SAND2007-2130C

Advanced Mass Spectrometry Methods

GNEP Safeguards Working Group Meeting
Salt Lake City, Utah

Ben Cipiti (Sandia)
Kathleen Holt (Sandia)
Tom Whitaker (Atom Sciences, Inc.)

April 12-13, 2007

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

- In mixed reprocessing samples, mass spectrometry is the only way to achieve high-precision accountability measurements.
- However, the lengthy measurement time limits how many measurements can reasonably be taken.
- Two advanced mass spectrometry techniques are being investigated for their promise to dramatically speed up the overall measurement time.
  - Ion Chromatography-Mass Spectrometry (IC-MS)
  - Thermal Atomization Resonance Ionization Spectroscopy (TARIS)

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

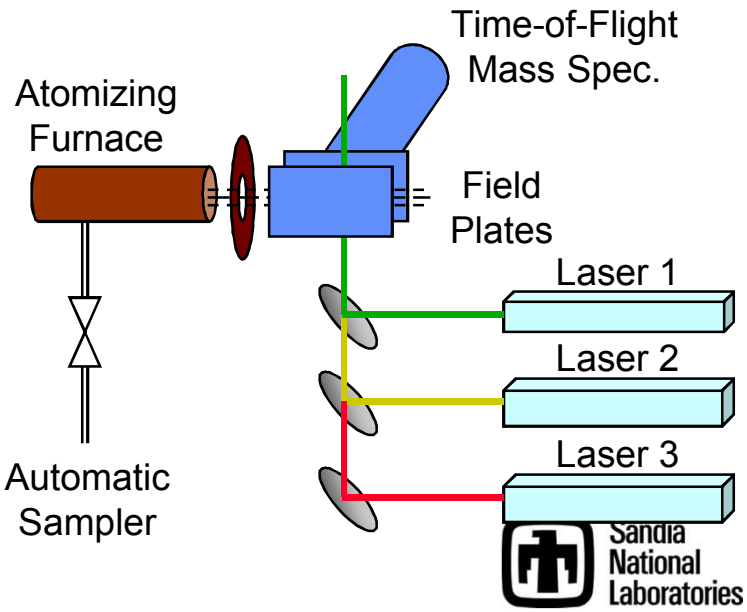
Measurement Precisions for Nuclear Material

	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.**

Advanced Mass Spec Concept: TARIS

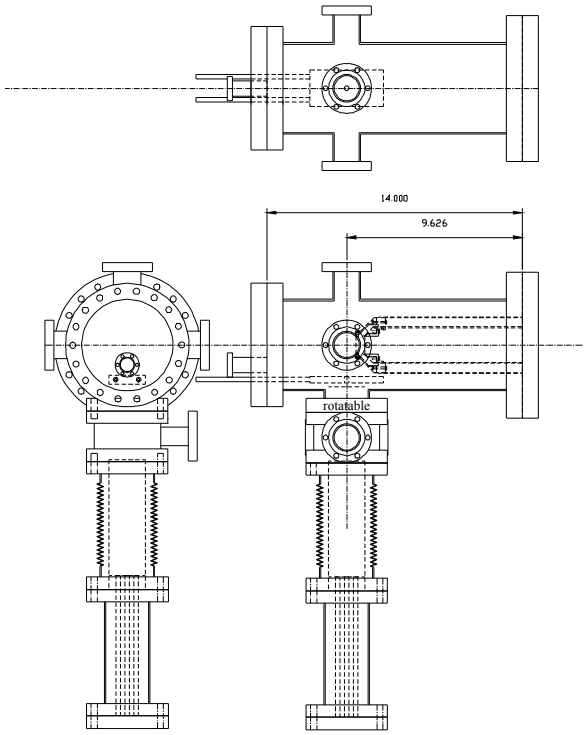
- **TARIS (Thermal Atomization Resonance Ionization Spectroscopy) is a combination of three technologies:**
  - **A liquid sample is placed on a hot tantalum plate in a vacuum chamber to atomize the solution (TA)**
  - **Lasers selectively ionize a particular element group (RI)**
  - **The ions are sent 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.**

