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# Materials Accountability in Next Generation Reprocessing Plants

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Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

# Objective & Outline

- One of the goals of the Global Nuclear Energy Partnership is to provide a proliferation resistant fuel cycle.  Part of achieving this goal involves building advanced materials accountancy into fuel cycle facilities.
- Sandia is investigating advanced instrumentation for MC&A measurements and advanced accountancy schemes that integrate all plant process data with MC&A measurements
- Outline
  - Current Accountability Practices & Goals
  - Instrumentation Types
  - Advanced Instrumentation
  - Advanced Accountancy Schemes
  - Safeguards Performance Model

# Current Accountability Practices

- The ultimate goal in reprocessing accountability is to get high precision near real-time measurements of Pu content.
- Current reprocessing plants measure bulk quantities of material entering and leaving the process in real-time, but the U and Pu content is measured in samples taken once or twice per day.
- Physical Inventories are taken upon plant shutdown.
- Near real time accountability of elemental content (Pu for example) is incredibly difficult because of the difficulties of measuring in-process Pu inventories with high precision.
  - We can examine increased sampling of in-process material (which requires advanced instrumentation).
  - We can also look at advanced accountancy schemes that take advantage of all plant processing data.

# Accountability Goals for a Next Generation Plant

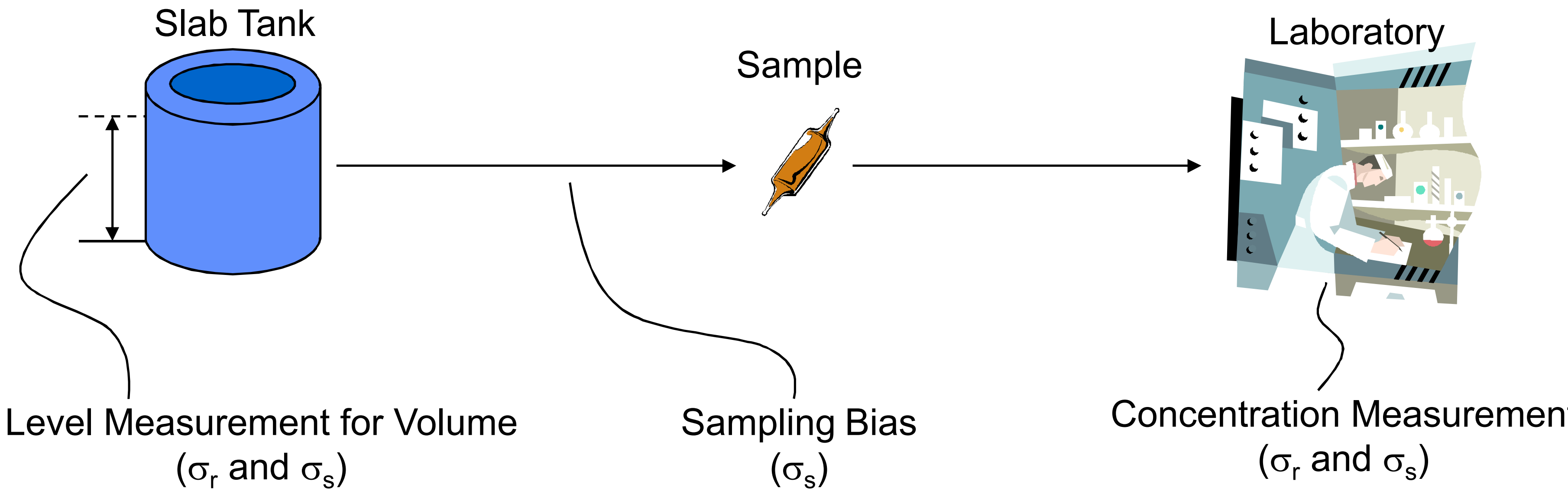
- **For an extended diversion, we want to be able to detect the loss of 8 kg of Pu in one month with a 95% detection probability, which means the error is 2.43 kg**
- **A 2,000 MT/yr reprocessing plant processes 2,330 kg Pu per month:**

**2.43/2330 = 0.1%**

**(This is the combined error we need to achieve for the major Pu-bearing streams)**

- **The good news is that for waste and recycle streams, the Pu content is low or near-zero, so we don't need as stringent measurement uncertainties.**
  - **For these streams/products, gamma spec and neutron counting techniques can work well enough**

