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

PI: David Jacobson

Co-PIs:

Daniel S. Hussey
Jacob M. LaManna
Eli Baltic

Physical Measurement Laboratory
National Institute of Standards and Technology
Gaithersburg, MD 20899

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 measure *in operando* the water transport in PEMFCs.

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. Neutron imaging has impacted fuel cell research from TRL 1 to TRL 7.

Presently, NIST maintains and operates the facility, with assistance from GM.

Barriers

(A) Durability

(C) Performance

(D) Water Transport within the Stack

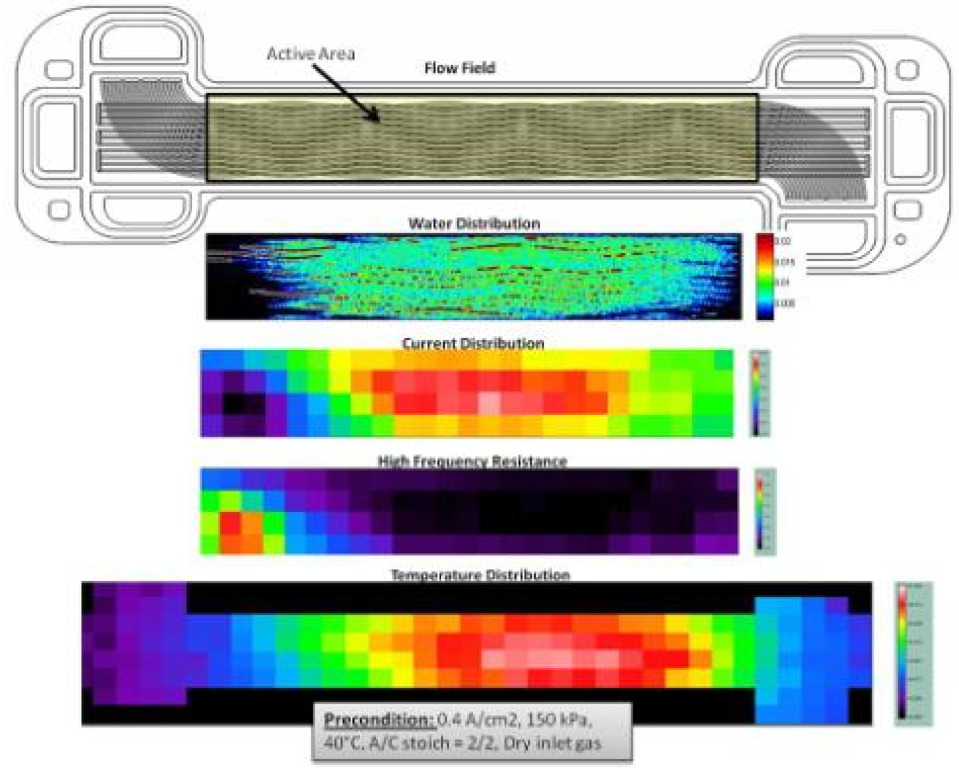
Partners/Users/Collaborators

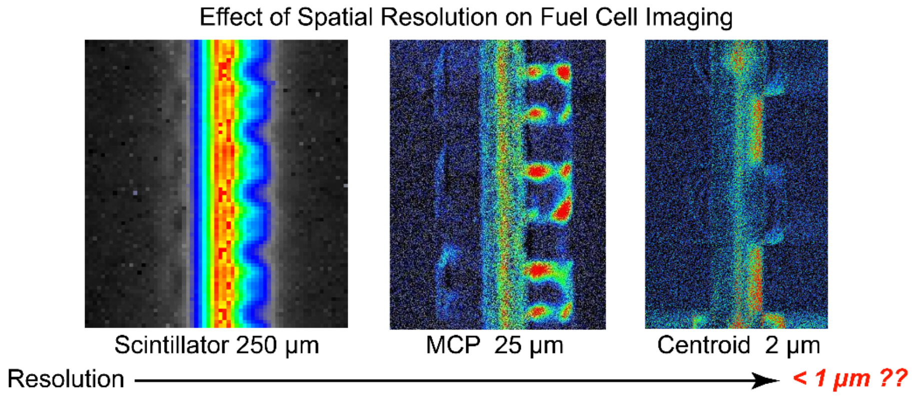
Project Lead: National Institute of Standards and Technology

- General Motors
- Honda
- Hydrogenics Corp.
- 3M
- HySA Infrastructure
- NASA, MSFC
- Lawrence Berkeley National Laboratory
- Los Alamos National Laboratory
- NECSA
- Oak Ridge National Laboratory
- Sensor Sciences
- Toyota
- CEA
- NREL
- 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
- University of Surrey
- Vanderbilt

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.

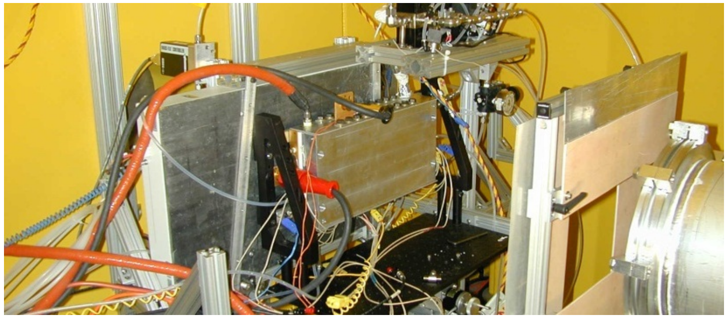




- In order to extend this capability to the catalyst layer, we are engaged in a continuous effort to enhance the image spatial resolution.
- We are constantly improving the image analysis to correct systematic effects and ensure accurate water content measurements.
- We provide to the fuel cell research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



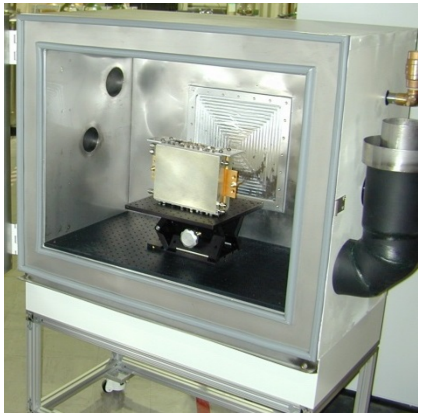
Approach

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

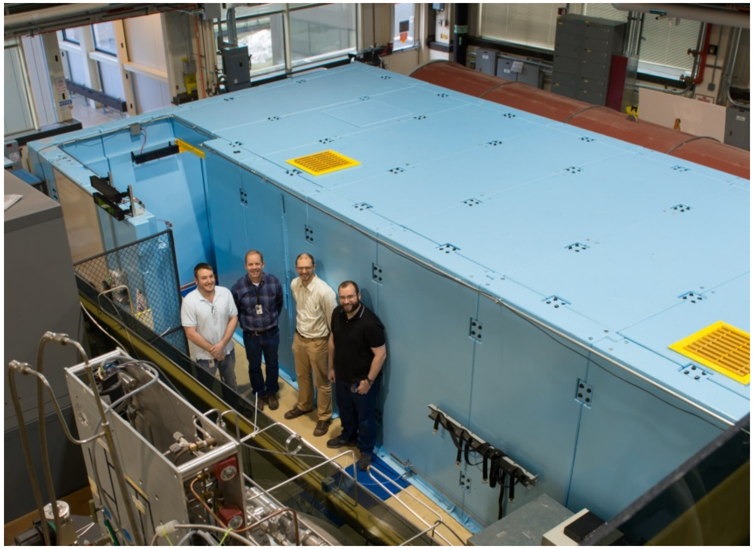
Full integration of EIS acquisition into scripting



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

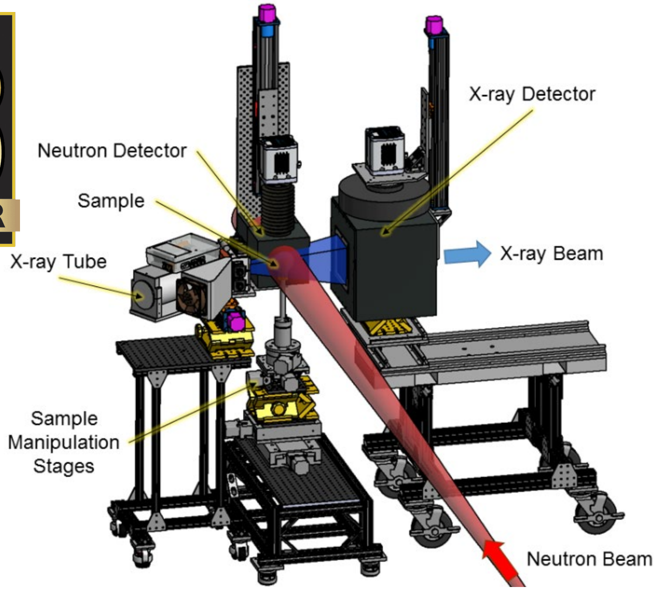
Recent Facility Advancements/Milestones

- Cold Imaging Facility Developments
  - Research focus is neutron imaging method development, including Bragg edge, phase, and magnetic imaging, will house the Wolter optic microscope in 2022
  - New guides installed 2023-2024 will increase the flux by factor 3 and eliminate fast neutron and gamma background
- Methods to improve image spatial resolution – Ongoing
  - Scintillation light centroiding with 1.5 μm resolution, 2 h image exposure time
  - Improved image reconstruction time from 1.5 days to 4 hours
  - 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
  - 2018 R&D 100 award winner.
  - Developments in improved volume reconstruction quality, registration and segmentation
- User program – Ongoing
  - New scriptable EIS is available for facility users
  - 6 new fuel cell proposals from last call for proposals
  - 70 % of open beamtime allocated to Fuel Cell and hydrogen storage experiments
  - Beam time limitations have forced sidelining of some proposals
  - Centroid imaging major strides towards real time detector
  - Results from AEMFC work highlighted by the Editorial Board of Energy & Environmental Science as a Hot manuscript in 2018, Omasta et al, DOI: 10.1039/C8EE00122G



NIST Cold Neutron Imaging Instrument.

