An Overview of Neutron Imaging

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Motivation: Hydrogen Economy

“Hydrogen Economy” is an energy system based upon hydrogen for energy storage, distribution, and utilization. The term was first coined at General Motors in 1970.

- No Greenhouse gases or other pollutants
- Efficiency of 50% to 75%
- Can be produced from solar, wind, nuclear. Natural gas short term reduces CO₂ emissions by 50% to 10%

How can we improve Fuel Cells for use in cars?
Motivation: Why Neutron Imaging For Fuel Cells ???

Neutrons see material differently than x-rays and other common probes

X-ray cross section

neutron cross section

Fuel Cells have lots of aluminum and carbon (and some stainless steel)
Motivation: Why Neutron Imaging For Fuel Cells ???

The fine details of the water distribution in these Lilies are clear to neutrons even in a lead cask
An Overview of Neutron Imaging

Motivation: Why Neutron Imaging For Fuel Cells ???

- Small changes in water distribution in fuel cells affect performance and durability (more about this for the hands on exercises)
- Neutron Imaging measures these small mass changes (60 nanogram above) with high precision using standard fuel cell construction
- The NIST imaging facility was built primarily to study fuel cells
Neutron Sources: Uranium Fission Reactors

- “Large” (~$10^{14}$ cm$^{-2}$ s$^{-1}$) flux of neutrons at the core
- Continuous spectrum or one wavelength for a long time
- Generally more stable operation than an accelerator-based source
Neutron Sources: Pulsed Spallation

- High energy (1 GeV) proton beam smashes into a heavy nucleus
- Historically lower integrated neutron flux than reactor (SNS x3 lower than NIST)
- ESS will change that, having similar integrated flux to a 60 MW research reactor
- Short pulsed proton beam, choppers, and time-of-flight yields energy information
- Can resolve all wavelengths within a large range in each pulse
Neutron Sources: Continuous Spallation

- SINQ at the Paul Scherrer Institute is the only major facility
- 3 accelerators feed a multi-use facility and direct a 570 MeV continuous proton beam with thermal power of ~0.75 MW onto SINQ’s lead spallation target
- Neutrons are produced “continuously”, no intrinsic time of flight information
- Comparable to a medium power reactor source in flux, but without the societal concerns of a fission reactor

http://aea.web.psi.ch/Urs_Rohrer/MyWeb/weha.htm
Neutron Sources: Facilities with Imaging User programs

- Dingo at ANSTO, Australia (building)
- NEUTRA, ICON, BOA at PSI, Switzerland
- ANTARES at FRM II, Germany
- CONRAD and PONTO at HZB, Germany
- IMAT* at ISIS, England (building)
- ERNIS* at JPARC, Japan (building)
- NRF at Hanaro, Korea
- SANRAD at NECSA, South Africa
- CG1 at HFIR, United States
- BT2 at NIST, United States (building a cold neutron line)
- ESS* (planning)
- Venus* at SNS, United States (planning)  
  * = pulsed source
Method: Transmission Neutron Radiography/Radioscopy

\[ I = I_0 e^{-N \sigma t} \]

\( I_0 \) and \( I \) = Incident (or reference) and transmitted fluence (dimension of length\(^{-2}\))

Note: fluence rate is a directed particle beam while flux has no preferred direction

\( T = I/I_0 \) = transmission, assuming no scattered beam from sample

\(-\ln(T)\) = Absorbance or optical density – for neutrons OD is better as scattering out of the beam is often a large contribution (such as with water)

\( \sigma \) = cross sectional area due to absorption and scattering that the atom presents to the neutron (“barn book” or on-line tools, dimension of length\(^2\))

\( N \) = atom density \((N_A \rho / M_w \) dimension of length\(^{-3}\))

\( N \sigma = \Sigma \) = Total macroscopic cross-section (dimension of length\(^{-1}\))

Note: \( \Sigma \) is similar to \( \mu \), the x-ray mass attenuation coefficient and is sometimes used

\( t \) = Sample (i.e. water) thickness (dimension of length)

Simply measure water from the OD and knowledge of \( \Sigma_w \): \( t_w = -\ln(T) / \Sigma_w \)
Method: Neutron Image Formation

- Pinhole optics is basis for images
- Poke hole in reactor wall, form image of core at detector
- Optimal resolution when object contacts detector
- Geometric blur is given approximately by:
  \[ \lambda_g \approx \frac{z d}{L} \]
- High resolution of finite objects requires small aperture (d) or large L/D
- Small d or large L \(\rightarrow\) small flux
- No magnification, so intrinsic detector resolution only path to higher resolution
Method: Neutron Imaging Fuel Cells