# How is Plutonium Measured?




- **It is incredibly challenging to get the combined total error < 0.1% due to propagation of error.**
- **It would take a combination of very precise and well-calibrated instruments.**

# Measurement Precisions for Nuclear Material

| | U Measurement | | Pu Measurement | |
|---|---|---|---|---|
| | Dissolver Solution | Pure Product | Dissolver Solution | Pure Product |
| Passive/Active Neutron Counting | N/A | N/A | 5% | 2% |
| High Resolution Gamma Spectroscopy | N/A | N/A | N/A | 0.5-2% |
| Isotope Dilution Gamma Spectroscopy | N/A | N/A | 0.50% | 0.50% |
| Gravimetry | N/A | 0.10% | N/A | 0.10% |
| Titrimetry | N/A | 0.10% | N/A | 0.1-0.5% |
| Constant Potential Coulometry | N/A | 0.1-0.2% | N/A | 0.1-0.2% |
| Calorimetry | N/A | N/A | N/A | 0.1-0.5% |
| Spectrophotometry | 1% | 0.2-0.3% | 1% | 0.2-0.3% |
| Hybrid K-Edge Densitometry | 0.20% | N/A | 0.70% | N/A |
| ICP Mass Spectrometry | <0.1% | <0.1% | <0.1% | <0.1% |

- **Mass Spectrometry is the only way (currently) to get the high precisions required for accountability, but it requires about 8 hours to measure one sample.**
- **This lengthy measurement time makes it costly to use mass spec measurements in multiple locations in the plant.**
- **An advanced accountability system will require more mass spec measurements: Techniques like TARIS (Thermal Atomization Resonance Ionization Spectroscopy) and IC-MS (Ion Chromatograph coupled Mass Spectrometry) could speed up the processing time.**



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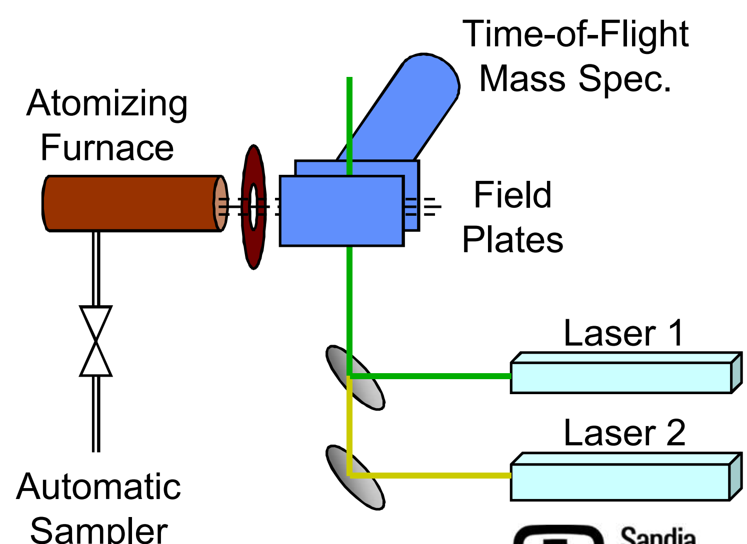
# Advanced Mass Spec Concept: TARIS

- TARIS (Thermal Atomization Resonance Ionization Spectroscopy) is a combination of three technologies:
  - A liquid sample is placed on a hot tantalum plate in a vacuum chamber to atomize the solution (TA)
  - Lasers selectively ionize a particular element group (RI)
  - The ions are sent through a mass spectrometer to determine the isotopic spectrum
- Atom Sciences, Inc. is investigating this technique over this next year
- The measurement time for Pu content could be as low as 20 minutes
- Other elements (U and other actinides) could be measured, but tunable lasers would be required.