2018 R&D 100 WINNER

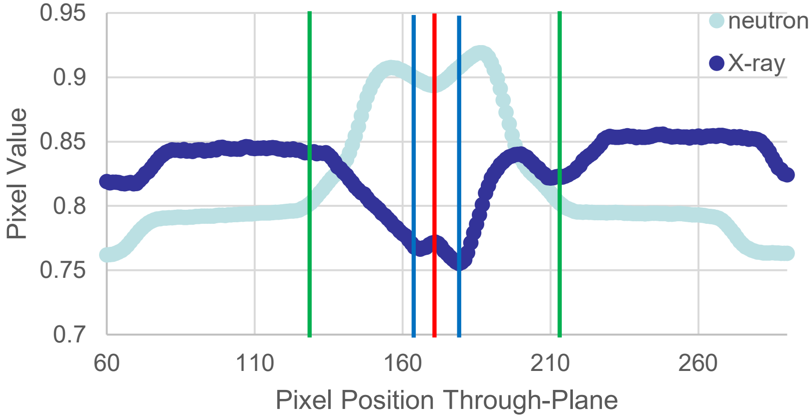


Dual x-ray/neutron tomography

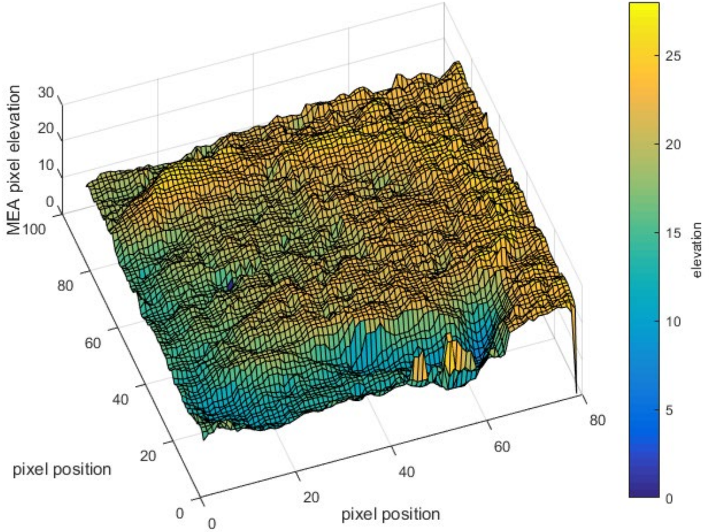
Improving interface detection with addition of X-ray imaging

- 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

2D profiles



3D Topology of MEA



Slice through 3D reconstruction, white area is CCM

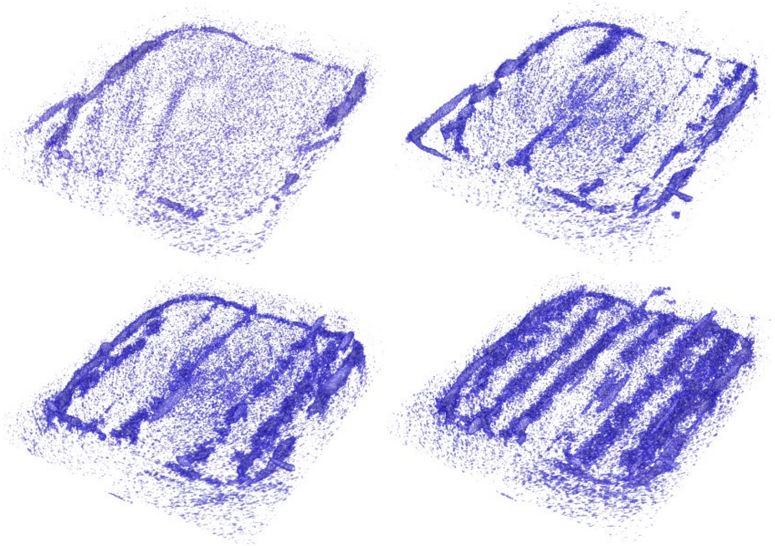
The topology of the CCM can be tracked and correlated to the interface boundary for the GDL

3-Dimensional, in-operando water distribution

2018 R&D 100 WINNER

0.1 Acm^{-2}
0.790 V
25.1 °C

0.5 Acm^{-2}
0.631 V
26.5 °C



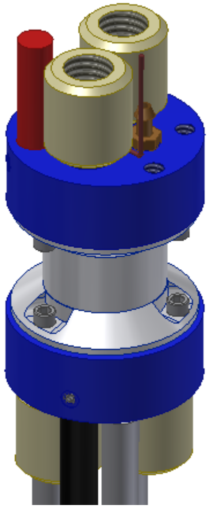
1.0 Acm^{-2}
0.520 V
28.4 °C

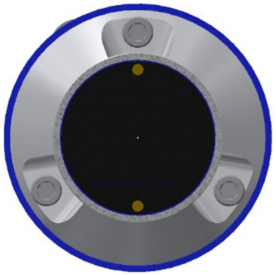
0.200 V
1.9 Acm^{-2}
34.0 °C

6 mm x 6mm active area, constant 200 sccm anode and cathode flow, 25.0 °C dew point

Neutron tomography captures water distribution variation across operating conditions even at long scan times

- New design has:
  - Full temperature control
  - Stiffer mounting
  - Selectable flow field patterns
  - Improved 3D reconstruction quality
  - Fiducials to track an/ca and inlet/outlet in reconstructions

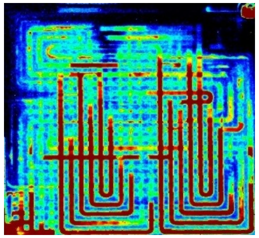




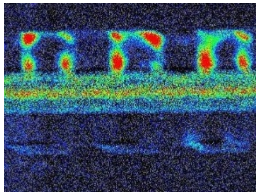
Cell design improvements to remove sources of artifacts and increase testing flexibility

Spatial Resolution Development Timeline

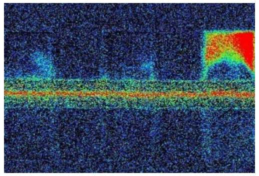
2001: 250 µm



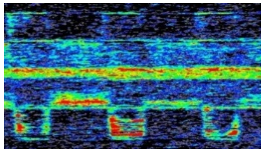
2006: 25 µm



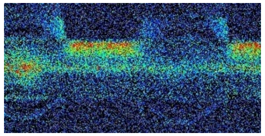
2009: 10 µm



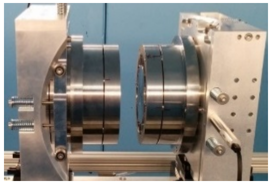
2016: 4 µm w/ slits



2017: 1.5 µm w/ centroiding

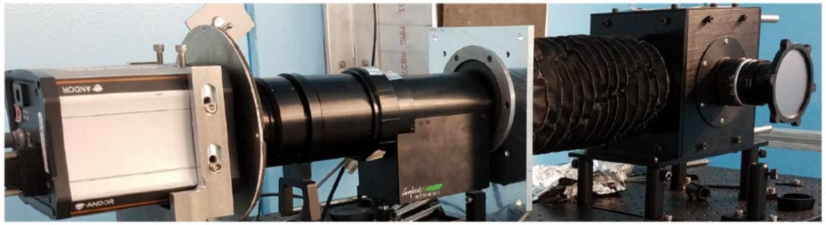


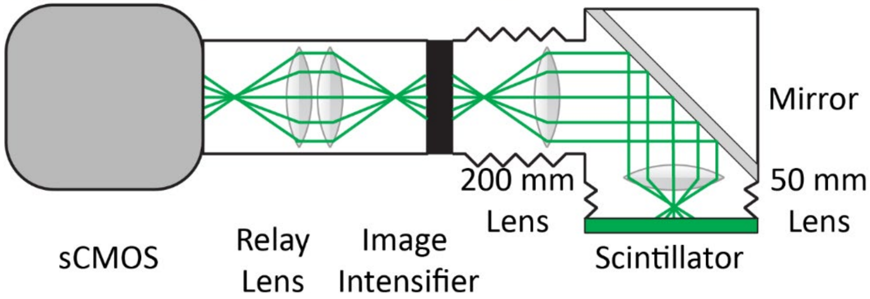
2022: 3 µm w/ Wolter Optics



- 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 is steadily improving exposure and image reconstruction times.
- **Goal 3 µm (20 min):** A neutron lens (Wolter optics) is under development which will improve ***both*** spatial (by factor 7) and time resolution (by factor 1000). Optics are expected to approach 3 µm by 2022.

Centroid Imaging, 1.5 μm Spatial Resolution





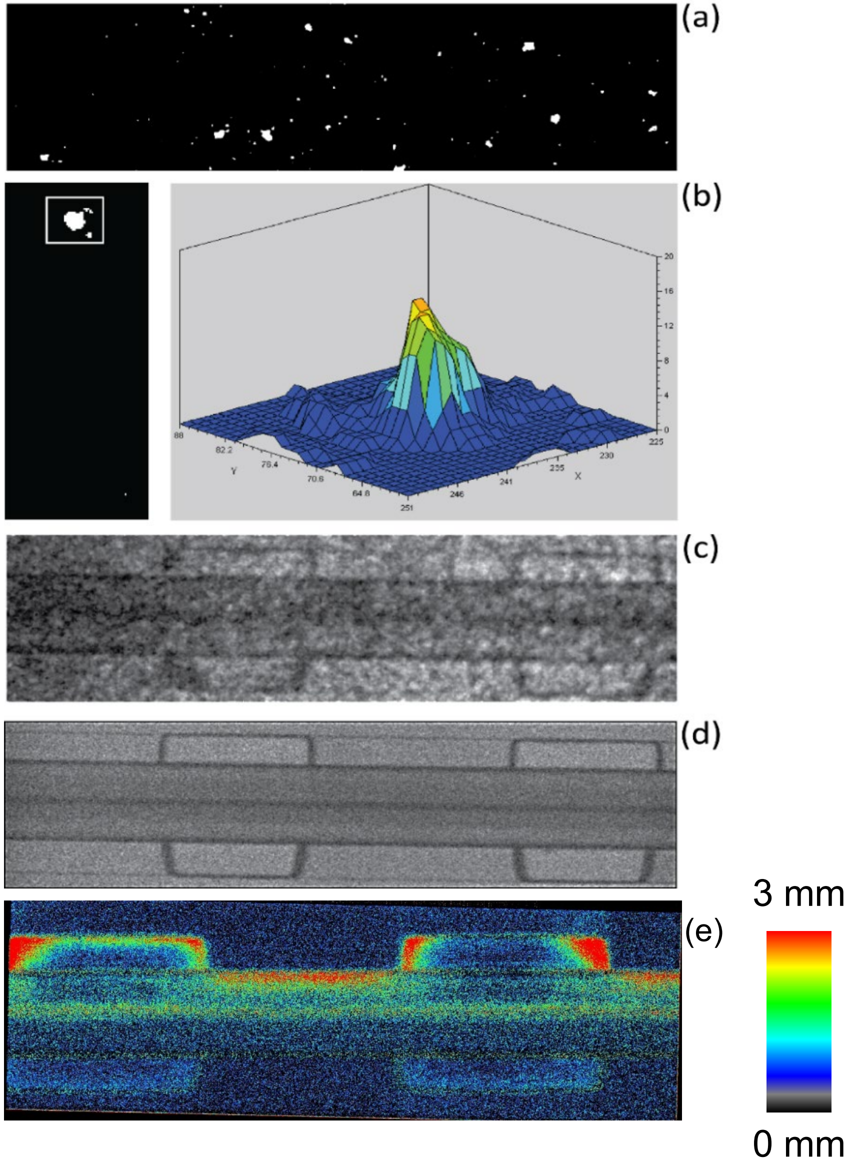
a) Use short exposure times (3 ms) to resolve individual neutron capture events
   - ➢ *30 Hz rate means ~90% deadtime*

b) Close up of a typical scintillation event

c) Sum of 3k frames after thresholding only

d) Sum of 80k frames and centroiding
   - ➢ *Reconstruction in software after acquisition, rate is 2 Hz*

e) Fuel cell imaged with 1.5 μm spatial resolution
   - ➢ *20 min “livetime”, 1.5 day reconstruction*