An Overview of Neutron Imaging

![Image of fuel cell setup]

- **Sample, i.e. Fuel cell**
- **Point Source formed by Instrument**
- **Neutron Detector**

Mathematical expressions:

\[ I(i,j) \]

\[ I_0(i,j) \]

\[ T(i,j) \]

\[ \frac{I(i,j)}{I_0(i,j)} = T(i,j) \]
Instrument Design: Schematic of NIST Imaging Facility

L ≈ 6 m

D = 2 cm to 1 mm
Instrument Design: Neutron Collimation at BT2

- Primary collimator tapers beam
- Bismuth filter
  - 10 cm long
  - Liquid nitrogen cooled to increase thermal neutron transmission
  - Reduces gamma and fast neutron background
- Apertures
  - 5 positions to optimize L/D for each experiment
- Local shutter
  - Heavy concrete filled
  - 60 cm in length
  - 3 through tubes for additional collimation
- Fast exposure control
  - Allows < 1 second exposure control
  - Stops thermal beam for gamma and fast neutron background measurements
Instrument Design: Primary Collimator

- Hollow steel frame
- Filled with heavy concrete
- 10 cm long steel rings taper beam down to 2 cm maximum aperture size
- Borated aluminum discs are used throughout to reduce long term activation
Instrument Design: Bismuth Filter

- Bismuth crystal
  - Filters gammas and high energy neutrons
  - Ideally single crystal
  - Here we have several large single crystals
- 5 cm reduces thermal neutrons by 57%
- 15 cm reduces neutron fluence to 19%
- Banjo Dewar
  - Provides insulated liquid nitrogen jacket
  - Sealed and evacuated during operation

Banjo Dewar
(named after the musical instrument)

Super insulation/liquid nitrogen jacket

10 cm dia. hole for bismuth to sit
Instrument Design: Aperture Assembly

- Can be any material that fits
- 5 positions
- Largest aperture diameter is 2 cm due to primary collimation
- Easily changed without major shielding manipulations
- Using slits optimize geometry for fuel cells and batteries

5 apertures (2 cm, 1.5 cm, 1.0 cm, 0.5 cm, 0.1 cm)
Instrument Design: Local Shutter

- Rotates to 1 of 4 positions
  - Position 0 beam is blocked
  - Position 1 beam is collimated for 1 cm effective aperture
  - Position 2 beam is collimated for 2 cm effective aperture
  - Position 3 no collimation currently
- Filled with heavy concrete
- 60 cm long
Instrument Design: Sample Area inside BT2

Flexible space for ancillary equipment
Neutron Detectors: $^6$Li-doped ZnS

- We don’t directly detect the neutron
- Special isotopes that are highly absorbing ($^6$Li, $^{10}$B, Gd) are used to capture neutrons.
  - $^6$Li + n$^0$ → $^4$He + $^3$H + 4.8 MeV
  - Gd + n → Gd + γ + β (β has ~0.05 MeV)
- Resulting nuclear reaction produces charged particles with Mega-eV energies.
- Converter (ZnS:LiF or GadOx) stops the particles
- Energy lost by particles is ~proportional to the amount of light generated (GadOx is about 100x darker than ZnS:LiF)
- The spatial resolution of the converter is limited by charge particle range and by the spread of light in the converter.
- For AST screens ZnS:LiF this is 100 - 300 µm for neutron stopping powers of ~10 - 20%.
- PSI ZnS:LiF are about 50 µm
- Resolution of GadOx screens are more limited by the range of the β, 20 µm thick screens have 80% thermal neutron stopping power
Neutron Detectors: Visible (Scintillation) Light Imagers

- **CCDs (charge-coupled device)**
  - Deep cooling (-100°C) to eliminate dark current
  - Large area sensors possible (up to 10 cm x 10 cm with 100 MegaPixels)
  - Slow readout times (impacts dynamic imaging)
  - Relatively large pixel pitch 10-13 µm, adjustable field of view with a lens
  - 45° mirror and lens to keep the sensor out of the beam to avoid radiation damage

