Nuclear Forensics at Los Alamos National Laboratory

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Overview

• Introduction to nonproliferation efforts
• Scope of activities at Los Alamos National Laboratory
• Facilities for radioanalytical work at LANL
• Radiochemical characterization capabilities
• Bulk chemical and materials analysis capabilities
Los Alamos National Laboratory

- Over 65 years of actinide nuclear science
- Continuing mission
  - Develop and apply science and technology to ensure the safety and reliability of the United States nuclear deterrent; reduce the threat of weapons of mass destruction, proliferation, and terrorism; and solve national problems regarding defense, energy, environment, and infrastructure.
Los Alamos National Laboratory

• Analytical chemistry established in 1943 – plutonium R&D
• Special nuclear material recovery and processing (recycle or stabilization)
• Nuclear reactor fuel R&D
• Safeguards accountancy
• Space programs nuclear materials (Cassini, Galileo, Mars Rover, Mars Science Lab)
• Environmental management and remediation
• Destructive and non-destructive analysis standards
• Bulk nuclear material forensics
LANL technical capabilities fostered by the nuclear weapons program support the evaluation of all categories of nuclear events:

- **Materials**
- **Device**
- **Post-detonation**
Nonproliferation Efforts

- Combat the illicit trafficking of nuclear materials
- Detect activities indicating nuclear proliferation
- Monitor adherence to treaties and international agreements
- Characterize and catalog US nuclear material attributes

Nuclear Material Analysis and Data Assessment
Scope of Technical Activities

Our ability to characterize nuclear materials and processes relies on analytical methods from:

Certification of materials in pit production

Support for international safeguards programs

Underground test experience

Our ability to characterize signatures of worldwide nuclear materials production relies on skills built in:

Our ability to characterize the origins of a nuclear explosion is based on:
Several facilities to work with materials of all quantities

All facilities house ongoing missions that exercise analytical capabilities routinely

- **TA-48**
- **TA-55**
- **Sigma**
- **RC-45**
  - $< 10^{11}$ atoms Pu-239
- **RC-1, Sigma**
  - $10^9 - 10^{22}$ atoms Pu-239
- **CMR**
- **Nuclear facilities (CMR, PF-4)**
  - $> 10^{22}$ atoms Pu-239
Sample Matrices

**Plutonium Metal**
- pure
- impure
- heat source
- power source
- precursors

**Uranium Oxide**
- pure (EU)
- targets (NU)
- MOX fuels (EU)
- reactor fuel (EU)
- metal precursor (EU)
- ore concentrate (NU)

**Plutonium Oxide**
- pure
- impure
- MOX fuels
- heat source

**Uranium Metal**
- pure (EU)
- targets (NU)

**Uranium Fluoride**
- tetrafluoride
- hexafluoride

**Rad Sources**
- PuBe neutron
Radiochemistry

- Single or multi-element
- Elementally separated radiochemistry
- Standardized procedures
- Refractory matrices
Count Room capabilities

Provides qualitative and quantitative assay of gamma, beta, and alpha-emitting radionuclides in a variety of matrices and over a wide range of activity levels,
  – Trace levels to $>10^{13}$ fissions

• Specializing in fission product measurements,
• Operates 24x7x365
• Sample receipt and handling protocols
• Makes ~ 70,000 measurements annually
  – 30,000 high resolution gamma collections
• 30 non-automated high-resolution gamma-ray spectrometers, some highly specialized
• 10 custom automated high-resolution gamma-ray spectrometers,
• 6 custom automated beta counters.
• 6 custom non-automated beta counters (3 ~0.2 CPM Bkg)
• ~90 Alpha spectrometers,
• Batch and interactive analysis codes,
• Relational database with web-based visualization tools.
Clean chemistry and mass spectrometry

-State-of-the-art mass spectrometry instrumentation
  • TIMS
    - NBS 1290
    - Sector 54
    - IsotopX Isoprobe-T
  • ICPMS
    - HR-ICPMS (Element XR and Element 2)
    - MC-ICPMS (Neptune and Neptune Plus)
  • IRMS
    - MAT-253
    - Delta V

-10,000 sq. ft. of clean lab space, ideally suited for low-level routine analyses
  - Perchloric acid hoods
  - Dry and wet ashing

- DOELAP accreditation; full quality envelope

- Many years of routine trace environmental and bioassay monitoring experience

- Expertise in the development and implementation of new procedures
Radioanalytical capabilities - application to nonproliferation forensics
# Nuclear Materials Characterization

