U.S. DEPARTMENT OF ENERGY | Nuclear Energy

Material Protection, Accounting and Control Technologies (MPACT) Campaign Overview and Advanced Instrumentation Development

Michael Miller, Ph.D.
National Technical Director
Los Alamos National Laboratory

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Argonne National Laboratory | INL Idaho National Laboratory | Los Alamos National Laboratory | Lawrence Livermore National Laboratory | Oak Ridge National Laboratory | Pacific Northwest National Laboratory | Sandia National Laboratories

Introduction

- **Preventing, deterring, and detecting misuse of nuclear materials and associated fuel cycle technologies is of paramount concern to both national and global security. Success in this area is critical for the existing and future nuclear energy enterprise.**
- **A large national and international infrastructure exists to address this potential threat involving many U.S. government agencies and the international community. For nuclear energy sector**
  - The IAEA and the NRC are largely focused on verification and maintaining control of nuclear material (respectively) within the civilian nuclear fuel cycle (non-R&D focus).
  - NNSA is primarily focused on international safeguards and security (civil and military) and supporting the IAEA through technology development. Programs supporting nuclear energy:
    - Defense Nuclear Nonproliferation Research and Development (NA-22) – Nuclear Weapons and Material Security (WMS)
    - Nonproliferation and International Security (NA-24) – Next Generation Safeguards Initiative (NGSI) and International Nuclear Safeguards and Engagement Program (INSEP)
  - US State Dept. – Member State Support Program to the IAEA provides technology and personnel expertise to strengthen international safeguards and the Nonproliferation Treaty.
  - DOE-NE Fuel Cycle Technologies R&D is focused on enabling the US civilian nuclear fuel cycle (domestic) by coupling material protection, accounting and control technology development with nuclear technology development.

MPACT is about Next Generation Nuclear Materials Management

- **Mission** – Develop innovative technologies and analysis tools to enable *next generation nuclear materials management* for existing and future U.S. nuclear fuel cycles, to manage and minimize proliferation and terrorism risk.

- **Objectives**
  - Develop and demonstrate advanced material control and accounting technologies that would, if implemented, fill important gaps
  - Develop, demonstrate and apply MPACT analysis tools to assess effectiveness and efficiency and guide R&D
  - Develop tools, technologies, and approaches in support of used fuel safeguards and security for extended storage, electrochemical processing, and other advanced nuclear energy systems
  - Perform technical assessments in support of advanced fuel cycle concepts and approaches
  - Develop guidelines for safeguards and security by design and publish guidance documents

Role of Instrumentation for MPACT

- Traditional instrumentation for nuclear materials accounting (NMA) is radiation (nondestructive assay) and chemistry (destructive assay) based and provides _quantitative_, _independent_ determination of U and Pu mass to meet regulatory timeliness requirements for diversion detection
- Measurements fit within an overall safeguards and security approach for each fuel cycle facility
  - Containment & surveillance provide continuity of knowledge
  - Physical security controls access
- Process monitoring augments NMA, could be more fully used (integrated system, transparency of operations) – challenge is quantitative use of the data and uncertainty propagation.

- MPACT R&D responsive to other FCT Campaigns and program priorities

MPACT Control Accounts

Management and Integration

- Technical and project management, maintain internal and external interfaces, provide technical support, international engagements

Safeguards and Security by Design – Echem

- Integrated safeguards and security for electrochemical process
- Systems approach (safeguards and security performance model, fundamental mass flow models, pattern recognition and statistical inference)
- Technology development (actinide sensor, level/density sensor, microfluidic sampler, voltametry)

Used Fuel Extended Storage

- Concepts and approaches for integrated safeguards and security for used fuel extended dry storage (best practices, risk-informed security framework, data gaps)
- Signatures and assessments (signature development, advanced monitoring techniques, vulnerability assessments, consequence analysis)

Exploratory Research/Field Tests

- Advanced instrumentation development and field tests for next generation nuclear materials management
- Microcalorimetry, multi-isotope process monitor, high-dose neutron detector

