Neutron Imaging Facility Development and Research Trend at NIST

M. Arif\textsuperscript{a}\textsuperscript{*}, D.S. Hussey\textsuperscript{a}, E.M. Baltic\textsuperscript{a}, D.L. Jacobson\textsuperscript{a}

\textsuperscript{a}Neutron Physics Group, Radiation Physics Division, Physical Measurement Laboratory, NIST, Gaithersburg, MD, 20899 USA

Abstract

The National Institute of Standards and Technology (NIST) maintains a sustained focus in the development of advanced neutron imaging facilities and hardware components to enable breakthrough research with vastly improved spatial and temporal resolutions, and to identify and employ research practices important to a wide variety of industrial and scientific applications. NIST’s main focus is to enable research with broad appeal and commercial impacts. In this article we will give a brief overview of the NIST facility, select examples of current research, and finally comment on emerging technologies including advance manufacturing where neutron imaging has the potential to play an important role.

Keywords: Neutron imaging; Fuel cell; Battery; Wolter Optics; Neutron Detector

1. Introduction

NIST has operated a thermal neutron imaging instrument at the NIST Center for Neutron Research (NCNR) since 2003. The primary science driver for the development of the instrument is energy related research that includes, among others, optimizing water management in proton exchange membrane fuel cells (PEMFCs) for its rapid development and commercialization. As such, PEMFC test and control infrastructure is tightly coupled to the instrument enabling external researchers to perform a wide array of PEMFC and electrochemical studies. NIST is also developing a Wolter Optics based state of the art highly efficient and very high resolution cold neutron imaging station to expand current research focus to meet anticipated future needs.
2. Facility

2.1. General Description

Shown in Figure 1 is the layout of the Beam Tube 2 (BT2) imaging facility which is located in the confinement building of the NCNR. Not shown is the primary collimation which is composed of graduated steel rings which create a 2 cm focal spot at the aperture position. After the primary collimator is 10 cm of a quasi-single crystal of Bi that is cooled by liquid nitrogen to eliminate thermal-diffuse neutron scattering. After the Bi filter is an aperture paddle that contains 5 positions that can be selected by a push-button control. The aperture paddle can be exchanged to provide an aperture geometry that better suits the experiment. A secondary collimator, which also serves as the local safety shutter, follows the aperture. There is a 1° and 1.5° divergence for the two collimations. A fast thermal neutron stop follows the local shutter which is composed of 1 mm of Cd and 5 mm of borated-aluminum which is used to measure the gamma and fast neutron response of imaging detectors. Evacuated aluminum tubes, 30 cm in diameter enclose the flight path to prevent air scattering. The tubes are about 75 cm in length to permit short flight paths to provide higher intensity for experiments requiring high frame rates. A motorized beam mask composed of lithiated plastic, borated aluminum and cadmium define the region of interest in the image, and the edge of the beam mask is always visible in an image to eliminate a systematic uncertainty due to the point spread function of the detector. The samples are rigidly mounted to a motorized 5-axis table with horizontal translations, pitch, roll, and yaw; a vertical axis can be motorized if required. Detectors are mounted on a platform that is mounted to the shield wall. The beam stop consists of a magnesium entrance window followed by 10 cm of boron carbide powder, and then alternating stacks of polyethylene and lead, which is encased in a shield composed of 20 cm of wax and steel shot. The wall and roof shields are 20 cm wide steel forms filled with wax and steel shot. A couple of common instrument configurations are for an L/D = 450 the fluence rate is $1.4 \times 10^7$ cm$^{-2}$ s$^{-1}$ and for a 1 mm wide slit with 1 cm height (L/D = 6000,600) the fluence rate is $8.0 \times 10^5$ cm$^{-2}$ s$^{-1}$. 