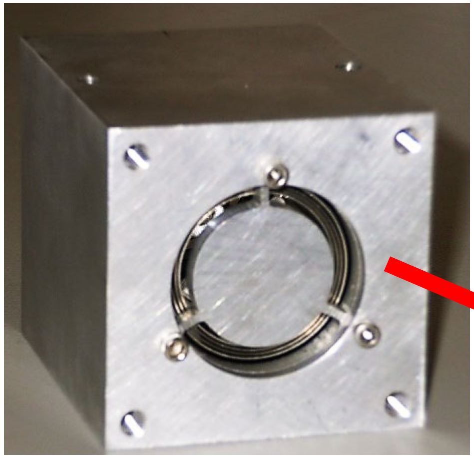


Future Work

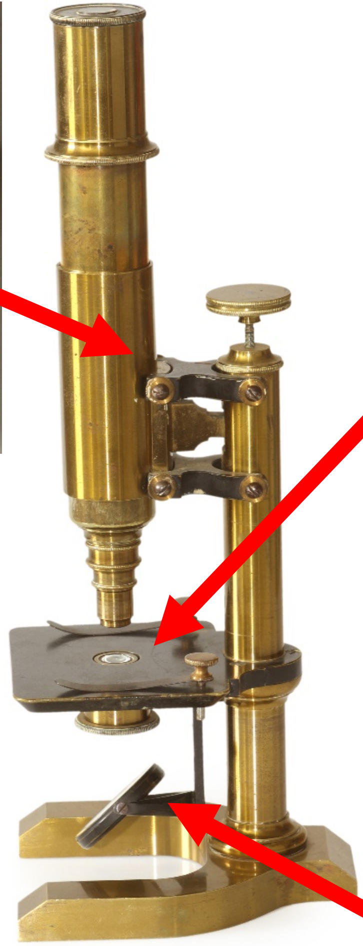
- Centroiding
  - Implemented reconstruction on GPU for 3x faster reconstruction time from disk
  - New camera with 100 Hz frame rate couple with GPU code would reduce deadtime to 70% and produce images in real time.
  - With sufficient funding, a new system, using existing technology, would have no deadtime and reconstruction the image in real time
- Fuel cell testing infrastructure at cold neutron imaging
  - With sufficient funding, install hydrogen infrastructure at cold imaging facility to support fuel cell experiments using Wolter optics
  - 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
  - First tests of the microscope are expected 2022
- In the most recent call for proposals: 6 new fuel cell related proposals
- Any proposed future work is subject to change based on funding levels

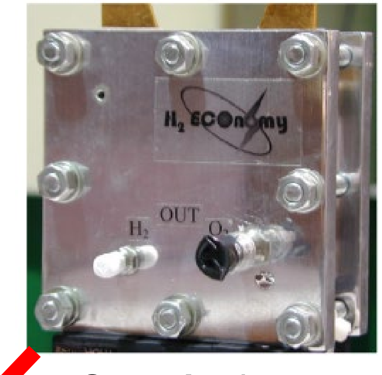
Wolter optics design

Objective optic:
Will be about 20 cm long,
4 to 10 cm in diameter,
consist of 10 shells,
magnify the image by 7x
and have 3 μm resolution



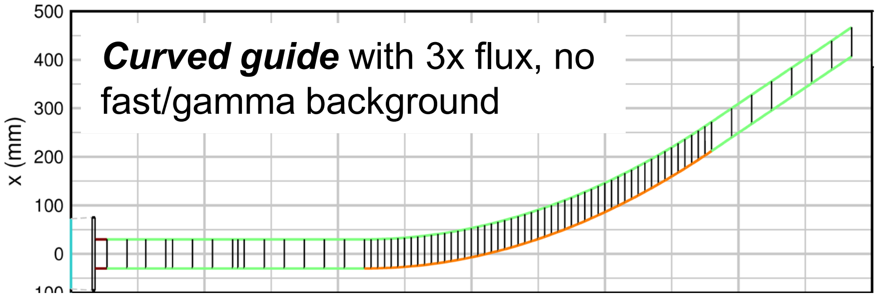
Prototype optic consisted of 3 shells, 3 cm in diameter, 6 cm in length, M=4 and 100 μm resolution





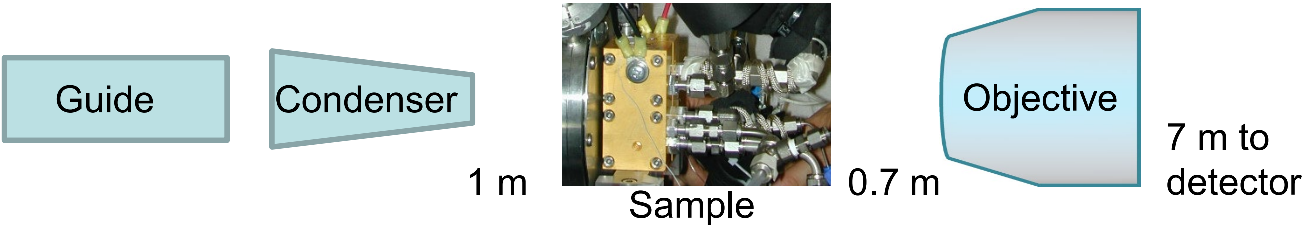
Sample sits at focus of objective, FOV is about 1 cm

Condenser optic: a standard x-ray telescope (5 shells, 60 cm long, 10 cm diameter) that will concentrate ~1% of guide into objective for 1000 flux