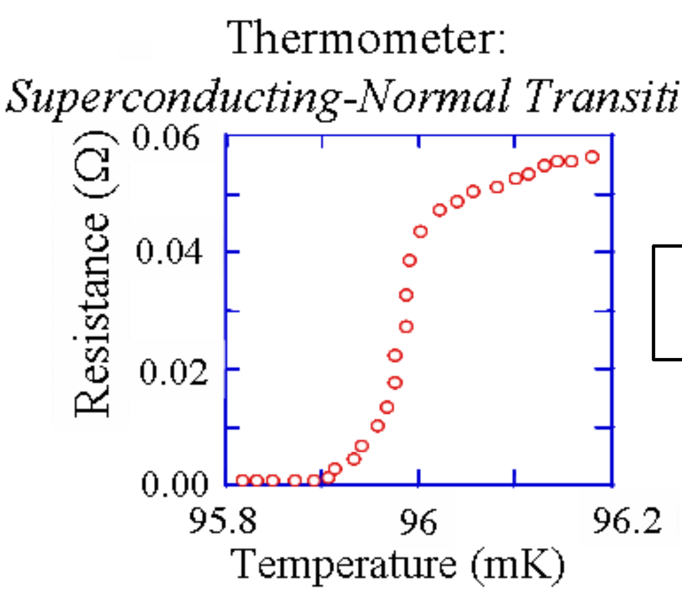
Level 2 Milestones – FY 2015

- **Mathematical basis for PM combination with NMA – LANL, 4/15**
- **Actinide sensor tests with PuCl3 – INL, 5/15**
- **Level/density probe design for field test (JFCS) – INL, 6/15**
- **Signature development for electrochemical processing – LANL, 7/15**
- **Characterization of microcalorimeter bandwidth limits – LANL, 7/15**
- **Security considerations for consolidated ISF – SNL, 8/15**
- **Demonstrate microfluidic sampling with spectroscopic data – ANL, 8/15**
- **MIP Monitor deployment readiness (H-Canyon)* – PNNL, 8/15**
- **Prototype instrument design for dry cask neutron fingerprint – LANL, 9/15**
- **Integration of PM with EChem SSPM – SNL, 9/15**

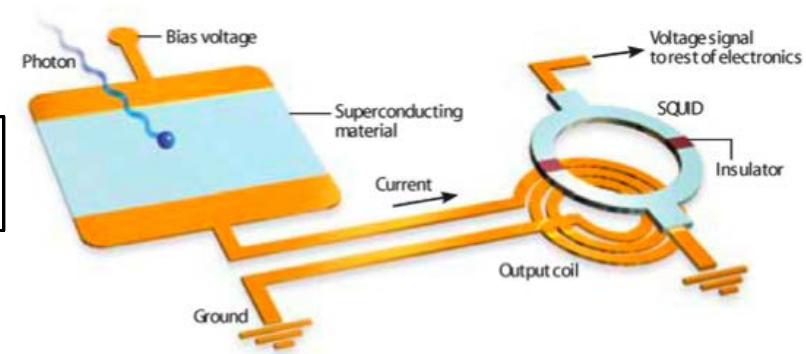
*actual schedule is dependent on facility run cycle

Microcalorimetry (LANL)

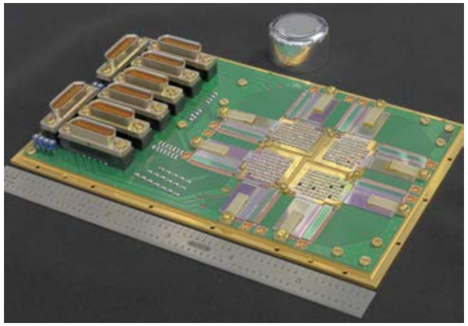
- Gamma spectroscopy combined with neutron or heat measurement is a staple of nuclear material accounting and control
- Increased resolution (10x) can enable improved accuracy and precision of Pu analysis by gamma spectroscopy
- Close gap between NDA and DA methods (<0.1% vs 1%)
- Reduce reliance on sampling and mass spectrometry