Fig. 1. Cartoon of NIST Neutron Imaging Facility
2.2. Infrastructure and Resources for Experiments

The facility provides researchers with two fuel cell test stands, one that can control automotive scale fuel cells and small stacks and one that is appropriate for controlling PEFMCs with active area of 50 cm$^2$ or smaller. A hydrogen generator (ProtonOnsite S40$^*$) provides pure hydrogen gas with a maximum flow rate of 19.8 slpm at a gauge pressure of 1.38 MPa (200 psi). A liquid nitrogen dewar supplies users with nitrogen gas, and there is a water purifier that supplies the instrument with 18 MΩ cm$^{-1}$ water. Hydrogen is directly vented into the confinement building through the hydrogen vent which is located 4 m above the floor. Extensive computational fluid dynamics modeling of the confinement room demonstrated that with the refresh rate of the confinement building under continuous operation of the hydrogen generator the maximum concentration of hydrogen as 20 times below the lower flammability limit in air, except in a small region above the release point. As such, it was deemed safer to vent the hydrogen into confinement rather than create a penetration out of confinement. In addition to the infrastructure permanently mounted to the instrument, users have access to Viasala humidity sensors and a Zahner IM6 electrochemical workstation.

Researchers have access to several different detectors which are optimized for different application. A Varian Paxscan 2520 that has the electronics folded out to enable them to be shielded provides a 25 cm x 20 cm field of view with a pixel pitch of 0.127 mm at a maximum frame rate of 10 Hz or 0.254 pixel pitch at a maximum frame rate of 30 Hz. One issue with so-called high energy flat panels is the need to incorporate active cooling inside the shielding to stabilize the dark current; this is done at NIST by blowing cold air (-10 °C) through the panel enclosure. A LiF:ZnS scintillator, 0.3 mm thick is used which yields a spatial resolution of about 0.25 mm. There are two high resolution microchannel plate (MCP) detectors with cross-strip anode readouts developed by Sensor Sciences. The first of these detectors is a 40 mm diameter active area with 5 µm pixel pitch, with an MCP that has 4 µm diameter pores on 6 µm centers, yielding a spatial resolution of ~13 µm and has been available to users since October 2009. The second MCP has a large active area of 9 cm x 9 cm with a 6 µm pixel pitch, with an MCP that has 6 µm diameter pores on 8 µm centers, yielding a spatial resolution of ~20 µm. A limitation of MCP detector technology is that they are event counters and one must limit the field of view to a few cm$^2$ in order to avoid dead time effects. In the case of high resolution PEMFC or Li-ion battery imaging, this field of view restriction is not limiting as the regions of interest are sufficiently small. The most versatile of the available detectors is the Andor NEO, a scientific complimentary metal-oxide semiconductor (sCMOS) camera that has read noise similar to that of an electron multiplied charged-coupled device (EMCCD) but has a larger format (5 MP), smaller pixel pitch (6.5 µm) and with higher frame rates (up to 50 Hz, up to 100 Hz with a different camera model is possible). The sCMOS camera views either a LiF:ZnS or GadOx scintillator using Nikon macro lenses including a 50 mm f/1.2, 85 mm f/1.8, 200 mm f/4 (max reproduction ratio of 1:1), and 105 mm f/2.4 (max reproduction ratio of 1:1). Higher reproduction ratios can be achieved using extension tubes. There are two GadOx screens available, 20 µm and 7 µm thick. The 20 µm screen has a thermal neutron stopping power of about 80 % with a resolution of about 20 µm and the 7 µm screen has a stopping power of about 30 % with a resolution <20 µm. Due to the low light output of GadOx, the 7 µm screen has not been widely employed. However, an imager intensifier will be incorporated into the detector system which will greatly improve the performance of the GadOx screens with the sCMOS camera. The LiF:ZnS screens include a 0.3 mm thick and 0.15 mm thick screens yielding spatial resolution of about 0.25 mm and 0.15 mm.

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$^*$ Certain trade names and company products are mentioned in the text or identified in an illustration in order to adequately specify the experimental procedure and equipment used. In no case does such identification imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the products are necessarily the best available for the purpose.
2.3. Facility Use

Since the instrument was built, the primary research focus has been PEMFCs. This focus has recently broadened to include other electrochemical applications, two phase flow in heat pipes, fluid flow in porous media (concrete, geothermal energy storage, oil and gas extraction), and strain mapping. Figure 2 shows the distribution of research fields at the instrument over the time period 2010 to 2014. The beam time percentage was allocated 33 % to industry (19 separate institutions), 32 % to national laboratories (5 separate institutions including NIST) and 35 % to academia (31 separate research groups).