Curved guide with 3x flux, no fast/gamma background

Microscope Beamline Layout



Guide

Condenser

1 m

Sample

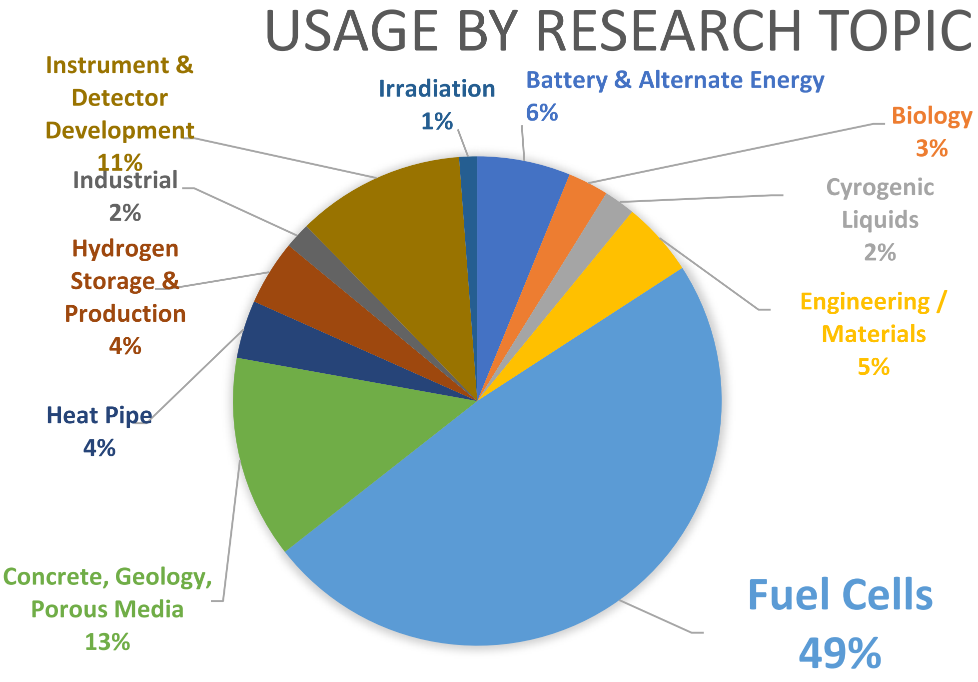
0.7 m

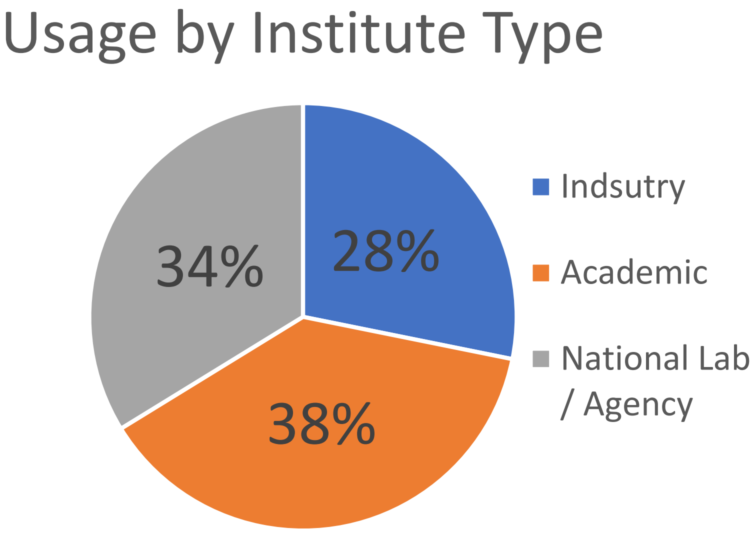
Objective

7 m to detector

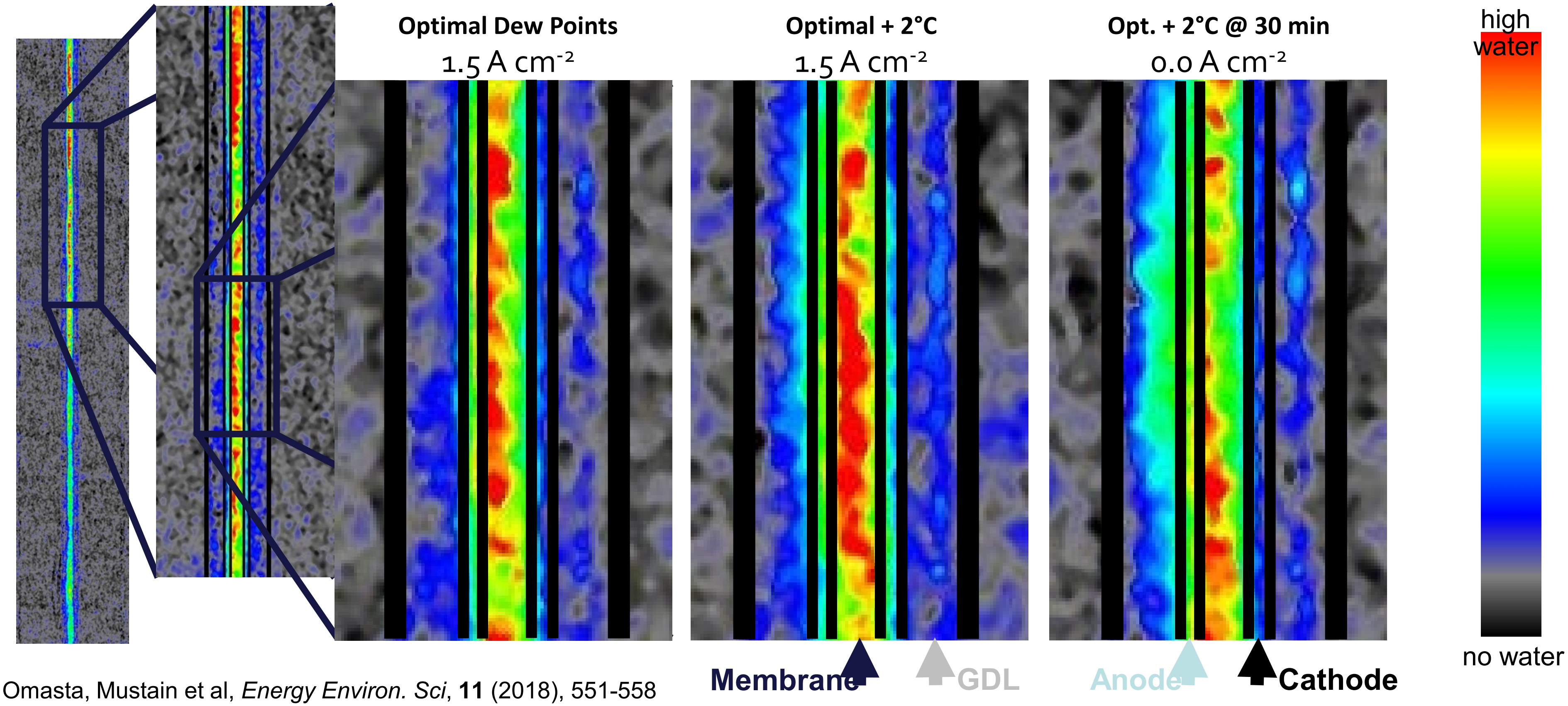
Broad Impact of Program

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



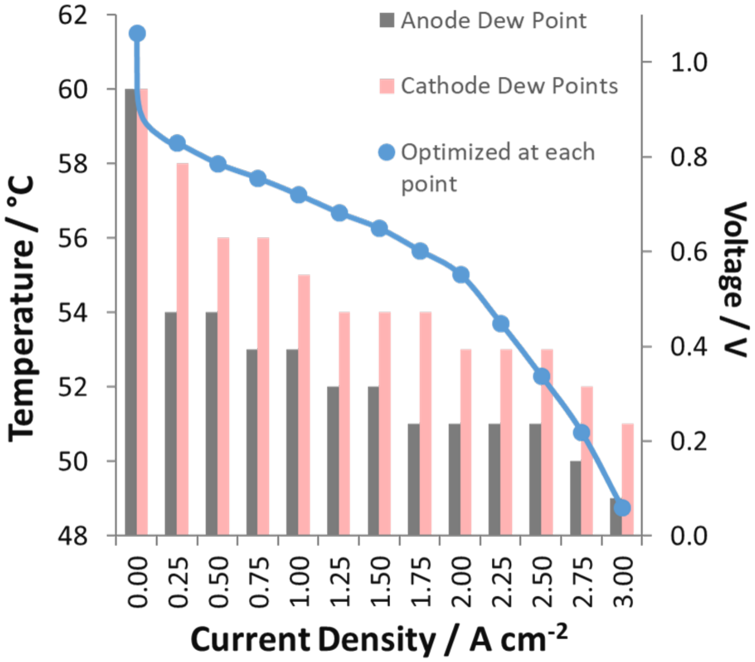


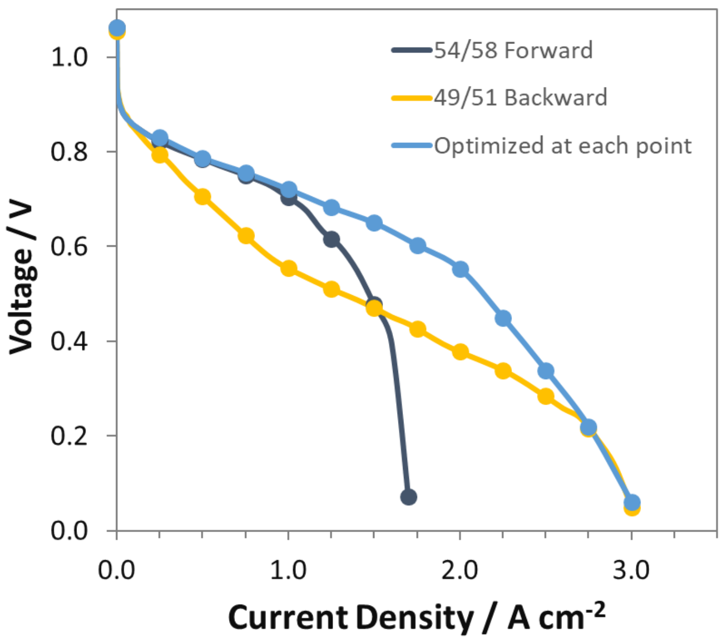
Neutron Imaging of Operating AEMFCs
Univ. of South Carolina,Univ. of Conn., NREL
Univ. of Surrey – ETFE-BTMA membrane



- Omasta, Mustain et al, *Energy Environ. Sci*, **11** (2018), 551-558

Dynamic (Point-by-Point) Optimization of Reacting Gas Water in AEMFCs

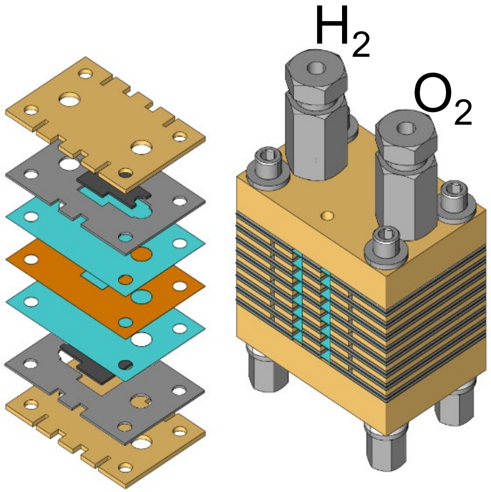




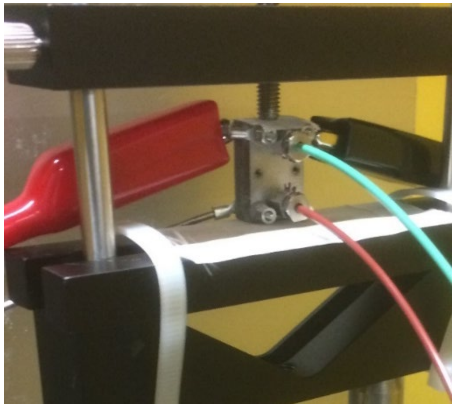
- Each condition sustained for 30 min

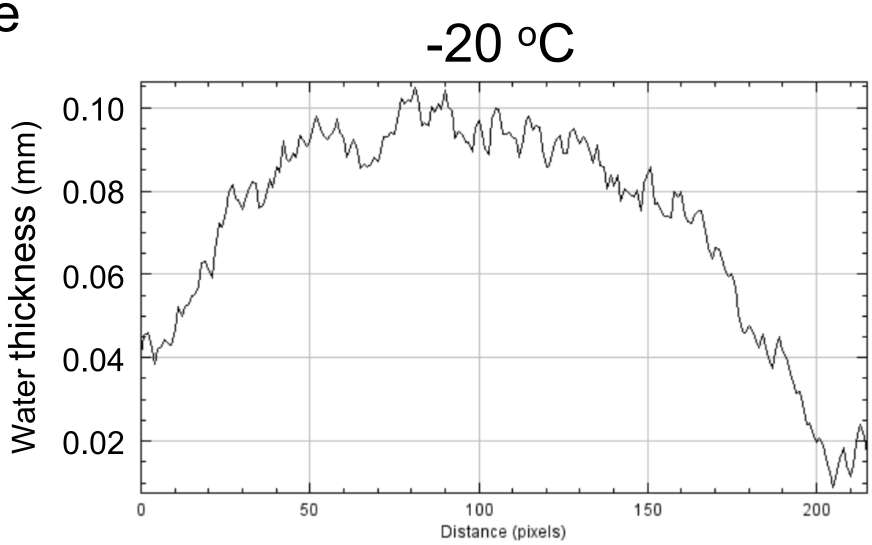
LANL/DOE (NNSA) Miniature Fuel Cell

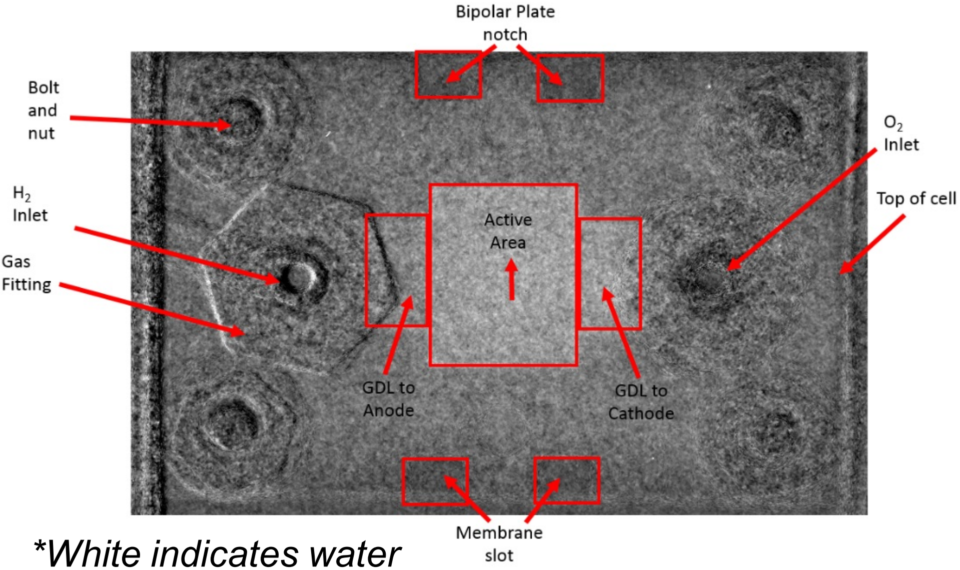
Stack with passive water management; Operation down to -55^{o}C