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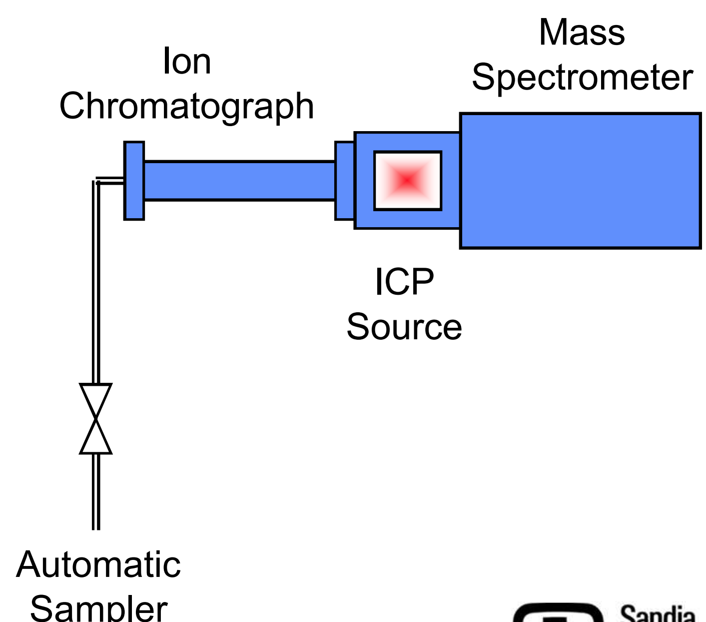
# Advanced Mass Spec Concept: IC-MS

- IC-MS (Ion Chromatography-Mass Spectroscopy) is an automated combination of two technologies:
  - A liquid processing sample passes through an ion chromatograph to separate the elements (IC)
  - Each group of separated elements then runs through the mass spectrometer (MS)
- Sandia is investigating this technique, though it has been used in the past at PNNL
- The measurement time could reduced due to the automation of the separation
- Other elements (U and other actinides) an easily be measured.



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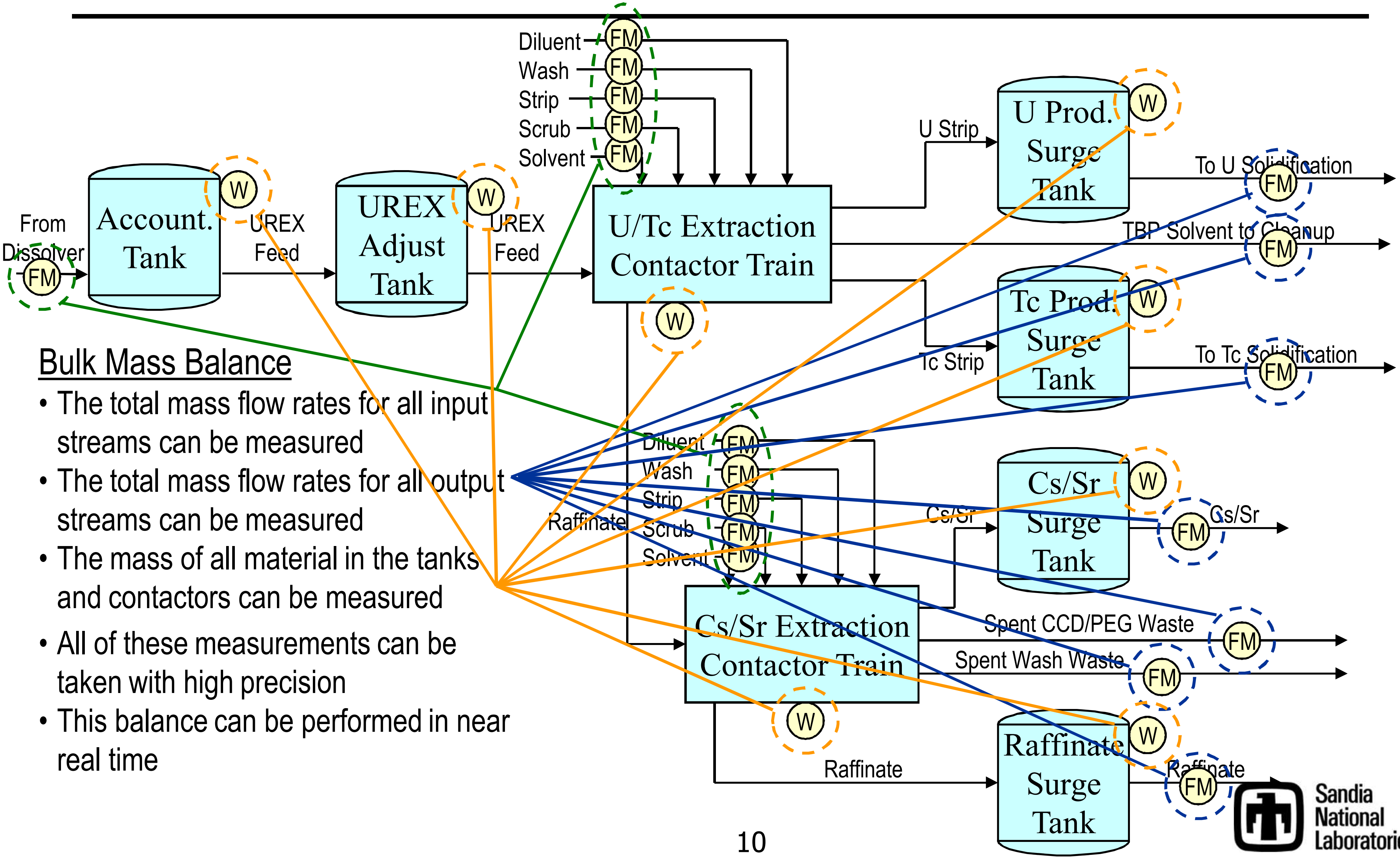
# Advanced Accountancy Schemes

