U.S. DEPARTMENT OF ENERGY | Nuclear Energy

Fuel Cycle Research and Development

Advanced Sensors and Instrumentation R&D – MPACT Campaign

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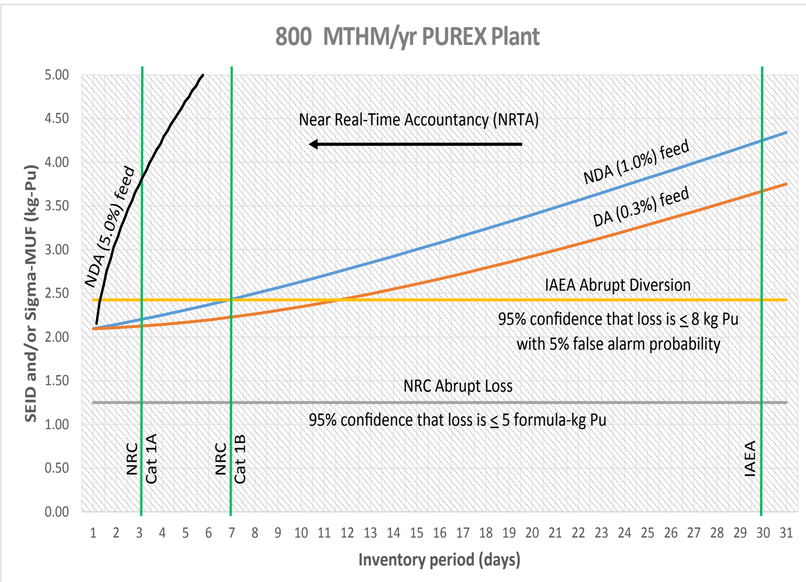
Advanced Sensors and Instrumentation Webinar

October 12, 2016

Outline

- Introduction
- Materials Protection, Accounting, and Control Technologies Campaign Overview
- Selected Research Highlights
- Summary

Large Throughput, Bulk Processing Facilities Drive Need for Improved Instrumentation



100 MT/yr EChem Facility

IAEA Regulation (8kg Pu in 30 Days)	Detection Probability Input: 1% ER: 1% Outputs: 1%	Input: 3% ER: 1% Outputs: 3%	Input: 1% ER: 3% Outputs: 1%	Input: 5% ER: 1% Outputs: 5%	Input: 3% ER: 3% Outputs: 3%
Scenario 1					
Scenario 2					
Scenario 3					
Scenario 4					

NRC Regulation (2kg Pu in 3 Days)	Detection Probability Input: 0.5% ER: 0.5% Outputs: 0.5%	Input: 1% ER: 0.5% Outputs: 1%	Input: 1% ER: 1% Outputs: 1%
Scenario A			
Scenario B			
Scenario C			

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*

Technology Development (first two objectives)
Applications (third objective)
Leadership (fourth objective)

Long Term Objectives (10 – 20 years)

- **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 safeguads 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

- **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

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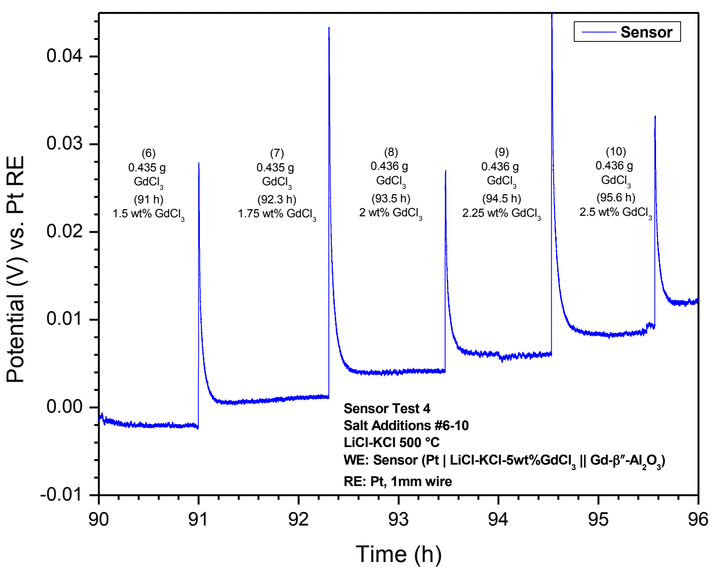
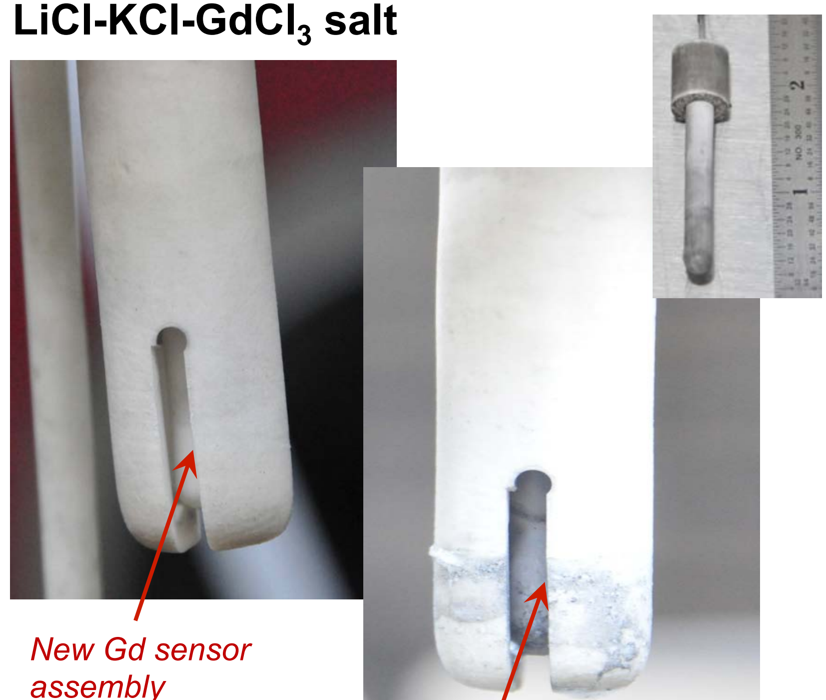
- **Potentiometric sensor in high temperature molten salt for on-line measurements**
- **Preparation of actinide ion conducting materials is the critical path**
- **Experimental results with surrogate sensors (Gd) have demonstrated sensitivity and stability in molten LiCl-KCl-$GdCl_3$ salt**