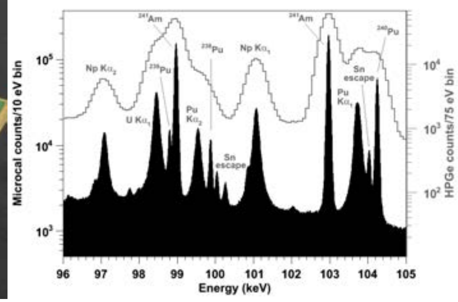


$$\Delta E \sim T(k_b C)^{1/2}$$

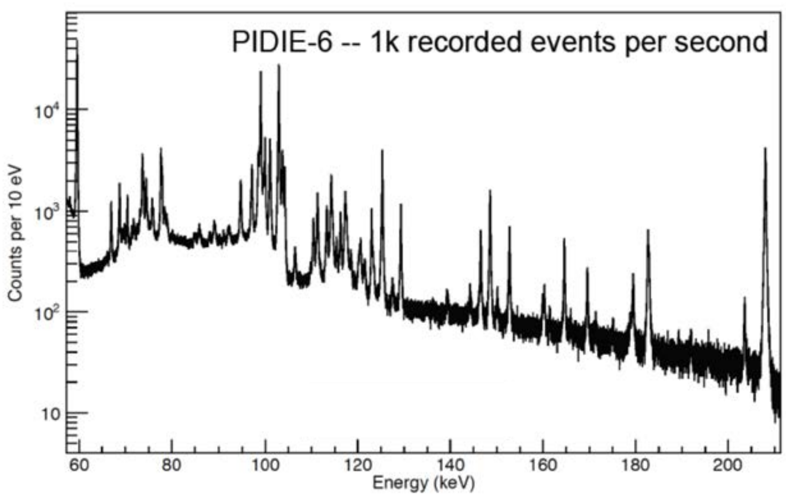


Microcalorimetry (LANL)

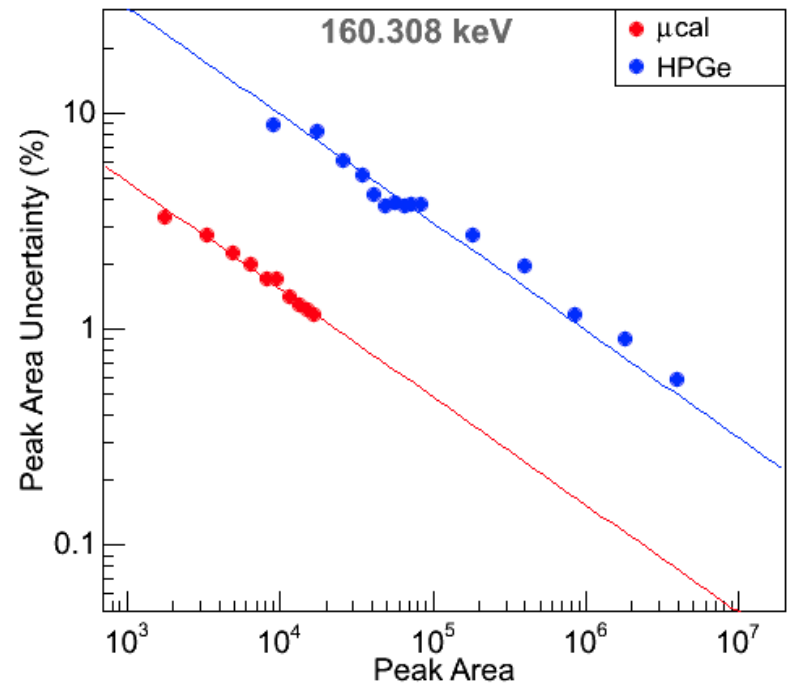




High yield (>90% of 256 pixels) arrays with <100 eV resolution



High rate data collection for practical applications



Demonstrated improvement over HPGe performance

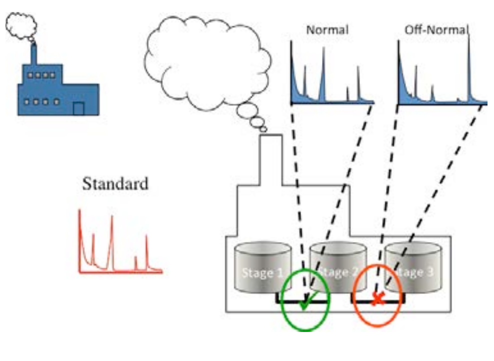
Recent publications

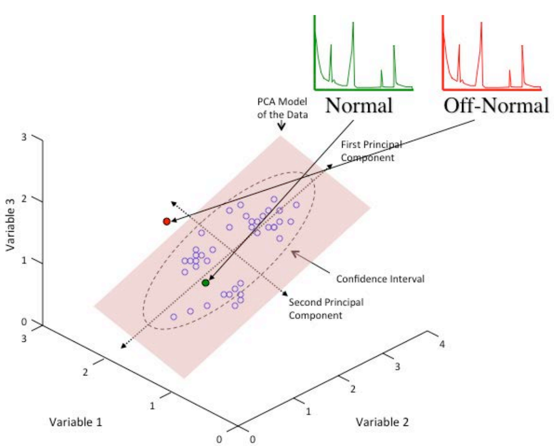
- A.S. Hoover, et al., IEEE Trans. Nucl. Sci. 61 2365-2372 (2014)
- R. Winkler, et al., Nucl. Instrum. Meth. (in press, 2014)
- T. Burr, et al., Nucl. Data Sheets (in press, 2014)
- B.K Alpert, et al., Rev. Sci. Instrum. 84(5) 056107 (2013)

