Proposed Techniques for Improved Cathode Product Sampling Within a Near Real Time Accountancy (NRTA) Framework

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Introduction

- Pyroprocessing face challenges with applying traditional international safeguards
  - Increased reliance on enhanced containment and surveillance
- Alternative methods for supporting safeguards being investigated at UNM
  - Near Real Time Accountancy (NRTA)
  - Process Monitoring (PM)
- Possible lessons in measurements of masses from other metal processing industries
  - E.g. copper or aluminum processing
- Improved process monitoring methods have the potential to *improve* operations and throughput of facility while decreasing offline maintenance and safeguards inspection requirements
- Previous UNM work has focused on ER and CP failure modes and their resultant process monitoring implications
- Research into sampling of metallic ingots for process monitoring is in the very early stages at UNM

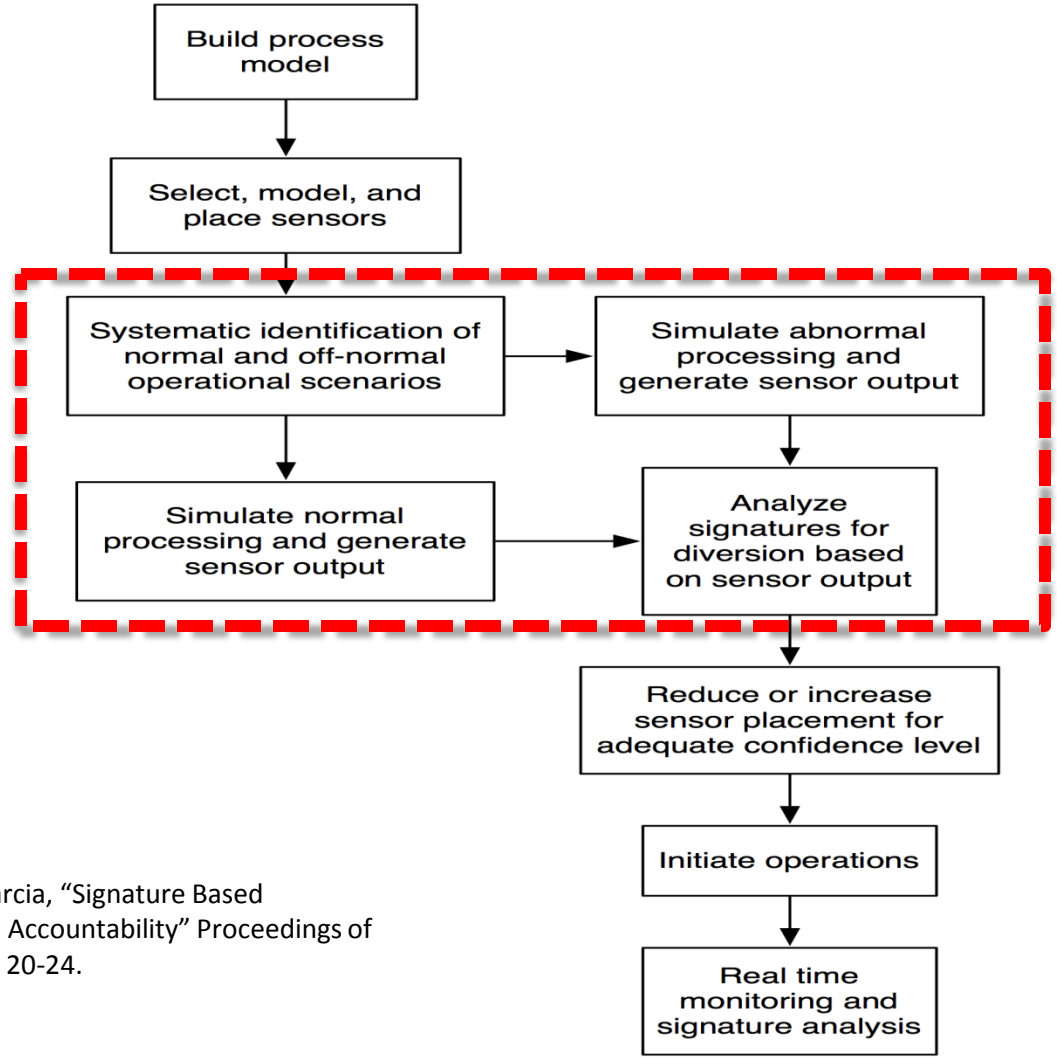
Near Real Time Accountancy Background

- Similar to traditional international safeguards as it utilizes a mass balance
  - "In" – "Out" – "Holdup" = MB
    - MB = 0 means all material accounted for
    - Occurs over a set time interval or balance period
- NRTA is different in that the balance period is shorter and more regular
  - Utilizing statistical means tests such as Page's test demonstrate that NRTA is more effective
  - Time interval must be finely tuned to reduce safeguards uncertainty sufficiently
- Sampling methods occur in real time
  - Improved sampling methods that provide results timely with the balance period a must.

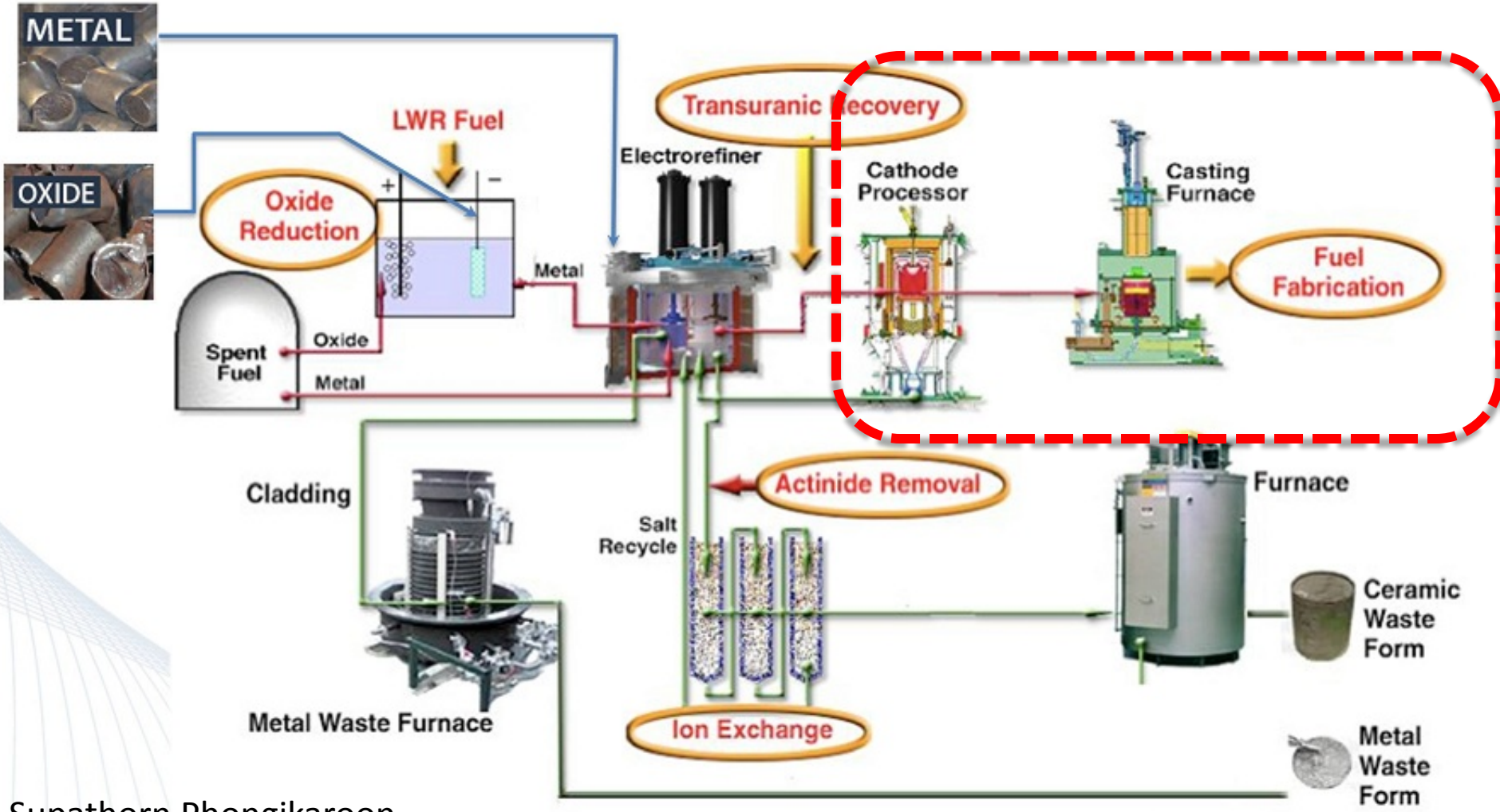
Signature Based Safeguards (SBS)