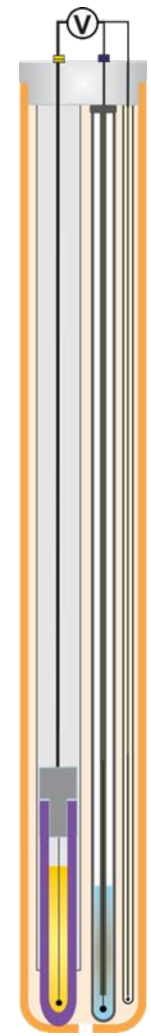


The sensor is stable, provides a clean signal and responds to change in GdCl3 concentration



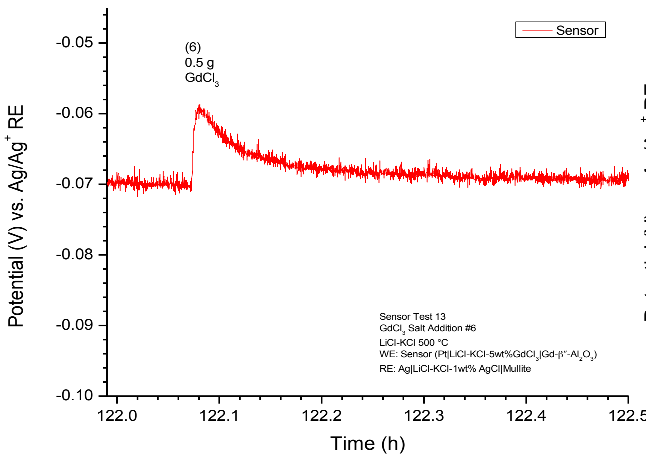
New Gd sensor assembly

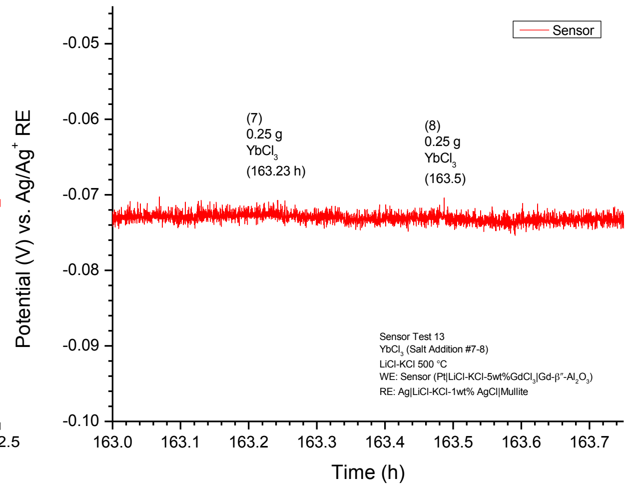
The Gd sensor assembly after exposure to LiCl-KCl based molten salt at 500 °C for 464.25 hours



Development of Actinide Sensor for Application in Molten Salt

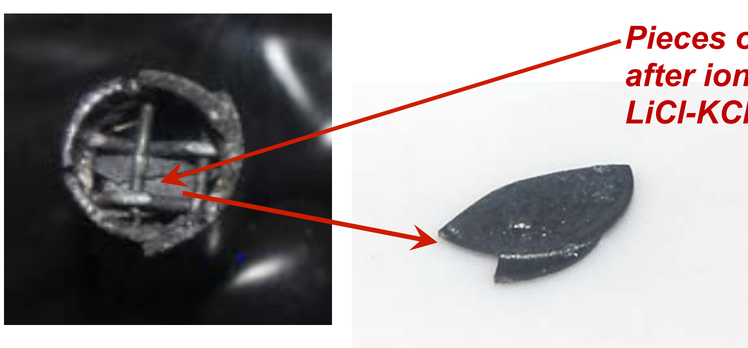
- **Selectivity of gadolinium surrogate sensor was tested in multicomponent molten salt**





