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Title: Developing a Signature Based Safeguards Approach for the Electrorefiner and Salt Cleanup Unit Operations in Pyroprocessing Facilities

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Developing a Signature Based Safeguards Approach for the Electrorefiner and Salt Cleanup Unit Operations in Pyroprocessing Facilities

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- Collaborators:
  - Philip Lafreniere, University of New Mexico
  - Michael Simpson's team, University of Utah

Outline

- Introduction & Motivation
- Literature Review
- Gaps in SBS Development
- Current and Proposed Work
- Novel Contributions
- Research Proposal Summary & Goals

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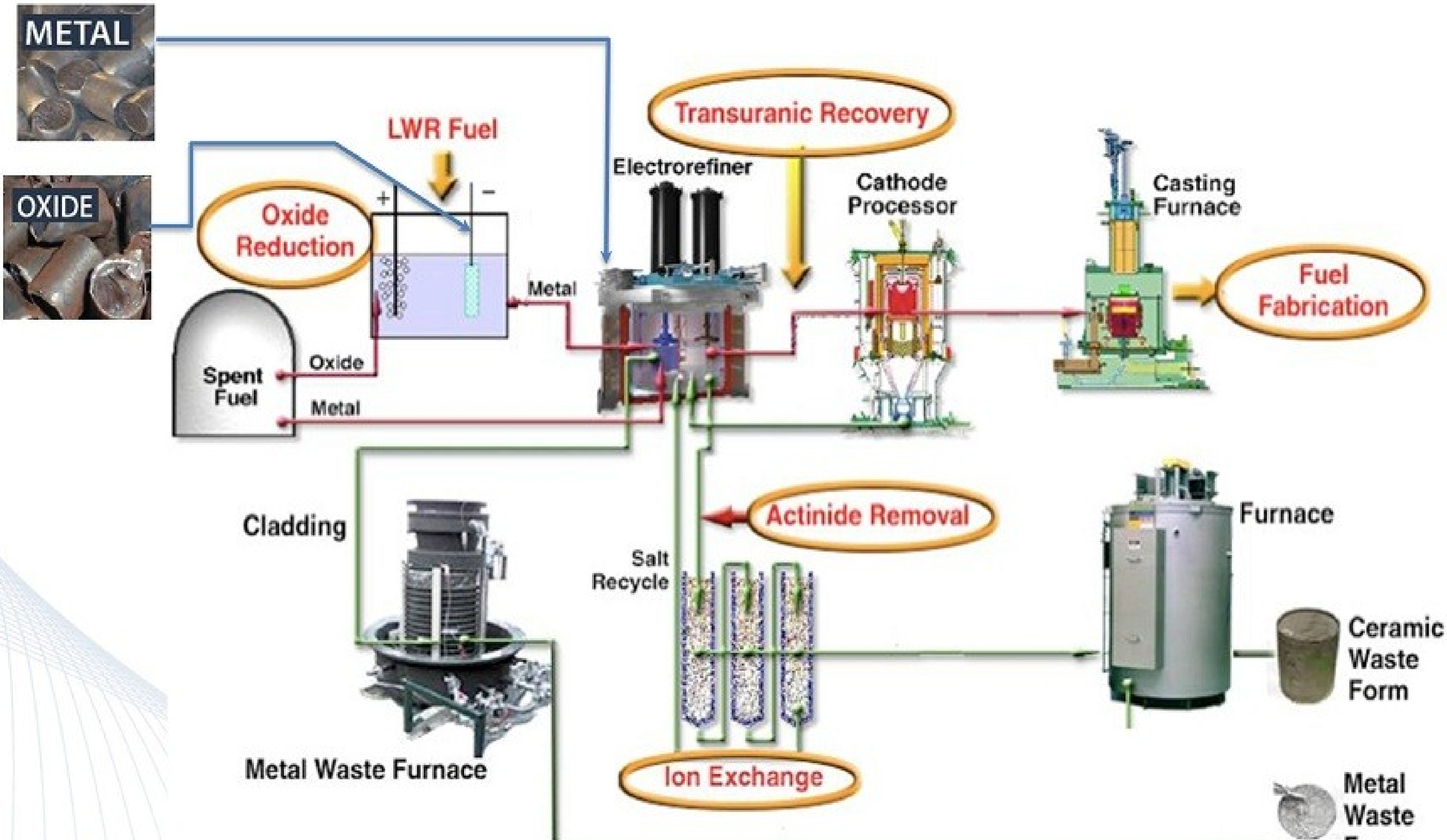
Purpose and Motivation

