Deconstructing Nuclear Materials


May 2, 2018
Outline

• Capabilities
• Developmental work
• Recent studies
Introduction – Los Alamos National Laboratory

• Over 70 years of actinide nuclear science

• Mission Statement
    Delivering science and technology to protect our nation and promote world stability

• 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

**LANL Analytical Techniques**

Pu, U – Assay/ Fe Spectrophotometry

Thermal Ionization Mass Spectrometry

Radio Chemistry – Np, Am, Pu

X-ray Fluorescence – Ga, U, (Fe)

Cold Vapor Atomic Fluorescence

LECO interstitial gas analyzer

ICP-Mass Spectrometry

ICP-Atomic Emission Spectrometry

Ion Chromatography

Gas Mass Spectrometry
Morphological Analysis Capabilities

- Optical microscopes
- Electron microscopes
  - SEM-EDS
  - TEM (in process)
  - Microprobe, Auger and XPS
- Analytical quantitative software development
## MAMA Analysis

<table>
<thead>
<tr>
<th></th>
<th>Count</th>
<th>Pixel Area</th>
<th>ECD</th>
<th>Ellipse Perimeter</th>
<th>Diameter Aspect Ratio</th>
<th>Circularity</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Known</strong></td>
<td>740</td>
<td>39.34 (7.10)</td>
<td>4.41 (0.20)</td>
<td>15.93 (0.79)</td>
<td>1.75 (0.02)</td>
<td>0.59 (0.01)</td>
</tr>
<tr>
<td><strong>Unknown</strong></td>
<td>339</td>
<td>251.28 (9.26)</td>
<td>17.04 (0.30)</td>
<td>57.52 (1.02)</td>
<td>1.38 (0.01)</td>
<td>0.55 (0.00)</td>
</tr>
</tbody>
</table>

Detection of Nuclear Material Morphology by Computer Vision

Compare morphology by creating multidimensional vectors that represent key points in the image
1. Pick Sample Image

2. Scan Database

3. Images are ordered by percent match

4. Morphology and Sample history are predicted based on highest correlated database images
Study I – Plutonium Fluoride Powder

• Pu fluoride samples
• Aged, heterogeneous material
• Used for bomb reduction to produce metal
Study I – Gamma Spectra

• $F(\alpha, n)$ and $F(\alpha, p)$ reaction signatures are clearly visible in gamma spectrum
• $^{22}\text{Na}$ measured from the sample
  • $^{19}\text{F}(\alpha, n)^{22}\text{Na}$
• Using 414 keV Pu photopeak and 583 keV $F(\alpha, n)$ Pu assay can be performed only using gamma-ray spectrum data
Study: Uranium Target Analysis
- Background on samples

• Records indicate that the samples are ~ 30 years old uranium targets that were part of irradiation experiments in a water cooled, light water moderated research reactor.

• The targets were irradiated for 5–85 h at thermal fluxes of ~ $1.0 \times 10^{14}$ n cm$^{-2}$ s$^{-1}$.

• Natural uranium metal and natural uranium oxides were used in the irradiation experiments.
Developing diagnostics for very-low burnup

- Some basic reactor physics:

  - Neutron exposure gives information about Pu production
  - Cooling time dates the capabilities
  - Depletion analysis uses reaction network to model the buildup of isotopes in a reactor environment