2.4. Planned Upgrade

A double monochromator assembly was installed in summer 2012 and has been used for several neutron-energy selective imaging experiments (see below). The monochromatic beam is displaced 30 cm vertically to ensure that the experiment is completely out of the direct beam, including the gamma ray and fast neutron beam emanating
Two highly oriented pyrolytic graphite (HOPG) crystals with 0.5° mosaic spread (ZYA grade) are used to monochromate the beam using a parallel reflection geometry. For the thermal neutron spectrum available at BT2, the wavelength resolution of the assembly is improved by using the HOPG (004) reflection, with a field of view of about 7 cm × 7 cm. The useful neutron wavelength distribution spans 0.1 nm to 0.3 nm, corresponding to the thermal neutron source available. Currently, images using the sCMOS with a 0.15 mm LiF:ZnS with an effective pixel pitch of 25 µm require about 6 min to 9 min exposure time (broken into three separate images combined with a median filter to reject noise from gamma rays, etc.) using an L/D of 450. Exposure times can be reduced by coarsening the spatial resolution or by using a new detector system described below.

A near-future upgrade will be the incorporation of an image intensifier with the sCMOS camera viewing a GadOx scintillator. Commercially available generation 3 intensifiers have high spatial resolution (50 line pairs per mm is typical), long operational lifetime of over 10,000 h, and provide gains of over 1,000. Since the light output from GadOx is about 1 % of that of the LiF:ZnS screens, the use of GadOx has been limited to reasonably high neutron fluence rate situations, for instance GadOx does not produce enough light for use in energy selective imaging. However, GadOx has much higher neutron stopping power with a thicker screen, a 20 µm thick GadOx deposit has about 80 % thermal neutron stopping power whereas a 0.3 mm thick LiF:ZnS screen has about 20 % stopping power. By amplifying the scintillation light and overcoming the read noise in the camera, one will be able to make use of the better neutron counting statistics possible with GadOx. As an example, shown in Figure 3 is the measured signal to noise ratio as a function of intensifier gain for 1 s exposures with a 20 µm thick GadOx screen in a neutron fluence rate of $6 \times 10^6$ cm$^{-2}$ s$^{-1}$. By enabling the use of scintillators with higher neutron stopping power and better spatial resolution, it is anticipated that the image intensifier will improve the image quality for a wide array of experiments, including tomography, high spatial resolution imaging, high frame rate imaging, and energy selective imaging.

3. Recent Measurements

A) Boeing 787 Battery Fire: On January 7, 2013, the auxiliary power unit of Boeing 787 caught fire at Logan Airport in Boston, MA. The National Transportation Safety Board (NTSB) conducted an extensive investigation of the incident and has recently issued its findings, NTSB (2013). The NTSB was looking for the root cause of the fire, and a potential mechanism was a lithium salt deposit might have shorted a battery terminal to the external case. Neutron tomograms of all the battery headers involved in the fire were acquired to exploit the high neutron absorption cross section for lithium. The resultant images indicated that there were no such lithium salt deposits on the case. Shown in Figure 4 are renderings from three battery headers, cells 5 and 6 were the cells in which the fire started and cell 2 suffered minimal damage from the incident.

B) Performance of carbon paper free diffusion media in PEMFCs: At mass production volumes, the carbon paper that composes the gas diffusion layer in PEMFCs represents a large fraction of the cost of a fuel cell stack. In addition, the thickness of the layer contributes to mass transport resistance. Researchers at Nissan and the University of Connecticut have devised a fuel cell that requires only the beneficial microporous layer (MPL) without the carbon paper substrate, Kotaka et al (2014). Without the carbon paper, the limiting current density is increased by nearly a factor of 3 over a fuel cell with carbon paper and no MPL. Using high resolution neutron radiography, it was confirmed that with only an MPL, there is no liquid water buildup between the cathode and the flow channels, which will reduce mass transport resistance.
C) Non-precious metal group PEFMC cathode catalysts: About half of the cost of a PEMFC stack is due to the platinum required for the oxygen reduction reaction in the cathode catalyst layer. While significant reductions in the amount of Pt required have been achieved, a catalyst that had nearly similar performance containing no precious metals is still desired by the PEMFC community. Researchers at Los Alamos National Laboratory have been developing an iron-based catalyst that is functionalized with nitrogen groups stemming from a polyaniline precursor (PANI). In small cell testing, severe mass transport and flooding limited the performance of the non-precious metal group (NPGM) catalyst. Since the NPGM catalyst is significantly thicker than conventional Pt on carbon catalyst (Pt/C), it was thought that the thickness was causing the flooding. Neutron radiography was used to compare the water content of two PEFMCs, one with a 50 µm thick Pt/C catalyst and one with an 80 µm thick PANI catalyst, Hussey et al (2013). Figure 5 compares the two cells at open circuit voltage, where no current is produced, with inlet gases with 100% relative humidity (RH), the PANI catalyst layer has about 2.5x more water than the Pt/C catalyst, including a large spike in the water content at the interface between the membrane and the PANI catalyst. This spike indicates that the surface is hygroscopic and is the primary reason for the flooding.

