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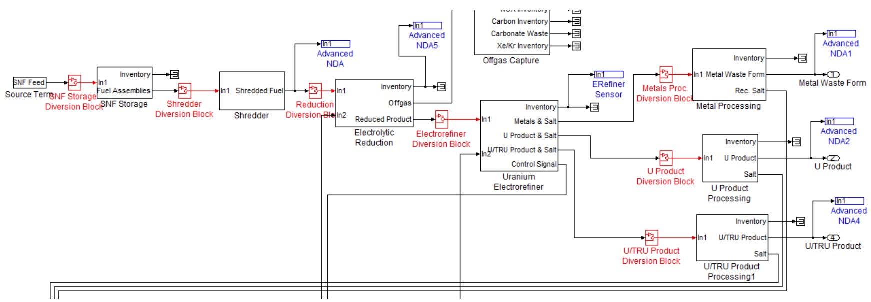

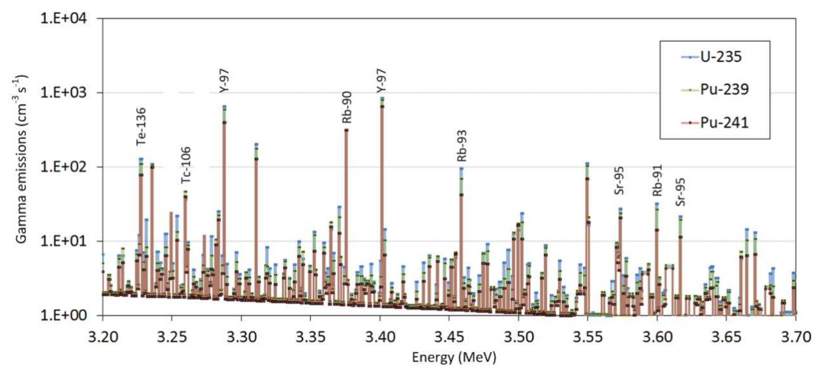

Pyroprocessing Safeguards Challenges

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U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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Motivation

- South Korea is strongly pushing to use pyroprocessing to close their nuclear fuel cycle.
  - Limited available land and public opposition to a repository.
  - Recycling through the use of fast reactors.
- Because a commercial-scale facility has never been built, there are many safeguards questions.
  - The material form (molten salts and metal products) are much different than what the international safeguards community is familiar with as compared to aqueous processing.
  - The unit operations are much different than aqueous processing.

