Challenges in Bulk Nuclear Forensics Sample Analysis

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Introduction - Los Alamos National Laboratory

- Over 70 years of actinide nuclear science

- Mission Statement
  To solve national security challenges through scientific excellence.

- 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.
Continuing Mission - Operational Requirements

- Nuclear Facilities
  - safe handling of actinide materials
  - actinide material processing capabilities

- Analytical Chemistry
  - actinide analysis capabilities
  - quality assurance program

- Quality Assurance Program
  - actinide product certification standard
Continuing Mission – Capability Requirements

- alpha spectrometry
- colorimetry
- controlled potential coulometry
- gamma spectrometry
- gravimetry
- combustion - infrared spectroscopy
- ion chromatography
- neutron counting
- thermal ionization mass spectrometry
- titrimetry
- icp - mass spectrometry
- icp - atomic emission spectroscopy
- x-ray fluorescence

Capability Application – Actinide Product Certification
Introduction - Los Alamos National Laboratory

Continuing Mission – Analytical Chemistry Samples

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

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

- **Neutron Source**
  - PuBe

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

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

- **Uranium Fluoride**
  - tetrafluoride
  - hexafluoride

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Introduction – Analytical Chemistry

Continuing Mission – Quality Assurance Standards

**QC-1**
Implements quality assurance requirements for domestic nuclear industry contractors performing work in relevant product life-cycles. Institutional quality requirements are implemented through Manufacturing Administrative Procedures.

**NQA-1**
Implements quality assurance requirements for nuclear facility applications including design and operation.
Nuclear Forensics - Analytical Chemistry

Nuclear Forensics Mission - Operational Requirements

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Nuclear Forensics - Analytical Chemistry

Nuclear Forensics Mission – Capability Requirements

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- x-ray fluorescence

- density
- particle size analysis
- optical microscopy
- secondary electron microscopy
- x-ray diffractometry
- x-ray radiography

Capability Application – Actinide Sample Analysis
Nuclear Forensics - Analytical Chemistry

- Nuclear Forensics Mission – Quality Assurance Standard

ISO/IEC 17025:2005

General Requirements for the Competence of Testing and Calibration Laboratories

ISO/IEC 17025:2005 specifies the general requirements for the competence to carry out tests and/or calibrations, including sampling. It covers testing and calibration performed using standard methods, non-standard methods, and laboratory-developed methods.

It is applicable to all organizations performing tests and/or calibrations.
New Mission – Quality Assurance Requirements

ISO-17025 Procedures

- NF-QA-001, *Nuclear Forensics Analysis Results Review, Approval and Release*
- NF-QA-003, *Calculation of Uncertainties*
- NF-QA-006, *Root Cause Analysis*
- NF-QA-007, *Management Review*
- NF-QA-008, *Review of Requests, Tenders, and Contracts*
- NF-QA-009, *Nuclear Forensics Case Notebook*
New Mission – Quality Assurance Standard

ISO-17025 Customer Interaction

NF-QA-008, Review of Requests, Tenders, and Contracts

Name of Customer: __________________________ Date of Request/Tender/Contract: ____________

Written or Oral? ________________ Reference Number: _________________

Are the customer requirements adequately defined, documented, and understood?
Can listed customer requirements be addressed by existing capabilities and resources?
Do personnel/equipment have the skills and expertise necessary to perform the tests?
Have appropriate test methods been selected to meet the customer requirements?

If any of the above are not met, consult with the customer to resolve, or if not possible, turn down the request, tender, or contract.

List any issues/concerns which need to be addressed prior to acceptance:
New Mission – Quality Assurance Requirements

ISO-17025 Quality Testing

- NF-PLAN-006 LANL Actinide Analytical Chemistry Proficiency Testing (PT) Plan

The laboratory policy is to run proficiency test samples (PTs) where applicable and when PTs are available. Applicability is based on the analyte/nuclide, available matrix, and concentration range of the PT.

When a PT is not available, appropriate quality control standards will be analyzed to ensure the accuracy and precision for quantitative methods. For the qualitative identification, other appropriate schemes within the laboratory test procedures will be implemented.
New Mission – Quality Assurance Requirements

ISO-17025 Quality Testing

- NF-PLAN-006 LANL Actinide Analytical Chemistry Proficiency Testing (PT) Plan

LANL analytical chemistry has one accreditation discipline, chemistry, with two sub disciplines: general or wet chemistry and spectroscopy. Note there are no ISO/IEC 17043 accredited domestic (U.S.) PT Programs for the bulk nuclear fuel or nuclear material programs.
New Mission – Quality Assurance Requirements

ISO-17025 Quality Testing

- Applicable Proficiency Test Programs Would Require...

