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

Presented by: David Jacobson

Daniel Hussey

Eli Baltic

Muhammad Arif

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

FC021

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Timeline

Project Start Date: Fiscal Year (FY) 2001
Project End Date: Project continuation and direction determined annually by DOE
Percent Complete: 100% for each year

Budget

DOE FY13:	$ 300 k
DOE FY14(Planned):	$ 300 k
NIST :	$ 900 k
Industry:	$ 250 k
Total	**$ 1,575 k**

Barriers

(A) Durability
(C) Performance
(D) Water Transport within the Stack

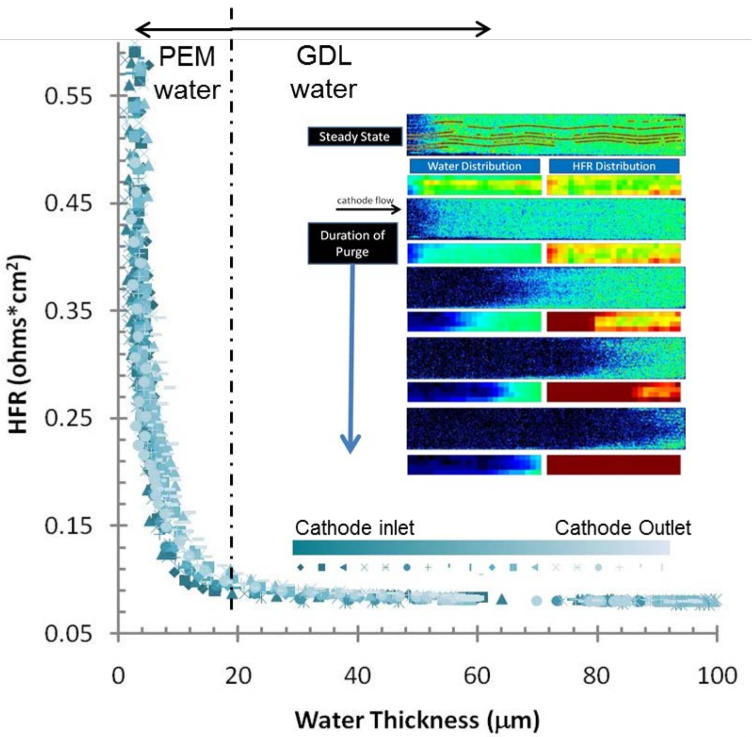
Partners/Users/Collaborators

Project Lead: National Institute of Standards and Technology

- Automotive Fuel Cell Corporation
- Ballard
- Ford
- General Motors
- Honda
- Nissan
- Nuvera
- Proton OnSite
- NASA, MSFC
- Lawrence Berkeley National Laboratory
- Los Alamos National Lab
- Massachusetts Institute of Technology
- Sensor Sciences
- NOVA Scientific
- CEA
- Michigan Technological University
- Oak Ridge National Laboratory
- Pennsylvania State University
- Rochester Institute of Technology
- Sandia National Laboratory
- Georgia Tech
- University of California, Berkeley
- University of Connecticut
- University of Kansas
- University of Michigan
- University of Tennessee
- Wayne State University

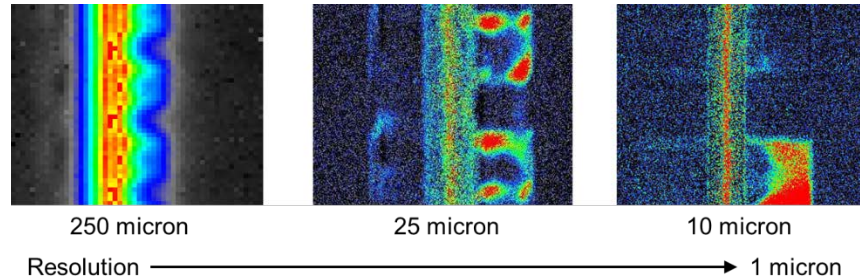
Relevance

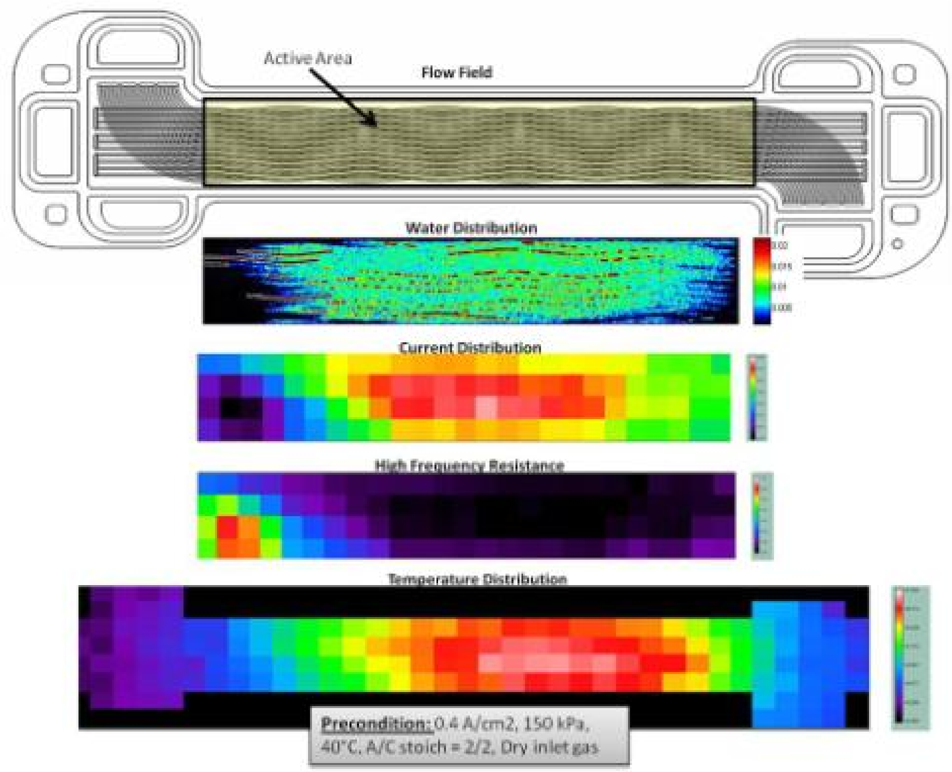
- Neutron imaging is the most powerful method to image water in the fuel cell *in situ* as neutrons readily penetrate common fuel cell hardware yet are extremely sensitive to liquid water
- This allows one to study a wide range of fuel cell water management questions:
  - Dynamic water transport in the flow fields and manifolds
  - Liquid water distribution anode versus cathode
  - Cold start and freeze-thaw effects
  - Catalyst degradation induced by liquid water
- 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 the needs of the fuel cell community
  - Improve the spatial resolution to provide more detail of the water content in commercial MEAs