- SBS goal:
  - Real time/ near real time detection of anomalous events in the pyroprocessing facility as they could indicate loss of special nuclear material
- My research:
  - Continue existing work on using the electrorefiner code ERAD to simulate new off-normal processes, e.g. changes in salt concentration, as this can lead to safeguards significant events
  - Benchmark these runs with proposed experiments, make improvements to code
  - Create a new code based off of existing literature on gas sparging techniques to simulate incomplete salt cleanup operations
  - Couple this code with ERAD to create a two-system dynamic model of the electrorefiner and the salt cleanup process

How Pyroprocessing Works



Challenges with IAEA Safeguards with Evolving Nuclear Technologies

- Nuclear Material Accounting (NMA) material balances (MB) require low uncertainty
  - 95% certainty for the detection of 1 SQ
  - Thus uncertainty of MB assuming normal distribution must be less than 1 SQ/3.3
- In high throughput reprocessing facilities, metric tons of separated material are processed that must be accounted for
  - E.g. Rokkasho Reprocessing Plant- 800Mt U/year, 4 Mt U/day, 8 Mt Pu/yr
- Even with very low uncertainties of accountancy measurements (<.1%) the uncertainty of the MB is still greater than the desired level.
- These challenges have caused issues at facilities such as THORP at Sellafield and Tokai Reprocessing Plant

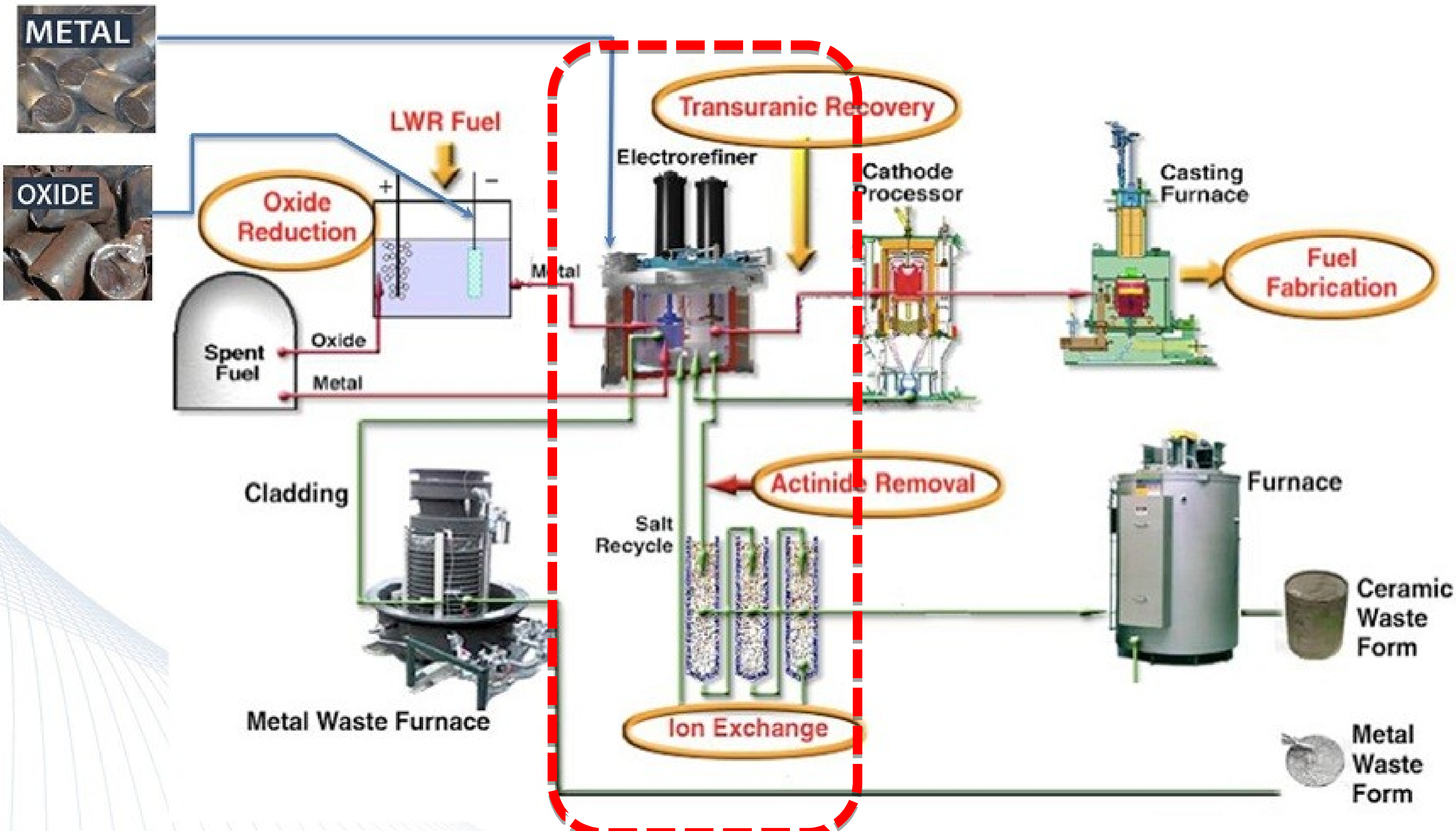
This picture shows the pipe that cracked at THORP. The pipe is 4 cm thick and leads highly active liquid into the IAT

