Fuel Cycle Research and Development

Advanced Sensors and Instrumentation R&D – MPACT Campaign

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Outline

- Introduction
- Materials Protection, Accounting, and Control Technologies Campaign Overview
- Selected Research Highlights
- Summary
In addition to improving instrument performance though, an advanced systems approach is needed to fully utilize all available information as part of an advanced safeguards and security system.
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 and support advanced integration capabilities
- Perform technical assessments in support of advanced fuel cycle concepts and approaches
- Develop guidelines for safeguards and security by design and apply to new facility concepts
Establish Safeguards and Security by Design as a standard paradigm for nuclear energy systems

Enabled by:

- Demonstrate and implement next generation nuclear materials management technologies and approaches, including advanced integration methods
  - Echem, H-Canyon, bilateral engagements, new fuel cycle facilities and demos …
- Address safeguards and security issues associated with technology development in other Campaigns
- Support NRC rulemaking through engagement and data generation
- International engagement to help influence and support the nuclear energy enterprise and demonstrate U.S. leadership
Research Thrusts for MPACT

Nuclear Energy

- **Safeguards and Security by Design – Echem**
  - Integrated safeguards and security for electrochemical process
  - Systems approach (safeguards and security performance model, fundamental mass flow models, signature development)
  - Technology development (actinide sensor, level/density sensor, microfluidic sampler, voltammetry)

- **Exploratory Research/Field Tests**
  - Advanced instrumentation development and field tests for next generation nuclear materials management
  - Microcalorimetry, high-dose neutron detector, in situ Pu probe for metal product, MIP monitor

- **Advanced Integration**
  - Methods to quantitatively integrate disparate data sets and associated field demonstrations
  - Pattern recognition and statistical inference, correlation analysis, modeling and simulation

Sensor and instrumentation development efforts range from advancing state-of-the-art for traditional nuclear material accountancy to novel applications such as process monitoring.
Development of Actinide Sensor for Application in Molten Salt - INL

- Potentiometric sensor in high temperature molten salt for on-line measurements
- Preparation of actinide ion conducting materials is the critical path
- Experimental results with surrogate sensors (Gd) have demonstrated sensitivity and stability in molten LiCl-KCl-GdCl₃ salt

The sensor is stable, provides a clean signal and responds to change in GdCl₃ concentration
Selectivity of gadolinium surrogate sensor was tested in multicomponent molten salt.

Uranium sensor development is under way.


**EDS elemental analysis of U-ion exchanged ceramic**

<table>
<thead>
<tr>
<th>Element</th>
<th>Weight%</th>
<th>Atomic%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Na K</td>
<td>-0.57</td>
<td>-1.17</td>
</tr>
<tr>
<td>Al K</td>
<td>29.59</td>
<td>51.27</td>
</tr>
<tr>
<td>Si K</td>
<td>0.16</td>
<td>0.27</td>
</tr>
<tr>
<td>Cl K</td>
<td>24.41</td>
<td>32.19</td>
</tr>
<tr>
<td>K K</td>
<td>8.32</td>
<td>9.95</td>
</tr>
<tr>
<td>U M</td>
<td>38.09</td>
<td>7.48</td>
</tr>
</tbody>
</table>
- 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

Top features of the bubbler system

Bubbler panel ready for hot cell installation
<table>
<thead>
<tr>
<th>Triple Bubbler Calibration Results</th>
<th>Triple Bubbler measurements</th>
<th>Expected</th>
<th>% Difference</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Density (kg/m³)</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>DI Water</td>
<td>997.13 ± 0.29</td>
<td>997.83</td>
<td>-0.1%</td>
</tr>
<tr>
<td>20% CaCl₂</td>
<td>1191.3 ± 0.1</td>
<td>1190.5 ± 0.2</td>
<td>0.1%</td>
</tr>
<tr>
<td>36% CaCl₂</td>
<td>1362.1 ± 0.1</td>
<td>1361.9 ± 0.9</td>
<td>0.0%</td>
</tr>
<tr>
<td><strong>Surface Tension (mN/m)</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>DI Water</td>
<td>72.3 ± 0.6</td>
<td>72.5 ±</td>
<td>-0.3%</td>
</tr>
<tr>
<td>20% CaCl₂</td>
<td>81.7 ± 0.4</td>
<td>81.5 ± 0.6</td>
<td>0.2%</td>
</tr>
<tr>
<td>36% CaCl₂</td>
<td>94.5 ± 0.1</td>
<td>Unknown</td>
<td></td>
</tr>
<tr>
<td><strong>Depth (cm) - EQ (1)</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>DI Water</td>
<td>16.01 ± 0.03</td>
<td>16.04 ± 0.03</td>
<td>-0.2%</td>
</tr>
<tr>
<td>20% CaCl₂</td>
<td>16.02 ± 0.05</td>
<td>16.00 ± 0.00</td>
<td>0.1%</td>
</tr>
<tr>
<td>36% CaCl₂</td>
<td>15.97 ± 0.01</td>
<td>15.99 ± 0.03</td>
<td>-0.1%</td>
</tr>
</tbody>
</table>