- *J. Jue and S. Li, "Actinide ion sensor for pyroprocess monitoring," US Patent 8,741,119 B1, 2014*
- *N.J. Gese, et al., "Potentiometric Sensor for Real-Time Remote Surveillance of Actinides in Molten Salts," Proc. of the 53rd Annual Meeting of INMM, Orlando, FL, 2012*

- **Uranium sensor development is under way**



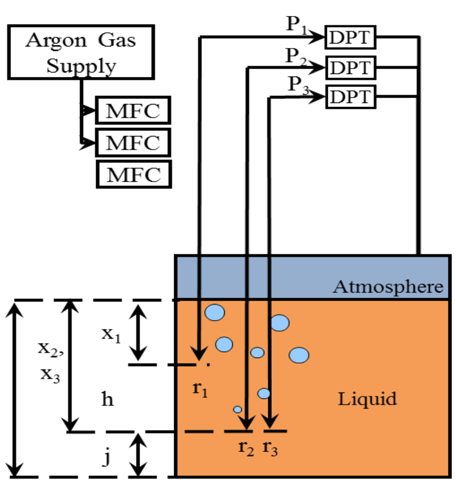
Pieces of ceramic disc after ion exchange with LiCl-KCl-UCl$_3$

Element	Weight%	Atomic%
Na K	-0.57	-1.17
Al K	29.59	51.27
Si K	0.16	0.27
Cl K	24.41	32.19
K K	8.32	9.95
U M	38.09	7.48

EDS elemental analysis of U-ion exchanged ceramic

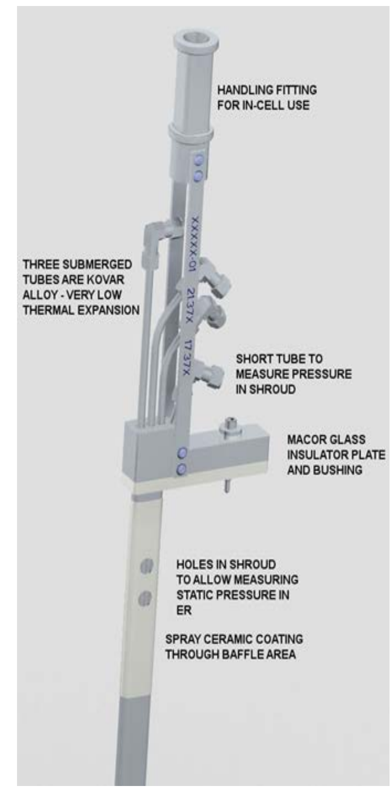
Level/Density Sensor (Triple Bubbler) - INL

- 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



Bubbler panel ready for hot cell installation

Top features of the bubbler system



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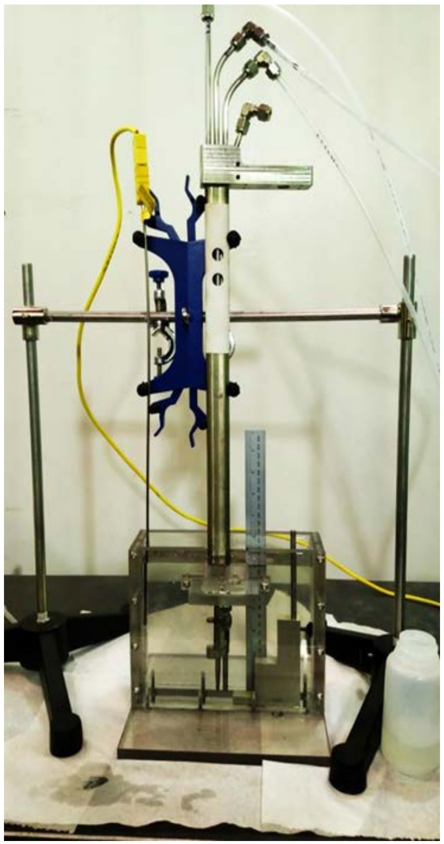
Triple Bubbler Calibration



Calibration Apparatus
5cm x 20cm x 18 cm

Triple Bubbler Calibration Results

	Triple Bubbler measurements	Expected	% Difference
Density (kg/m^3)			
• DI Water	997.13 ± 0.29	997.83	-0.1%
• 20% $CaCl_2$	1191.3 ± 0.1	1190.5 ± 0.2	0.1%
• 36% $CaCl_2$	1362.1 ± 0.1	1361.9 ± 0.9	0.0%
Surface Tension (mN/m)			
• DI Water	72.3 ± 0.6	72.5 ±	-0.3%
• 20% $CaCl_2$	81.7 ± 0.4	81.5 ± 0.6	0.2%
• 36% $CaCl_2$	94.5 ± 0.1	Unknown	
Depth (cm) - EQ (1)			
• DI Water	16.01 ± 0.03	16.04 ± 0.03	-0.2%
• 20% $CaCl_2$	16.02 ± 0.05	16.00 ± 0.00	0.1%
• 36% $CaCl_2$	15.97 ± 0.01	15.99 ± 0.03	-0.1%