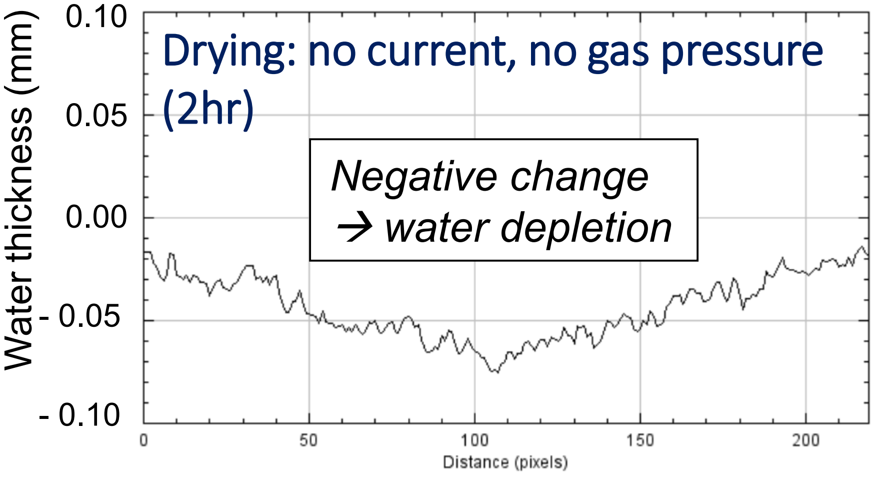
Stack in Beam Line







*White indicates water

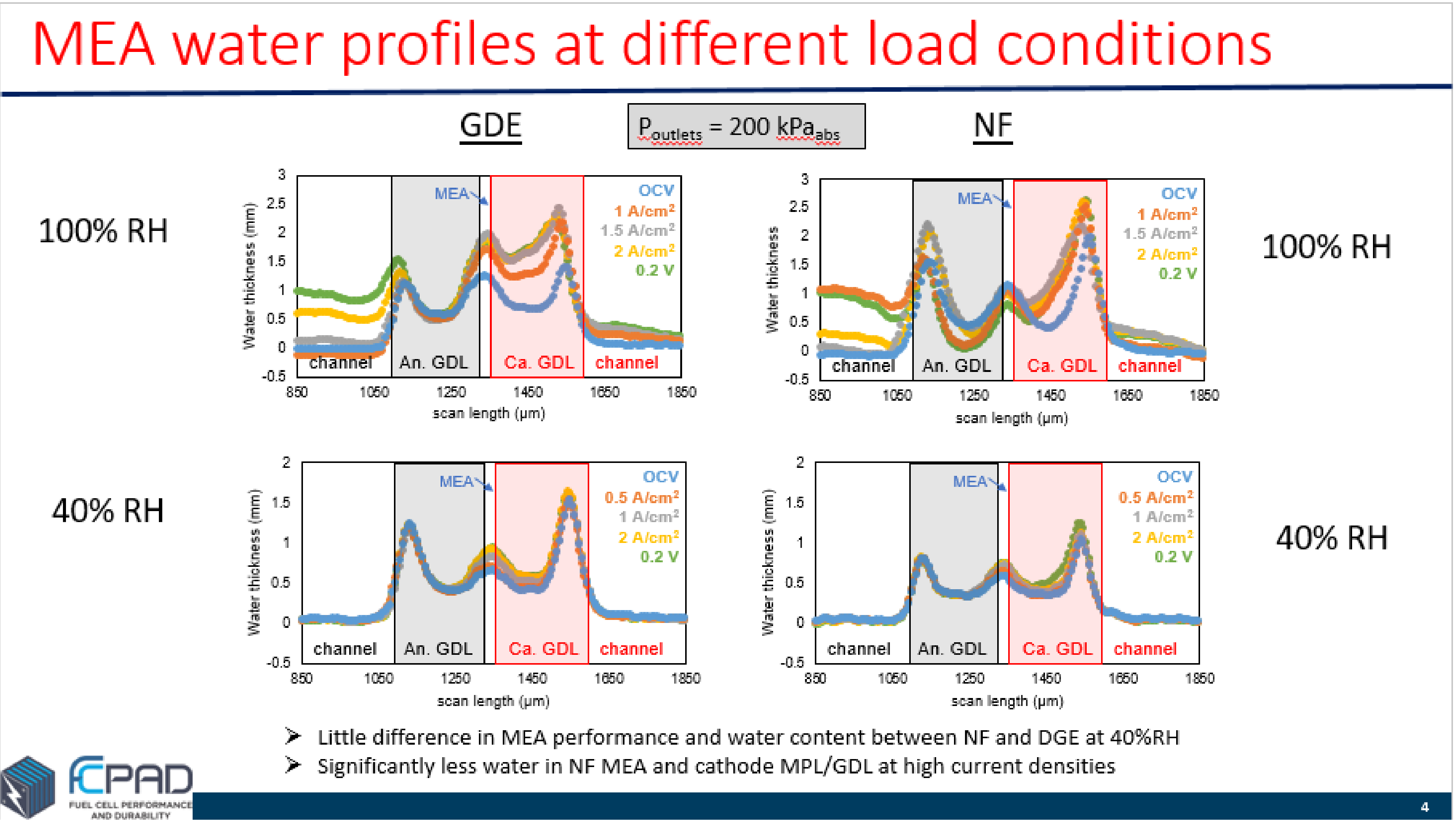


Drying: no current, no gas pressure (2hr)

Negative change
→ water depletion

-20C, gases off

Vanderbilt/LANL (FC-PAD): Membrane-Electrode-Assemblies with Ultra-Low Pt Nanofiber Electrodes



MEA water profiles at different load conditions

GDE

$P_{outlets}$ = 200 kPa_{abs}

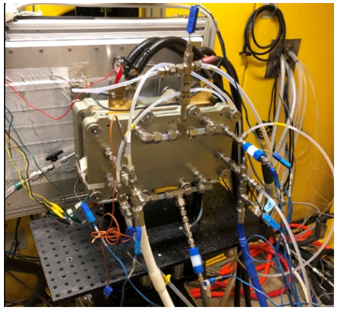
NF

100% RH

40% RH

- ➢ Little difference in MEA performance and water content between NF and DGE at 40%RH
- ➢ Significantly less water in NF MEA and cathode MPL/GDL at high current densities

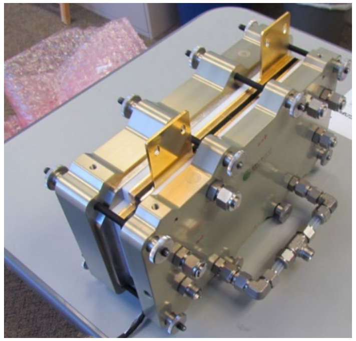
Water Imaging in Mirai Short-Stack provided by USCAR



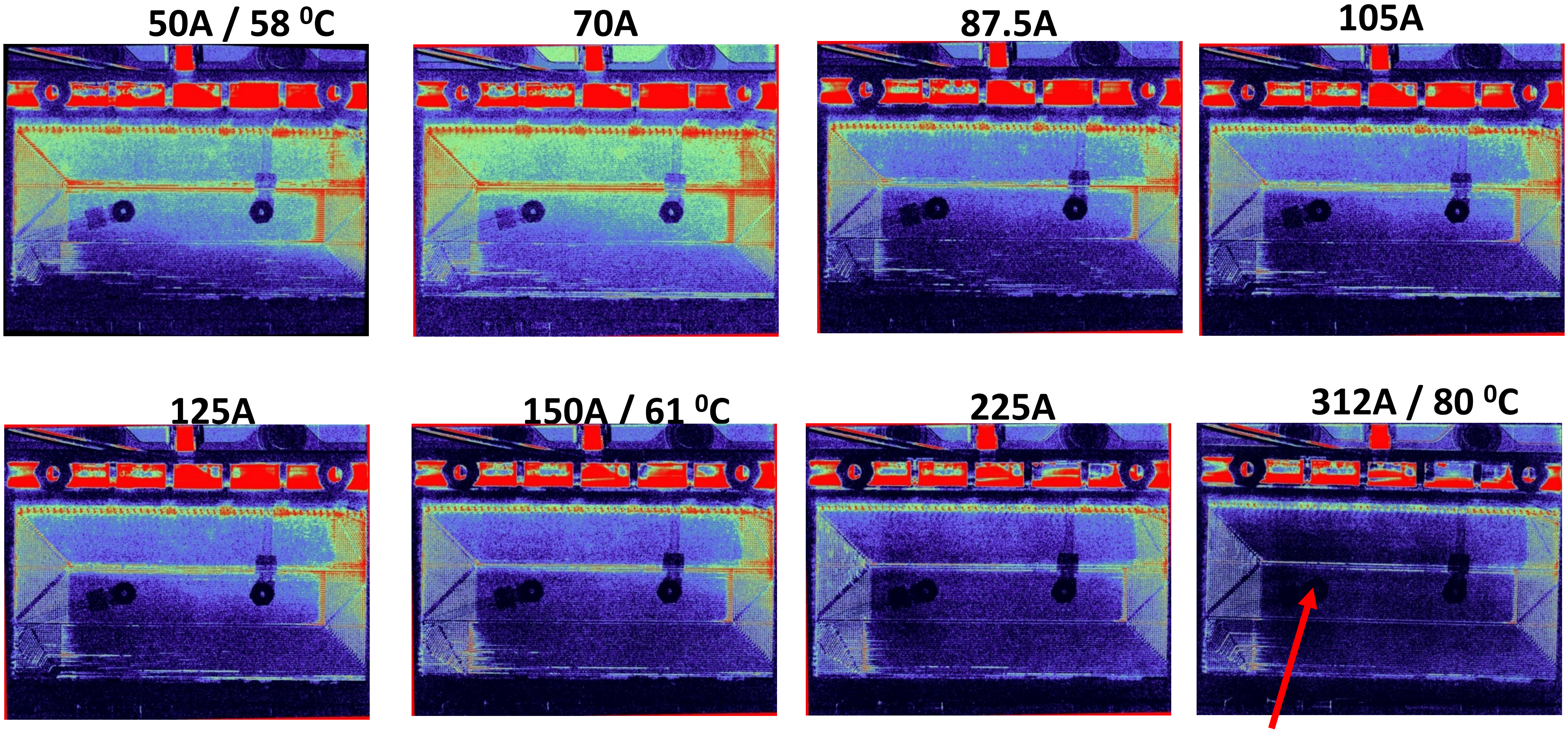
USCAR/LANL (FC-PAD) Collaboration

Water Measurements of State-of-Art Commercial Stack

LANL: Kavi Chintam, Derek Richard, Andrew Baker, Rod Borup
USCAR: Dave Masten, Sinichi Hirano, Chunchuan Xu
GM: Joe Fairweather



USCAR Matrix of Operating Conditions



50A / 58 ^{0}C

70A

87.5A

105A

125A

150A / 61 ^{0}C

225A

312A / 80 ^{0}C

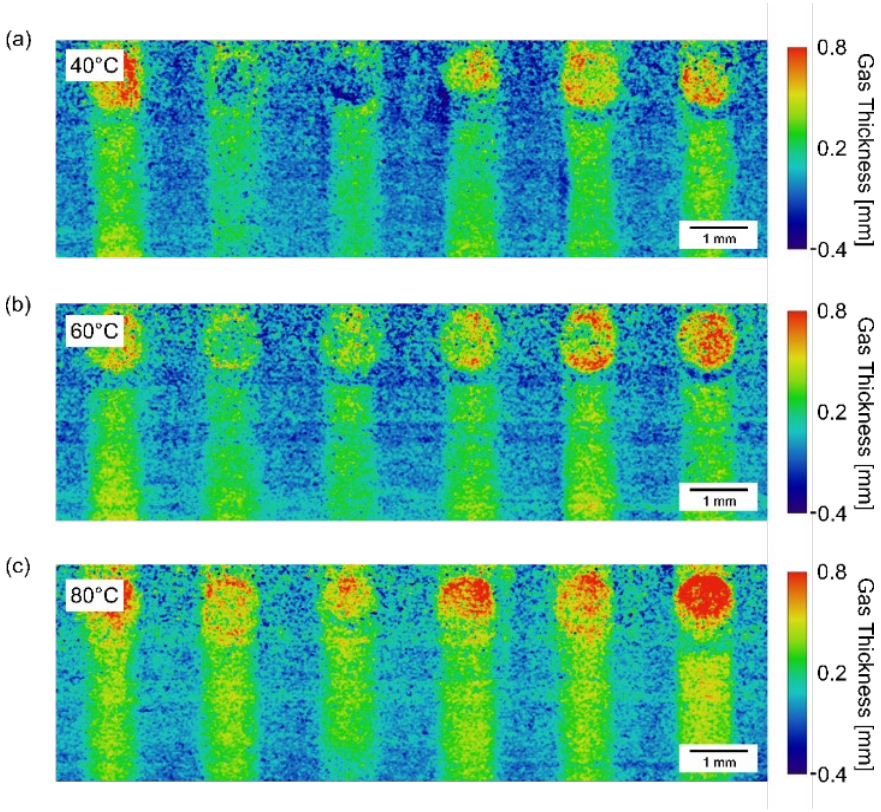
High Current & Flowrates show much less liquid water than low current/flowrates

Notes: *12.5 Amp point, unstable*
450 Amp point insufficient H2

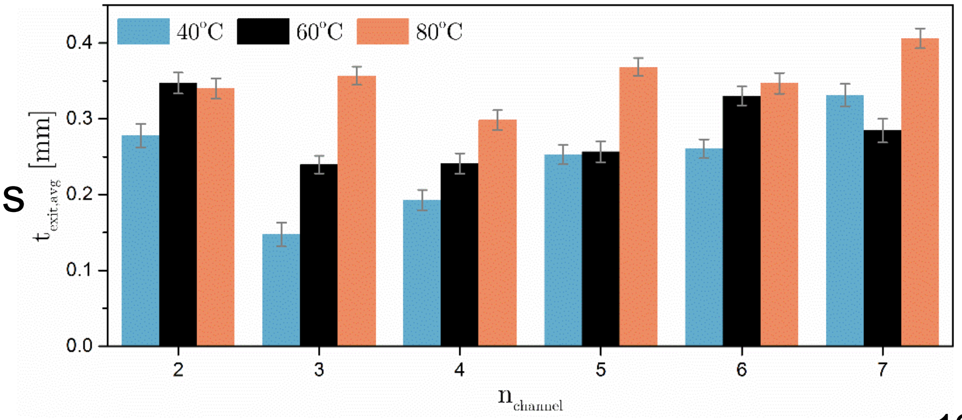
The Effect of Temperature on Two-Phase Flow in Anode Flow Channels of PEM Electrolyzers

ChungHyuk Lee, Jason K. Lee, Michael G. George, Kieran F. Fahy, Aimy Bazylak, University of Toronto
(Manuscript in Preparation)

- Operating temperatures of 40 (a), 60 (b), and 80 °C (c)
- Operating current density of 3.0 A/cm^2; (d) Quantified gas thickness in each channel shown in (a),(b), and (c). The mean exit gas thicknesses were 0.24, 0.28, and 0.35 mm for operating temperatures of 40, 60, and 80 °C, respectively. The uncertainties show 95 % confidence intervals based on the variation of thickness within each channel.
- Observed an increase in gas thickness with increasing temperature in most channels.