- Although high precision near real-time mass balances of elemental content is difficult, we can do it for bulk mass flows.
- To do a near real-time material balance, you have to know all mass flows in and out along with the amount of material in process.
- Any diverted material should be detected almost immediately
- However, an integrated NRTA of bulk materials flows along with elemental measures from key samples could yield an advanced accountability system
- The key is to use all of the process monitoring and MC&A measurements together

# U, Tc, Cs/Sr Extraction Bulk Mass Balance




## Bulk Mass Balance

- The total mass flow rates for all input streams can be measured
- The total mass flow rates for all output streams can be measured
- The mass of all material in the tanks and contactors can be measured
- All of these measurements can be taken with high precision
- This balance can be performed in near real time



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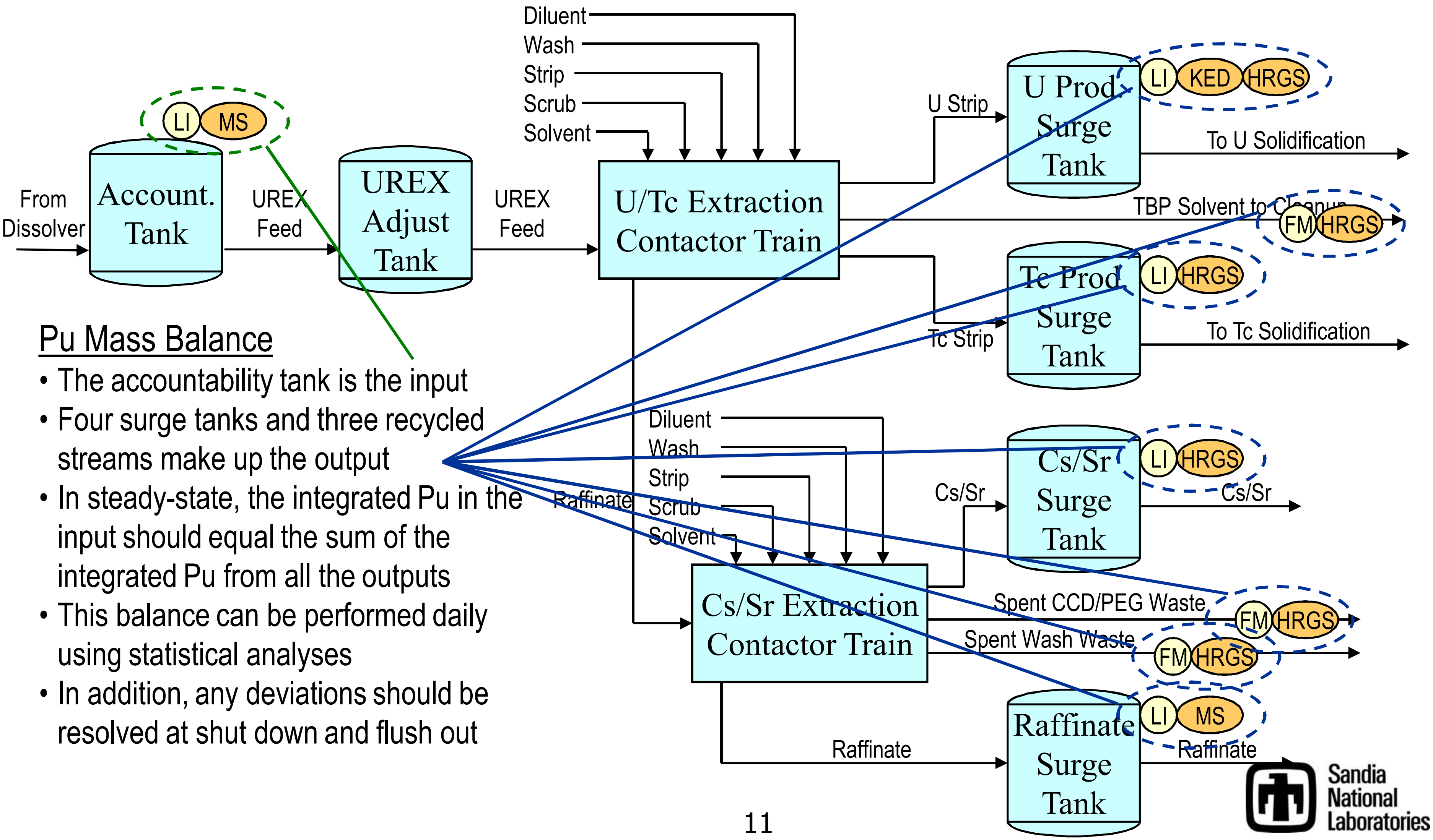
# U, Tc, Cs/Sr Extraction Plutonium Balance