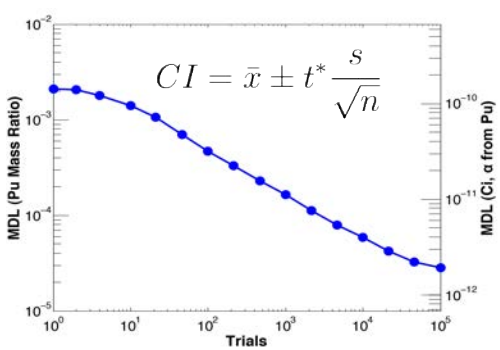
- **System has been operated in molten salt system, calibration in molten salt is under way**

G. Galbreth et al., "The Application of a Triple Bubbler System for Accurate Mass and Volume Determination," 57th INMM 57th Annual Meeting, Atlanta, GA, 2016

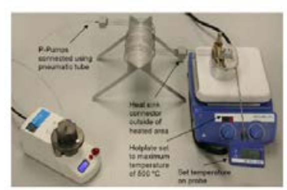
Micro-Analytic Sampling - ANL

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

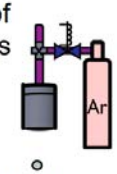
- **Facilitate the analysis of large numbers of samples**
  - 'High throughput micro-sampling"
  - Achieved through droplet generation
  - Analyze each droplet
    - *1000's of trials with one mL salt*
  - Improve confidence interval
  - Lower limit of detection



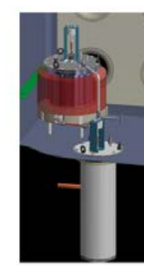
- **Generation 1: Microchip**
  - Too delicate for process deployment
  - Better suited to process development and analytical applications



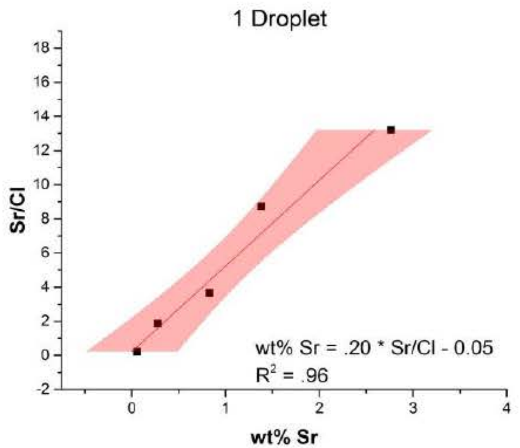
- **Generation 2: Pneumatic Spotter**
  - Widest range of droplet volumes
  - Not ideal for continuous operation
  - Best for intermittent sampling for off-line analysis

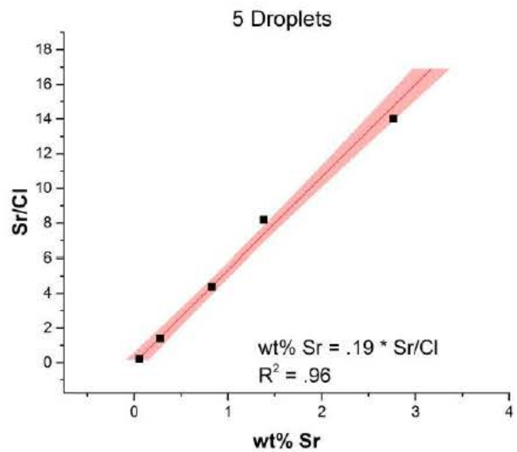


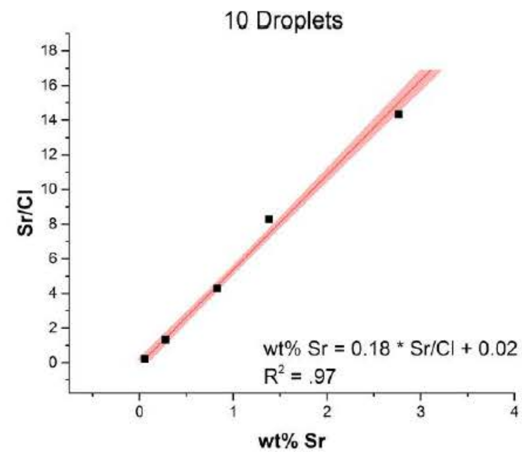
- **Generation 3: Flow Cell**
  - Better suited for continuous operation
  - Best choice for on-line monitoring

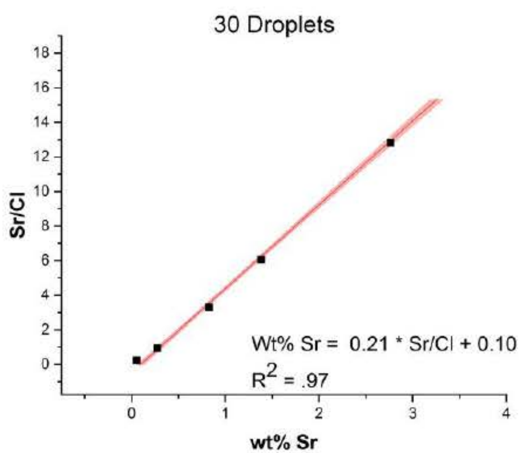


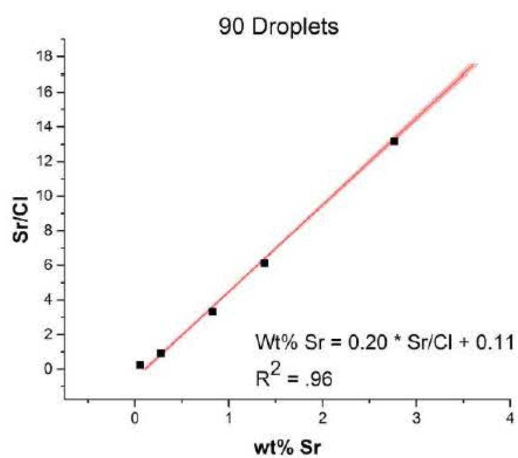
Known wt% vs Averaged Peak Height with Confidence Intervals