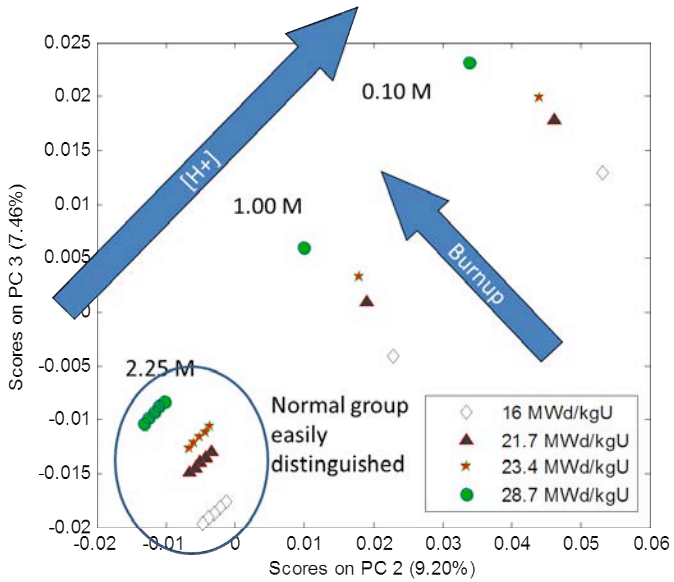
Out-Year Focus: Prototype demonstration of Microcal for MPACT in 2016 - 2017

Multi-Isotope Process Monitor (PNNL)

- Changes in process chemistry may be indicative of facility misuse and detectable via monitoring of fission product distribution by gamma spectroscopy (high or low resolution)
- Principle components analysis used to compare spectra with baseline







Multi-Isotope Process Monitor (PNNL)

Preparing for field tests:

- Spent fuel measurements at PPNL hotcell
- H-Canyon scoping and background measurements

Recent publications

- K. Dayman, et al., Nucl. Instrum. Meth. 735 624-632 (2014)
- S. Bender, et al., J. Radioanal. Chem. 296(2) 647-654 (2013)
- C. Orton, et al., Nucl. Instrum. Meth. 672 38-45 (2012)

Out-Year Focus: MIP demonstration(s) in fuel cycle facilities (e.g., H-Canyon) in 2015 - 2016

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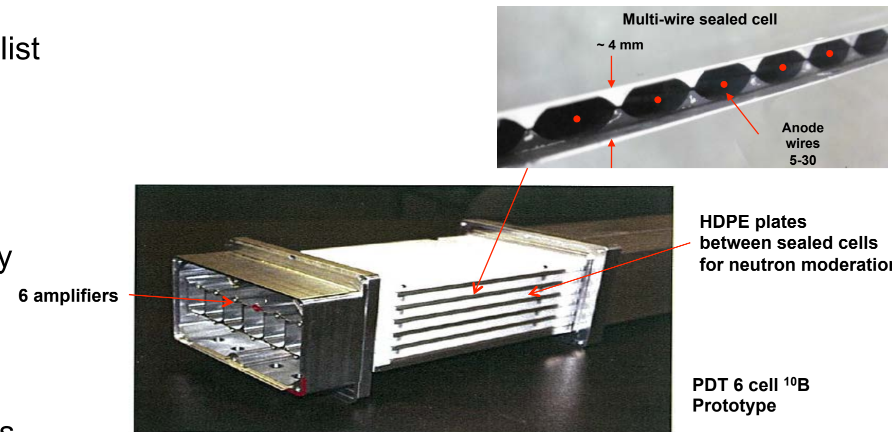
High-Dose Neutron Detector (LANL)

New Project

- Neutron counting in a high radiation environment (n,γ) using ^{10}B plate detector
- Target application is Echem product assay

- Multi-plate design provides neutron energy measurement with list mode readout of each gas cell
- The front/back cell ratio provides a measure of the neutron multiplication
- The ^{144}Cm balance method will require a multiplication measurement from the TRU ingot. The multiplication can verify the Pu content

- Narrow gas cells (4 mm) for minimal gamma ionization signal
- Fast amplifier shaping time parameters for enhancing neutrons versus gamma
- <u>List mode</u> readout for the 6 parallel plate cells
- List mode potential rejection of gamma events via multiple cell coincidences. (The charged particles from the neutron reactions are limited to a single cell.)



