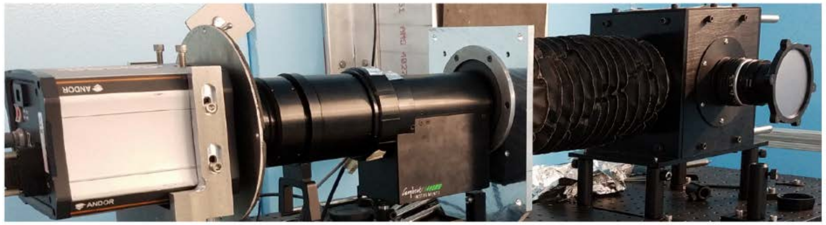
ChungHyuk Lee, Rupak Banerjee, Nan Ge, Jason Keonhag Lee, Benzhong Zhao, Aimy Bazylak
University of Toronto Institute for Sustainable Energy

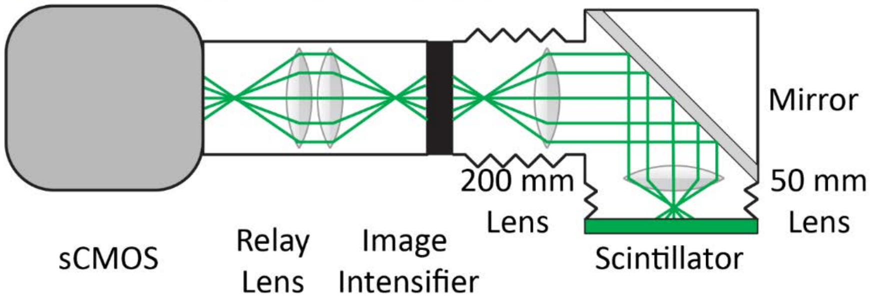
Rami Abouatallah, Rainey Wang
Hydrogenics Corporation, Advanced Stack Technology

Lee et al, DOI: 10.1016/j.electacta.2018.05.066

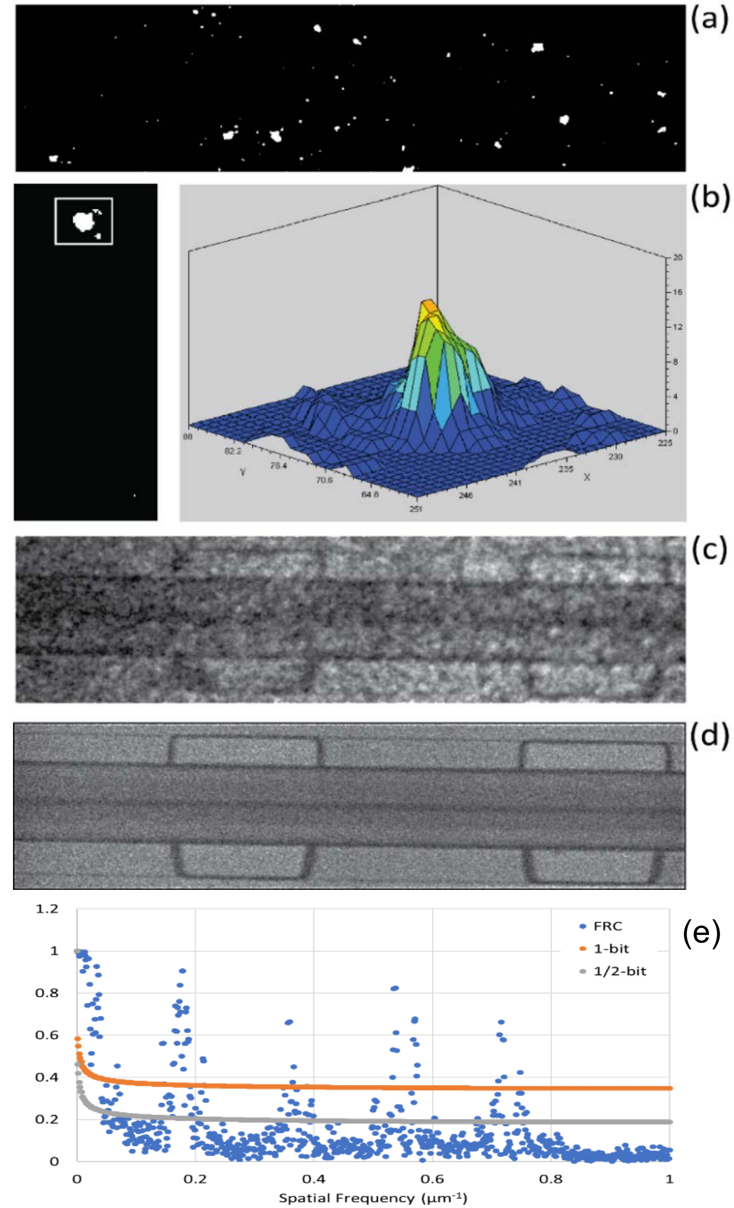


Centroid Imaging, 1.5 µm Spatial Resolution





a) Capture Frames with short exposure times (3 ms) to see individual neutron capture events
b) Close up of a typical scintillation light bloom
c) Sum of 3000 frames after thresholding only
d) Sum of 80k frames and centroiding
e) Fourier Ring Correlation which shows spatial resolution of ~1.5 µm