Specific Safeguards Challenges for Pyroprocessing Facilities

Cipiti et al. (2012):

- Traditional NMA does not work well for safeguards when applied to pyroprocessing
  - Lack of input accountability tank (IAT)
  - Plant flushouts not economically feasible
  - Sampling methods do not meet required level of confidence
- Alternate methods such as Signature Based Safeguards (SBS) are being investigated

Research Focus



SBS Research Plan for ER and Salt Cleanup Unit Operations

1. Build process model for salt cleanup technology

2. Select and place sensors to monitor operating conditions of the salt cleanup technology

3. Identify off-normal operational scenarios of the electrorefiner and its effect on the salt cleanup operation that could lead to safeguards significant events

4. Simulate off-normal processes for both systems and generate sensor output

5. Analyze signatures for safeguards significance using sensor output

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Summary of Literature Review

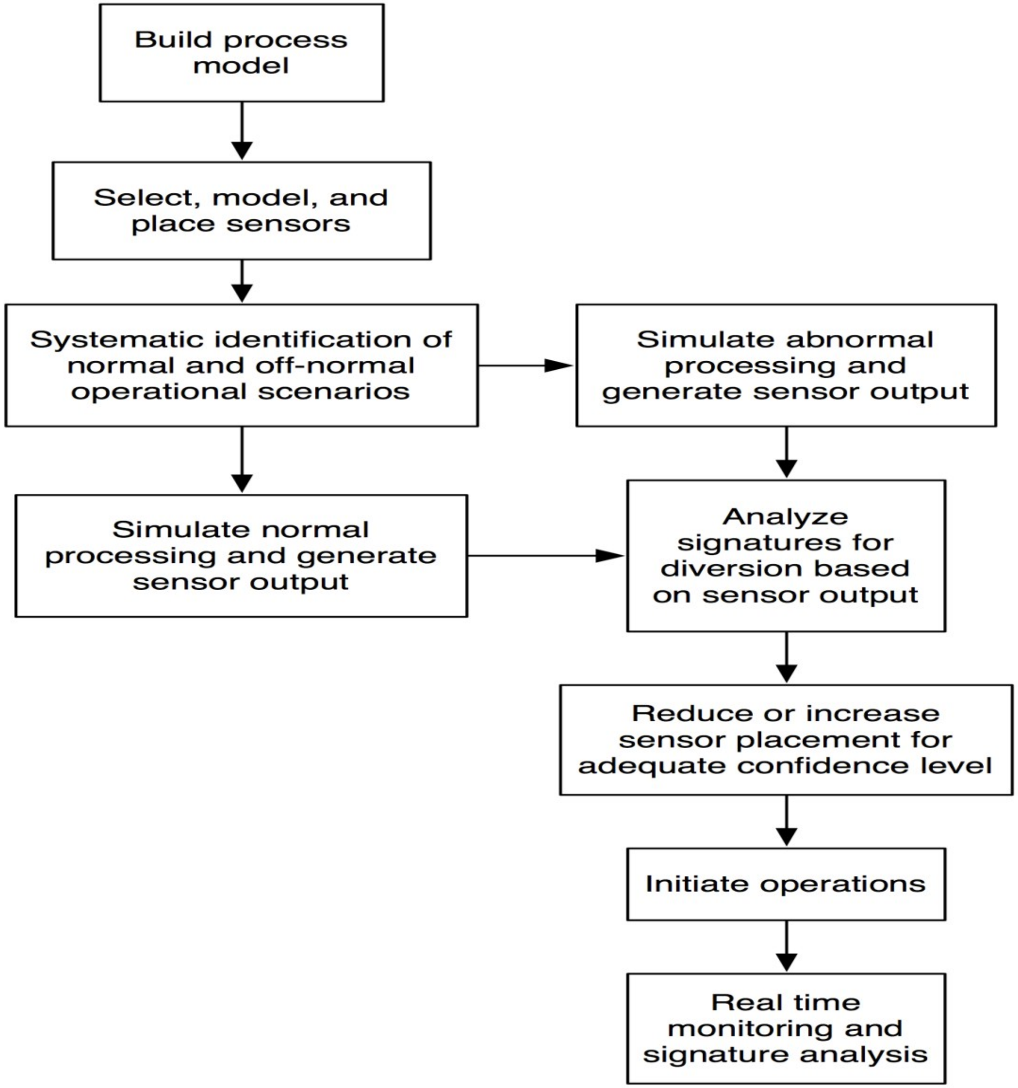
- Signature Based Safeguards
- Electrorefiner
  - Modeling Work
  - SBS Work
- Salt Cleanup
  - Experimental Work
  - Modeling Work