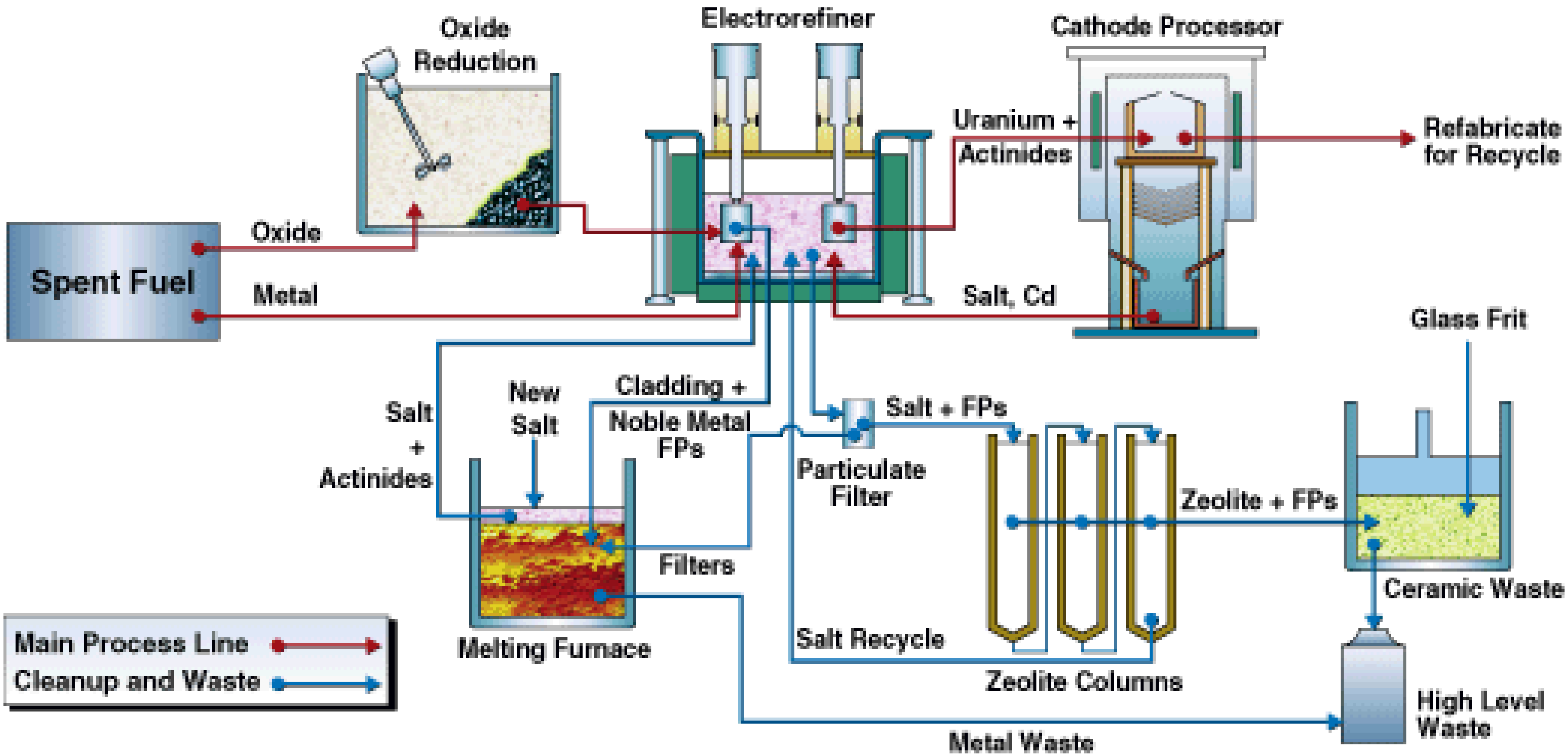
Objective

- The goal of this work is to develop a safeguards system design for electrochemical plants and examine how the system will perform under diversion scenarios.
- Progress:
  - Safeguards challenges have been identified.
  - An accountancy structure for a commercial echem plant has been developed that relies on a ten-day balance period.
  - Measurement technologies for four key measurement points have been evaluated to identify research gaps
  - Diversion scenarios have been evaluated using both NRC and IAEA regulations as a basis

History

- Electrochemical processing has been examined for several decades going back to the Experimental Breeder Reactor II (EBR-II) program at INL
  - EBR-II was a fast reactor that used electrochemical processing to recycle the fuel—melt refining was performed from 1964-69.
  - Current operations are focused on research and treating the old fuel.
- Recently, the Korea Atomic Energy Research Institute (KAERI) has been developing electrochemical technology.
  - The Pyroprocess Integrated Inactive Demonstration Facility (PRIDE) is located in Daejeon, and was designed to demonstrate operations using depleted uranium.