<table>
<thead>
<tr>
<th>Analytical Method</th>
<th>Information</th>
</tr>
</thead>
<tbody>
<tr>
<td>High Resolution Autoradiography</td>
<td>Number density of radioactive, approximate size</td>
</tr>
<tr>
<td>High Resolution Gamma Spectrometry (Radiochemistry)</td>
<td>Isotopic composition of Np, Am, U, and Pu, fission products</td>
</tr>
<tr>
<td>Micro X-ray Fluorescence</td>
<td>Elemental Distribution</td>
</tr>
<tr>
<td>Optical/Electron Microscopy</td>
<td>Particle size, elemental distribution, grain size, porosity, surface roughness.</td>
</tr>
<tr>
<td>X-ray Diffraction</td>
<td>Composition/phase</td>
</tr>
<tr>
<td>Pycnometry</td>
<td>Density</td>
</tr>
<tr>
<td>Laser-Based Particle Size Analysis</td>
<td>Particle size distribution</td>
</tr>
</tbody>
</table>
Analysis and characterization of actinide materials

Onsite Analytical Chemistry and Sample Management

- Coordinate sample receiving, shipping, and distribution at TA-55 and CMR
- Onsite radiochemical and trace analysis

Assay and Classical Chemistry

- Coulometric titration
- Ceric titration
- Pu (III) and Pu (IV)
- U Assay by Davies Gray
- Fe and Si determination
- Loss on Ignition (LOI)
- Free acid determination
- Standard preparation

Plasma Spectroscopy

- Inductively Coupled Plasma-Mass Spectrometry
- Inductively Coupled Plasma- Atomic Emission Spectrometry

- DC Arc Emission
- Cold-Vapor Atomic Fluorescence

X-Ray Fluorescence (XRF) and X-Ray Diffraction (XRD)

- Fingerprint Detection Technology
Analysis and characterization of actinide materials

**Mass Spectrometry**
- High-Precision Gas Mass Spectrometry

**Radiochemistry and Nondestructive Analysis**
- Alpha and gamma spectrometry
- Gross alpha, liquid scintillation

**Interstitial Analysis & Ion Chromatography**
- Fluoride, chloride, nitrite nitrate, phosphate, sulfate oxalate and perchlorate
- Carbon, oxygen, hydrogen sulfur, moisture, and tritium

**Laboratory Information Management System and Quality Assurance**
- Oracle SQL*LIMS
- Sample/nuclear material tracking and data management

**Quality Assurance and Control**
- Record Management
- Document Control
- Training
Physical characterization tools for bulk materials

Materials processing techniques leave unique signatures in microstructures:
- Grain Size/Morphology
- Inclusion Distribution/Morphology
- Microstructural Texture
- Mechanical Properties
- Surface Features

100 µm
Materials analysis

Origin?
Production Method?
Age of Material?
Intended Use?

History

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Sample (µg/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{234}$U</td>
<td>$5.5 \times 10^1$</td>
</tr>
<tr>
<td>$^{235}$U</td>
<td>$5.7 \times 10^3$</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>$7.9 \times 10^5$</td>
</tr>
<tr>
<td>$^{238}$Pu</td>
<td>$7.4 \times 10^4$</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>$2.5 \times 10^1$</td>
</tr>
<tr>
<td>$^{240}$Pu</td>
<td>$1.0 \times 10^2$</td>
</tr>
<tr>
<td>$^{242}$Pu</td>
<td>$2.5 \times 10^4$</td>
</tr>
</tbody>
</table>
Sample management, QA/QC

• Bulk analysis and radiochemistry analytical capabilities operate under specific quality programs funded by sponsors. These include DOELAP, NQA-1

• Some operations will move toward ISO 17025 compliance

• Need for expanded SRM program, laboratory intercomparisons
Investments in modern capabilities

- Neptune MC-ICP-MS
- IsotopX Isoprobe-T MC-TIMS
- FEI SEM w/WDS & EDS
- Element XR ICP-MS

- Dual-Clover high resolution gamma-ray spectrometers
Conclusions

• Analytical chemistry measurements on plutonium and uranium matrices are critical to numerous defense and non-defense programs including safeguards accountancy verification measurements.

• Los Alamos National Laboratory operates capable actinide analytical chemistry and material science laboratories suitable for nuclear material forensic characterization.

• Actinide analytical chemistry uses numerous means to validate and independently verify that measurement data quality objectives are met.

• Numerous LANL nuclear facilities support the nuclear material handling, preparation, and analysis capabilities necessary to evaluate samples containing nearly any mass of an actinide (attogram to kilogram levels).