Signature Based Safeguards

[Simpson et al. 2014]

- The goal of SBS is to identify signatures associated with anomalous operations in a facility
- Define complementary process monitoring (PM) approaches to traditional safeguards that use a wide range of sensors capable of taking real-time process control measurements
- Response from sensors helps corroborate NMA measurements and help indicate abnormal operating scenarios
- Relies on physics-based models and predicted signatures for diversion

Signature Based Safeguards (SBS)



Past Work in Electrorefiner Modeling

- Equilibrium model developed by [Johnson, 1988]
  - Improved upon by [Nawada and Bhat, 1998]
  - Reanalyzed by [Ghosh, 2009]
  - Did not capture the kinetic features related to the evolution of elements
    - e.g. partial current.
- Kinetics model developed by [Hoover, 2010]
  - Unreasonable results -> over potential was not properly used.
- Diffusion controlled model developed by [Kobayashi and Tokiwai, 1993]
  - Improved by [Li, 1999]
  - The activity coefficients were assumed to have a value of 1
  - Used the same thickness for the mass transfer boundary
- Improved diffusion control model developed by [Zhang]
  - Predictive model for electrochemical processes, reaction kinetics, mass and ion transport, current and potential distributions, and material flows in the system as functions of operating conditions and time.
- Enhanced REFIN Anodic Dissolution (ERAD) developed by [Cumberland]

Past Work in Electrorefiner SBS

- Involves the identification of failure modes and modeling their detection computationally
  - INL’s ExtendSim
  - SNL’s Echem SSPM
  - Problems:
    - Accurately reflect transport during normal operation but do not allow for the possibility of failure modes upsetting the material balance

Past Work in Electrorefiner SBS (2)

[Hoover et al. 2014]

- Potential electrorefiner failure modes were identified
  - Focused on failure modes that reduced limiting current density
    - Poorly characterized feedstock (concentration gradient)
    - Change in electrode rotation speed (diffusion layer thickness)
    - Decrease of electrode submersion depth (electrode surface area)

[Lafreniere et al 2014]

- Computational ER code ERAD used to simulate off normal conditions in ER.
  - Performed studies varying the diffusion layer thickness, cathode surface area, initial concentration of salt, and operating current
  - Resulting product from ER analyzed using the HLNCC model
- In all studies performed, once limiting current exceeded, resulted in co-deposition of plutonium

[Lafreniere et al. 2015]

- Detector response (measured in MCNP) was proportional to the amount of Pu codeposited
  - Singles followed linear relationship and Doubles followed exponential as expected

Past Work in Electrorefiner SBS (3)

[Lafreniere 2014]

Failure Mode Identified	Safeguard Significance	Detectability
ER Failure Modes		
Poorly Characterized Anodic Feedstock	L	L
Change in Electrode Submersion Depth	M	M
Change in Electrode Rotational Speed	H	H