Pyroprocessing Technology



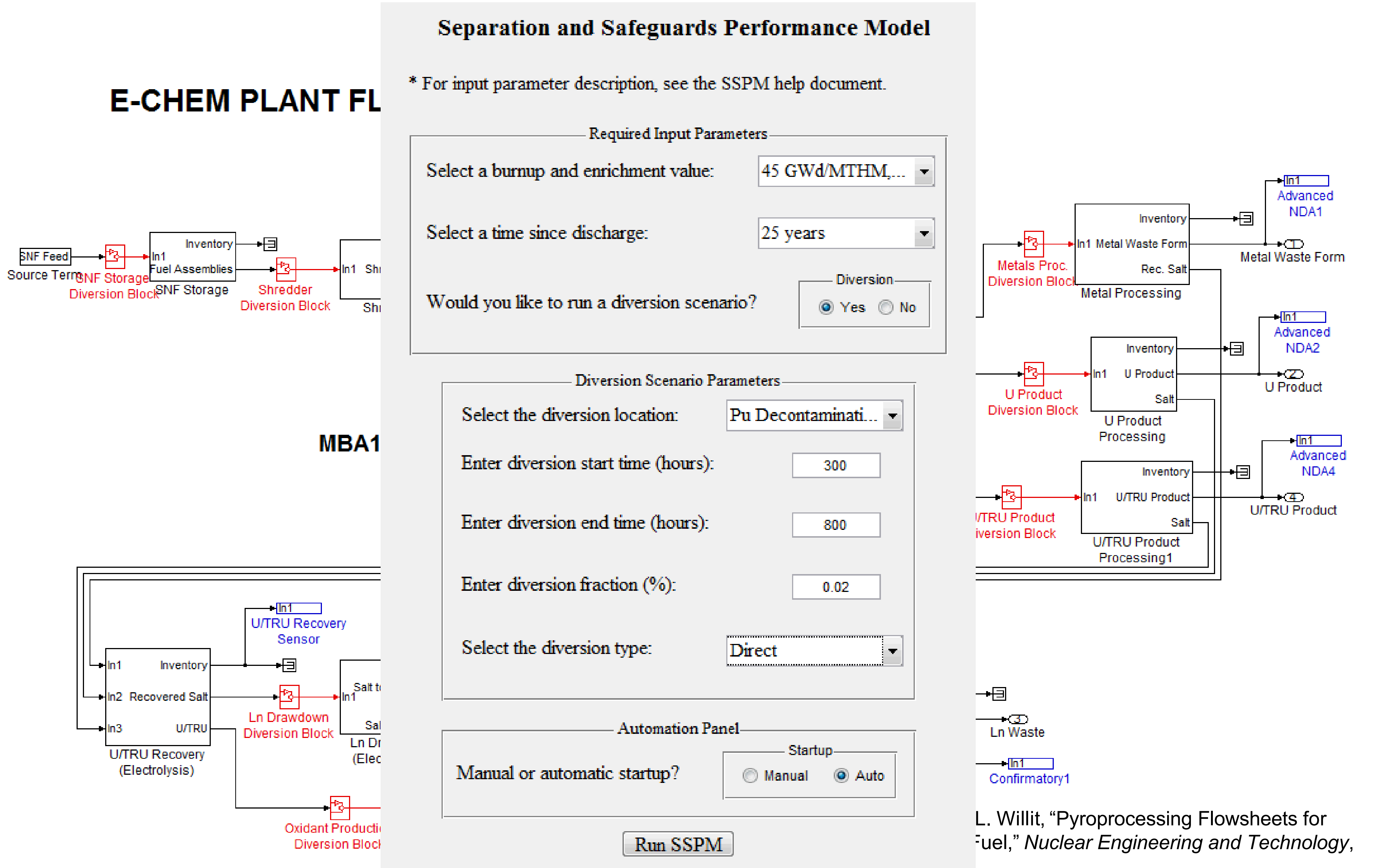
Safeguards at Aqueous Plants

- Materials accountancy is a combination of containment and surveillance on solid materials (fuel assemblies at the front end and oxide products at the back end) and accountancy measurements throughout the plant.
- Accountancy measurements are based mainly on measurements of the material dissolved in nitric acid at the input and output accountability tanks—we can achieve low measurement uncertainties (0.2-0.8%).
- Plant flushouts occur once or twice a year to close out the material balance.
- Periodic interim inventory measurements may be made monthly through sampling of tanks to determine inventory.