Approach

Apply quantitative neutron radiography to measure the water content in an operating fuel cell to provide a complete picture of heat and mass transfer and cell performance



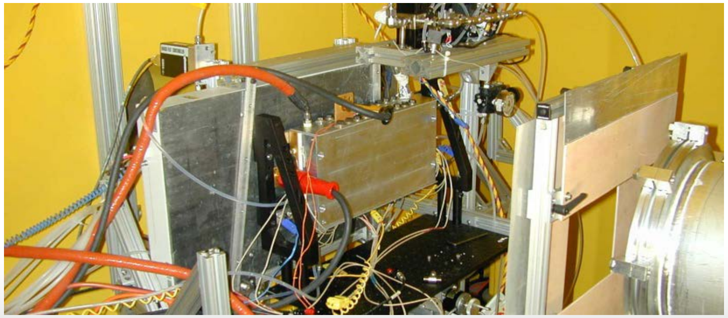


Continuous effort to:

- Enhance image spatial resolution to provide detailed measurement of water content in MEAs
- Improve image analysis to correct systematic effects and ensure accurate water content measurements
- Make state-of-the-art detectors, methods, and analysis available to the fuel cell research community

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
- Fee based access for proprietary research
  - Contact NIST for details
  - Proprietary users trained to use the beam
- 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
  - Fuel cell test stands fully integrated with GUI and scripting
  - Image analysis software is tailored to fuel cell user needs

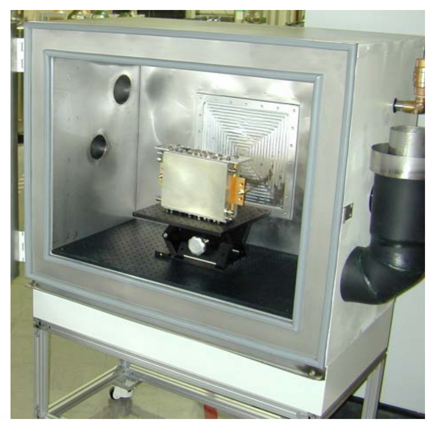


Approach

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

Small scale test stand:
Cell area ≤50 cm^2,
dual & liquid temperature control,
absolute outlet pressure transducers

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)



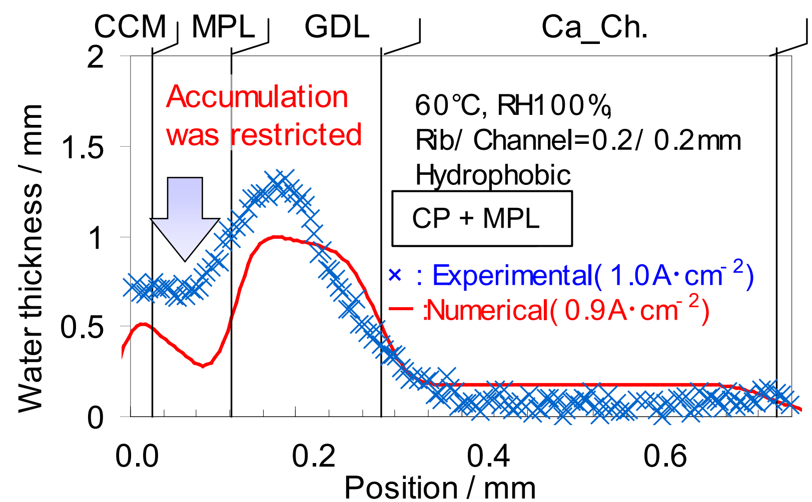
Environmental Chamber:
-40 °C – 50 °C
RH 20-90% above 20 °C
1 kW cooling at -40 °C

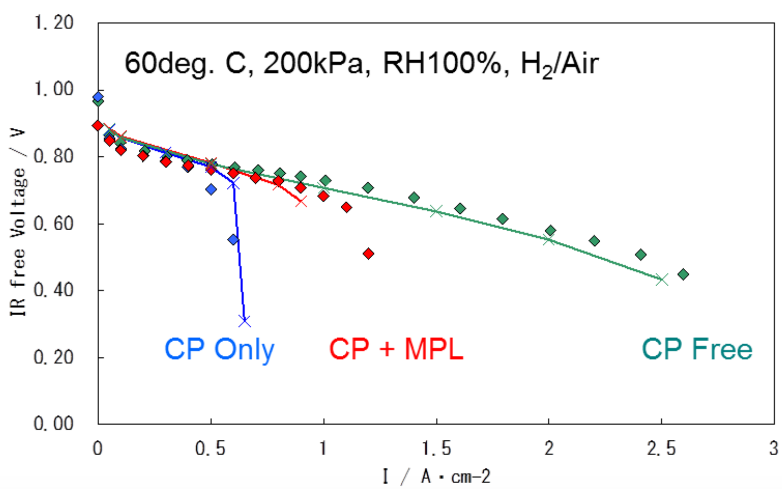
Accomplishments: Milestones

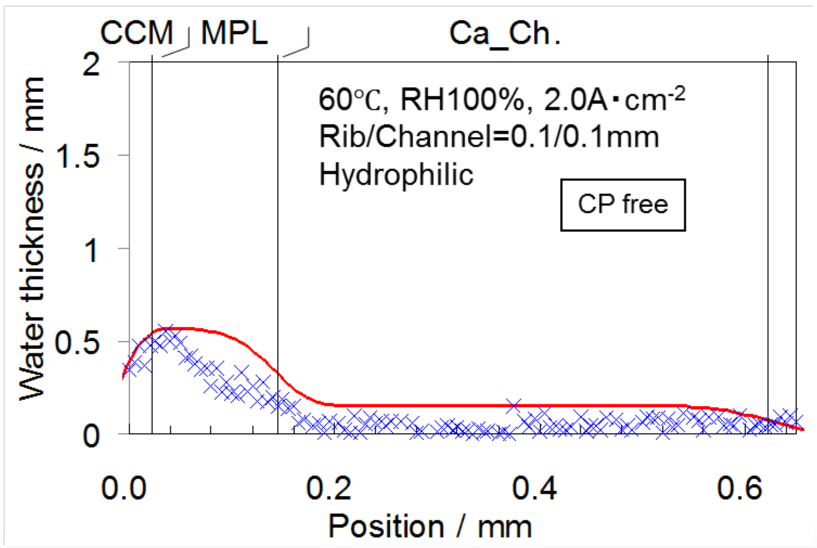
Milestones	Progress	Comments	FY13 %
1) High resolution MEA water content Employ a high resolution imaging method to achieve resolution approaching 1 μm	Ongoing	Details in presentation	100 %
2) Neutron image analysis and corrections Deblurring algorithms for images of the in-plane water content of fuel cells are demonstrated and published	Complete	Details in presentation	100 %
3) Fuel cell infrastructure for automotive-scale testing Build fuel cell infrastructure to enable testing of automotive-scale test sections; large-scale fuel cell test stand is available to users	Complete	Details in approach, stand is available to all users	100 %
4) Improve fuel cell high resolution image time Build neutron detector system to improve the time resolution of the high resolution imaging method in milestone 1	Tested concept procurement of system in progress	Full completion expected by Oct 2014	50 %