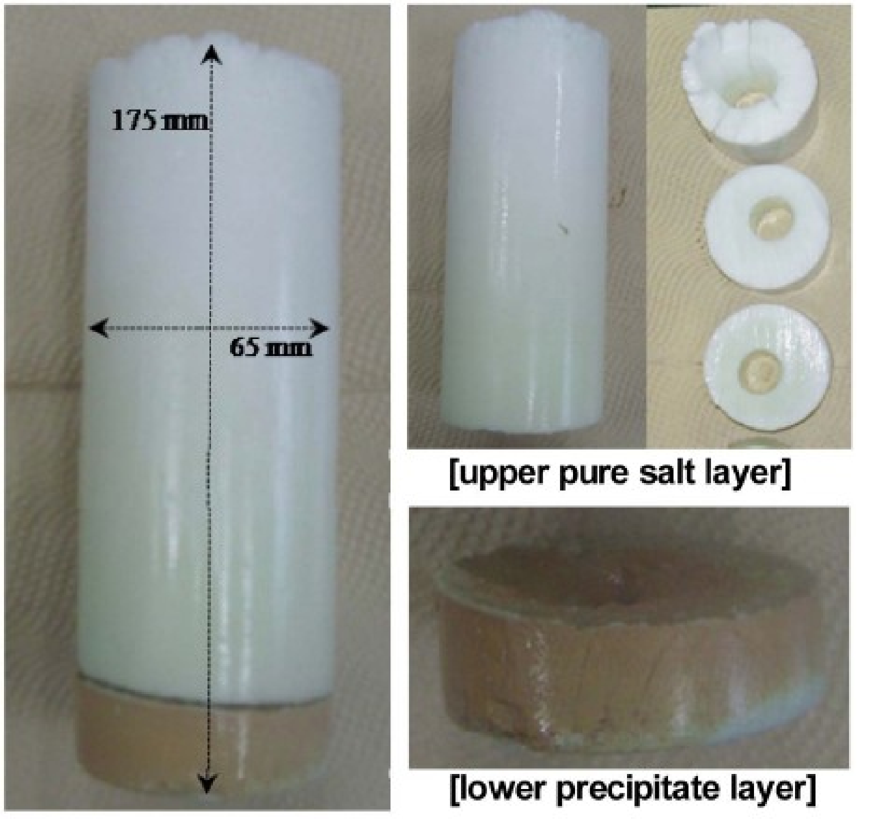
Legend: H-High, M-Medium, L-Low

1. The safeguards significance reflects the failure modes significance in producing a loss of Continuity of Knowledge (COK).

2. The detectability reflects the amount that the results diverge from the normal operating state and the number of sensors that can detect it.

Salt Cleanup: Technology Options

- Ion exchange
- Gas sparging
- Zone freezing
- Electrowinning



Past Work in Salt Cleanup Experiments

- Ion exchange [Pereira et al., Lexa and Johnson, Gouger et al.]
  - Effect of several species ion exchanging in the zeolite in a flow system.
  - Three-component system and an experimental setup with a small sample of zeolite in contact with non-flowing salt.
    - Cesium takes up much more space in the zeolite than other species.
  - Combined the two approaches above in experimental setups involving fission products in the salt as well as uranium and transuranics.
- Gas sparging [Cho et al. 2012, Phongikaroon et al. 2013]
  - Optimum oxygen sparging rate by using a differential pressure analysis method using a lab-scale apparatus
  - Measurements and analysis of oxygen bubble distributions
  - Bubble sizes and rise velocities increased with an increase in oxygen sparging rate.
  - Bubble equivalent diameters are normally distributed.
- Zone freezing [Williams 2012, Cho et al. 2013]
- Electrowinning [Lee et al. 2013, Song et al. 2010, Yoon et al. 2010]
  - Engineering-scale development in R&D conducting heat transfer analysis

Past Work in Salt Cleanup Modeling

Ion Exchange [Ahluwalia et al. 1998]

- Model that accounted for ion-equilibria and kinetics
  - Relied upon an exchange factor approach
  - intracrystalline diffusion was assumed to be the rate limiting step for determining the kinetics of the ion exchange reaction.

Ion Exchange [Simpson and Gouger 2003 and later Phongikaroon and Simpson 2006]

- Two-site equilibrium model for ion exchange between monovalent and then divalent and trivalent cations and zeolite-A in a molten salt.
  - Closely fits with Lexa and Johnson's experimental data

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Gaps in SBS Development

- Work has not been conducted on the salt cleanup unit operation
- Work is still in early phases for electrorefiner
  - More simulation work of off-normal scenarios to generate sensor output
  - More experimental work remains to get a better understanding of lab-scale behavior of the ER under different operating conditions
- More work remains on analyzing signatures for safeguards concern using statistical methods, NDA, and DA, etc.

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1. Build process model for salt cleanup technology

- This model will focus on the gas sparging technique
- Develop a physics based model
- Use existing experimental data to help validate model

2. Select and place sensors to monitor operating conditions of the salt cleanup technology

- Using the new model, determine operating conditions that would need to be monitored under normal operation
- Select appropriate sensors and detectors

3. Identify off-normal operational scenarios of the electrorefiner and its effect on the salt cleanup operation that could lead to safeguards significant events

- Electrorefiner
  - Salt composition changes leading to Pu depositing on the solid cathode without Cm or entering the salt cleanup process
- Salt Cleanup
  - Events leading to the incomplete purification of actinides from the ER salt resulting in Pu in the ceramic waste products without Cm or actinides recycling back into the ER changing ER salt composition

Identification of Off-Normal Scenarios

[...]over et al. 2014, Murphy et al. (JNMM) 2016]

- Electrorefiner
  - Poorly characterized anodic feedstock
  - Change in electrode submersion depth
  - Change in electrode rotational speed
  - Change in salt concentration
  - Changes in temperature

[Murphy et al. (INMM) 2016]