Pyroprocessing Safeguards Challenges

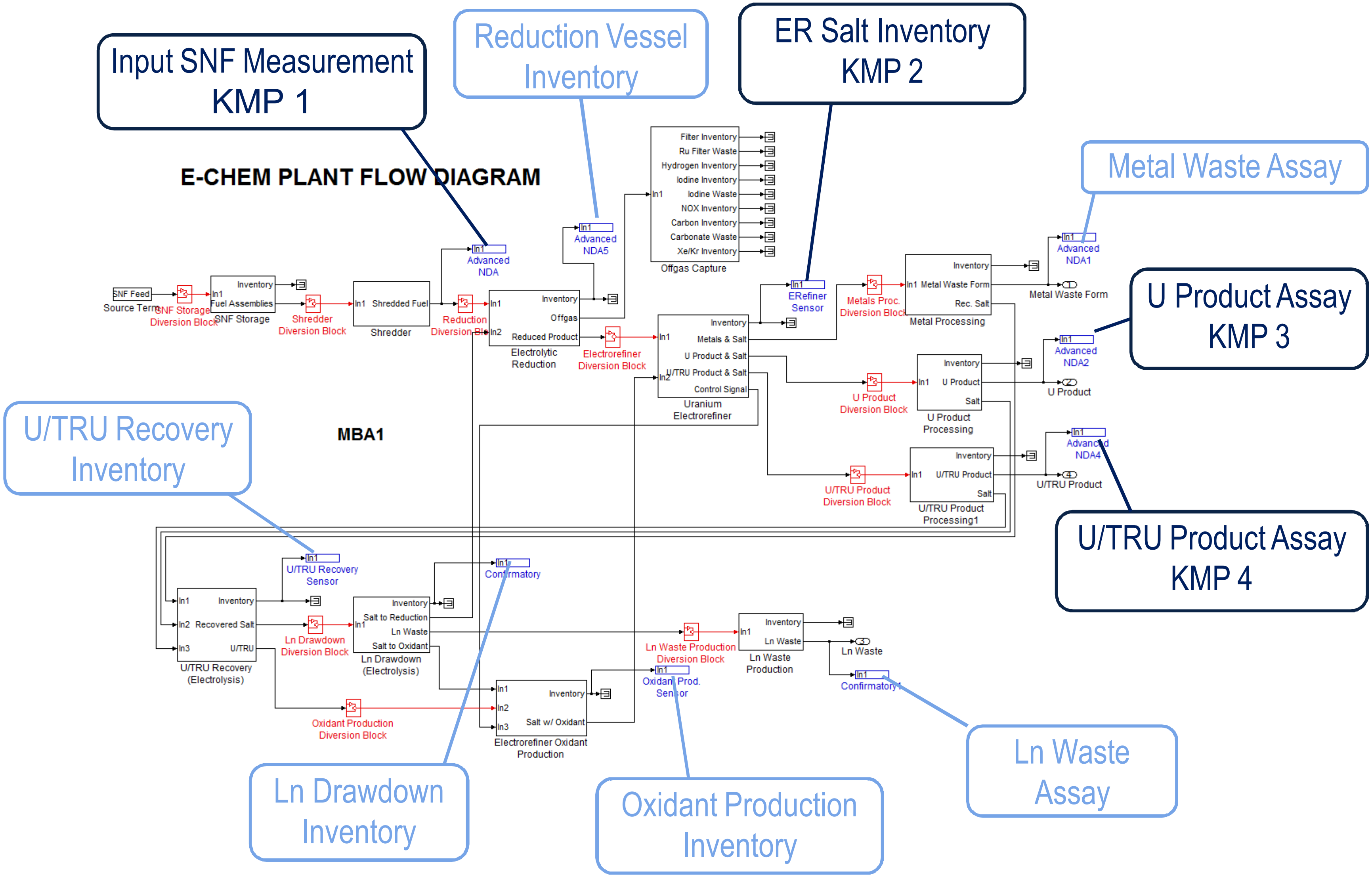
- Lack of Accountability Tank
  - In electrorefining, the extraction onto the cathode occurs as the material dissolves into the salt; the process cannot be decoupled, so the electrorefiner cannot be used to determine input accountability—*need alternative input measurements*.
- Inability to Flushout the Plant
  - Because actinides must buildup in the electrorefiner, it is not feasible to flushout the plant periodically—*requires more extensive use of inventory measurements and near real time accountability*.
- Electrorefiner Inventory
  - The electrorefiner contains a very large TRU inventory, much larger than any other vessel in the plant—*measuring the salt content will be important.*
- Product Measurements
  - Dendrite structures with entrained salt will be difficult to measure—*need to develop measurements for metal products*.