- **System has been operated in molten salt system, calibration in molten salt is under way**

Replace manual sampling with automated sampling
- Exact metering
- Integration with automated analysis

Facilitate the 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
- Improve confidence interval
- Lower limit of detection

**Generation 1: Microchip**
- Too delicate for process deployment
- Better suited to process development and analytical applications

**Generation 2: Pneumatic Spotter**
- Widest range of droplet volumes
- Not ideal for continuous operation
- Best for intermittent sampling for off-line analysis

**Generation 3: Flow Cell**
- Better suited for continuous operation
- Best choice for on-line monitoring
Known wt% vs Averaged Peak Height with Confidence Intervals

Despite wide distributions of individual XRF measurements, the averages of large numbers of measurements converge neatly into a calibration curve with a tight confidence interval.
Cyclic Voltammetry is Ideally-suited for In-situ Process Monitoring – ANL

- Voltammetric techniques can be used to monitor actinide concentrations in molten salts
  - Technique does not require use of standards
  - Allows rapid, real-time measurements
  - Equipment not affected by high radiation background
  - Compatible with remote operations
  - Well-developed theory for voltammetric response for given redox reaction
  - Analyze for multiple components with single indicator electrode
  - Multiple voltage perturbation waveforms and methods of analyzing resultant current available

- Concentration determined from peak currents / fit to i-v curve

![Graph](image-url)
Excellent agreement between numerical and experimental results for single species at low concentrations
- Peak current closely matched with previously reported relative errors in measurements of ~1%

Non-ideal behavior arises at concentrations > 1 wt%
- Behavior identified during methodology development
- Reduction in effective diffusion coefficient makes predictions from CVs low?

Non-ideal behavior arises with multi-component salts
- i-v curve does not conform to Berzins-Delahay equation

Predicted concentrations follow the parity line when iR and cylindricity effects are included
Transition edge sensor (TES) technology coupled to superconducting quantum interference device preamplifier (SQUID) yields resolution 10x better than best HPGe detectors currently available.

This translates into potential performance enhancement of greater than 10x.
High-Dose Neutron Detector for High γ Environments - LANL

PDT sealed-cell concept with corrugated boron coated cells

- Each cell individually sealed
- No organic materials inside sealed cell
- High temperature cleaning treatment for high gas purity
- Each cell contains 15 anode wire channels
- Stability equal to 3-He tube system

Detector has been fabricated and new fast preamplifier completed (up to 10MHz)

System has undergone a series of bench top tests – efficiency profile (compared to MCNP), stability

Ready for demonstration in relevant environment – neutron performance with high gamma dose

Boron Particle Size Distribution via Aerodynamic Separation Analysis

Stability measurements for boron-10 plates and He-3 tubes
In Situ Measurement of Pu Concentration in U/TRU Ingot - INL

- U-TRU Product is primarily U-Pu with minor actinides (Am and Np) and rare earths (Nd, Ce, La, Pr)
- U-Pu phase Diagram established by multiple researchers
- Liquidus curve represents the melting point of the alloy on solidification
- Determine melting point of U-Pu Alloy → Determine Pu concentration

- B. Westphal and S. Li, “Experimental Investigations in the U-Rich Region of the U-Pu Phase Diagram,” submitted to NuMat 2016
**In Situ Measurement of Pu Concentration in U/TRU Ingot – current research activities**

- **Goal:** design, install, and calibrate instrumentation to determine Pu concentration in U/TRU products (~100 g)

- **Experimental**
  - Establish internal/external thermocouple configuration at 100g U/TRU scale
  - $\text{Y}_2\text{O}_3$ crucible (20 cc)
  - 8g Al 6061 alloy (~1 wt. % Mg), heat of fusion (750 calories) similar to 100 g U-Pu

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**Graph:** 
![Al 6061 alloy cooling curve](image)
- Gamma-ray based instrument where subtle changes in spectrum (not peak areas) are correlated to process/sample conditions with principal components analysis

- Field test in real operating facility brings practical knowledge and lessons learned

Field Test: Tank A (red), Tank B (green) and mixture of Tank A and B (black) in PCA space
MPACT campaign continues to make progress in advancing technologies and analysis tools to support advanced safeguards and security systems.

Advanced sensors and instrumentation span a range from advancing the current state-of-the-art in traditional nuclear material accountancy to novel applications such as process monitoring.

Facilities in the DOE complex provide unique opportunities for test and evaluation.