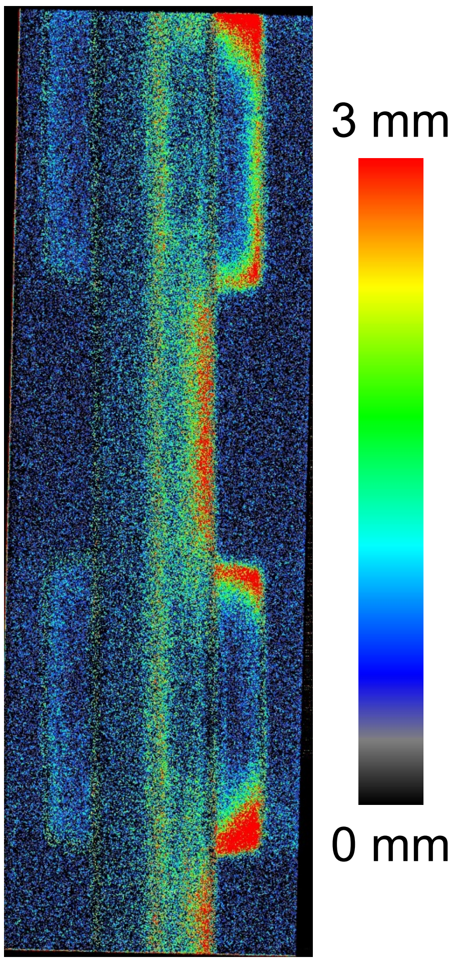


Evaluation of Centroiding Method for Imaging Fuel Cell Water Content

Yang Yue, Thomas Trabold,
Rochester Institute of Technology
Joseph Fairweather,
General Motors
R. Mukundan, R. Borup
LANL

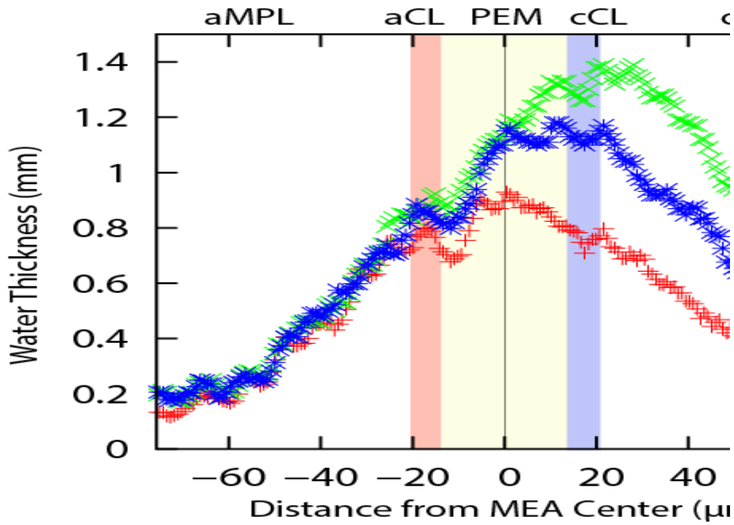
- The square fuel cell active area was 1cm2, with both anode and cathode flow fields comprised of five straight parallel channels, each with width and depth of 1 mm and 254 μm, respectively.
- Anode and cathode land widths were 1 mm. The fuel cell materials listed below were provided by General Motors to LANL, where they were then die-cut prior to shipment to NIST for the cell build.
- MEA: MEA-7992, ESR15-2048, Type 2/XL 4a
- GDL: SGL 24BC
- Gasket: Stacked 2 mil and 5 mil PTFE
- The total thickness of the soft goods assembly was (7 mil) * 2 + 20 μm = 376 μm.