Out-Year Focus: Proof-of-concept in 2015 – 2016, prototype field test (JFCS) in 2017

Actinide Sensor and Voltametry (INL)

Nernstian potentiometric cell

- The voltage generated across the ion conducting electrolyte is proportional to the logarithm of the concentration of the mobile species.

$$V = \Delta E = \frac{RT}{zF}\ln a_{Gd^{3+}(ref)} - \frac{RT}{zF}\ln a_{Gd^{3+}(ws)} = A - B\ln a_{Gd^{3+}(ws)}$$

E = electrical potential (V) of the cell

R = molar gas constant (8.3144 $Jmol^{-1}K^{-1}$)

T = absolute temperature (K)

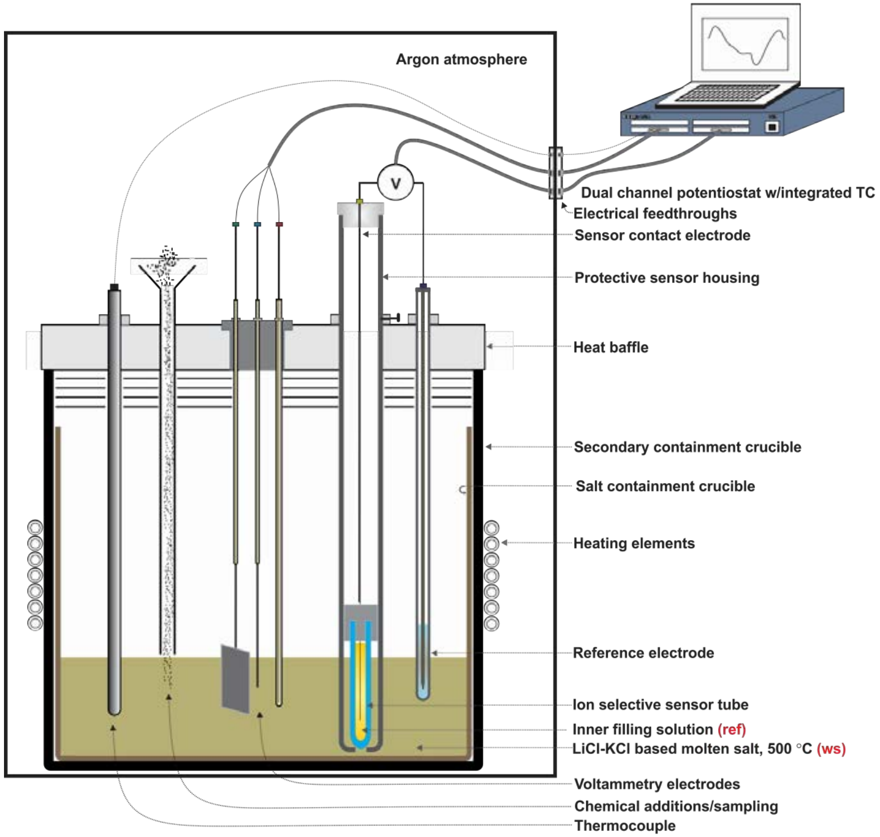
z = charge of the measured ion

ref = reference solution (constant)

ws = working solution (unknown)