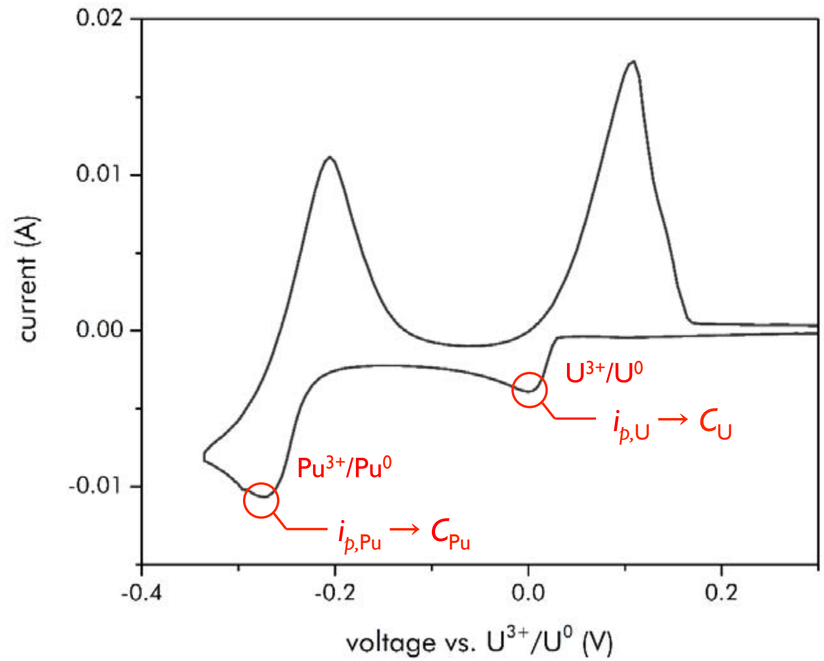




Despite wide distributions of individual XRF measurements, the averages of large numbers of measurements converge neatly into a calibration curve with a tight confidence interval.

Cyclic Voltammetry is Ideally-suited for In-situ Process Monitoring – ANL

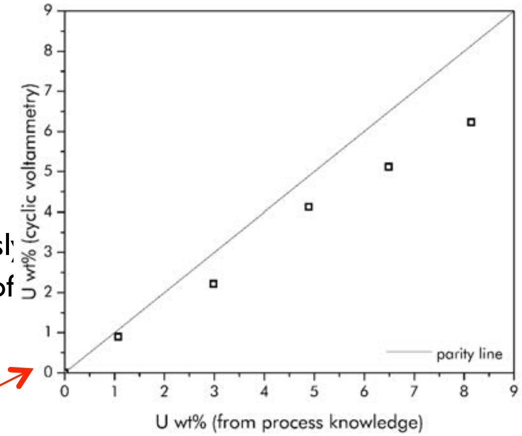
- **Voltammetric techniques can be used to monitor actinide concentrations in molten salts**
  - *Technique does not require use of standards*
  - *Allows rapid, real-time measurements*
  - *Equipment not affected by high radiation background*
  - *Compatible with remote operations*
  - *Well-developed 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*



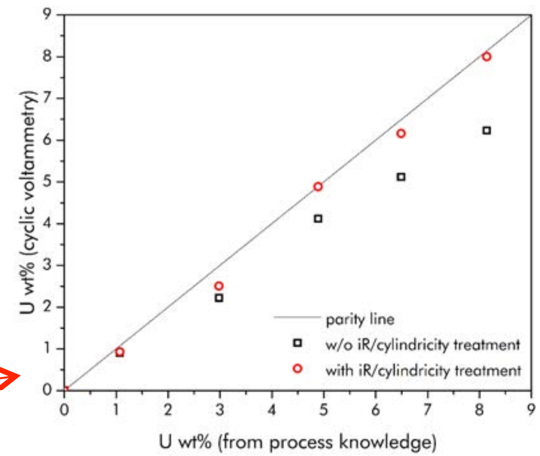
- **Concentration determined from peak currents / fit to *i-v* curve**

Non-Ideal Behavior in Experimental CVs

- **Excellent agreement between numerical and experimental results for single species at <u>low concentrations</u>**
  - 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**



Parity plot between known U wt% and U wt% determined by CV techniques (using Berzins-Delahay equation)



Parity plot comparing predicted uranium concentrations (with and without *iR*/cylindricity treatment) versus known concentrations (from process knowledge)