## Pu Mass Balance

- The accountability tank is the input
- Four surge tanks and three recycled streams make up the output
- In steady-state, the integrated Pu in the input should equal the sum of the integrated Pu from all the outputs
- This balance can be performed daily using statistical analyses
- In addition, any deviations should be resolved at shut down and flush out



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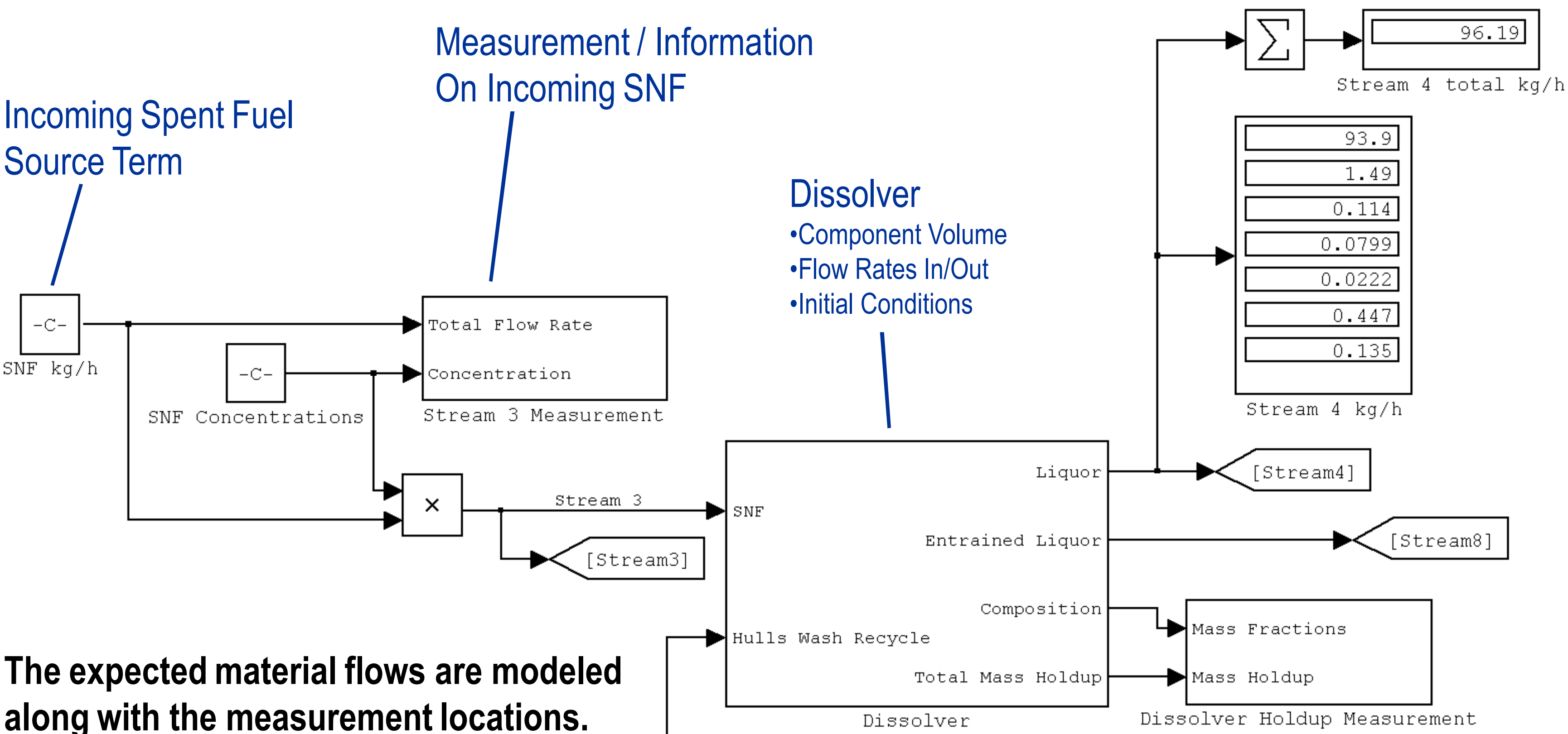
# Safeguards Performance Modeling