User Program Highlights: Effect of GDL and MPL

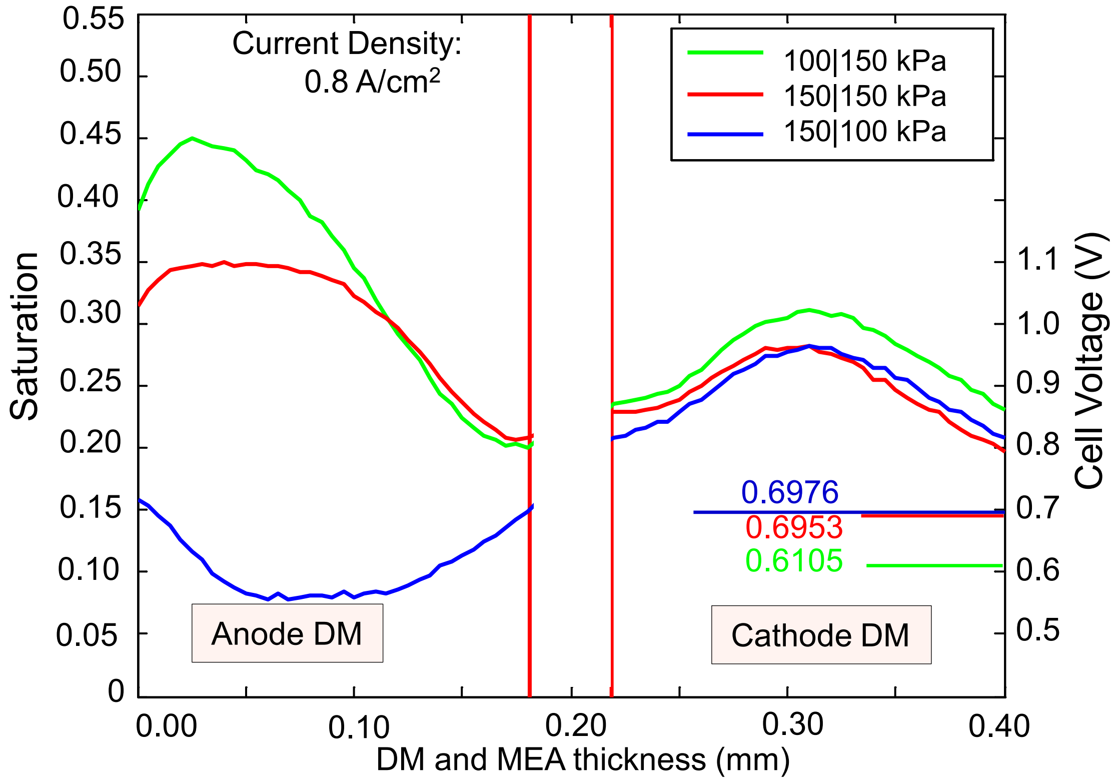
- Modeling and imaging work carried out by Nissan and University of Connecticut
- Compared performance of single cells with carbon paper GDL (CP), CP+ MPL, and MPL only
- MPL-only cells show highest limiting current and least amount of water hold-up
- Points are measurement, lines are model predictions