Separation and Safeguards Performance Model

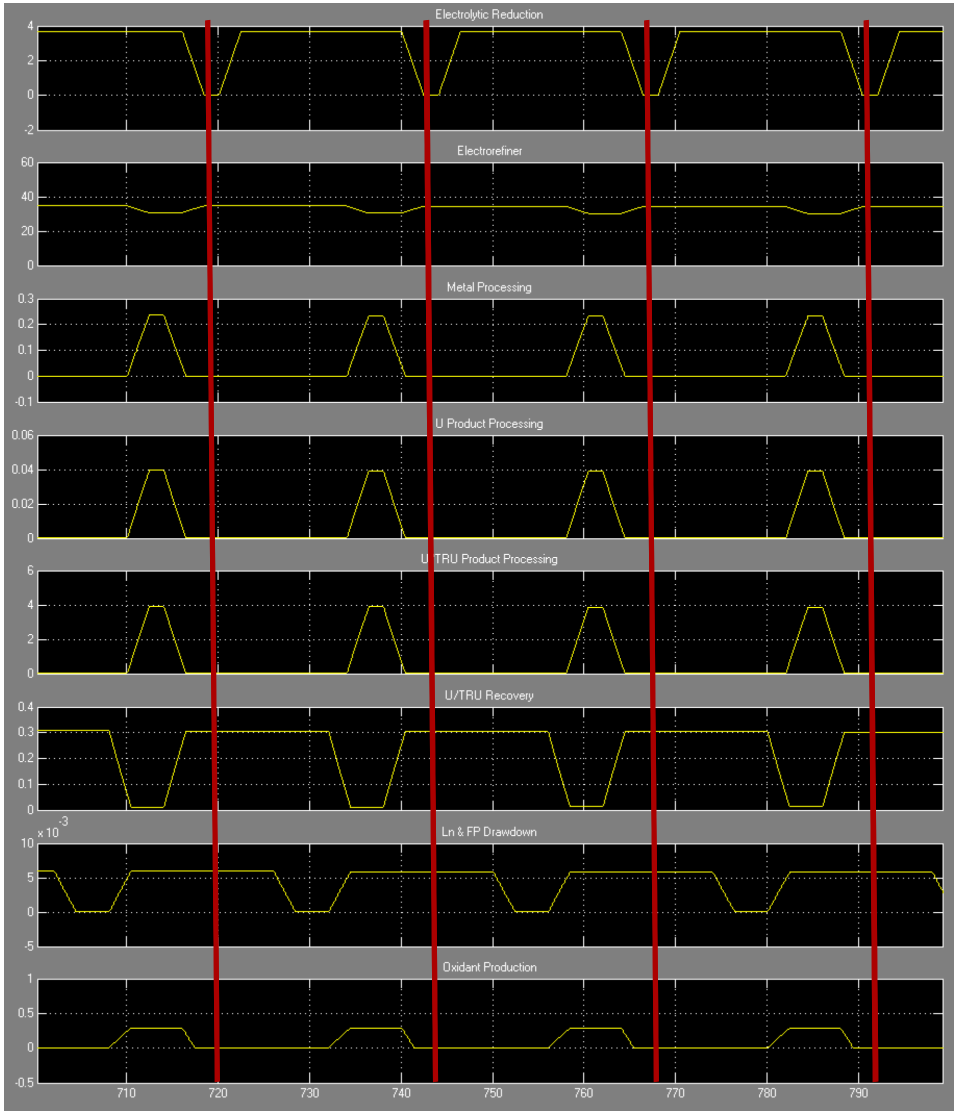


L. Willit, "Pyroprocessing Flowsheets for ... Fuel," *Nuclear Engineering and Technology*,

Accountancy Structure



Inventory Measurement Timing Sequence



← Reduction Vessel
(trace actinide inventory)

← **Electrorefiner**
(high actinide inventory)

← Metal Processing
(in between batches)

← U Product Processing
(in between batches)

← U/TRU Product Processing
(in between batches)

← **U/TRU Recovery**
(small actinide inventory)

← Ln & FP Drawdown
(trace actinide inventory)

← Oxidant Production
(in between batches)