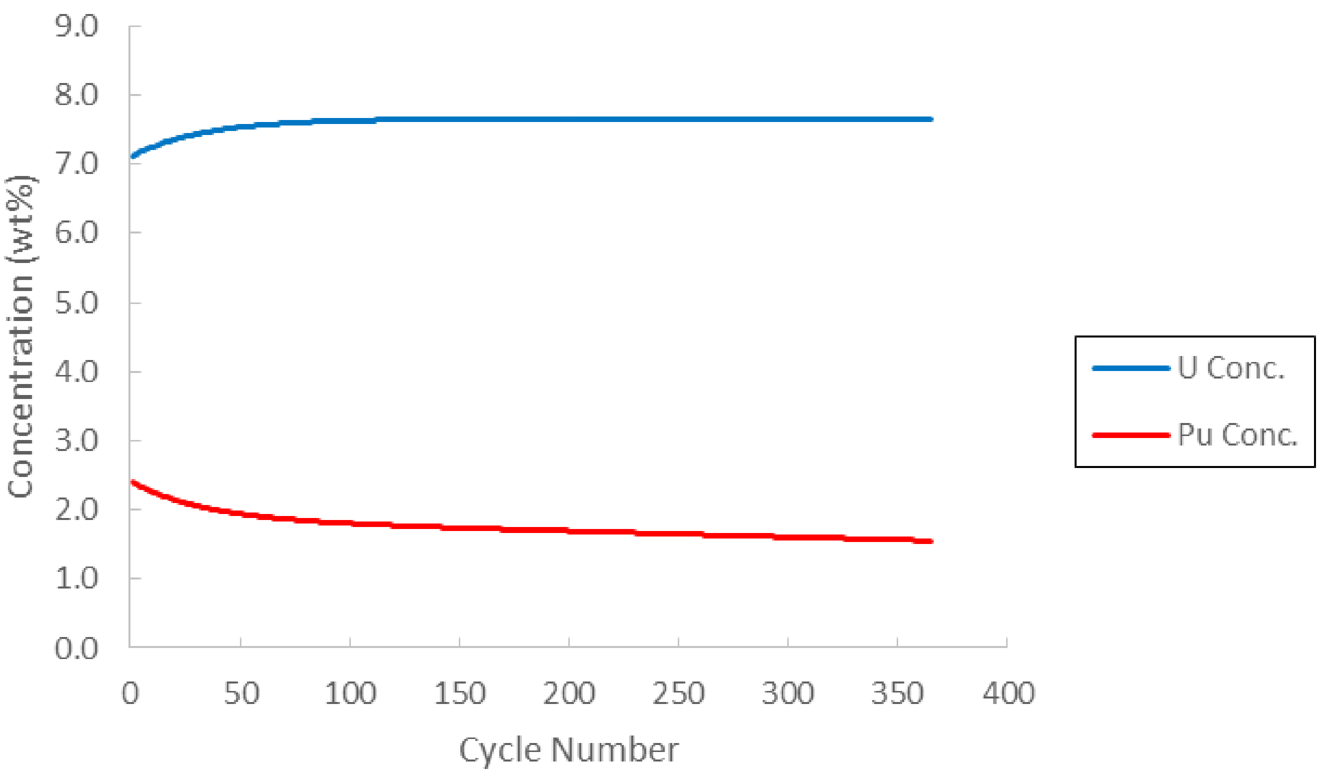
- Salt Cleanup Process
  - Incorrect input salt composition
  - Changes in temperature
  - Changes in pressure
  - Oxygen sensor failure
  - Flow meter failure
  - Vacuum and pressure sensor failures

4. Simulate off-normal processes for both systems and generate sensor output

- Enhanced REFIN with Anodic Dissolution (ERAD) ERAD is a basic 1-D electrochemical transport code
  - Butler-Volmer
    - determines the rate at which a given species will be oxidized or reduced
  - Nernst
    - determines equilibrium potential of a species $E_{eq,j}$ based on concentration
  - Nernst-Plank
    - calculates mass transfer of each species
  - Not benchmarked against any ER design
  - Experiments need to be performed to demonstrate that ERAD results are applicable in their application to SBS development
- New Gas Sparging Model

Previous Long Term Operation Studies

- Electrorefiner is operated for 365 24-hour cycles using a surface area of 90 cm^2
- Long term electrolyte data