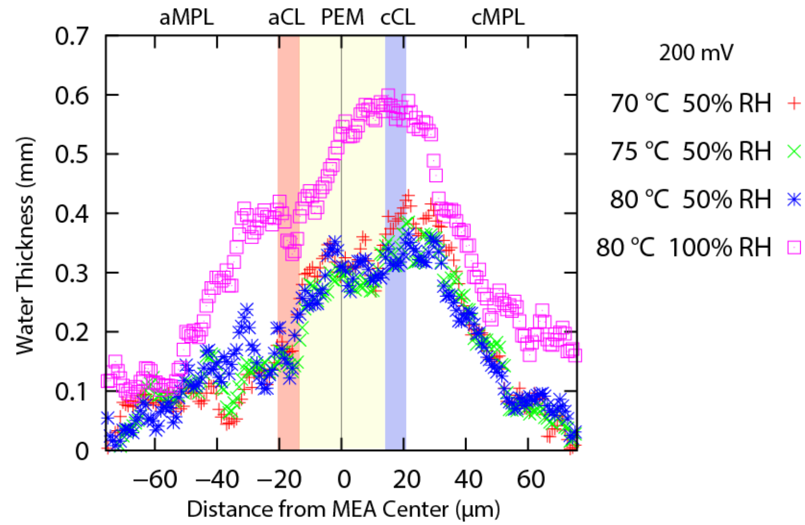
1.5 μm Spatial Resolution

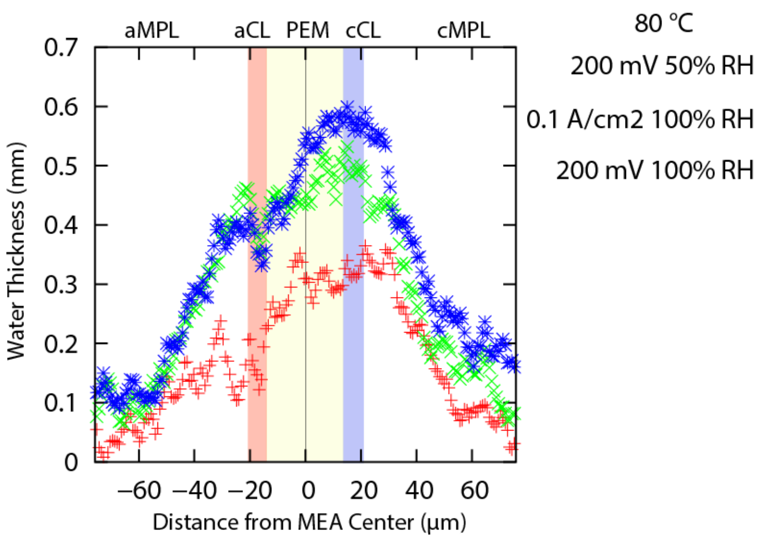


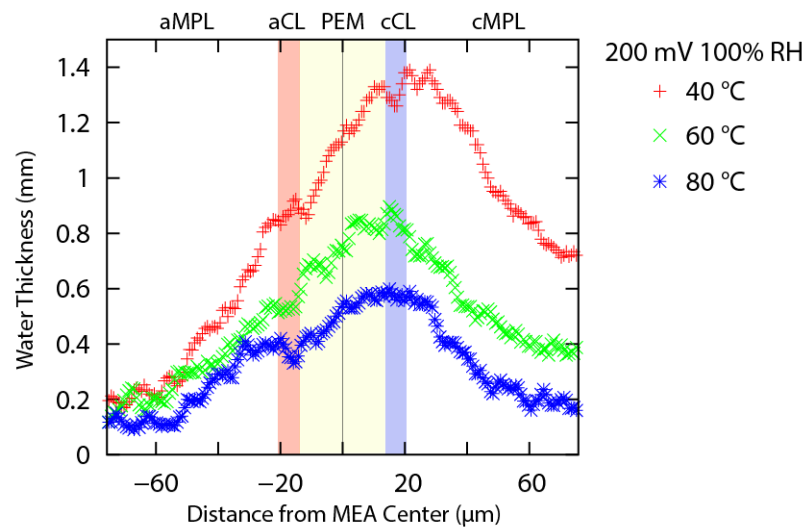
Summed Through Plane Water Thickness Profiles