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-Life</th>
<th>Fractional Abundance</th>
<th>Decay Mode</th>
</tr>
</thead>
<tbody>
<tr>
<td>238Am</td>
<td>98 M</td>
<td>e: 100.00% α: 1.0E-4%</td>
<td></td>
</tr>
<tr>
<td>239Am</td>
<td>11.9 H</td>
<td>e: 99.99% α: 0.01%</td>
<td></td>
</tr>
<tr>
<td>240Am</td>
<td>50.8 H</td>
<td>e: 100.00% α: 1.9E-4%</td>
<td></td>
</tr>
<tr>
<td>241Am</td>
<td>432.6 Y</td>
<td>e: 100.00% α: 4E-10%</td>
<td></td>
</tr>
<tr>
<td>242Am</td>
<td>16.02 H</td>
<td>β^-: 82.70% e: 17.30%</td>
<td></td>
</tr>
<tr>
<td>237Pu</td>
<td>45.64 D</td>
<td>e: 100.00% α: 4.2E-3%</td>
<td></td>
</tr>
<tr>
<td>238Pu</td>
<td>87.7 Y</td>
<td>α: 100.00% SF: 1.9E-7%</td>
<td></td>
</tr>
<tr>
<td>239Pu</td>
<td>24110 Y</td>
<td>α: 100.00% SF: 3.3E-10%</td>
<td></td>
</tr>
<tr>
<td>240Pu</td>
<td>6561 Y</td>
<td>α: 100.00% SF: 5.7E-6%</td>
<td></td>
</tr>
<tr>
<td>241Pu</td>
<td>14.329 Y</td>
<td>β^-: 100.00% α: 2.5E-3%</td>
<td></td>
</tr>
<tr>
<td>236Np</td>
<td>153E+3 Y</td>
<td>e: 86.30% β^-: 13.50%</td>
<td></td>
</tr>
<tr>
<td>237Np</td>
<td>2.144E+6 Y</td>
<td>α: 100.00% SF: 2E-10%</td>
<td></td>
</tr>
<tr>
<td>238Np</td>
<td>2.117 D</td>
<td>β^-: 100.00%</td>
<td></td>
</tr>
<tr>
<td>239Np</td>
<td>2.356 D</td>
<td>β^-: 100.00%</td>
<td></td>
</tr>
<tr>
<td>240Np</td>
<td>61.9 M</td>
<td>β^-: 100.00%</td>
<td></td>
</tr>
<tr>
<td>235U</td>
<td>7.04E+3 Y</td>
<td>e: 0.7204% α: 100.00% SF: 7.0E-9%</td>
<td></td>
</tr>
<tr>
<td>236U</td>
<td>2.342E7 Y</td>
<td>α: 100.00% SF: 9.4E-8%</td>
<td></td>
</tr>
<tr>
<td>237U</td>
<td>6.75 D</td>
<td>β^-: 100.00%</td>
<td></td>
</tr>
<tr>
<td>238U</td>
<td>4.468E9 Y</td>
<td>99.9242% α: 100.00% SF: 5.4E-5%</td>
<td></td>
</tr>
<tr>
<td>239U</td>
<td>23.45 M</td>
<td>β^-: 100.00%</td>
<td></td>
</tr>
</tbody>
</table>

Primary fuel sources: LEU (~3%), nat U (0.71%)
Developing diagnostics for very-low burnup

- **Objective**: determine the total neutron exposure and the age of reactor samples
  - Neutron exposure $\Phi_n$ relates to burnup
  - Cooling time of sample helps date the capabilities

- **Issues**: Typical methods are not ideal in very-low burnup
  - Not enough $^{241}$Pu/$^{241}$Am produced for age estimate
  - Not enough $^{134}$Cs or $^{154}$Eu produced for measurement
  - Graphite Isotope Ratio Method (GIRM) too invasive
  - Additional problems if the samples are very old!
Uranium Target Analysis

Chemical Separation: Flow Chart

1. Load sample in 2 mL 7M HNO₃
2. UTEVA 2mL pre-packed
   - Elute FPs 4 mL 7M HNO₃
   - Elute U 15 mL 0.02M HNO₃
3. ½ to Sr-Spec for Cs-Ba separation
4. ½ to ICP-MS for Ru isotope analysis
5. Load sample in 7 mL 3M HNO₃
6. Sr-Spec 2mL pre-packed
   - Elute Cs 2 mL 3M HNO₃
   - To ICP-MS for Cs Isotope analysis
Chemical separation

• Start with U-metal and UO$_3$ archived samples

- Dissolve in 7M HNO$_3$
- Separate Pu from U with anion-exchange columns
- Pu/U isotopics via TIMS

Original Sample → UTEVA (2mL) → Actinide Analysis

Gamma-ray spec (HPGe) → Analyze peak data with SNAP/FRAM → Product Analysis

B. Byerly et al., J. Radioanal Nucl Chem 307 (2016)
Applying the diagnostics

- Start with U-metal and UO$_3$ archived samples
  - Separate Pu and U then TIMS for isotopics
- Uranium isotopics indicate nat U for $\varepsilon_0$

PJ et al., PRApplied 8 044025 (2017)
Applying the diagnostics

- Gamma spectrometry to identify fission products
  - Measured $^{85}\text{Kr}$, $^{125}\text{Sb}$, $^{137}\text{Cs}$, $^{152}\text{Eu}$, $^{154}\text{Eu}$, $^{155}\text{Eu}$
  - Perform diagnostics on linear fission products

- Multiple $T_C$ diagnostics required!
- $^{137}\text{Cs}/^{125}\text{Sb}$ matches declared $T_C$
Conclusions

• Extensive sample analysis capability based on over 70 years experience in actinide material characterization
• Large investment in new capabilities for analytical chemistry and material science
• Robust research and development program for material characterization and deconstruction
• Ability to use protocols to address diverse actinide samples
Acknowledgements

• U.S. Department of Energy, National Nuclear Security Administration (DOE NNSA)
• U.S. Department of Homeland Security, Domestic Nuclear Detection Office National Technical Nuclear Forensics Center (DHS DNDO NTNFC)
• LANL Laboratory Directed Research and Development Exploratory Research (LDRD ER)
Developing diagnostics for very-low burnup