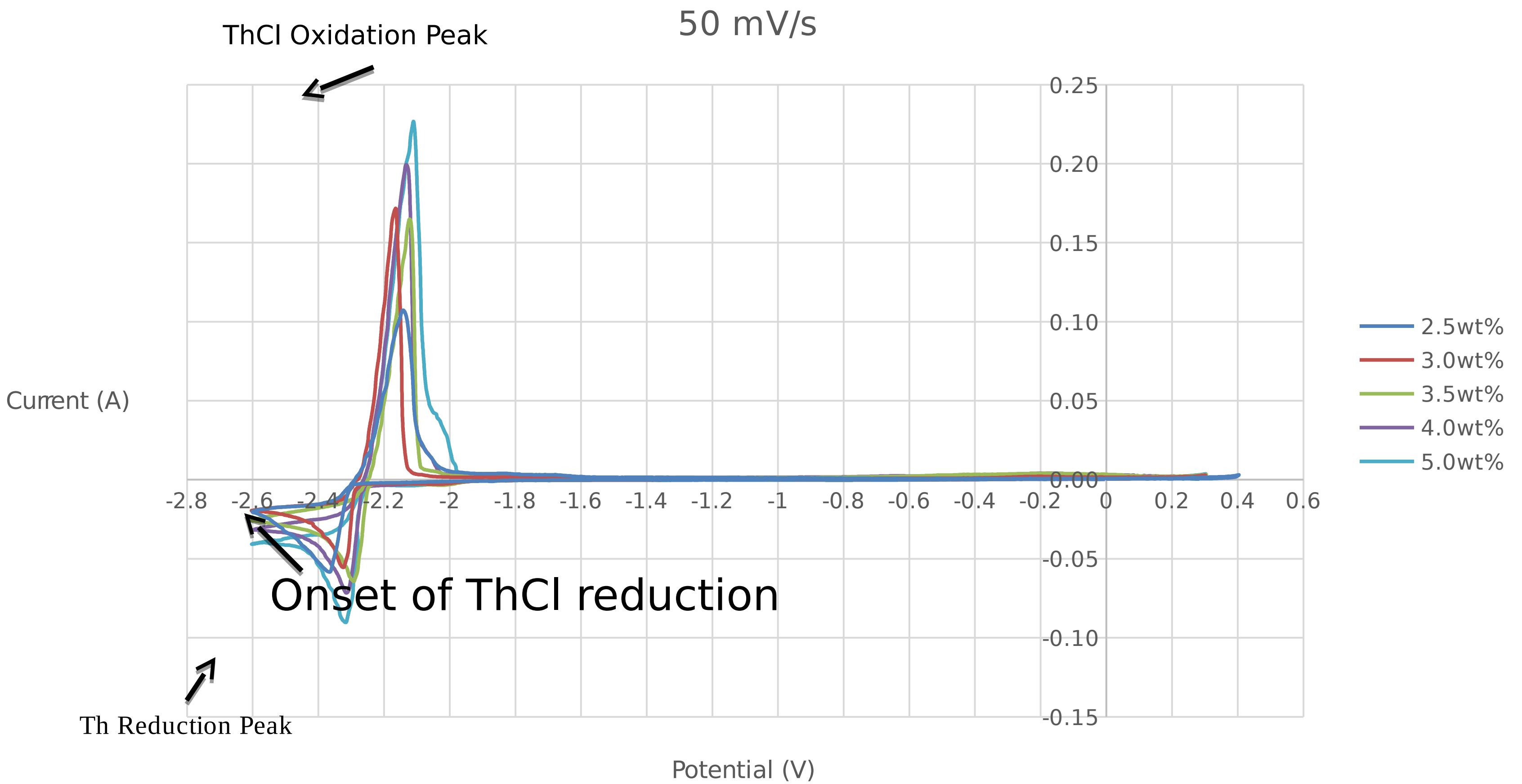
Experimental Work

- Create an experimental data set used for comparison with computational results
  - Performed on a lab scale
  - Attempts to replicate salt concentration changes observed computationally
  - Preliminary work performed in December at University of Utah
- Proposed experimental work
  - Perform multi-component experiments changing the concentration of one element
  - Collect CV and potential data for ERAD comparison