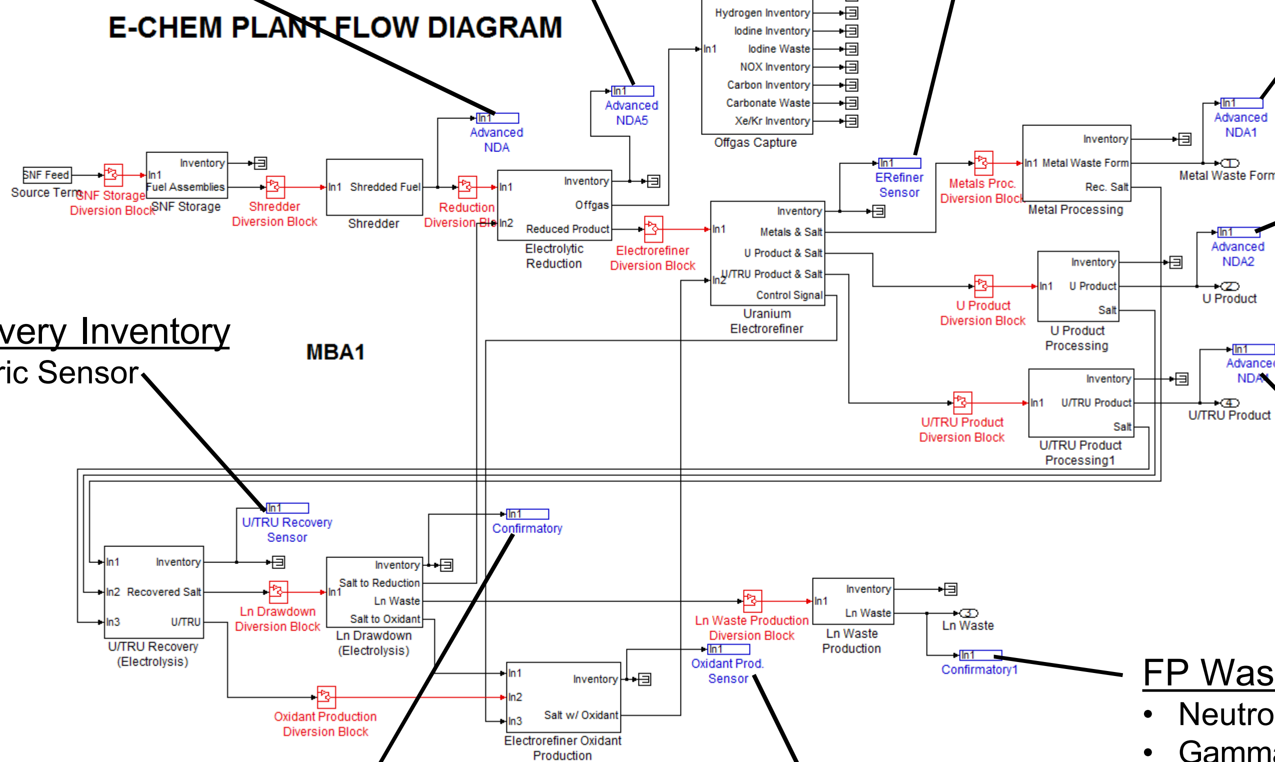
Accountancy Structure

Input Measurement
- NDA/DA of Volox Powders
- LIBS of Volox Powders
- NDA of Shredded Fuel

Reduction Inventory
- Neutron

Electrorefiner Inventory
- Sampling, TIMS & Double Bubbler
- Potentiometric Sensor
- LIBS
- Voltammetry

Metal Waste
- Neutron
- Gamma



U Product
- NDA
- Melt Sampling
- LIBS

U/TRU Recovery Inventory
- Potentiometric Sensor

U/TRU Product
- Melt Sampling
- LIBS

FP Waste
- Neutron
- Gamma

Ln Drawdown Inventory
- Neutron

Oxidant Production Inventory
- Neutron

Delayed Gamma NDA for Input Accountability

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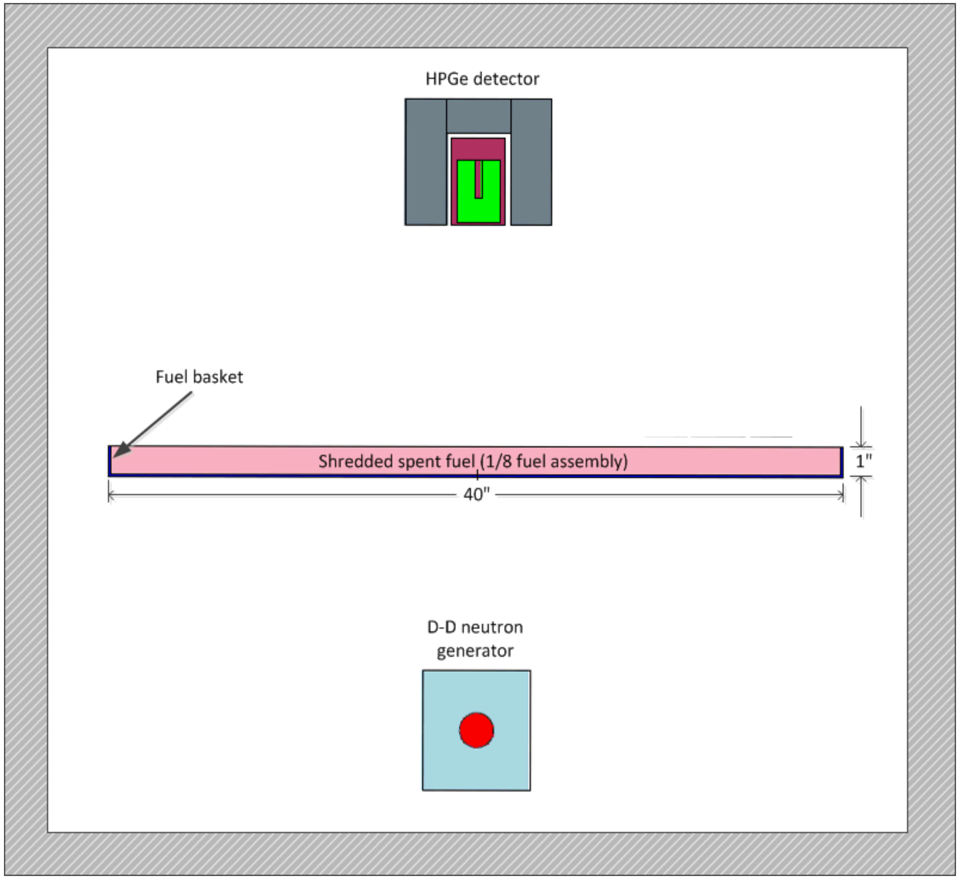