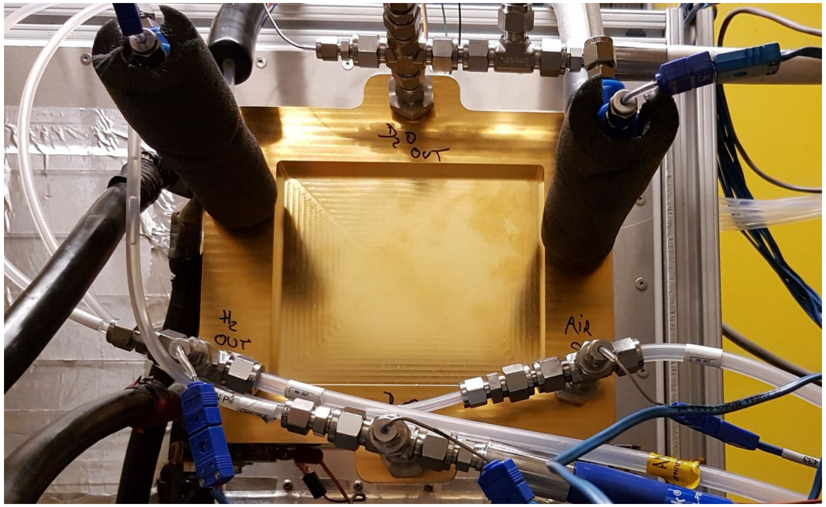


(d)

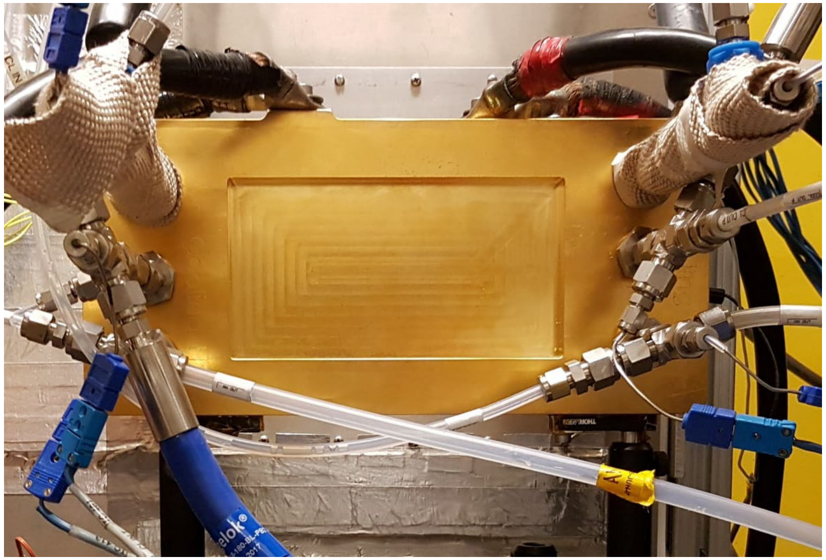


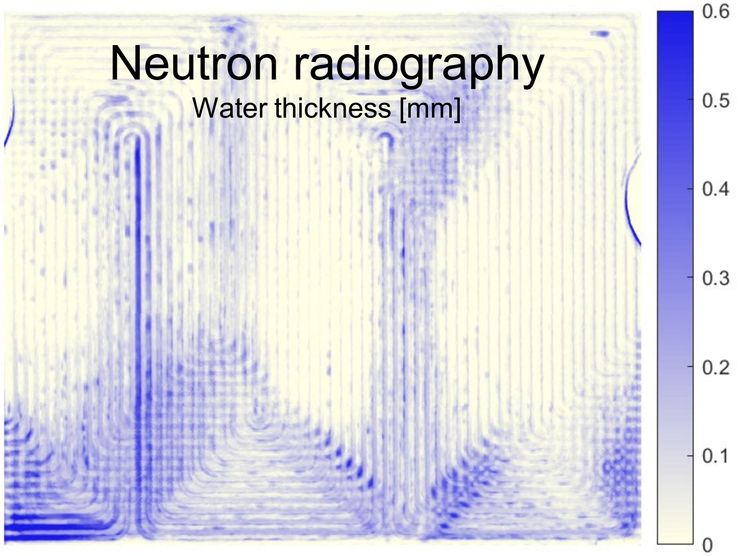
CEA: Effect of coolant flow Low and High T gradient between inlet and outlet for two RH's

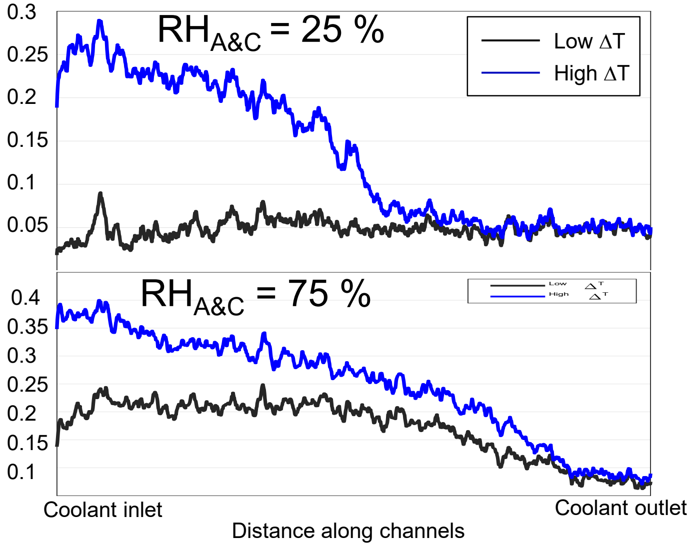
Generation 2009 (220 cm²) (Open)



Generation 2016 (~160 cm²) (Proprietary)







H_2/Air, i = 1 A/cm²; $P_{inlet\ A\&C}$ = 1.5 bar; St_c = 2; St_a = 1,5; T_{out} = 80°C; ΔT_{Low} = 5°C; ΔT_{High} = 25°C

Acknowledgements

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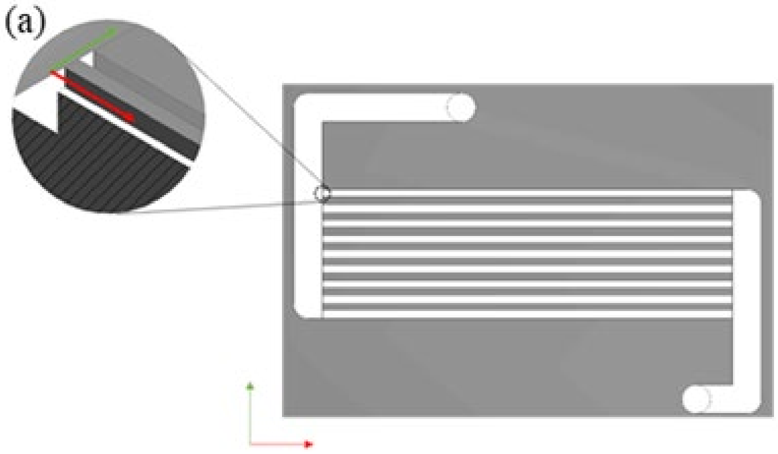
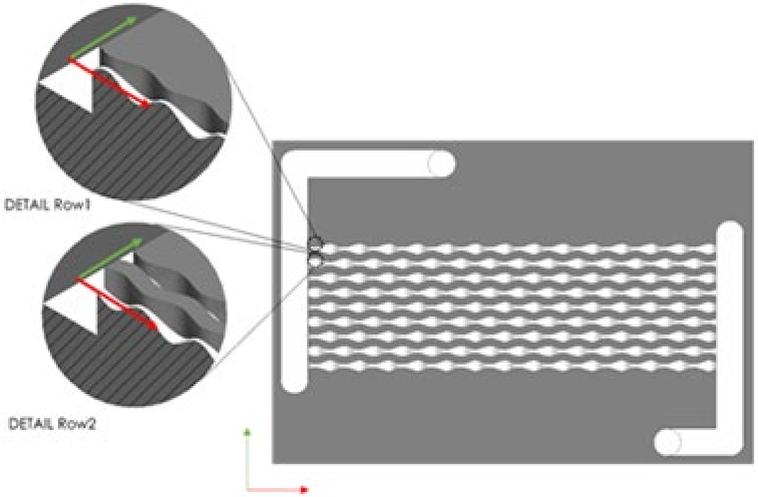
Muhammad Arif
January 24, 1954 ~ November 27, 2018 (age 64)

3-D Nozzle Flow Field Design by UC Merced

Felipe Mojica, Po-Ya Abel Chuang, U.C. Merced





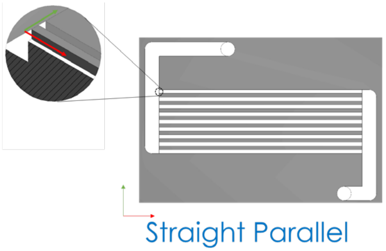
Straight Parallel

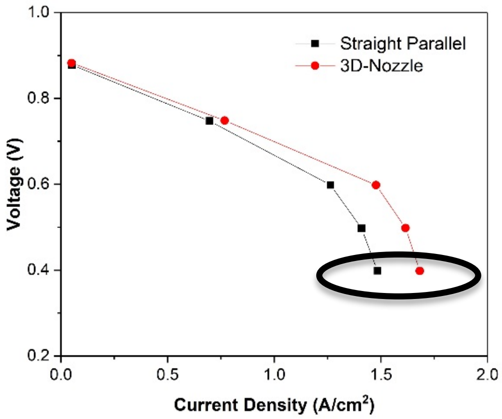
- Baseline design
- Convective flow in channel
- Diffusion transport in porous media

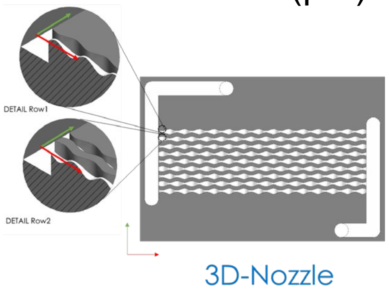
3-D Nozzle

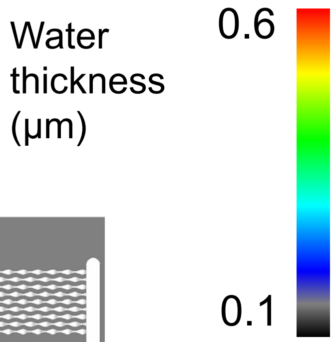
- Novel design with constant changing flow area down the channel
- Promote channel flow mixing and direct convective flow into gas diffusion layer
- Induce cross flow in gas diffusion layer