A = constant

B = constant

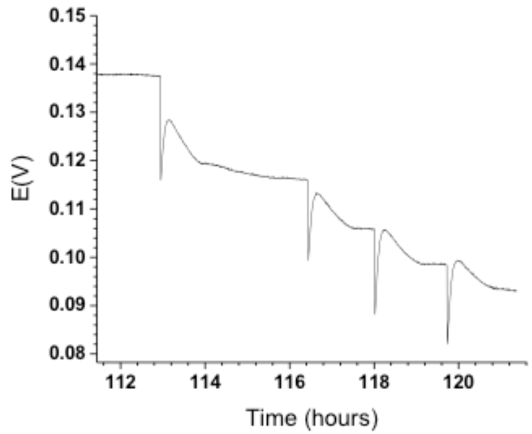


3- electrode cell for voltammetry

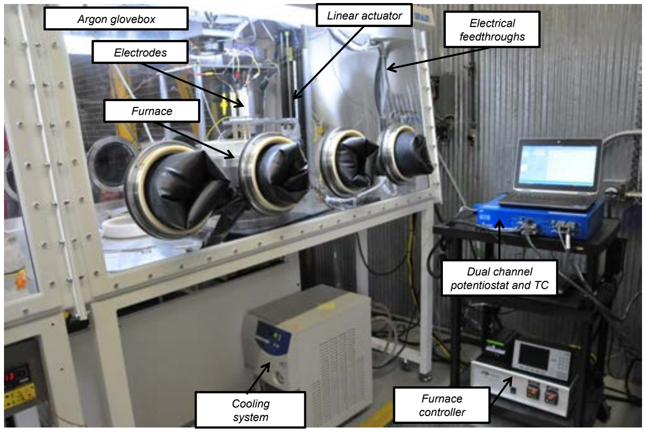
- Electrochemical changes to molten salt composition
- Can add or remove ions electrochemically to simulate ER operations

Actinide Sensor (INL)

Gd - β″-alumina electrode before (left) and after 215 hours immersion in molten LiCl-KCl-$GdCl_3$ salt at 500 °C (right)

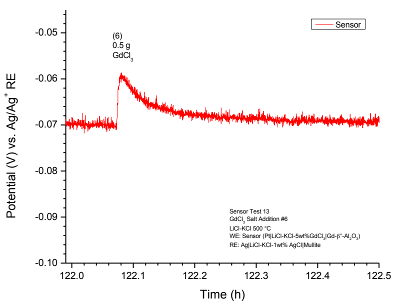


Results of sensor testing, open circuit voltage vs. time response for $GdCl_3$ addition (0.25 wt%)

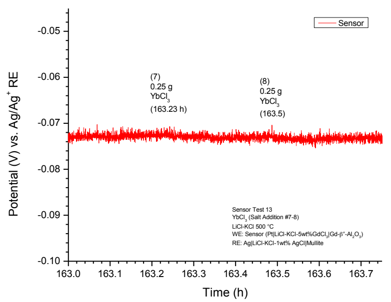


Recent publications

- M. Simpson, et al., TMS 142nd Annual Meeting (2013)
- N. Gese, et al., Inst. Nucl. Mater. Manage. (2012)



Sensor signal response for 0.5 g $GdCl_3$ addition.



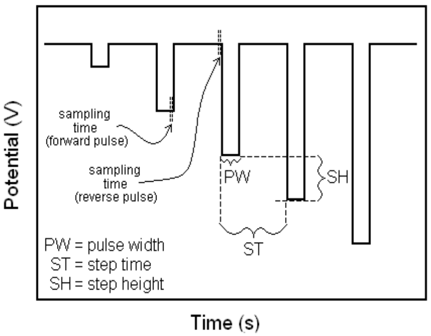
Sensor signal remained unresponsive during $YbCl_3$ additions.

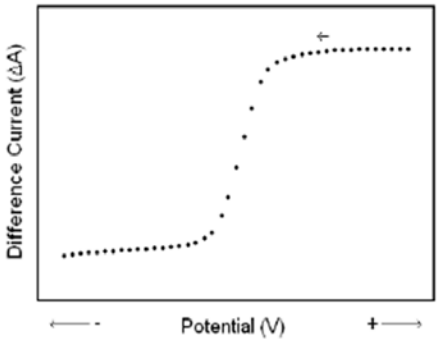
Out-Year Focus: Component validation in a relevant environment (JFCS) 2016 - 2017

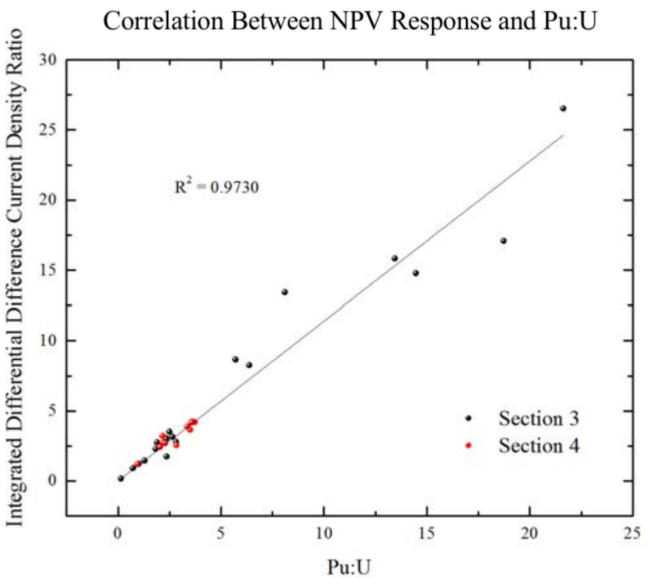
Voltametry (INL)