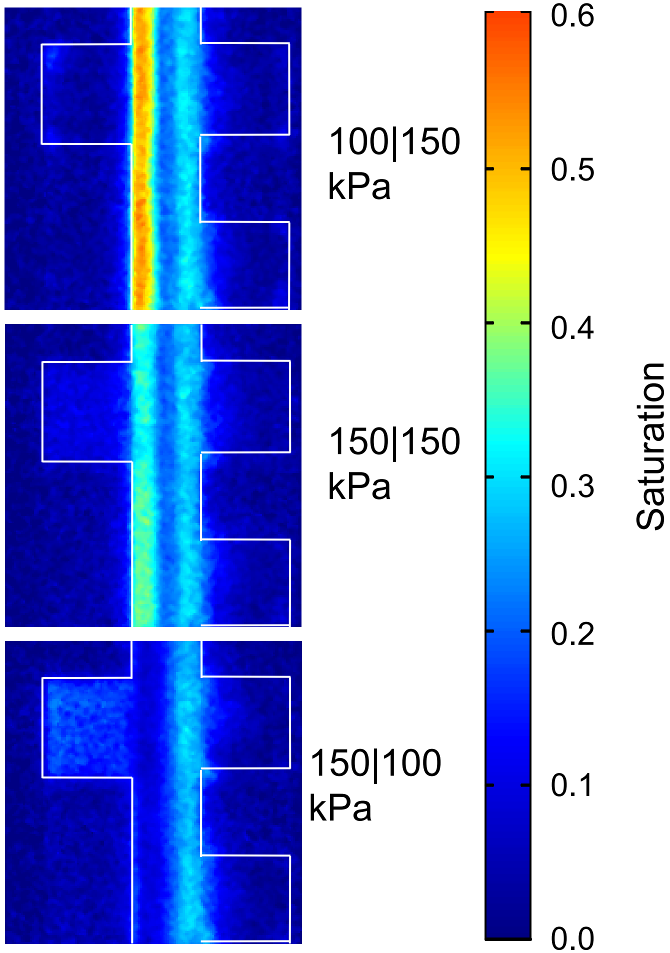






User Program Highlight: Exhaust Pressure Effects



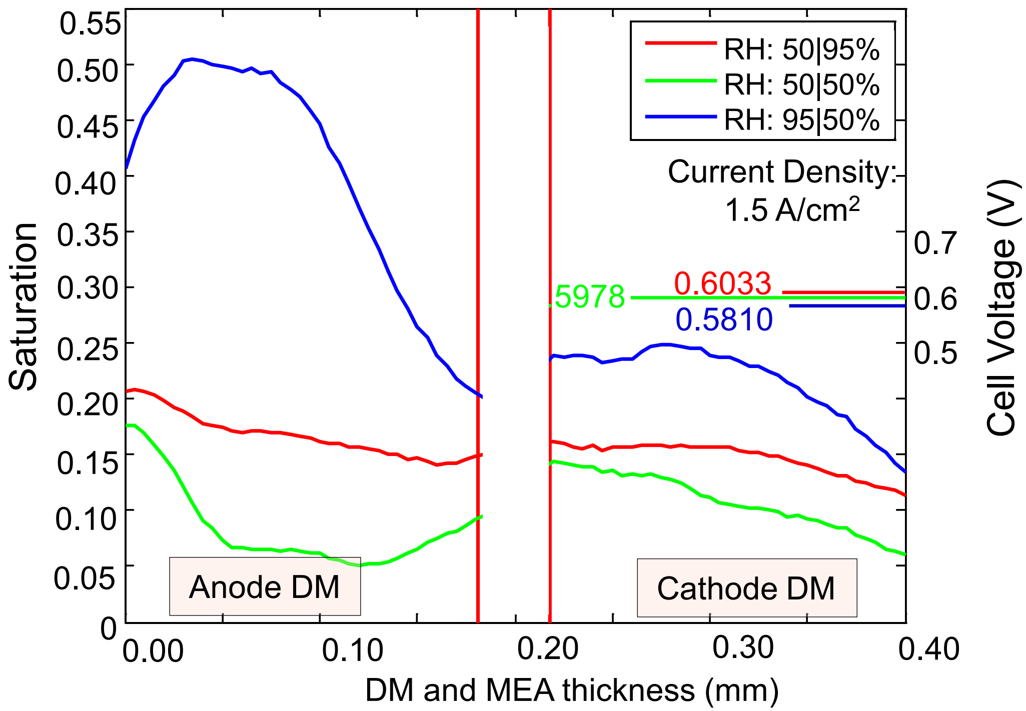


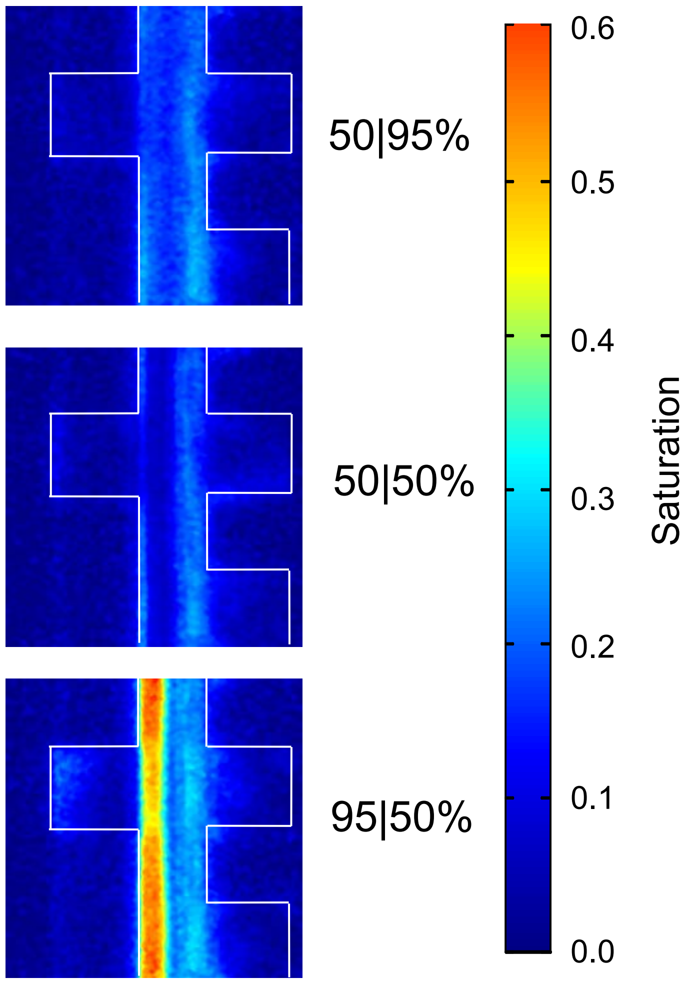
- Different pressures in Anode and Cathode lead to water convection to the lower pressure side.
- Anode is much more responsive to changes in back pressure due to difference in flow channel geometry.

Test Conditions: 40°C, 95% RH, 2:2 Stoic @ 1 A/cm^2

University of Tennessee-Knoxville
Data at www.PEMFCdata.org

User Program Highlights: RH Effects on Anode Water Content





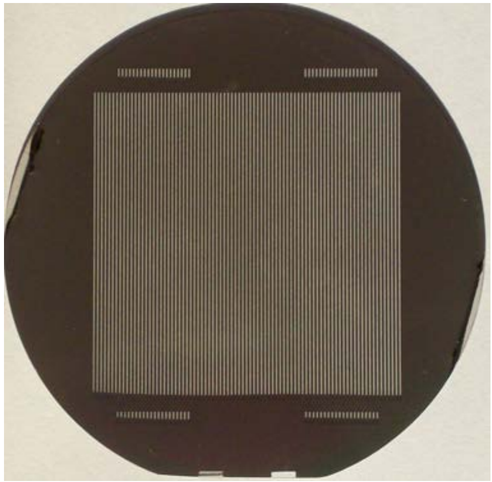
- Asymmetric Relative Humidity in Anode and Cathode gases has pronounced effect on the water distribution.
- Anode is still more responsive to changes in RH than Cathode.

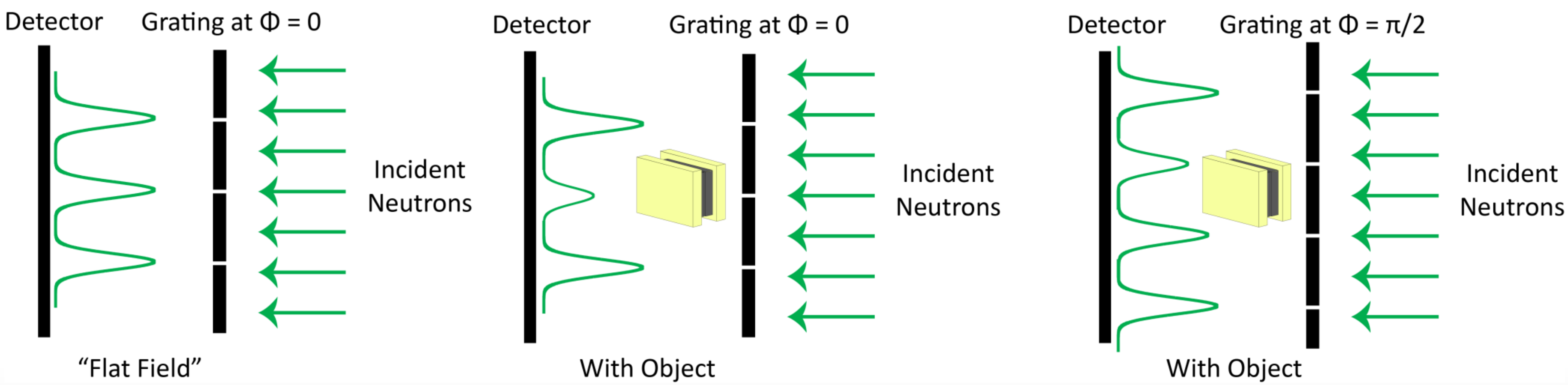
Test Conditions: 60°C, 50 kPag, 2:2 Stoic @ 1 A/cm^2