- **EMCCDs (electron-multiplier CCD)**
  - On pixel amplification reduces read noise compared to CCD
  - Small sensor areas (1k x 1k)
  - Fast Readout times possible (~30 Hz)
  - Pixel pitch of 8.5 µm, adjustable field of view with a lens
  - 45° mirror and lens to keep the sensor out of the beam to avoid radiation damage
Neutron Detectors: Visible (Scintillation) Light Imagers

- **sCMOS (scientific CMOS)**
  - New architecture developed by Fairchild, Andor and PCOImage
  - Read noise similar to EMCCD
  - Faster readout rates possible (up to 100 Hz) and larger sensor areas (5 MP) than EMCCD
  - Small Pixel pitch 6.5 µm, adjustable field of view with a lens
  - 45° mirror and lens to keep the sensor out of the beam to avoid radiation damage

- **α-Si (Amorphous Silicon)**
  - Developed for digital x-ray medical imaging
  - Amorphous, so sensor is very radiation hard (lifetime of 1 MegaRad)
  - Scintillator in direct contact with sensor, more efficient light collection
  - No lens – fixed field of view and spatial resolution
  - Large area sensor (~20 cm x 25 cm) with moderate spatial resolution (127 µm pixel pitch)

- All light imagers have some dark / read noise
Neutron Detectors: CCD Mirror Box

Water cooling lines to better reject heat from the Peltier cooler

Light-tight, flexible bellows

Motorized translation stage to adjust the focal distance for different field of view

With the scintillator mounted

With the scintillator removed to reveal mirror and lens
Neutron Detectors: Amorphous silicon

Varian Paxscan 2520, high energy model with readout electronics folded out from behind sensor. Picture taken in a class-100 clean hood.
Neutron Detectors: MicroChannel Plates

- Similar principle of photo-multiplier tube
- $^{10}\text{B}$ or $^{nat}\text{Gd}$ in wall glass absorbs neutron
- Reaction particles and high electric field create an electron avalanche
- Charge cloud detected with position sensitive anode
- No read noise due to large amplification, but small gamma-ray sensitivity
- Spatial resolution limited by channel separation and range of charged particle
- Ultimate resolution of $\sim 10 \ \mu\text{m}$
- Each detected neutron event is reconstructed, which introduces a deadtime
- Deadtime limits useable field of view to about 1-2 cm$^2$, which is OK for PEMFCs
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Neutron Detectors: MCP Detector Photo

- Sapphire window for UV transmission to condition detector
- High voltage requires oil-free high vacuum ($10^{-6}$ mtorr)
Neutron Detectors: Qualitative Impact of Resolution

A fuel cell was imaged at the same operating conditions and imaged with three different detectors.
Neutron Detectors: Spatial Resolution and the PSF

- Resolution described by point spread function (PSF) or its Fourier transform the Modulation Transfer Function (MTF)
- Common PSF models are Gaussian, Lorentzian, Voigt
- Quoted Resolution is from the spatial frequency at which MTF = 10% of its maximum
- Resolution typically measured from image of a knife edge

True, Sharp Image    **    Blurring function    Measured Image
Neutron Detectors: PSF Models

- The observed image is the convolution of the system PSF ($g$) with the true image ($f$):
  $$h(x,y) = f * g(x,y)$$
- There are at least 2 consequences of a PSF:
  1. Sharp features are smoothed
  2. There is an additive background
- Not every line shape is well-modeled by a Gaussian or a Lorentzian
- Looking only in the region of the edge, it can be difficult to discriminate between appropriate models
- A Gaussian decays rapidly relying on this as a PSF model blinds one to important systematic effects
- For more slowly decaying PSFs (i.e. Lorentzian) there’s an additive background that varies with the illuminated FOV:
  $$b(i,j) = \sum_{k,l} f(k,l)g(i - k, j - l) - f(i,j)g(0,0).$$
Neutron Detectors: Image restoration of a Cd edge

- The NIST a-Si has a PSF that is well-modeled by:
  \[ g(x) = 1 - \frac{x}{(x^2 + L^2)^{1/2}} \]
  \( L \) was found to be \( \approx 11 \) µm
- Ignoring noise, perform simple restoration in the Fourier domain
- Both the raw Edge image and raw flat field image are restored
- The edge profile is now sharper, and indicates in some instances restoration can improve image quality
- As well, the background is correct under the cadmium
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Neutron Detectors: Image restoration of Borated-aluminum

- Borated Aluminum coupon was inspected for the reactor – raw data was taken with a full field of view
- The nominal composition was expected to yield an optical density of about 3.2 for 1.8 Å neutrons
- From the raw data, the average optical density was 2.7; a 15% error
- From the corrected images, the average optical density is 3.1; beam hardening is likely to account for the difference
Neutron Detectors: Image Restoration of Dry Water Wedge

- Compare two data sets, full & masked fields of view
- Scattering backgrounds should be the same in both
- Steps are milled into a 2.5 cm thick Al block, max step size 2 cm
- Cuvette is placed 5 cm from the scintillator surface
- Images to the right are the masked image divided by full field of view image with no restoration, “optimal” restoration, and a truncated version of the PSF.