- A Safeguards Performance Model is being developed at SNL that has several valuable capabilities:
  - Testing of advanced instrumentation schemes
  - Investigation of data correlation strategies
  - Testing of diversion scenarios
- The model can be used for Safeguards Performance Testing (for licensing) of a reprocessing plant
- Ultimately it can form the basis for the materials accountability system

# Front End Simulink Model




- The expected material flows are modeled along with the measurement locations.
- The model simulates measurements based on the random and systematic errors
- A statistical package is used to calculate the SEID or MUF over a mass balance area



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# Scripts were developed to calculate SEID for different mass balance areas

**Component: Pu**

**Inventories:**

| Tag | Value (kg) | Rand Err (kg) | Syst Err (kg) |
|---|---|---|---|
| Dissolver | 4.575 | 0.065 | 0.000 |
| Hulls Wash | 0.015 | 0.000 | 0.000 |
| Feed Adjust | 4.616 | 0.065 | 0.000 |
| Centrifuge | 0.755 | 0.011 | 0.000 |
| UDS Leach | 0.763 | 0.011 | 0.000 |

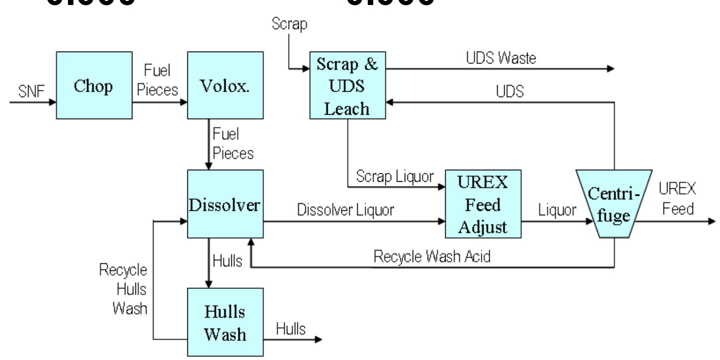
**Inflows:**

| Tag | Value (kg) | Rand Err (kg) | Syst Err (kg) |
|---|---|---|---|
| SNF | 14.620 | 0.737 | 0.000 |
| Scrap | 0.286 | 0.014 | 0.000 |

**Outflows:**

| Tag | Value (kg) | Rand Err (kg) | Syst Err (kg) |
|---|---|---|---|
| UREX Feed | 15.217 | 0.144 | 0.000 |
| Washed Hulls | 0.001 | 0.000 | 0.000 |
| UDS Waste | 0.000 | 0.000 | 0.000 |

**SEID = 0.762 kg = 5%**
**Inflow Total = 14.906 kg**
**Outflow Total = 15.218 kg**
**Inventory Total = 10.724 kg**
**Time Period = 10.00 h**