University of Tennessee-Knoxville
Data at www.PEMFCdata.org

Milestone 1: High resolution MEA water content
Higher Resolution Inspired by Structured Illumination

- Spatial resolution in any neutron detector is fundamentally limited by the range of the charged particles from the neutron capture
- Using a slit we can further define the spatial resolution by width of the slit
- Technique:
  - Illuminate object with narrow slits on a period that allows detector to resolve the image
  - Translate grating across the object
  - Stitch image together in software

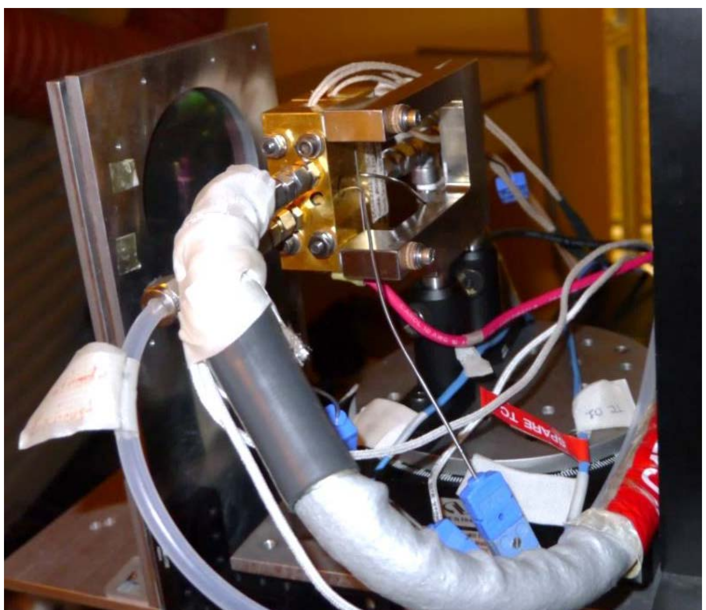


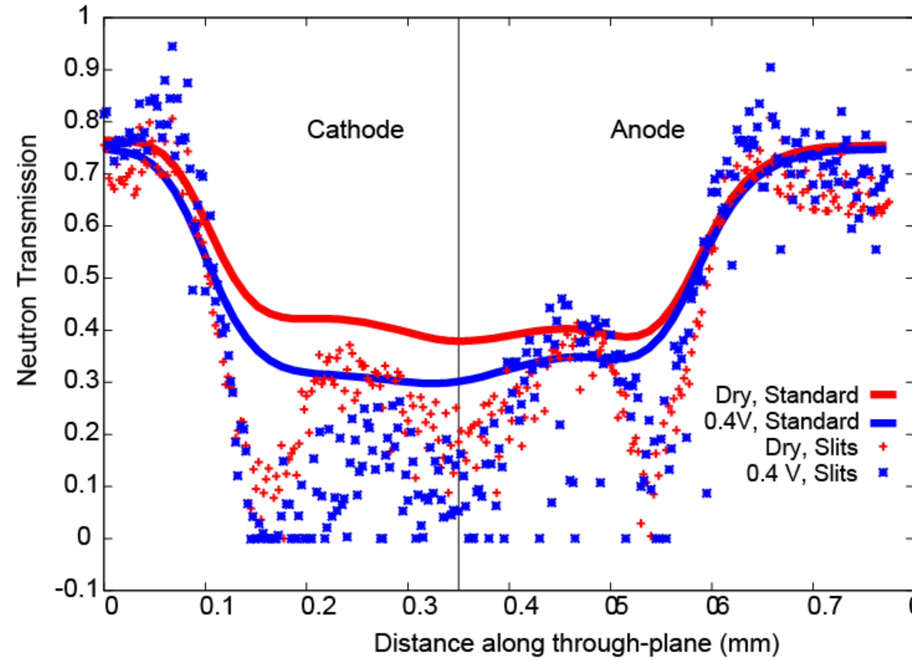


Milestone 1: High resolution MEA water content

First results sub 10 μm

- Used two gratings with ~3 μm of Gd, that allowed us to create an small (5 μm) opening
- Line profiles show clearly sharper features and demonstrate need for better signal-to-noise
- Images acquired over period of 15 h



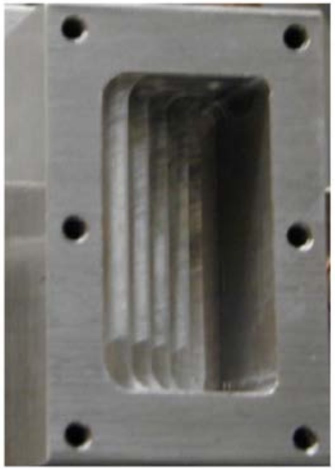


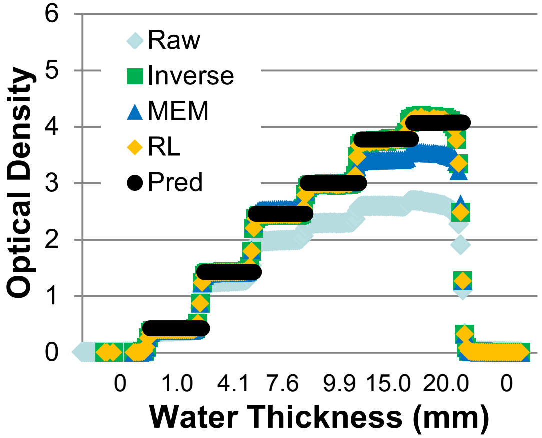
Line profiles from standard imaging with resolution 25 μm compared to slit method for a dry fuel cell and during operation at 0.4 V (about 2 A/cm^2)

Milestone 2: Neutron image analysis and corrections

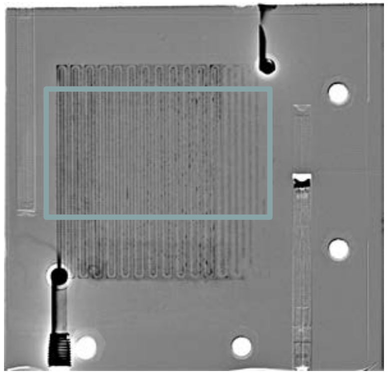
Improve fuel cell water measurement uncertainty:

- Cross-talk from detector resolution function results in a small additive background that can be corrected with image deconvolution
- Neutron scattering also results in a small additive background; correction is appreciable only for thicker sections of water than observed in fuel cell imaging
- Important for calibration, there is good agreement between model with scattering and restored images
- Correction is smaller than statistical uncertainty for typical fuel cell data
- Deconvolution available to users in NIST's image analysis routines