- Procedure: use Bateman equations to develop simplified depletion calculations

\[
\frac{dN_{B,2}}{dt} = \vec{Y}_{B,2} \cdot \vec{F}
\]

\[\vec{F} = \{F_{U235}, F_{U238}, F_{Pu239}, F_{Pu241}\}\]

Production via fission

Production/Depletion via β-decay

\[
\lambda = \frac{\ln(2)}{t_{1/2}}
\]

Production/Depletion via n-capture

\[
\phi_n \sigma_{A,2} N_{A,2} - \phi_n \sigma_{B,2} N_{B,2}
\]

- Need decay constants (\(\lambda\)), cross-sections (\(\sigma\)), the fission rate (\(F\)), the fission yields (\(Y\)), and neutron flux (\(\phi_n\))
Developing diagnostics for very-low burnup

- Example: Neutron exposure $\Phi_n$ from Uranium ratio

<table>
<thead>
<tr>
<th>$^{235}\text{U}$</th>
<th>$^{236}\text{U}$</th>
<th>$^{237}\text{U}$</th>
<th>$^{238}\text{U}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\lambda = 1\text{e-9s}^{-1}$</td>
<td></td>
<td></td>
<td>$\lambda = 2\text{e-10s}^{-1}$</td>
</tr>
<tr>
<td>$\sigma_n = 99\text{b}$</td>
<td>$N = 0$</td>
<td>$N = 0$</td>
<td>$\sigma_n = 2.7\text{b}$</td>
</tr>
<tr>
<td>$\sigma_f = 585\text{b}$</td>
<td></td>
<td></td>
<td>$\sigma_f = 2\text{e-5b}$</td>
</tr>
</tbody>
</table>

$^{235}\text{U}$ not produced in fission

$$\frac{dN_{U235}}{dt} = \vec{f}_{U235} \cdot \vec{F}$$

No $^{235}\text{Pa}$ and $\lambda_{U235} \sim 0$

$$+ \lambda_{Pa235} N_{Pa235} - \lambda_{U235} N_{U235}$$

No $^{234}\text{U}$

$$+ \phi_n \sigma_{U234} N_{U234} - \phi_n \sigma_{U235} N_{U235}$$

- $^{235}\text{U}$
  - $\sigma_f >> \sigma_n$

$$N_{U235}(t) = N_{U235}^0 e^{-\sigma_{U235} \Phi_n t}$$

- $^{238}\text{U}$
  - $\sigma_n >> \sigma_f$

$$N_{U238}(t) = N_{U238}^0 e^{-\sigma_{U238} \Phi_n t}$$

$$\varepsilon = \varepsilon_0 e^{-\Phi_n (\sigma_{U235}^T - \sigma_{U238}^T)}$$

$\Phi_n = \phi_n t$
Developing diagnostics for very-low burnup

- **Neutron Exposure: Uranium ratios**

\[
\Phi_n = \frac{\ln(\varepsilon_0 / \varepsilon)}{\sigma_{U235}^T - \sigma_{U238}^T}
\]

\[\varepsilon = \frac{^{235}U}{^{238}U}\]

\[
\Phi_n = \frac{1}{\sigma_{U235}^T - \sigma_{U236}^T} \ln \left( \frac{\sigma_{U235}^C - \rho(\sigma_{U236}^T - \sigma_{U235}^T)}{\sigma_{U235}^C} \right)
\]

\[\rho = \frac{^{236}U}{^{235}U}\]

- **\( ^{235}U/^{238}U \):**
  - Relies on knowing initial enrichment \( X \)
  - More accurate as concentration is higher \( \checkmark \)

- **\( ^{236}U/^{235}U \):**
  - Trouble when \( ^{236}U \) is very low \( X \)
  - Independent of initial enrichment \( \checkmark \)
Developing diagnostics for very-low burnup

- **Cooling Time**: Ratios of linear fission products
  - Linear in $\Phi_n$
  - Long half-life w.r.t. $T_{irr}$ and $T_C$
  - Short precursor half-lives

- $Z_{Cs137} = Y_{Cs137} + Y_{Xe137} + Y_{I137}$

\[
T_C = \frac{1}{\lambda_2 - \lambda_1} \ln \left( \frac{\alpha_{1/2} \lambda_2 \tilde{Z}_2 \langle \Sigma_{fiss} \rangle_\Phi}{\lambda_1 \tilde{Z}_1 \langle \Sigma_{fiss} \rangle_\Phi} \right)
\]

- $\tilde{Z} = \{Z_{U235}, Z_{U238}, Z_{Pu239}, Z_{Pu241}\}$
- $\langle \Sigma_{fiss} \rangle_\Phi$
- Cumulative yields

- Activity ratio of products 1 & 2

\[\alpha_{1/2} = \frac{\lambda_1 N_1}{\lambda_2 N_2}\]

- Flux-weighted macroscopic fission cross-section