Microcalorimetry – Super-High Resolution Gamma-Ray Spectrometry - LANL

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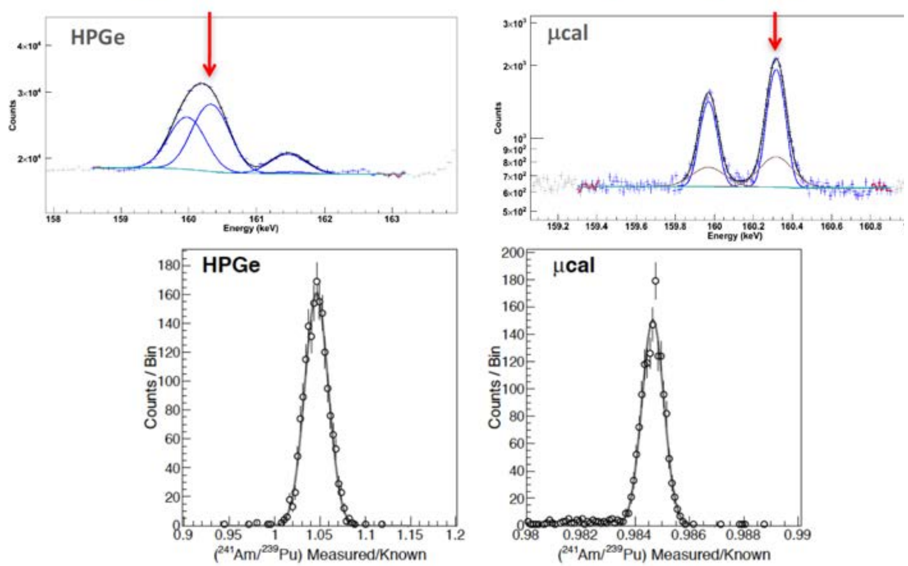
- Transition edge sensor (TES) technology coupled to superconducting quantum interference device preamplifier (SQUID) yields resolution 10x better than best HPGe detectors currently available
- This translates into potential performance enhancement of greater than 10x

Uncertainty in result due to uncertainty in basic nuclear data

HPGe highly sensitive to database or "book values" of gamma-ray energies



$\sigma_{HPGe} = 1 - 5\%$ $\sigma_{\mu Cal} \approx 0.05\%$

Contributions to uncertainty in systematics-dominated limit

Isotope ratio	Branch ratios	Half lives	Center energy	
			HPGE	µCal
^{238}Pu/^{239}Pu	0.82	0.17	0.2-6.9	0.015-0.15
^{240}Pu/^{239}Pu	0.98	0.16	1.0-3.2	0.024-0.051
^{241}Pu/^{239}Pu	0.74	0.13	0.2-1.7	0.013-0.051
^{241}Am/^{239}Pu	1.24	0.19	0.4-5.4	0.041-0.052

Values in %RSD

A striking contrast

µCal has factor of 10-to-60 lower sensitivity to peak center energy

µCal has a strong immunity

High-Dose Neutron Detector for High γ Environments - LANL

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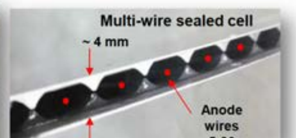
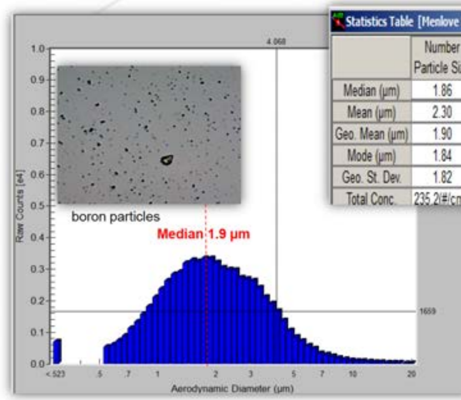
PDT sealed-cell concept with corrugated boron coated cells

- 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 3He tube system

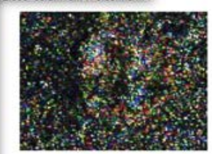


Boron Particle Size Distribution via Aerodynamic Separation Analysis



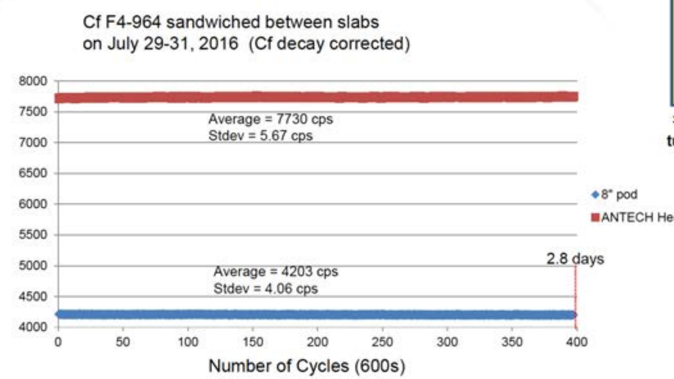
Statistics Table [Menlove H 11-16-2015 Boron B1.A21]

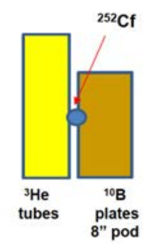
	Number Particle Size	Surface Particle Size	Mass Particle Size
Median (µm)	1.86	4.17	7.12
Mean (µm)	2.30	5.52	8.54
Geo. Mean (µm)	1.90	4.36	6.96
Mode (µm)	1.84	4.07	17.2
Geo. St. Dev.	1.82	1.98	1.95
Total Conc.	235.2(#/cm³)	2.39e+03(um²/cm³)	3.33(mg/m³)