Masked and full field of view (20 cm by 25 cm) images of an empty Al cuvet
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Neutron Spectrum: Temperatures

- **Fast Neutrons**: Neutrons freed from the nucleus are very energetic, they need to be slowed to be useful.
- **Epithermal Neutrons**: Many isotopes have strong absorption resonances enabling isotopic selective imaging.
- **Thermal Neutrons**: A few isotopes have strong, low energy absorption; useful for scattering; commonest.
- **Cold Neutrons**: Have the strongest refractive effects; useful for phase imaging; beams can be transported 100+ m.
Neutron Spectrum: Moderated/thermal spectrum & rule of 2’s

- Fast neutrons inelastically scatter from water and come to thermal equilibrium at a temperature ~40 C for research reactors
- Near the peak of the thermal flux neutrons move at \( \approx 2000 \text{ m/s} \)
- This speed is about 20 meV in energy
- The thermal neutron DeBroglie wavelength is about 2 Å
  - Like X rays, neutrons can diffract from an atomic lattice
- Further moderation with liquid cryogens produce “cold” neutrons, >1000 m/s, >4 meV <4 Å
Neutron Spectrum: Neutron scattering from a free atom

Nucleus can be modeled as having an effective cross section for both scattering events and absorption.
Not included in this cartoon is spin effects.
Absorption increases with decreasing energy as $\propto 1/E^2$, or $\propto \lambda$.
Absorption cross-sections typically given for neutrons at 25 meV or 1.8 Å.
# Neutron Spectrum: Attenuation Coefficients for 25meV

<table>
<thead>
<tr>
<th>Material</th>
<th>Attenuation Coefficient</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>~ 3 mm$^{-1}$</td>
</tr>
<tr>
<td>Carbon</td>
<td>~ 0.6 mm$^{-1}$</td>
</tr>
<tr>
<td>Aluminum</td>
<td>~ 0.09 mm$^{-1}$</td>
</tr>
</tbody>
</table>

![Graph showing attenuation coefficients for various materials](image)

### Attenuation Coefficients, $N \sigma = \mu$ (cm$^{-1}$) or $\Sigma$

Assuming standard mass density.

- **$^{113}$Cd**
- **$^{10}$B** (Natural Boron)
- **$^6$Li**
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Neutron Spectrum: Some on-line resources:


### Scattering Length Density Calculator

<table>
<thead>
<tr>
<th>Compound</th>
<th>Neutron SLD</th>
<th>Cu Ka SLD</th>
<th>Mo Ka SLD</th>
<th>Neutron Inc. XS</th>
<th>Neutron Abs. XS</th>
<th>Neutron 1/e length</th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂O</td>
<td>7.31E-7 (Å⁻²)</td>
<td>9.03E-6 +3.19E-8i (Å⁻²)</td>
<td>8.99E-6 +5.99E-9i (Å⁻²)</td>
<td>3.04 (cm⁻¹)</td>
<td>0.0118 (cm⁻¹)</td>
<td>0.3262 (cm)</td>
</tr>
<tr>
<td>Density (g/cm³)</td>
<td>1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Wavelength (Å)</td>
<td>1.8</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

[Scattering Length Density Calculator](http://www.ncnr.nist.gov/resources/sldcalc.html)
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Neutron Spectrum: Changes in Scattering Effects

- **Epithermal**: Resonance absorption, can identify isotopes
- **Thermal**: coherent and incoherent S-wave scattering, Bragg scattering, and 1/v absorption
- **Cold**: coherent and incoherent S-wave scattering, Bragg cut-off, “large” refractive index for phase imaging, 1/v absorption
- 1/v absorption gives rise to Beam Hardening

http://www.psi.ch/niag/neutron-interaction-with-matter
Neutron Spectrum: Beam Hardening