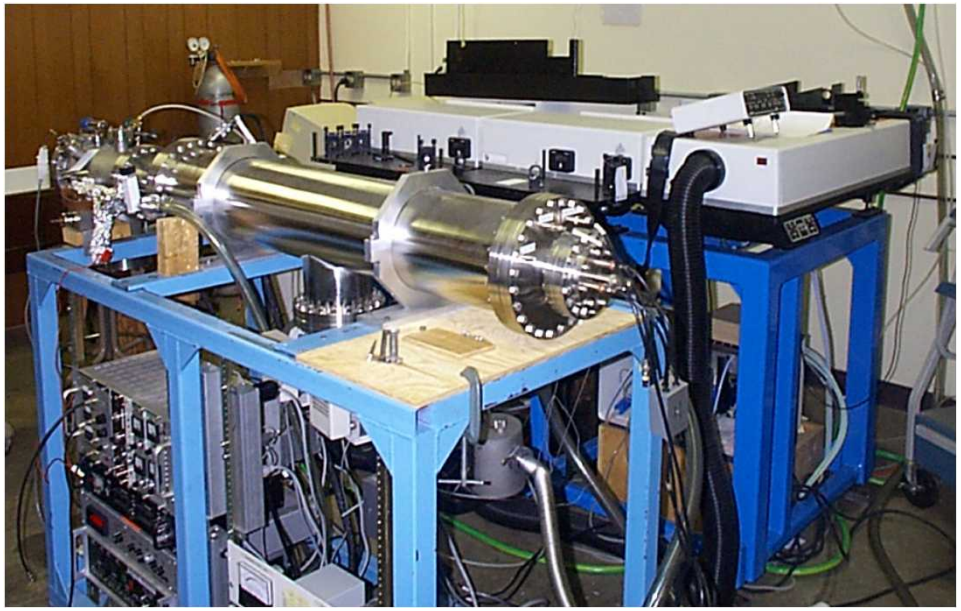


Sandia National Laboratories

TARIS Experiment at Atom Sciences

- A new source chamber was ordered to allow TARIS to be performed on an existing SIMS Time of Flight Mass Spectrometry





- Preliminary testing is expected to begin this month
- The lab will be licensed to work with U, Pu, and Th

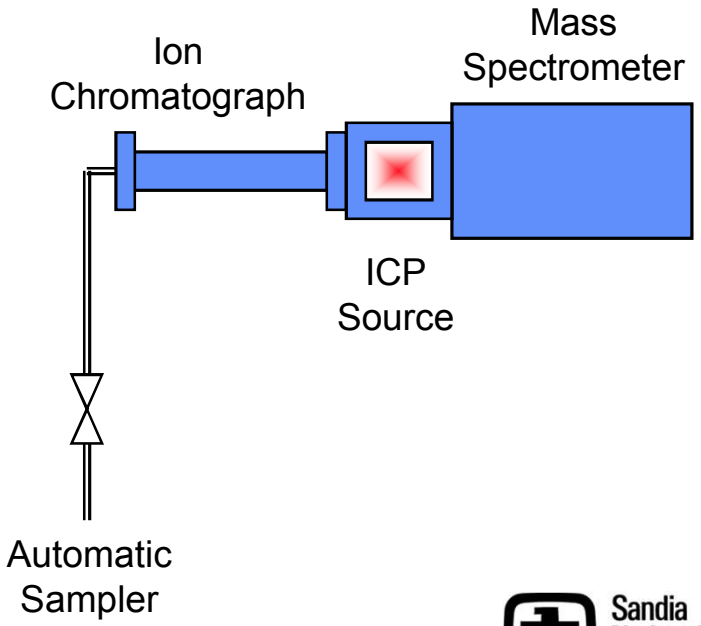
Sandia National Laboratories

TARIS Experimentation

- The first transition in U will be excited using a 319.9 nm dye laser, and the second transition will be excited using a second 591.5 nm dye laser. The second excited state can be photoionized by a 1064 nm Nd:YAG pump laser.
- The parameters of interest for this study are the overall measurement time, precision, effect of interferences, robustness for the reprocessing environment, and cost.
- Cost is likely to be an issue due to the number of laser wavelengths required—it may be too difficult to use TARIS for multiple elements.
- The waste stream produced by the residuals will also be important to quantify.
- Export control will likely be an issue for other potential international use.

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 & SRNL**
- **The measurement time could be reduced due to the automation of the separation. Previous work has shown analysis times of 30 minutes.**
- **Other elements (U and other actinides) could easily be measured.**



Sandia National Laboratories

IC-MS Experimentation

- A radioactive materials lab at Sandia is equipped to run the Ion Chromatograph with actinide samples in a glove box. Liquid samples available for use include small quantities of ^{238}U, ^{237}Np, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am, ^{241}Am, ^{99}Tc, ^{90}Sr, and ^{137}Cs.
- Initial work will examine solutions of U and Pu only.
- Separation of the actinides will be accomplished using Chelation Ion Chromatography.
- Currently the lab is not set up with a mass spectrometer, so this capability may need to be added later. The separated samples may need to be sent out for mass spectrometer measurements or will be analyzed using other radioanalytical techniques.
- The parameters of interest for this study are the overall measurement time, precision, effect of interferences, robustness for the reprocessing environment, and cost.

Future Work

- We expect experimental results for both techniques within the next month or two. The final analysis will appear in the end of the year report.
- We also plan to analyze the implications of these new types of measurement instrumentation using the Safeguards Performance Model.