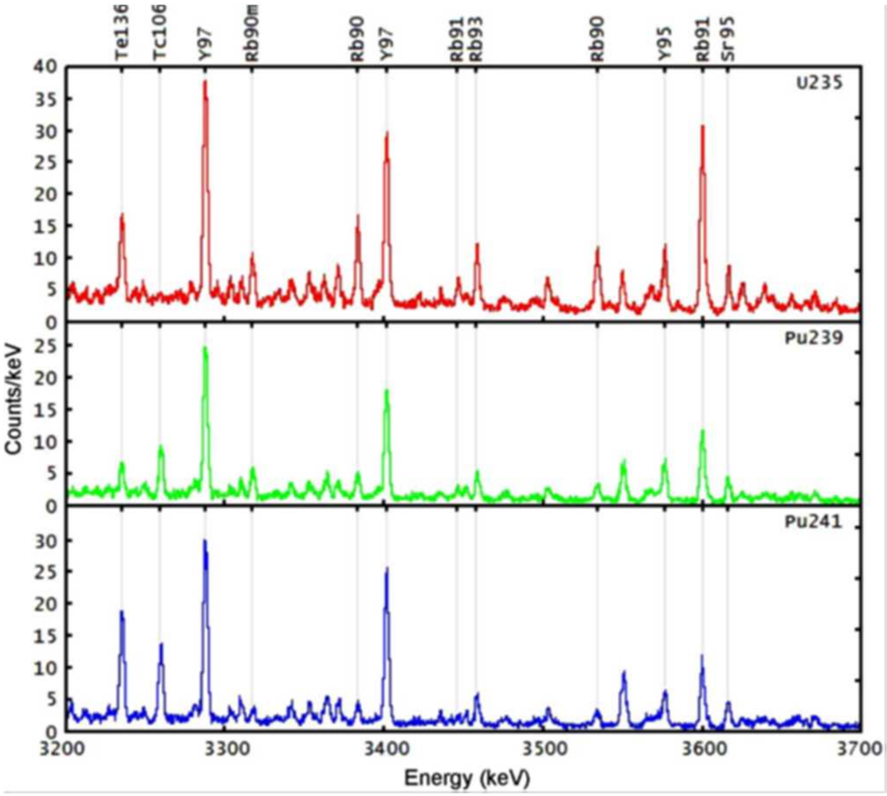
- Measurement is assumed in the front-end process cell (air environment and significant shielding required).
- LWR spent fuel disassembled and shredded
- Spent fuel basket is approximately 40”x40”x1” with porous stainless steel sides. Each basket contains about 75 kg of spent fuel (1/8 of a PWR assembly).
- Neutron source: D-D neutron generator (2.5 MeV) with polyethylene moderator.
  - Isotopic point source (10^{10} n/s, 2.5 MeV)
  - 10 second irradiation period
  - 1 second cool down
  - 30 second acquisition time
- HPGe detector system, 3.2 cm radius, 40% efficiency