3-D surface layer

Stability measurements for boron-10 plates and He-3 tubes





- 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

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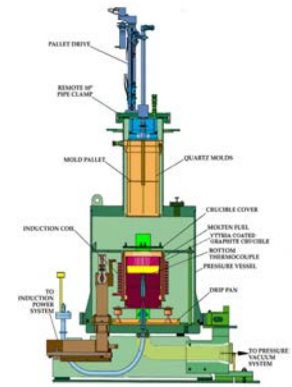
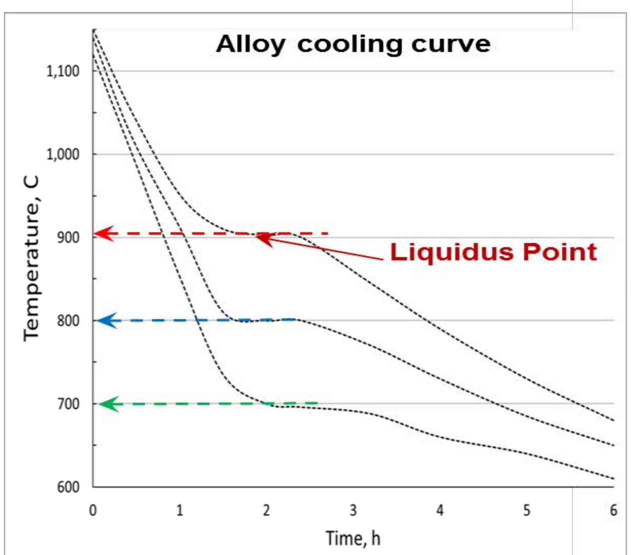
In Situ Measurement of Pu Concentration in U/TRU Ingot - INL

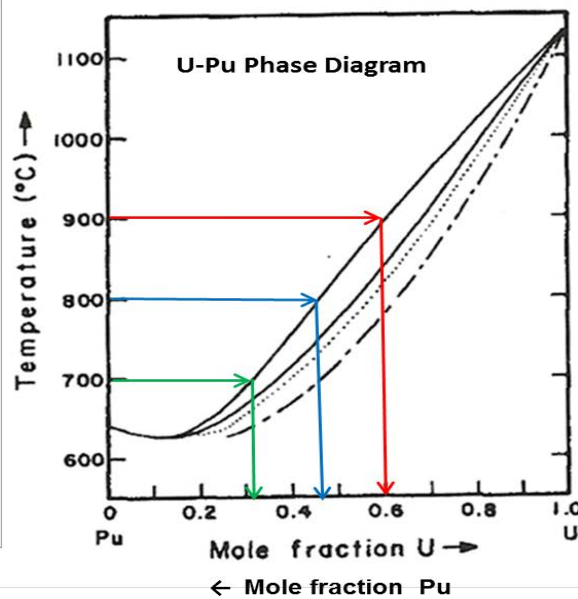
- U-TRU Product is primarily U-Pu with minor actinides (Am and Np) and rare earths (Nd, Ce, La, Pr)
- U-Pu phase Diagram established by multiple researchers
- Liquidus curve represents the melting point of the alloy on solidification
- Determine melting point of U-Pu Alloy → Determine Pu concentration