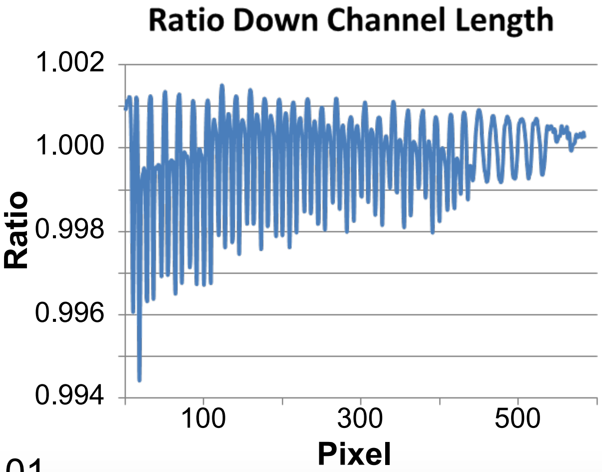




Predicted attenuation compared to inferred attenuation from raw and restored images with 3 deblurring methods: Fourier inversion, Maximum Entropy and Richardson-Lucy



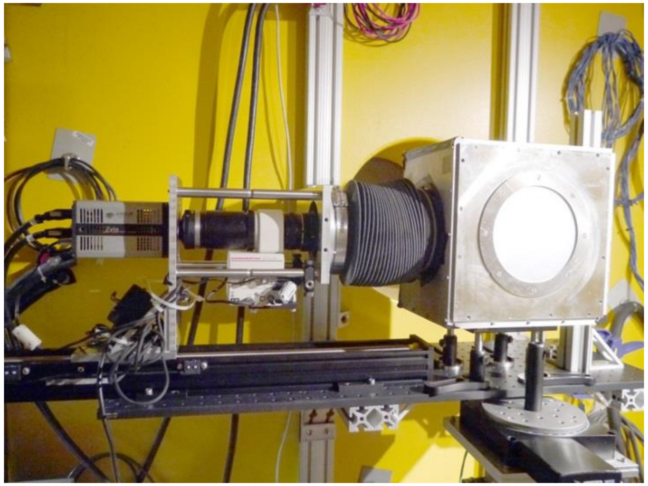
Ratio restored to raw.
Contrast:
Black = 0.99, White = 1.01

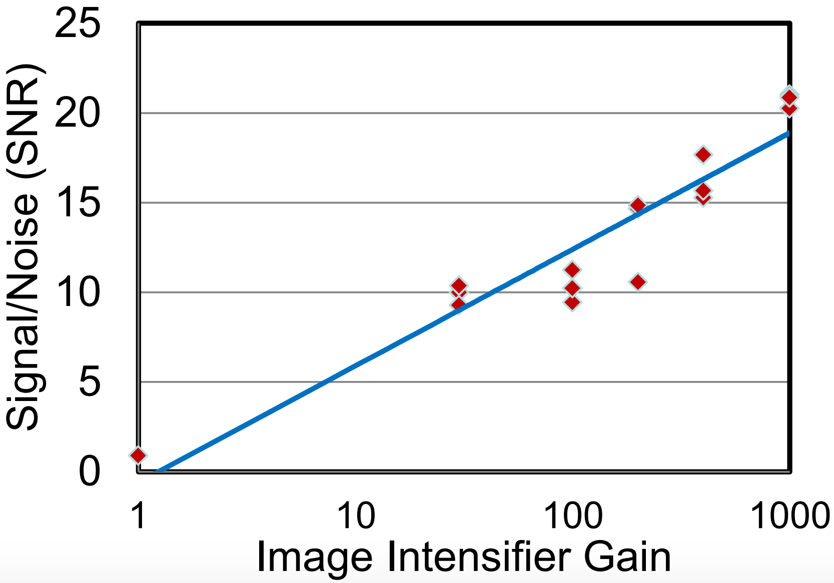


Milestone 4: Improve fuel cell high resolution image time

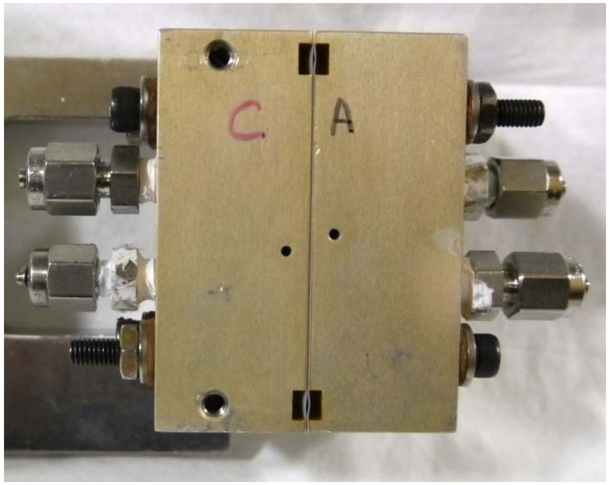
Low light, high resolution Gadox screens can be improved

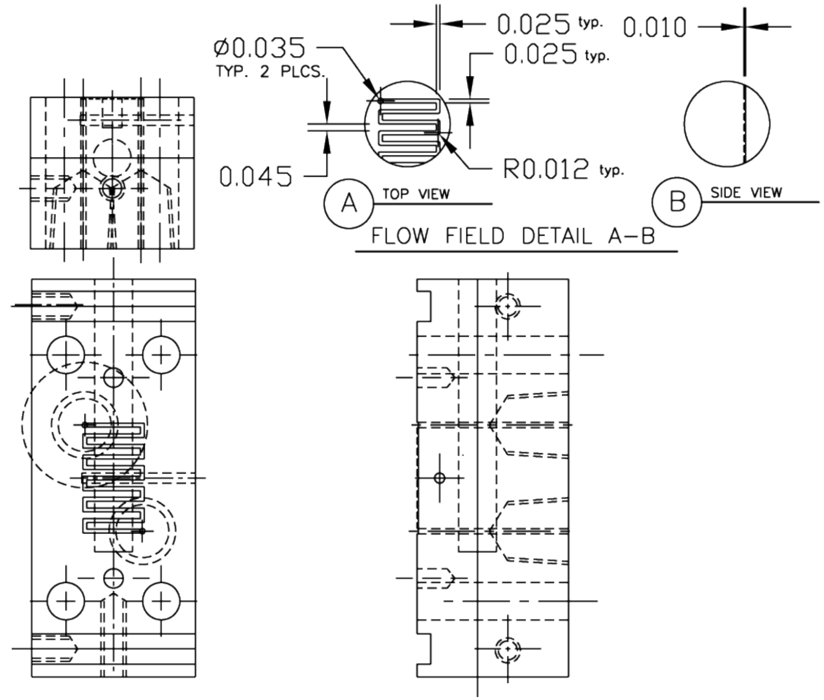
- 20 μm spatial resolution
- 80 % neutron detection efficiency is 4x larger than the current 20 % efficiency for MCPs
- Noise overwhelms very low light signal
- Only usable with an image intensifier and ultra-low noise camera
- New system realizes 4x improvement in image acquisition time
- Final system available by October – November 2014