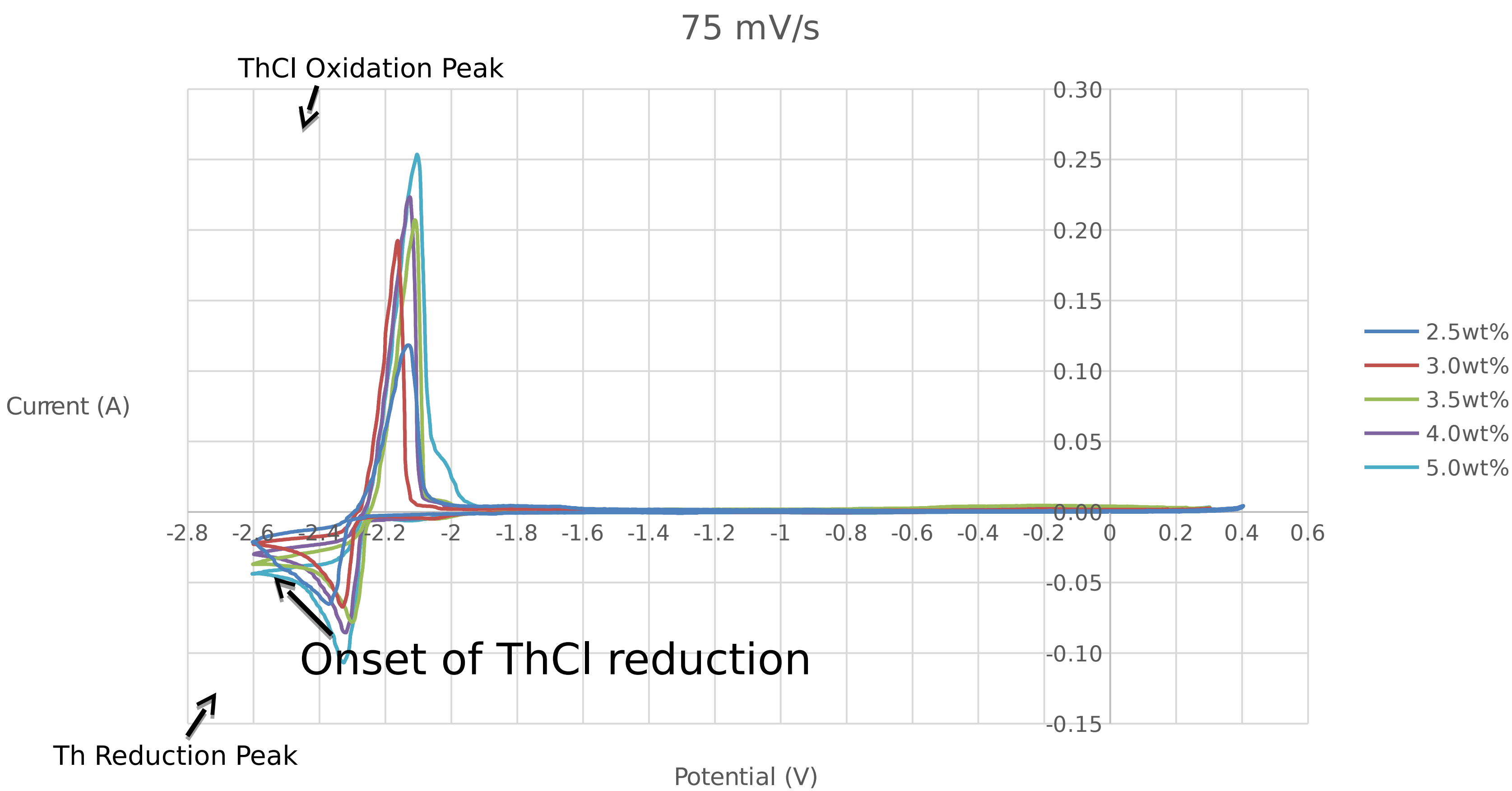
Previous Experimental Work

- University of Utah winter 2015
- Learning experience
- Collect baseline informatio on changing ThCl concentration
  - 2.5, 3.0, 3.5, 4.0, 5.0 wt. %

CV Results i vs E at 50 mV/s



CV Results i vs E at 75 mV/s



5. Analyze signatures for safeguards significance using sensor output

- Will have new data sets from off-normal operations of the ER and the salt cleanup process
- Statistically analyze these data sets to identify which measurements lead to safeguards significant events versus normal operation
  - Uncertainty quantification (UQ) and sensitivity analysis

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Novel Contributions

- Significantly enhance SBS development for salt cleanup process
  - Creating a new gas sparging process model
  - Selecting sensors to monitor normal operation
  - Identifying safeguards significant off-normal scenarios
  - Simulating those off-normal events and generating sensor output
- Further enhance SBS development for the electrorefiner
  - Simulating off-normal events caused by changes in salt concentration and identifying which conditions lead to Pu and Cm not tracking throughout the rest of the system
- New contribution in applying statistical techniques to analyze the signatures gained from these two models to help draw real time conclusions on anomalous events

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Research Plan Summary

- Simulations (winter, spring 2016)
  - Compare results from ThCl experiment
  - Conduct more long term, continuous operation runs with different starting fuel compositions
  - Conduct lab scale simulations used to compare with experimental results
  - Create gas sparging model
- Signature Analysis (winter, spring 2016)
  - Perform statistical analysis of sensor output from models
- Experiments (summer 2016)
  - Concentration variation with multicomponent setup
  - Perform DA and possibly NDA to determine deposits and salt constituents
- Defend dissertation (2017)

Questions?