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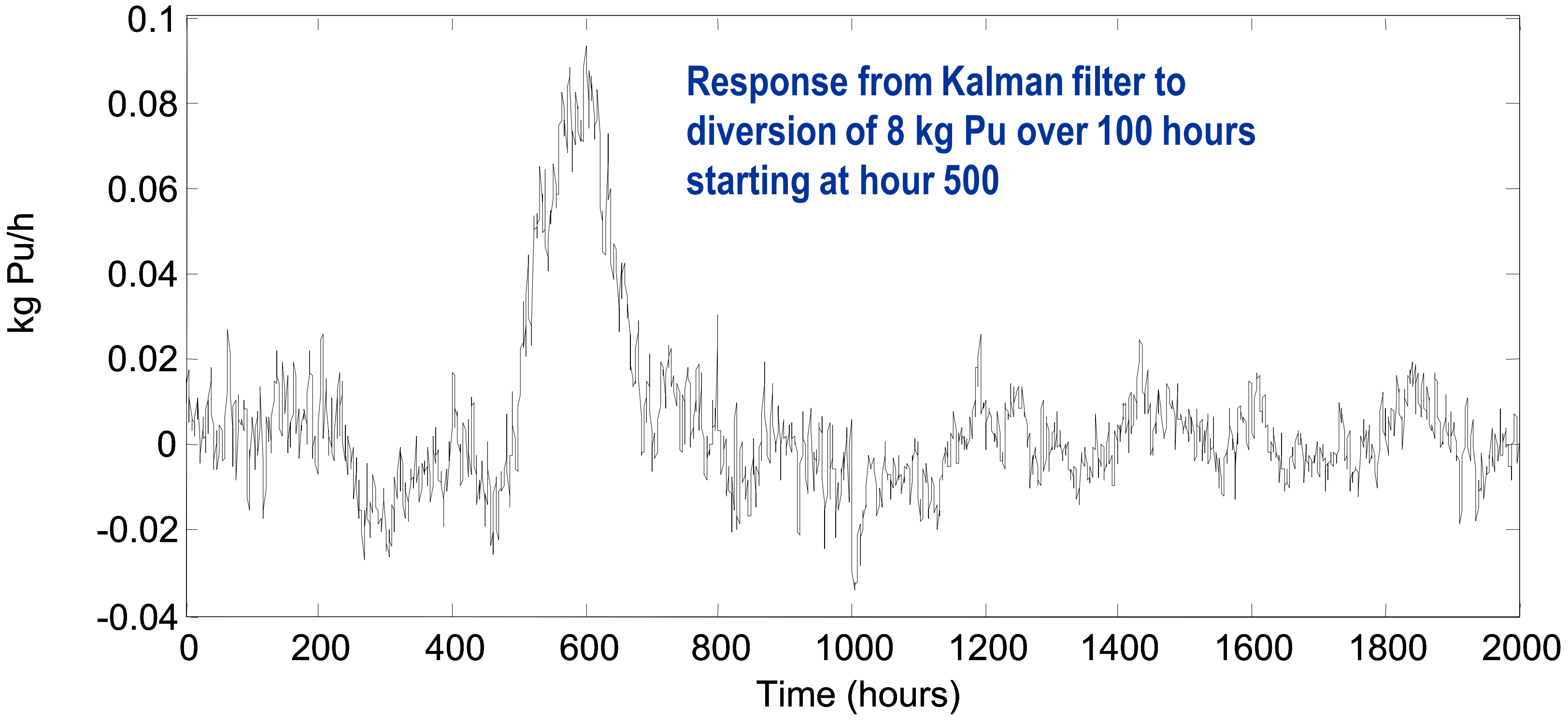
# Various statistical tests and filters are being investigated to compare detection thresholds



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# Future Work

- The Safeguards Performance Model will be updated as the process design for UREX+1a evolves, or as changes in the GNEP program occur.
- The process and instrumentation design work from a number of collaborators will be incorporated into the model.
- We will be producing SEID calculations over mass balance areas to help determine the instrumentation mapping—the end goal is to have a full instrumentation mapping by the end of the year.
- It is hoped that the testing of the TARIS and IC-MS techniques will give us new capabilities in materials accounting—the model will help us determine the impact of these technologies.