- Scattering cross-sections can depend on energy (wavelength)
- In general, Imaging beams are polychromatic
- For water, the rotational and vibrational modes affect the cross-section
- Increasing $\sigma$ with increasing $\lambda$ (or decreasing energy) gives rise to beam hardening, the beam transmitted through the sample has a more energetic spectrum than the incident beam
Neutron Spectrum: Bragg Scattering

- For crystalline materials there is an increase in transmission for neutron wavelengths above the maximum lattice spacing – “Bragg Edge Imaging”
- This can be used to:
  - Measure strain
  - Map chemical phase transitions
  - Locate crystalline species in a sample

Fe Cross section

K. Iwase, NIMA, 2009
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Water Metrology: Reminder of the setup

Sample, i.e. Fuel cell

Point Source formed by Instrument

\[ I(i,j) \]

\[ I_0(i,j) \]

\[ T(i,j) \]
Water Metrology: What to measure

- Measure a dry image (or an image of a reference state)
- Measure a wet image (or at some operating state)
- Measure the additive background image
- Use open beam or unchanging region to scale images for neutron source flux variation (i.e. orange box)
- Form the transmission image from the scaled, background corrected images

\[
\frac{(\text{Wet I} - \text{Background})}{(\text{Dry I} - \text{Background})} = T = e^{-\Sigma t}
\]
Water Metrology: Calibration of Optical Density with $t_w$

Transmission

$T = e^{-\Sigma t}$

Optical Density

Colorized O.D.

$$OD(t_w) = \Sigma w t_w + \beta w t_w^2$$
Water Metrology: Random Uncertainty Analysis

- Uncertainty in the liquid water thickness is limited by Poisson Counting Statistics:
  \[ \delta t \approx \sqrt{\frac{2}{\mu^2 I_0}} \]

- \( I_0 \) increases with
  - Neutron Intensity
  - Neutron Detection Efficiency
  - Area (pixel area, # pixels)
  - Exposure time

Calculate statistics using different areas and integration times

Can nanostructured synthetic polymers mimic the form and function of natural tissue?

Bob Moore, Jong Keun Park
Application: Actuation of an Oriented Nafion® IPMC

Square wave voltage input of 0.1Hz, 3 V

How does the membrane move: Dipole rearrangement? Hydraulic transport?
The observed gradient in water distribution upon electrical stimulation directly supports a \textit{hydraulic} contribution to the actuation mechanism.
Application: Water Gradient Across the IPMC Thickness

IPMC thickness = 1.4 mm (fully hydrated)

Further Refinement: Maximum attenuation occurs in hydrated zones, thus actuation is stimulated by zones of dehydration not differential swelling of clusters across membrane.
Application: Optimum Fuel Cell Channel Geometry

- Water slugs trapped in channels impede reactant flow
- Active water purges require additional energy
- Passive water removal mechanisms (surface coating or geometry) are preferred
Application: Channel Geometries and Surface Treatments

- **Rectangular channels**
  - Water flow is laminar tending to constrict and plug the channels
  - Water plugs form as large slugs and can be difficult to remove.

- **Triangular channels**
  - Water stays at the corner interface with the diffusion media leaving the apex of the channel more clear.
  - Water tends to come out in smaller droplets instead of large slugs, which require a high pressure differential to remove.
Application: Rectangular Comparison at 0.5 A/cm²
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Application: Triangular Comparison 0.5 A/cm²

Uncoated

PTFE Coated
Application: Geometry Comparison 0.5 A/cm²
Application: Lithium intercalation in HOPG

“stills” showing growth of Li- hot spots during charge

Movie during charge/discharge cycles
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Application: Tomography Water retention in (un)treated soil

Treated soil – higher retention

Untreated soil – lower retention

R. Murphy, Rhodia/Solvay
Application: Transmission vs. Phase Imaging

- Small variations due to stamping are not visible in the transmission image.
- The quarter details are very apparent in the phase gradient image.
- Phase imaging is useful to measure small density fluctuations.
Key Points

- Neutron imaging techniques can measure the concentration of water and lithium in metallic matrices and some applications were discussed.
- Pinhole optics is simple but requires one to optimize the imaging beam line for the experiment.
- Currently “best” resolution is about 10 µm, though several groups are looking into ways to improve this.
- The detector point spread function is important to measure and if possible use to deblur images to correct for an “additive background”.
- The spectrum of neutrons from spallation and reactor sources can be used for material identification, but is also the cause of an important systematic: beam hardening.
- The hands-on tutorial on water sorption in membranes will explore the impact of beam hardening and other systematic measurement effects.