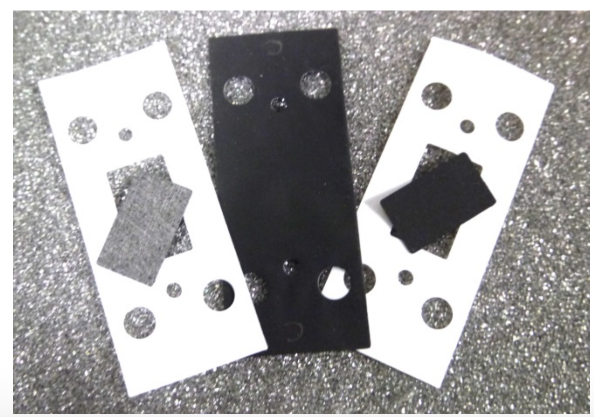


Future Work: Standard High Resolution Fuel Cell

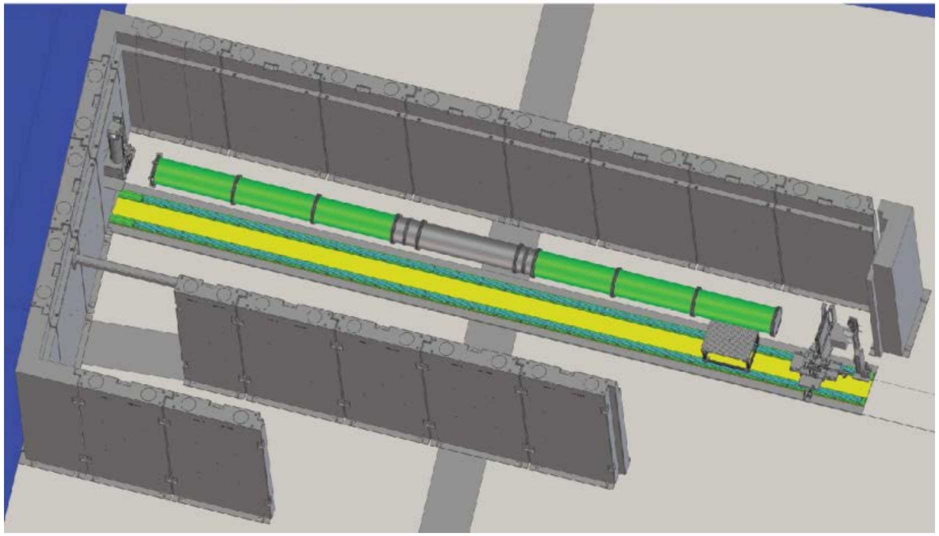




- Robust design (LANL)
  - Lots of use with neutron imaging
  - Easy to build, good performance
  - MEA can be cut from existing 50 cm^2 soft goods
- High resolution fuel cells
  - porous metal foam flow fields
  - No non-uniformities from rectangular flow fields
- Gauge block spacer
  - Avoids wedge, improves plane parallel



Future Work: Cold Neutron Imaging Facility

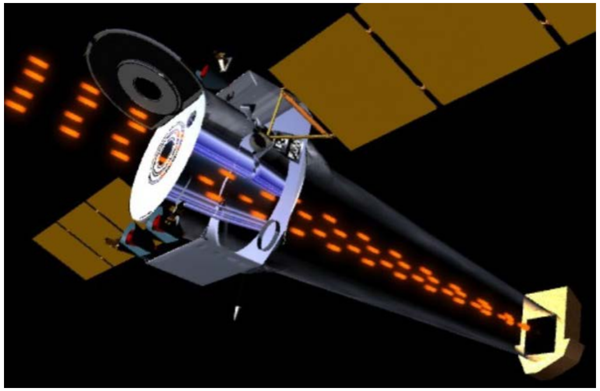


- Reactor maintenance and government shutdown cost about 80 days
- Delays also due to sluggish budget approval
- A second cold neutron imaging facility will provide additional beam time to develop new high resolution methods
- Cold neutron spectrum may allow discriminating ice from water
- Installation starts Summer 2014

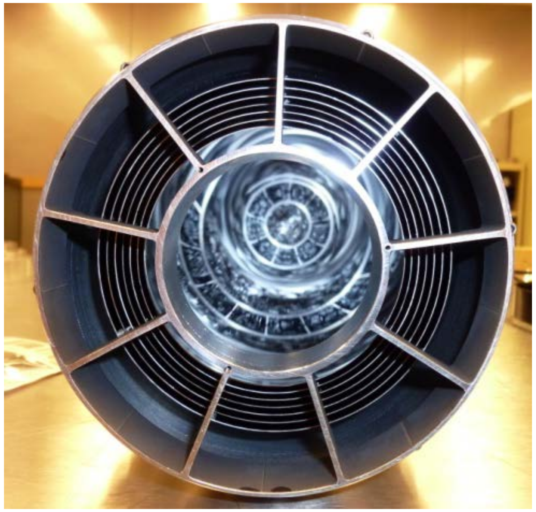
Progress: High speed, High resolution MEA water content

NASA's CHANDRA Inspired the Neutron Microscope

- High resolution neutron images require strong collimation resulting in low flux and long exposures
- Neutrons are hard to focus, the refractive index (n) is small and wavelength (λ[Å]) dependent: $n \sim 1 - 10^{-6} \lambda^2$
- Neutrons are neutral and neutron beams are large, not points, many x-ray tricks don't translate
- Faint x-ray sources (nebula, etc.) need to be focused for good imaging
- NASA is developing a new fabrication technique to create reflective mirror optics from nested Ni-foils– light for space telescopes and perfect for neutrons
- Resolution from the lens not collimation
- No collimation for 10 μm resolution can yield 100x flux increase for imaging with image times ~10 s
- Magnification of 10x can improve spatial resolution to 1 μm with image times ~20 minutes
- Ongoing collaboration with MIT and NASA to further develop the technology for fuel cells