New Project

- A normal pulse voltammogram is a plot of the difference between the *forward* current and the subsequent *reverse* current (the "difference current") versus the potential of the forward pulse.
- A complete return to a base potential prior to each pulse is the reason for the non-destructive nature of NPV.
- Small amount of species are reduced at each *forward* pulse, but these are immediately returned (oxidize) to solution when the pulse is *reversed*.
- With each pulse, larger and larger numbers of ions are reduced near the electrode until, eventually, steady state is reached where the number of ions reduced on each pulse is diffusion limited.



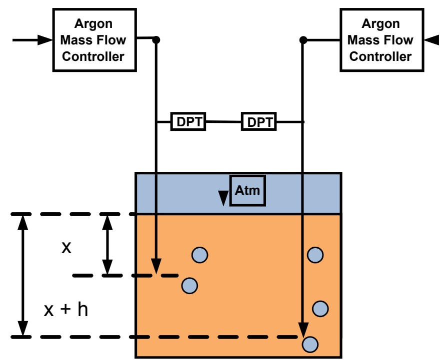




Out-Year Focus: Prototype demonstration in a relevant environment (JFCS) 2017

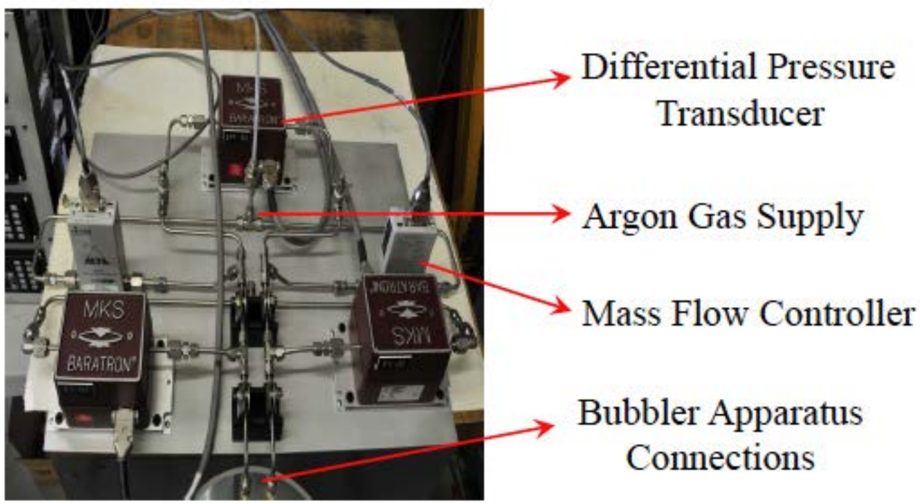
Level/Density Sensor (INL)

- Bubblers have a long history of use in aqueous systems
- Project goal is to develop multiple bubbler system for level and density measurement, in a molten salt environment



Double bubbler principle

- ***Static Pressure at Depth***
  - $P = \rho g h$
- ***Laplace Pressure***
  - $P_{int} = P_{ext} + \frac{2\gamma}{r}$
- ***Static Pressure w/ Laplace***
  - $P = \rho g h + \frac{2\gamma}{r}$



- ***Density***
  - $\rho = \frac{P_1 - P_2}{g \Delta h} + 2\gamma[\frac{1}{r_1} - \frac{1}{r_2}]$
  - *When $r_1 = r_2$*
  - $\rho = \frac{P_1 - P_2}{g \Delta h}$
- ***Level***
  - $X_{ave} = \frac{P_1 + P_2}{2\rho g} + \frac{\Delta h}{2} - \frac{\gamma}{\rho g}\left[\frac{1}{r_1} + \frac{1}{r_2}\right]$
  - *To find X_{ave}, must know surface tension*