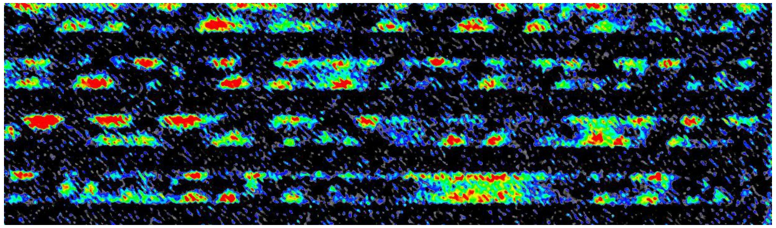
Wet Opearting Condition (100% RH; 80C; 300 kPa)

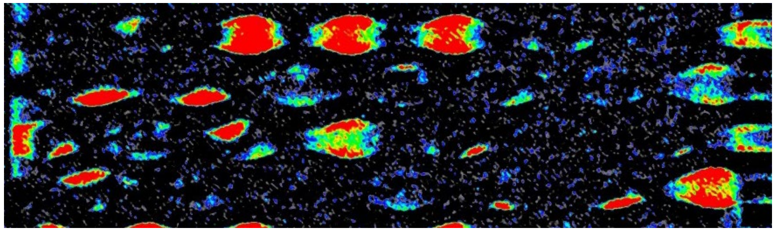






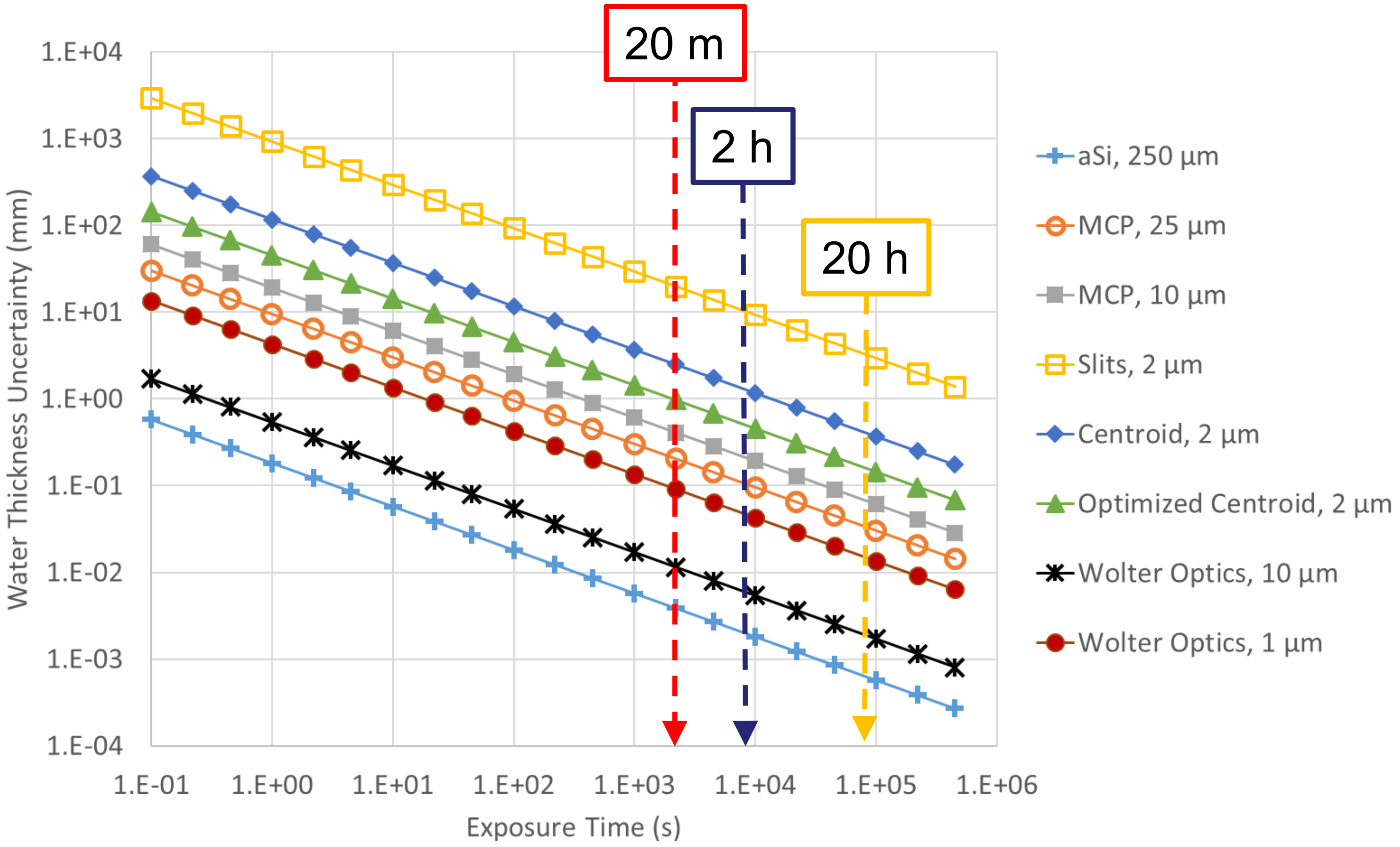




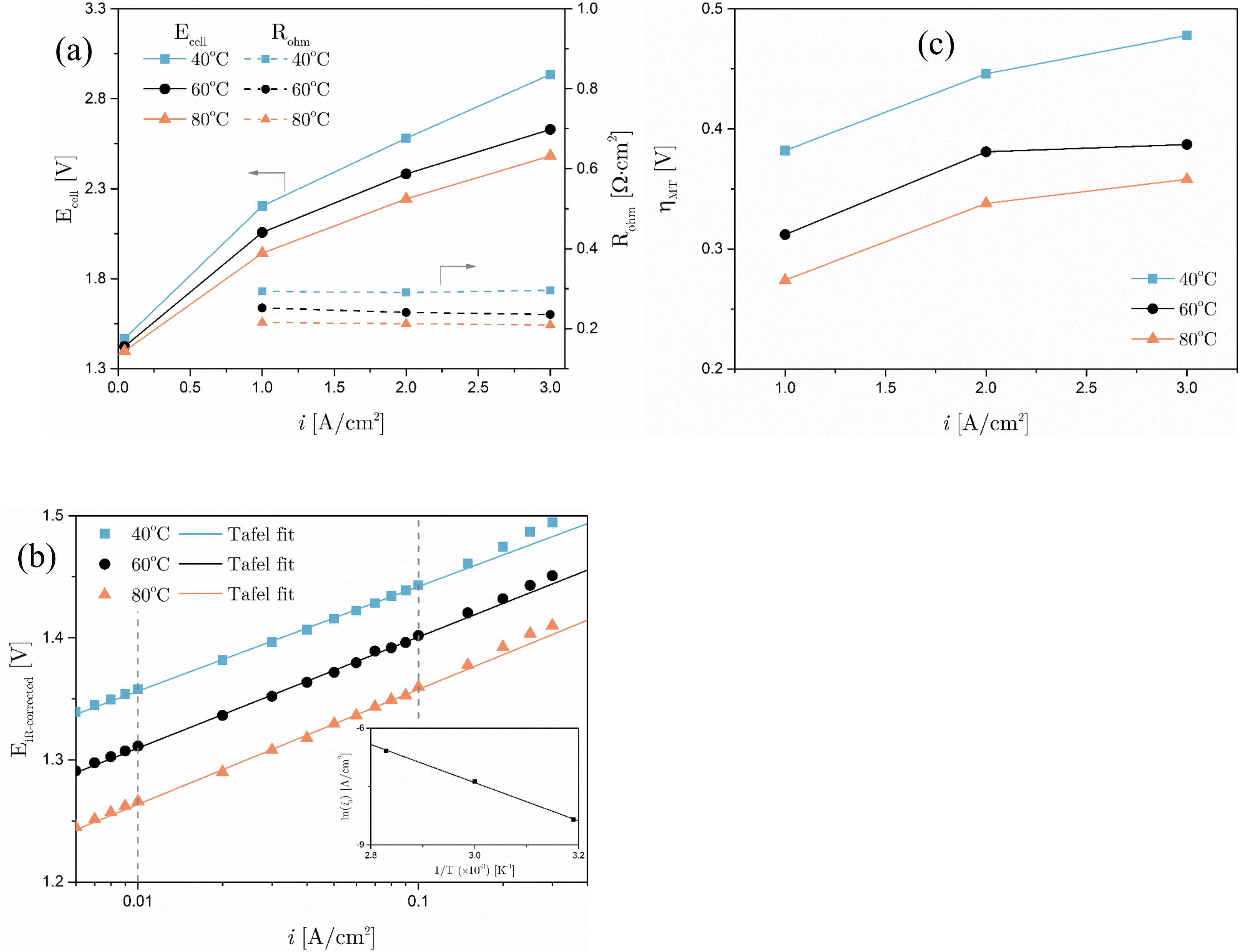


- 3-D Nozzle channel design showed improved wet performance resulting from lower level of liquid water flooding, especially in diffusion media under the land and near the throat.
- In contrast, straight parallel channel design shows higher liquid water saturation and transport resistance in diffusion media.

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


(a)
E_cell
R_ohm
40°C
60°C
80°C
E_cell [V]
R_ohm [Ω·cm²]
i [A/cm²]
3.3
2.8
2.3
1.8
1.3
1.0
0.8
0.6
0.4
0.2
0.0
0.5
1.0
1.5
2.0
2.5
3.0
(b)
40°C Tafel fit
60°C Tafel fit
80°C Tafel fit
E_iR-corrected [V]
i [A/cm²]
1.5
1.4
1.3
1.2
0.01
0.1
ln(i_0) [A/cm²]
1/T (×10⁻³) [K⁻¹]
-6
-9
2.8
3.0
3.2
(c)
η_MT [V]
i [A/cm²]
0.5
0.4
0.3
0.2
1.0
1.5
2.0
2.5
3.0
40°C
60°C
80°C

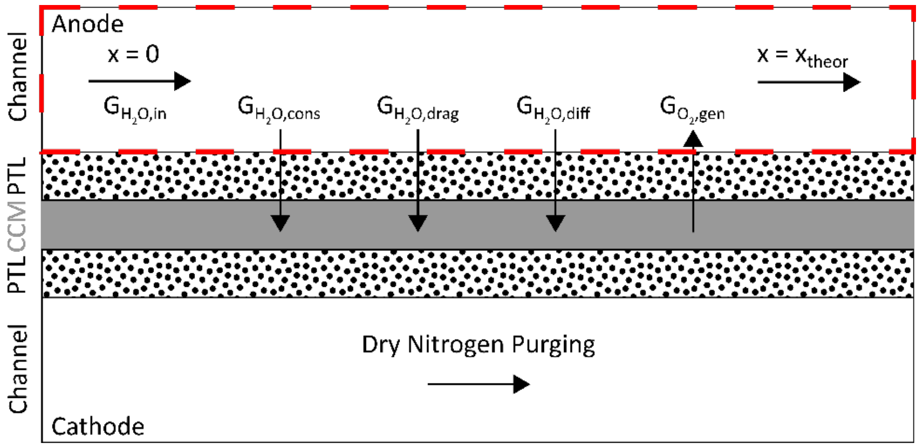


Figure 4: Schematic outlining the parameters considered in the analytical model. The red-dashed box indicates the anode flow channel, the region of interest for the calculation.