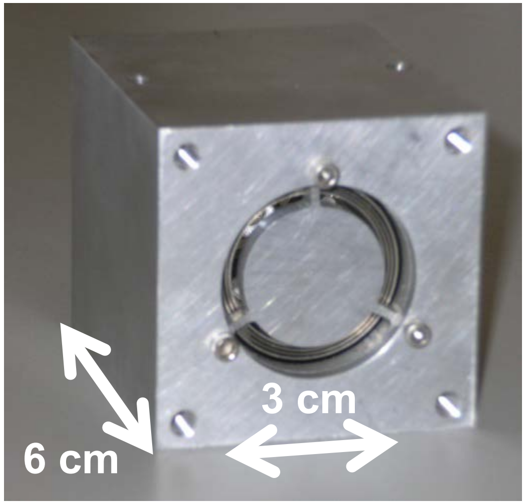
Wolter Optics power CHANDRA



Ni-foil Focused X-ray Solar Imager

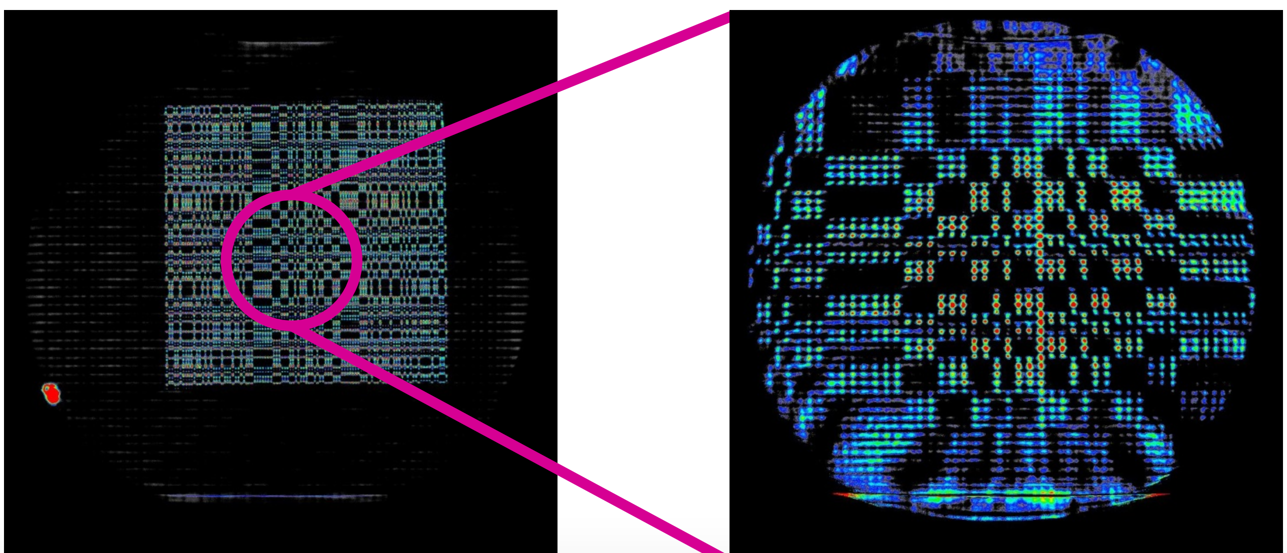
Progress: High resolution MEA water content

Image Magnification



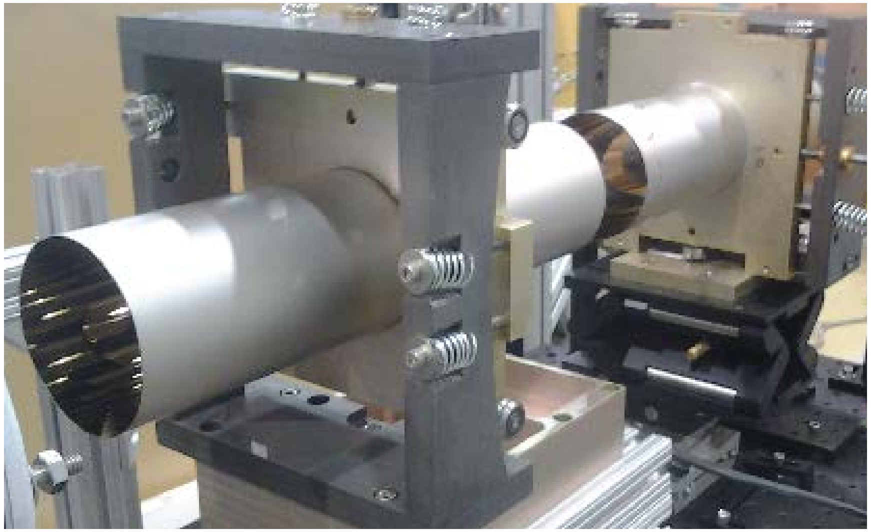
- 3 nested Ni mirrors w/ellipsoid and hyperboloid sections
- Overall focal length of 3.2 m
- This prototype lens truly formed neutron images with:
  - 1 cm FOV & 4x magnification
  - 75 μm spatial resolution, 5 mm depth of focus

More work: x100 resolution, x100 flux, x5 depth of focus



2 cm x 2 cm Pinhole mask, with 0.1 mm diameters on 0.2 mm centers
Left: Contact Image
Right: Lens Image

Progress: Prototype microscope tested for characterization data



- Prototype 1:1 microscope lens characterized with neutrons March 2014
- Measured focused intensity agreed with design model predictions
- Expect x50 over BT2 in final form

- NIST is providing internal funding to develop a cold neutron microscope.
- Project timeline (conservative estimate):

2014:

- Test a new prototype lens that is targeted for neutron imaging

2015:

- Demonstrate 10 μm image resolution
- Finalize 1:1 optic design and begin fabrication

2016:

- Begin fuel cell user operation for lens-based imaging at 10 μm resolution
- Finalize design for 10x magnifying lens

2018:

- Begin fuel cell user operation for 1 μm imaging with 10x lens

Response to 2013 Reviewers' Comments

- The membrane/catalyst layer and catalyst layer/gas diffusion layer interfaces are not distinguishable
  - We are working with our testing partners to develop a cell that can take advantage of our improved spatial resolution capabilities
- It is unclear if the technology can be used for a stack diagnosis.
  - Small stacks have been tested and previously reported on at 2011 AMR: http://www.hydrogen.energy.gov/pdfs/review11/fc021_jacobson_2011_o.pdf
- There is considerable risk in the proposed path forward for the high-resolution detector. An analysis of the technique in comparison to other approaches (x-ray or nano-CT) should be included to cast the risk of the technique improvement compared to the current state of the art of this and other techniques
  - Neutrons are truly non-destructive and do not affect the performance or function of the fuel cell. X-rays adversely affect the operation of a fuel cell (see: J. Eller and F. N. Büchi, "Polymer electrolyte fuel cell performance degradation at different synchrotron beam intensities", *J. Synchrotron Rad.* (2014) **21**, 82-88); thus x-rays are unsuited for many important in operando studies such as durability

Conclusions

- High resolution MEA water content
  - Employing a grating method to achieve resolution approaching **1 μm with 12 hour acquisition time** (end of 2014)
  - Developing a magnifying neutron lens to reach **1 μm with 20 min acquisition time** (2018)
- Neutron image analysis and corrections
  - Deblurring algorithms for images of the in-plane water content of fuel cells are demonstrated and published
- Fuel cell infrastructure for automotive-scale testing
  - Test stand to control automotive-scale cells is available to users
- Improve fuel cell high resolution image time
  - Time resolution for through plane water content measurements improved by a factor of 4 with 20 μm scintillator detector
  - Future neutron lens will increase time resolution by 50x to achieve image times of 10 s with 10 μm resolution
- Robust fuel cell user program with 11 publications and 8 presentations in 2013

Acknowledgements

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DOE Technology Development Manager

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