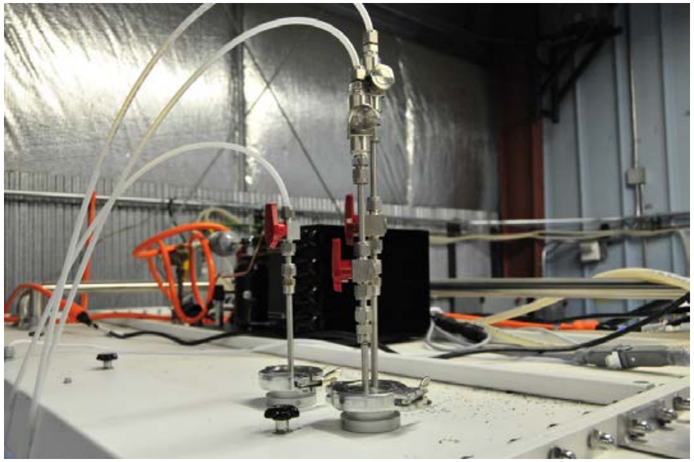
Level/Density Sensor (INL)

Molten Salt Results

- Density: good accuracy~ indications of less than 0.5% error.
- Level: good accuracy~indications of less than 0.5% using *buoyancy surface tension correction*.
- Null hypothesis is accepted, use of Reverse flow prevention is not statistically different from without.
- May need to install shield and use spacing to prevent bubble to bubble interference.
- Triple bubbler?

Furnace

Glovebox Penetration



Out-Year Focus: Prototype demonstration in a relevant environment (JFCS) 2015 – 2016

Microfluidic Sampling (ANL)

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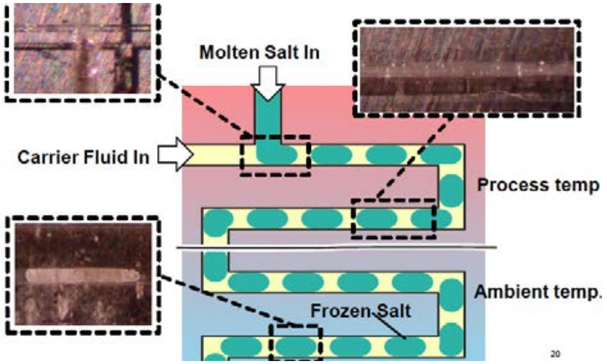
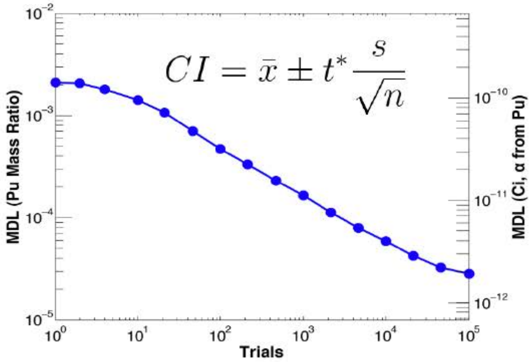
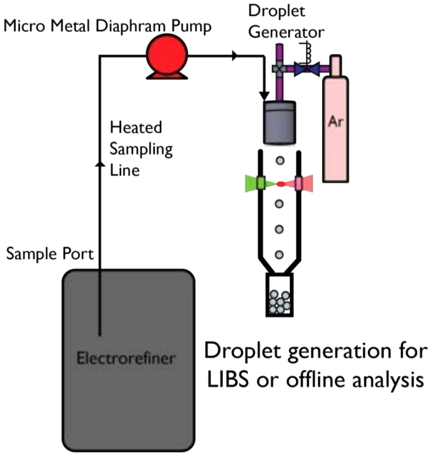
- **1. Replace manual sampling protocol w/ automated sampling**
  - Exact metering
  - Integration with automated analysis



- **2. Facilitate analysis of large numbers of samples**
  - High Throughput Micro-Sampling
  - Achieved through droplet generation
  - Analyze each droplet
    - *1000's of trials with one mL salt*
  - Reduce confidence interval
  - Lower limit of detection





Out-Year Focus: Deployable microfluidic sampling device for electrochemical 2016

MPACT-Related NEUP Instrumentation Projects

- **Current active projects (instrumentation)**
  - Mandal (SC) – B-doped amorphous Se neutron detector
  - McDeavitt (TAMU) – Gamma-blind metastable neutron detector
  - Phongikaroon (VCU) – LIBS for pyroprocessing
  - Ullom (CO) – Microwave readout for very large sensor arrays
  - Nino (FL) – BI_3 gamma spectrometer
  - Simpson (U Utah) – Advanced Voltametry Development

Summary and Future Plans

- The MPACT Campaign has a significant effort in the area of advanced instrumentation
- General approach is to develop technologies with field testing as a goal (as opportunities arise)
- Signature development activities are ongoing that may feed new instrumentation projects in Echem and dry cask monitoring
- NEUP projects have potential to feed instrumentation needs in the future – challenge is lack of continuity