D) Oscillating/Pulsating Heat Pipes: Electronics miniaturization is increasing to the point that the power density is too great for conventional heat pipes (CHP’s), which are used to cool many portable, high heat density generating electronic chips. For large heat inputs the pressure generated from the vapor phase overcomes capillary pressure pumping liquid to the evaporator shutting down the CHP capillary-phase change cycle. Currently heat fluxes reached 10 W/cm² to 40 W/cm² with total heat power of 10 W to 150 W. Next generation chips are expected to hit 80 W/cm² with 300 W total power; laser diodes can achieve 500 W/cm². Silicon chip reliability decreases 10% for every 2°C temperature rise with a limit of 125°C. Approximately 55% of all electronics failures are attributed to superheating of chips. Heat sinks are heavy and fail at 500 W/cm² due to low thermal conductivity. One technology that could operate at higher energy densities is the oscillating/pulsating heat pipe (OHP) invented in the 1990’s. OHPs are sealed metal pipes evacuated and filled with a driving fluid like water (high latent heat), acetone, ammonia, ethanol and others depending on the desired fluid properties.
The pipes are filled with multiple slugs of 50 % liquid phase and 50 % vapor phase. The pipe is then bent to snake through an evaporator region and back to a condenser region several times. Evaporation of liquid films in the evaporator and condensation of vapor in the condenser creates a pressure differential that sets the in motion oscillations or pulsations of the slugs inside the pipes. As the liquid slugs pulsate they make thermal contact with the walls of the tube in the evaporator region absorbing heat through the specific heat capacity of the liquid. Upon pulsating back to the cooler evaporator region the liquid then transfers the heat. This process was not well understood until neutron imaging, shown in Figure 6, was applied to directly visualize and correlate the liquid/vapor slug motion with measured heat transfer in the OHP. Neutron imaging has been applied to both guide and validate the development of nonlinear thermo-mechanical finite element models of OHP operation, Yoon et al (2012). In addition to model development the effect of varying pipe diameters, fluid properties and orientation can be directly measured and correlated with heat transfer properties to experimentally understand and improve the design of these devices, Peng et al (2014). Due to the high velocity of the slugs the motion was captured at a thermal neutron fluence rate of $3.2 \times 10^7 \text{ cm}^{-2} \text{s}^{-1}$ with $L/D = 300$, using the Varian Paxscan 2520 detector with 300 µm $^6\text{Li:ZnS (Cu,Al,Au)}$ screen running at 30 fps.

### 3.1. Future Trend

Future research will be aided by Wolter Optics based cold neutron imaging facility currently in development, Liu et al (2013). NIST is expect to continue and expand focus in the following research areas that has demonstrated and potential high commercial impacts:

- **Fuel Cells**: Water in catalyst in layers, Water transport in the GDL
- **Batteries**: Automotive batteries for propulsion
- **Geology**: Enhanced oil recovery, Fracking, Geothermal energy
- **Nuclear Fuel life cycle**: Fuel rod inspection
- **Heat Pipes**: Fluid properties and surface treatments
- **Advanced Manufacturing**: Hybrid welds, additive metal manufacturing
- **Porous Media**: Soil additives and root growth, Concrete
- **Law Enforcement**: Neutron Forensics
4. Conclusions

NIST has maintained a very successful goal oriented neutron imaging program for more than a decade. NIST focus on energy related research has contributed immensely in the development of fuel cells and to meet industrial needs of commercial significance in many other areas. The NIST neutron imaging program continues to develop state of the art imaging related hardware and methods and applies these new tools to a wide range of energy, engineering, and materials science applications. In addition, NIST maintains close contact and association with industry, academia, and government agencies that contributes in identifying potential pivotal role of neutron imaging in research areas that benefits the general neutron imaging community.

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