Delayed Gamma Background

- Passive measurements of signature isotopes lead to high measurement uncertainties
  - Cs-137, Cs-134, Cm-244, Eu-134
  - Dependent on reactor history and initial fuel composition (supplied by operator) and subject to high uncertainties
- Delayed gamma assay
  - Delayed gammas from fission products serve as signature
  - Gamma line intensity ratios can differentiate between fission of U-235, Pu-239, Pu-241.
  - Intense radiation from spent fuel complicates ability to distinguish fissile isotopes
  - Mitigated by identifying high energy emitting isotopes as signatures (Campbell et al.)
    - 3 – 4 MeV range



Modeling Approach

TINDER

(Computational Shell/Driver)

MCNP6

- 3D geometry, material compositions, and neutron source specification
- Performs neutron transport calculations in the irradiated material (spent fuel)
- Produces neutron and gamma flux in fine energy groups

CINDER2008

(Modified to include photon induced reactions)

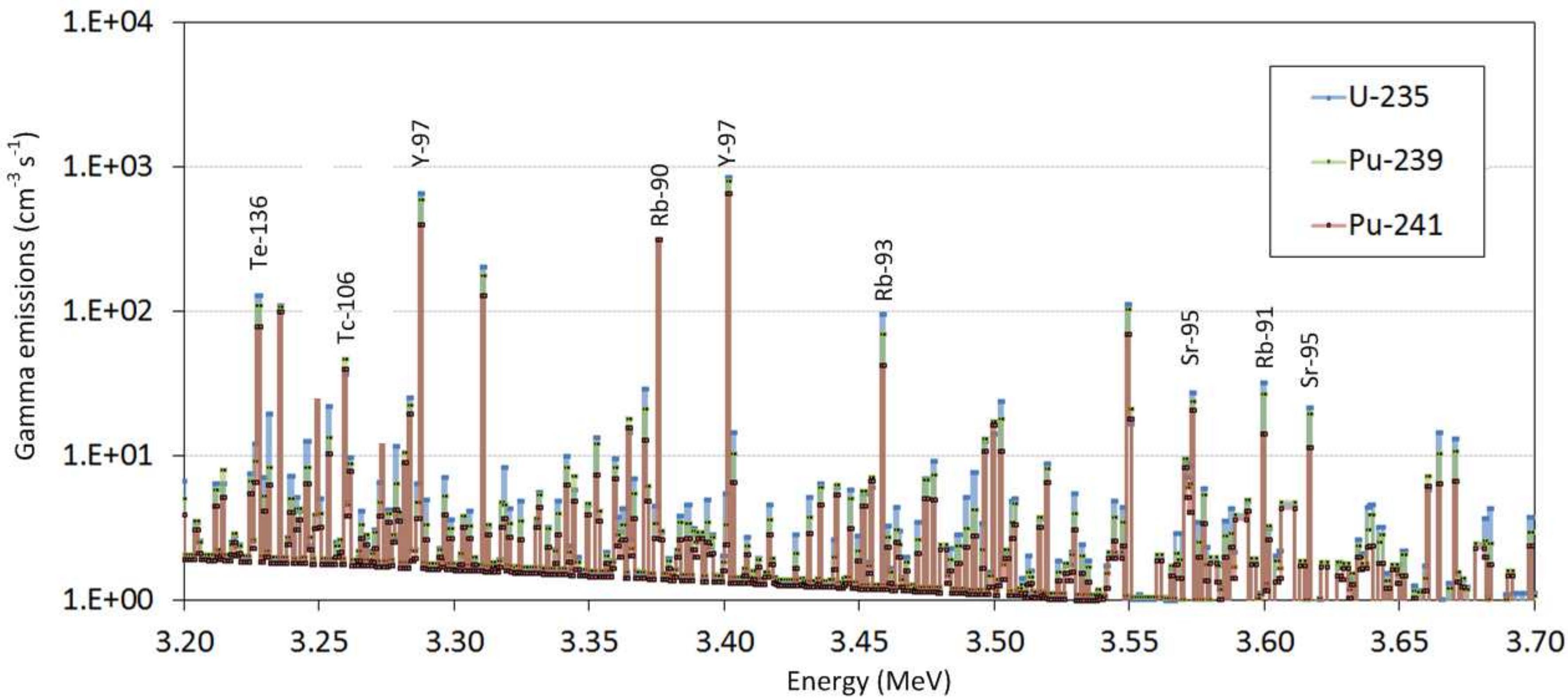
- Using predefined neutron/gamma fluxes and isotopic inventory
- Performs dual particle transmutation calculations in the sample material
- Produces discrete gamma spectrum (delayed and passive emissions)

MCNP6

- The photon source is applied back into the sample and propagated to the acquisition system
- Detector system parameters are applied (detector response, resolution, Gaussian energy broadening)
- Produces expected gamma spectrum as measured by HPGe detector

Simulation Results

- PWR spent fuel, 4% initial enrichment, 35 GWd/MTHM
- Gamma emission intensities for fissile isotopes vary slightly, but Tc-106 peak will be important measure.
- The Tc-106 peak will be ~10^5 counts, so at ~1% counting statistics, but this could be optimized more.



Simulation Results (Decrease of U-235 content from 40% of fissile makeup to 38%)

Percent change in emission intensity for selected isotopes

Isotope	Tc136	Tc106	Y97	Rb90	Rb91
Energy (MeV)	3.235	3.260	3.288	3.383	3.600
Percent change (%)	-2.418	2.872	-3.112	-3.148	-3.562

- Pu content increased by 0.62/0.6 = 1.033
- U-235 content decreased by 0.38/0.4 = 0.95
- Fission product yields for Tc-106:
  - U-235: 0.402, Pu-239: 4.4

$$\frac{(1.033)(4.4)+(0.95)(0.402)}{4.4+0.402}=1.026=2.6\%$$

Delayed Gamma NDA Discussion

- Count rates appear to be in the range needed for 1% counting statistics
- More work will be required to determine expected measurement uncertainties
- This geometry seems to be more desirable as compared to a fuel assembly (no axial variation, less self-shielding, less volume)
- Calibration could be a challenge, but probably easier than developing a spent fuel assembly calibration standard.
- Future work will focus on measurement uncertainty and can also examine the determination of fuel burnup and initial enrichment.

Conclusions

- A number of engineering challenges exist for nuclear material accounting in an electrochemical processing facility.
- Electrochemical facilities will require a new safeguards approach (as compared to existing aqueous plants).
- We're using modeling and simulation to determine measurement needs and performance in diversion scenarios.
- Future work will examine the integration of process monitoring information to help fill in the gaps in traditional accounting.