- SBS is a PM method that uses a system-centric framework.
  - Being developed by UNM and University of Utah
- The goal of SBS is to identify signatures associated with anomalous operations in a facility
  - Intentional (e.g. diversion scenarios) or unintentional (e.g. failure modes from normal operation)
  - The signatures that may have safeguards-significance are the combinations of process measurements

Signature Based Safeguards (SBS)

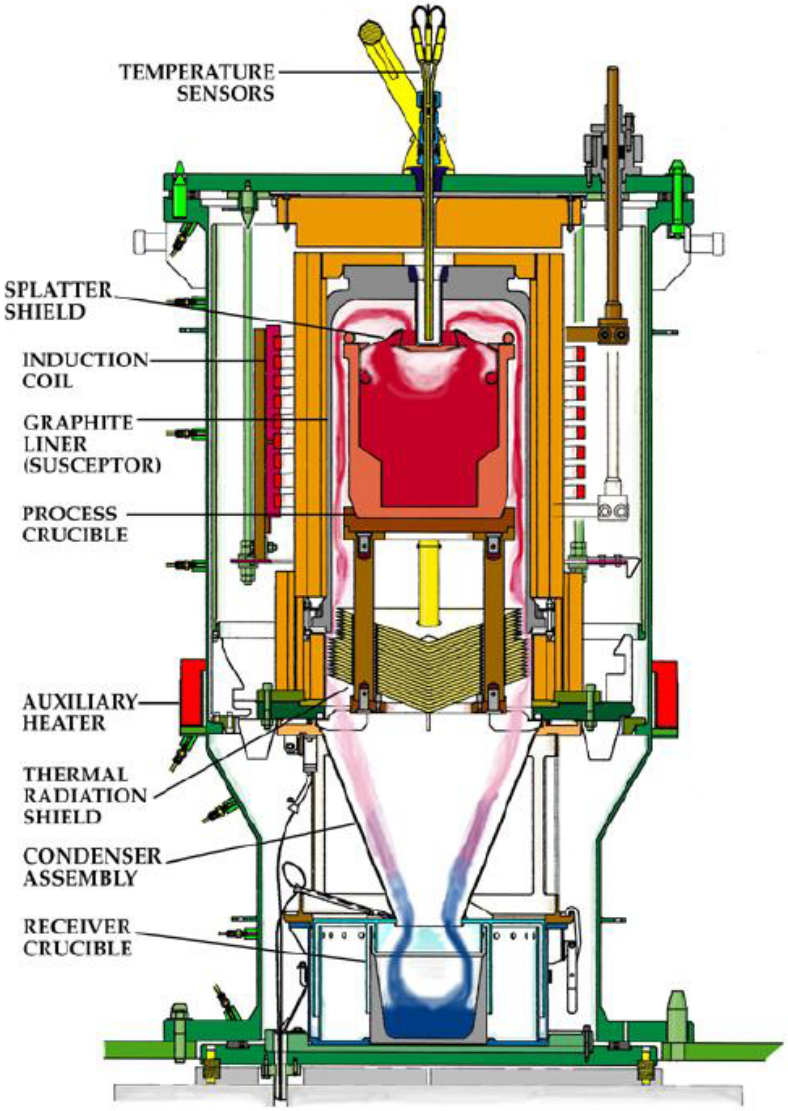


M. Simpson, E. D. Blandford, H. Garcia, "Signature Based Safeguards Alternative to Material Accountability" Proceedings of INMM 2014, Atlanta, GA, USA July 20-24.