1. Appropriate nuclear (Pu and U) materials for distribution
   - Certified Reference Materials
   - Working Reference Materials
   - Viable Control Materials

2. Accurate and precise measured reference values for materials

3. GUM compliant reference value uncertainties

4. Accredited Proficiency Test program administrator

5. Infrastructure and skill to package and ship bulk quantities of nuclear materials
Nuclear Forensics - Analytical Chemistry

- New Mission – Quality Assurance Requirements

ISO-17025 Quality Testing Options

1. Proficiency Test Programs
2. Round-Robin Nuclear Material Exchanges
3. Measurement Quality Control Samples
New Mission – Quality Assurance
Round-Robin Uranium Material Exchange
- Safeguards Measurement Evaluation (SME) Program - NBL
New Mission – Quality Assurance
Round-Robin Uranium Material Exchange

- Safeguards Measurement Evaluation (SME) Program - NBL
Nuclear Forensics – Reference Materials

- New Mission – Quality Assurance
- Round-Robin Plutonium Material Exchange
  - Plutonium Standards Metal Exchange Program - LANL
Nuclear Forensics – Reference Materials

- New Mission – Quality Assurance
  Round-Robin Plutonium Material Exchange
  - Plutonium Standards Metal Exchange Program - LANL

Validate Method and Trend Detection

![Graph showing average Pu concentration over time]

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Nuclear Forensics – Reference Materials

- New Mission – Quality Assurance Measurement Quality Control Sample
  - Plutonium Standards Metal Exchange Program - LANL

**Certified Atom Ratio**

\[ 0.062711 \pm 0.000016^* \]

* Decay corrected

**LANL Atom Ratio**

\[ 0.062719 \text{ (%RD = -0.013)} \]
New Mission – Quality Assurance Requirements

ISO-17025 Uncertainties

- NF-QA-003, *Calculation of Uncertainties*

There are various methods of calculating uncertainties, any of which may be used. If requested by customer, the uncertainty calculation may be performed through the use of **GUM** compliant methods.

**GUM**
## New Mission – Quality Assurance Requirements

### ISO-17025 Uncertainties

- **NF-QA-003, Calculation of Uncertainties**

<table>
<thead>
<tr>
<th>Measurement</th>
<th>Result</th>
<th>n</th>
<th>Replicate Standard Dev. (n=12)</th>
<th>Method Uncertainty (k=2)</th>
<th>GUM Uncertainty (k=2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu Assay</td>
<td>87.60 wt%</td>
<td>12</td>
<td>0.07 wt%</td>
<td>0.08 wt%</td>
<td>0.08 wt%</td>
</tr>
<tr>
<td>$^{238}$Pu content</td>
<td>0.0120 at%</td>
<td>12</td>
<td>0.0003 at%</td>
<td>0.0004 at%</td>
<td>0.0009 at%</td>
</tr>
<tr>
<td>$^{239}$Pu content</td>
<td>93.9645 at%</td>
<td>12</td>
<td>0.0008 at%</td>
<td>0.0007 at%</td>
<td>0.0021 at%</td>
</tr>
</tbody>
</table>
Nuclear Forensics – Reference Materials

- New Mission – Quality Assurance

New Reference Material Production – Pu Oxide via Recycle

Nitric Acid Process

- Batch dissolution: Impure oxides, MoO3, CaO, As, and other salts
- Feed treatment: OXCI, CI, CO3, and other anions
- Anion exchange: Multiple configuration paired column
- Oxalate precipitation: OXCI effluent
- Evaporation: Concentrated evaporator bottoms
- Calcination: Oxide (PuO2)
- Fixation in portland cement: Low nitrate and alpha to liquid waste treatment facility

LA-UR-14-24844
New Mission – Quality Assurance

New Reference Material Production – Pu Oxide to RM

Nuclear Material Production • Production Records • Packaging • Shipping (Send/Receive) • Stabilization • Homogenization • Blending • Material Analysis
## Nuclear Forensics – Reference Materials

### New Mission – Quality Assurance

New Reference Material Production – Sample Analysis

<table>
<thead>
<tr>
<th></th>
<th>CRM</th>
<th>WRM</th>
</tr>
</thead>
<tbody>
<tr>
<td>Form</td>
<td>Pu oxide</td>
<td>Pu oxide</td>
</tr>
<tr>
<td>Sample Size</td>
<td>160-200 mg</td>
<td>160-200 mg</td>
</tr>
<tr>
<td>Attributes</td>
<td>Pu Isotopics (238-244)</td>
<td>Pu Isotopics (238-244)</td>
</tr>
<tr>
<td>Pu assay</td>
<td>Pu assay</td>
<td>Pu assay</td>
</tr>
<tr>
<td>U assay and isotopics</td>
<td>U assay and isotopics</td>
<td>U assay and isotopics</td>
</tr>
<tr>
<td>Am assay (isotopics if appropriate)</td>
<td>Am assay (isotopics if appropriate)</td>
<td>Am assay (isotopics if appropriate)</td>
</tr>
<tr>
<td>Np assay</td>
<td>Np assay</td>
<td>Np assay</td>
</tr>
</tbody>
</table>
Conclusions

- Los Alamos National Laboratory operates capable analytical chemistry and material science laboratories suitable for nuclear material forensic measurements.

- LANL analytical chemistry has numerous ISO 17025 accredited measurement processes to support nuclear forensic customers.

- LANL analytical chemistry uses numerous means to validate and independently verify that ISO17025 measurement data quality objectives are met.
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