Engineering-scale casting furnace

U/TRU ingot with over 1kg Pu

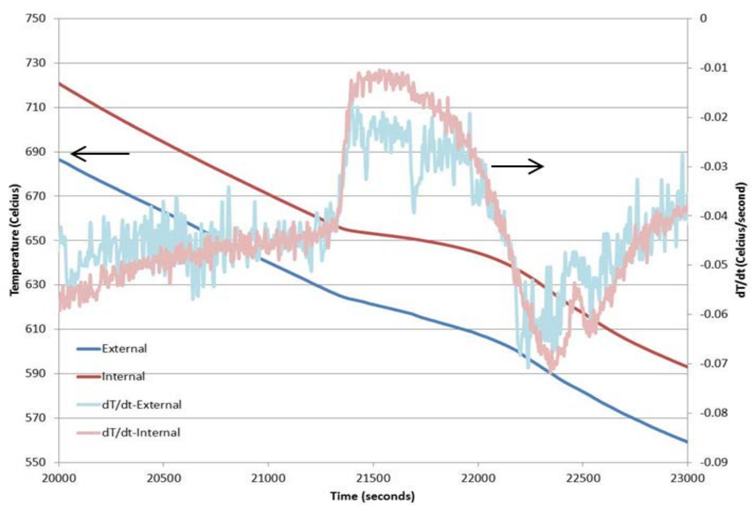




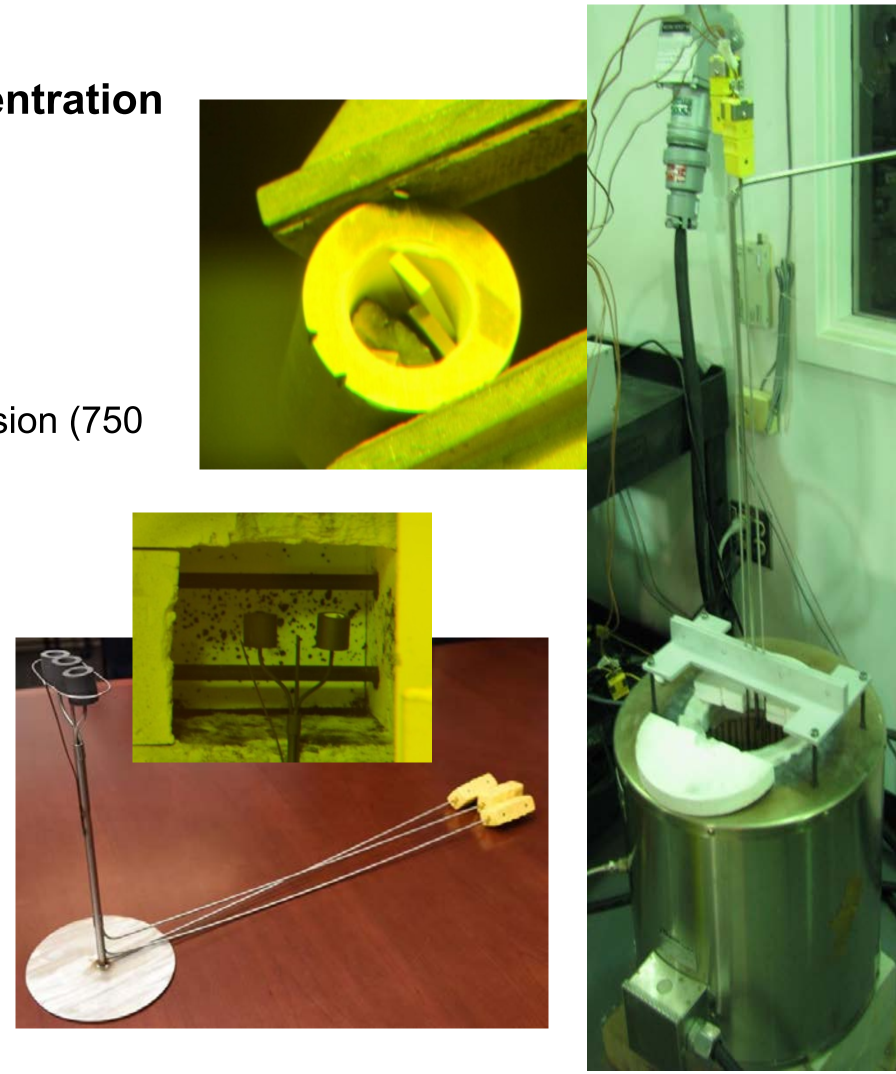
- S. Li, B. Westphal, and S. Herrmann., "Real-Time Monitoring of Plutonium Content in Uranium-Plutonium Alloys." US Patent , 9,121,807 B1, 2015
- B. Westphal and S. Li, "Experimental Investigations in the U-Rich Region of the U-Pu Phase Diagram," submitted to NuMat 2016

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
  - Y_2O_3 crucible (20 cc)
  - 8g Al 6061 alloy (~1 wt. % Mg), heat of fusion (750 calories) similar to 100 g U-Pu

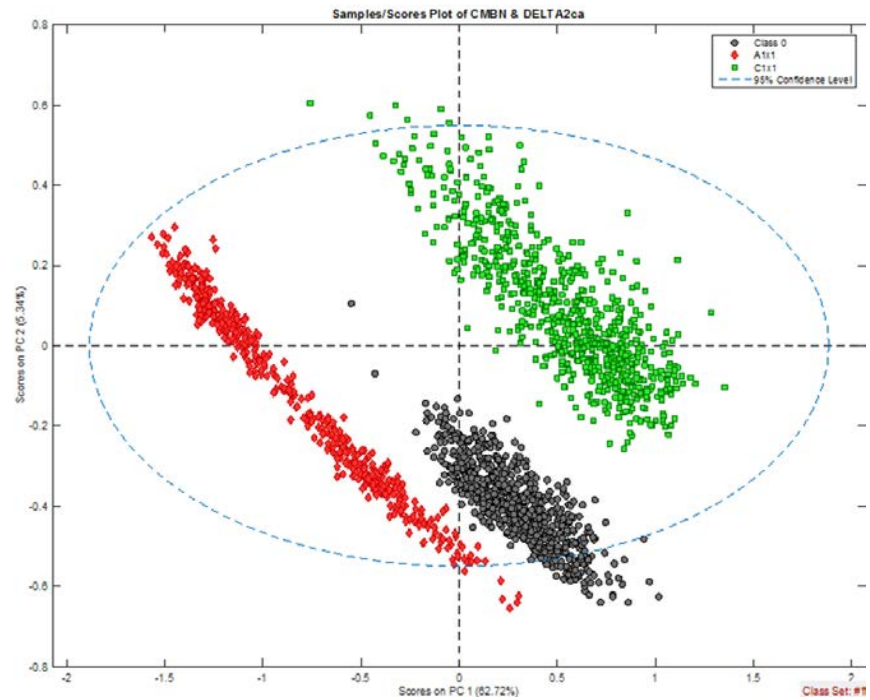


Al 6061 alloy cooling curve



Multi-Isotope Process (MIP) Monitor Field Tests at H-Canyon – PNNL

- 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

Summary

- 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