Supathorn Phongikaroon, http://www.if.uidaho.edu/~phonst/research.html

Cathode Processing: ANL Design



- Under vacuum and high temperature, salt is distilled from the U dendrite product
  - Occurs in process crucible
  - Salt distilled into receiver crucible
- Metal pre-consolidated for melt into fuel ingot in casting furnace
- Material processed in batch form

Westphal et.al., Proceeding to the DOE Spent Nuclear/Fuels and Fissile Materials Management, Charleston, 2002

Continuous Cathode Processing: KAERI Design

- Distillation of uranium dendrites occurs in distillation column
- Separate consolidation unit for forming ingots
- Uses continuous process to form metal ingots

Jang et al. *J. Radioanal. Nucl. Chem.*, **295** (2013)

Cathode Processing Failure Modes

- Failure modes in commercial of cathode processing equipment can *possibly* result in loss of Continuity of Knowledge (CoK)
- Several failure modes in CP at a commercial scale have been identified
  - Distillation Process
    - Uncertainties in feedstock of dendrites
    - Insufficient temperature for distillation
    - Loss of vacuum
  - Ingot Consolidation
    - Insufficient temperature and pressure for formation of ingots
    - Hold up in feed mechanism for continuous processing

Focus of Research

- UNM research is investigating a comprehensive sampling method for final metallic fuel ingots for integration into an SBS and NRTA compatible framework
  - Sampling methods must be timely, accurate, and representative of the bulk material expected in a commercial facility
    - Sampling methods should not lead to reduced throughput of material
- Derive sampling technologies and corresponding methodologies from past manufacturing quality assurance work on copper and other commercial metallurgical processing industries
  - Instead of detecting impurities for commercial standards, utilizing these approaches for process monitoring applications

Example: Sampling Research for Copper

- Copper is a metal for which electrorefining and electrowinning methods are commonly applied
- Much past work has been put into place to detect impurities to ensure that copper product is up to industrial application standards
- Sampling methods involving punching and drilling investigated
- Bulk of investigation dealt with best way to sample cathodes
  - Various chemical analysis methods
  - Various geometric patterns of drilling and punching

Metallurgical Sampling Methods

- Metallurgical sampling typically involves 3 step:
  1. Random selection of a representative number of cathodes
     - Multiple cathodes/cathode processors expected for commercial throughput… what are acceptable limits across different cathodes/cathode processing units?
  2. Obtain representative sample from collection of cathodes
     - Reducing possible variability between cathodic deposition axial/radial and ingot melt heterogeneities?
  3. Prepare homogeneous portion of samples for chemical analyses
     - What is required to infer the overall composition of the cathodes processed?

- Various methods of sampling metal for the purpose of QA have been developed
  - Drilling
  - Punching
  - Sectioning
  - Sawing
  - Melt sampling
    - Facilitated by advanced ceramics

Chemical Analysis Methods

- Various methods for the determination of impurities in metal
  - Emission spectrometry
  - Atomic absorption spectroscopy
  - ICP-MS
  - Laser Induced Breakdown Spectroscopy (LIBS)

Proposed Method for Sampling

- Sampling can't be applied directly to used nuclear fuel
- For this reason surrogate metals at UNM will be utilized in reduce-scaled experiments ensuring dynamic similitude
  - Use metals with similar thermo-physical and chemical properties to metals in the cathodic deposits and ER salt
    - Generate similar heterogeneity in metal for sampling
  - Utilize furnace to consolidate samples in similar geometric fashion to pyro products
- Perform sampling on melts and on final metallic ingots
- Generate array of metal ingots with similar geometric and mass properties to metal ingots from ER

Next Steps for UNM Sampling Approach

- Look into approaches for using ICP-MS for chemical analysis prior to casting process
  - Expertise on hand at UNM
  - Majority of elements can be resolved
- Possible utilization of X-ray Computed Tomography (CT) scans of surrogate ingot formation for complementary signature assessment

Summary

- Alternative methods for monitoring nuclear material could benefit from a timely, accurate sampling method for pyroprocessing end products
- Experience with well established industry quality assurance methods for electrolytically separated metals may help in the development of new sampling methods that may have safeguards applications
- UNM intends to perform reduced-scale experiments using surrogate materials to determine possible attractive methods of sampling cathodic process products and fuel ingots for process monitoring applications
  - Work in early stages of development but feedback is encouraged

QUESTIONS, COMMENTS, AND SUGGESTIONS?