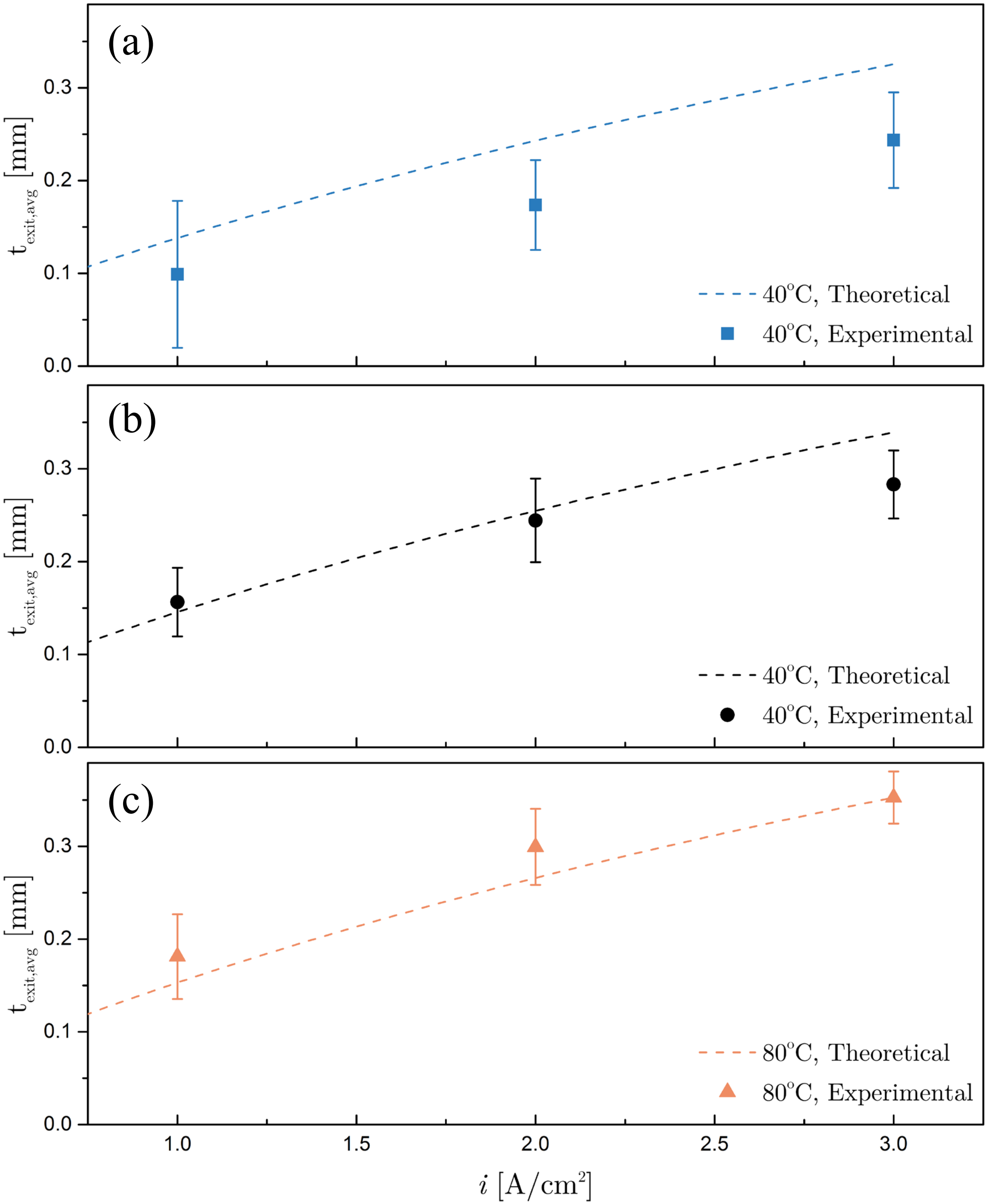
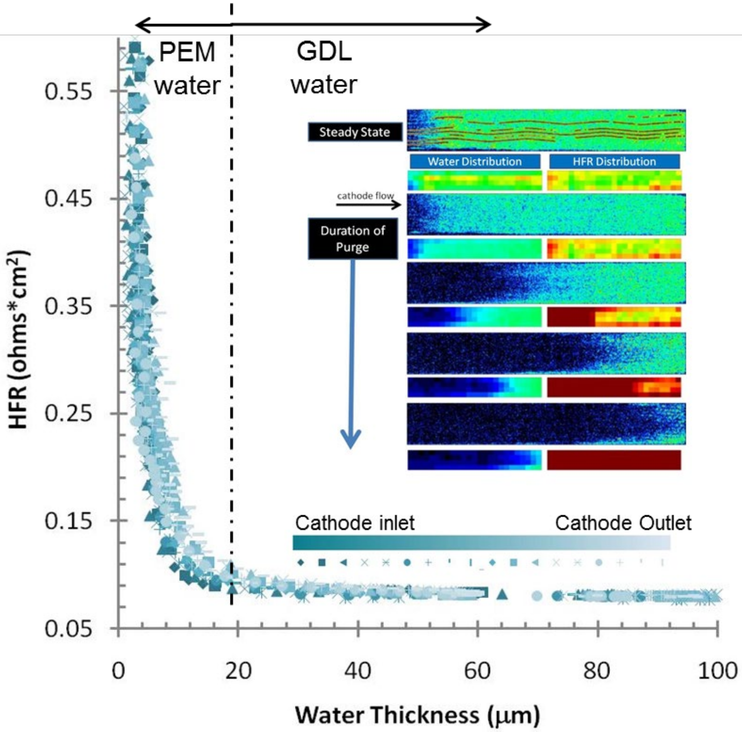


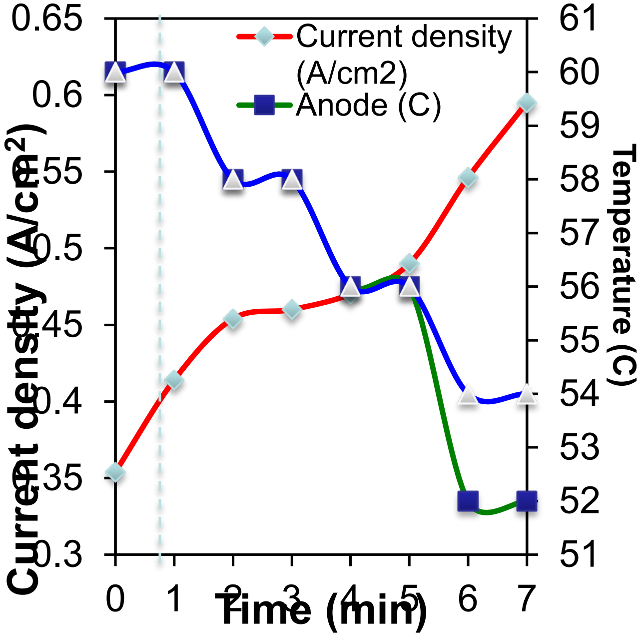
Figure 5: Mean exit gas thickness determined from *in operando* neutron radiography visualization at operating temperatures of (a) 40, (b) 60, and (c) 80 °C. The dashed lines indicate the theoretical exit gas thickness determined from the analytical model, assuming uniform liquid water distribution to each parallel flow channel and uniform current density operation (Fig. 4). The root-mean-square deviations between the experimental and theoretical data are 0.066, 0.034, and 0.025 mm for 40, 60, and 80 °C, respectively. We observed closest agreement between the theoretical and the experimental gas thicknesses at 80 °C, and the worst at 40 °C, indicating a relatively uniform reactant

Relevance

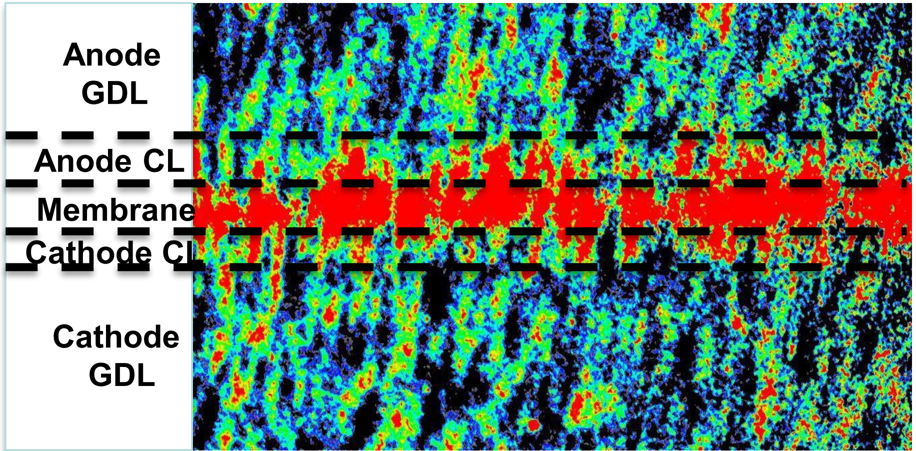
- Neutron imaging has supported fuel cell development from TRL levels 1-7
- Neutron imaging is the most powerful and sensitive method to *non-destructively* image water in the fuel cell *in operando* as neutrons readily penetrate common fuel cell hardware yet accurately measure small volumes of liquid 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



Real-time Visualization of Relieving Anode Flooding with Reduced Reacting Gas Dew Points

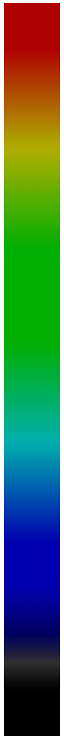


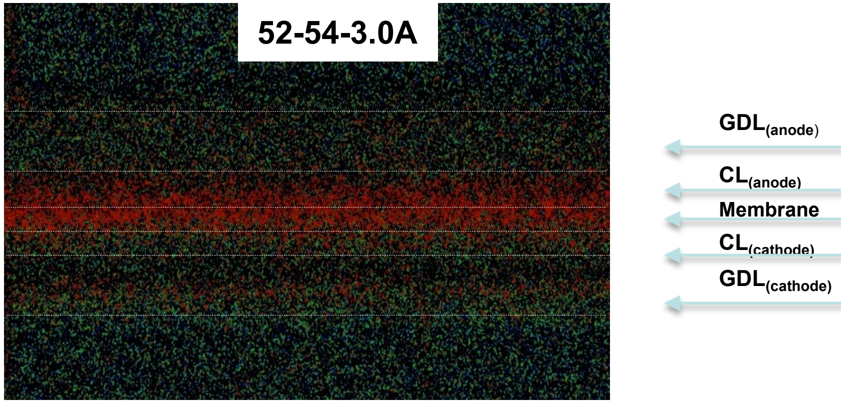
- Cathode: 0.60 mg/cm^2 of Pt (Pt/C)
- Anode: 0.70 mg/cm^2 of PtRu (PtRu/C)
- BP: 0.14 MPa backpressure for both sides
- T_{cell} = 60 °C, H_2-O_2 @ 1L/min



- Decrease dew points help remove water and relieve flooding
- After flooding events, water balanced by dew points and current density

High Resolution Neutron Images Showing How the Amount of Water Changes with Dew Points and Current Density





- At the same high current density, high dew points leads to more water accumulation in CLs and membrane
- Reducing the current density lowers the amount of water in the cell
  - Cathode can suffer from reduced back diffusion
- There is a clear difference in the volume of anode vs. cathode water, and there is a gradient through the membrane

Summary

- 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