1.5 µm Spatial Resolution









Future Work

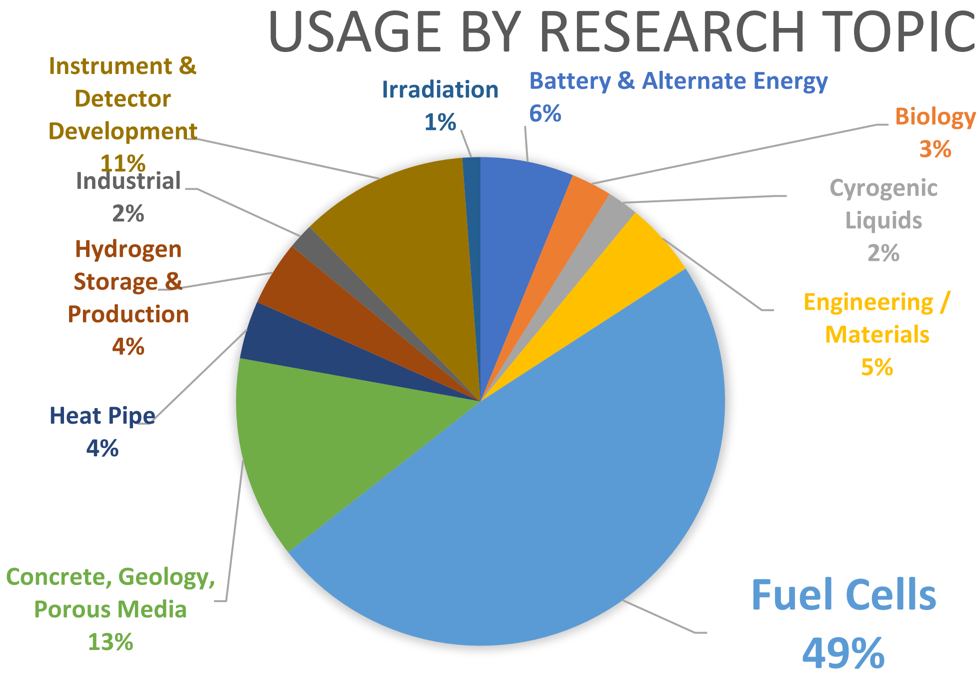
- Centroiding
  - Pursue detector package optimization that reduces light losses and incorporates hardware based centroiding to:
    - Reduce deadtime to near zero
    - Provide images in real time
  - Fabricate planar test section to avoid resolution issues due to fuel cell
- Fuel cell testing infrastructure at cold neutron imaging
  - Model hydrogen release for facility safety analysis
  - Install hydrogen infrastructure at cold imaging facility to support fuel cell experiments
  - Identify test stand for use at the new facility
- Neutron microscope
  - NIST is fully funding upgrades to neutron guides that will enhance image intensity with the new neutron microscope
  - Work with NASA and collaborators to fabricate 1:1 optic
  - First tests of the 1:1 optic are expected 2022
- Last Call For Proposals: 6 new fuel cell related proposals
- Any proposed future work is subject to change based on funding levels

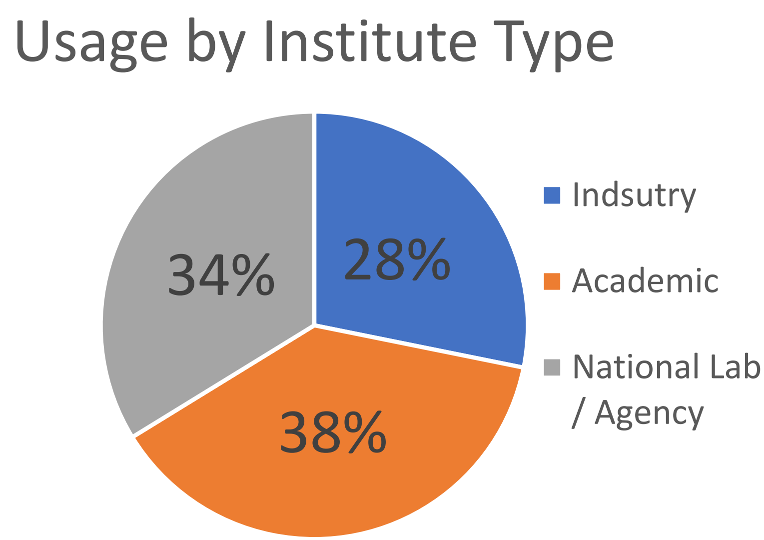
Summary

- We have made excellent progress towards measuring liquid saturation values in the catalyst and membrane
  - Centroiding 1.5 μm resolution demonstrated
    - Method has been improved using available resources
    - Available through the user program
    - Future: develop hardware based centroiding to
      - Improve throughput by reducing acquisition time from 2 hours current to 20 minutes
      - Allow real time image reconstruction versus current 1.5 day post acquisition reconstruction time per image.
  - Wolter optics
    - NIST will design and fabricate mechanical lens fixtures for beam line
- Interface identification porous media
  - New in operando x-ray imaging capability will allow higher resolution studies of porous materials with in operando neutron measurement of water transport
  - Due to the complimentary nature of x-rays we will be able to easily distinguish interfaces in the MEA using x-ray tomography
  - Critical for fully interpreting high resolution neutron water transport images.
- User program
  - User proposals has increased due to interest in new spatial resolution
  - New cold imaging facility is currently being upgraded to include full support
  - Including EIS into the scripting of the test stand will be a great benefit to the users

Broad Impact of Program

- Total Number of Beam Days: **1931**
- **416** Unique Experiments
- **40** Patents (Fuel Cells only)
- **200** papers
- **66** PhD Students





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Thanks to

Nancy L. Garland

DOE Technology Manager

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