Nuclear Forensics & Bulk Special Nuclear Material (SNM) Characterization

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Analysis of Major and Minor Constituents

- **Major – accurate and precise assay of plutonium (Pu), uranium (U), other actinides with traceability to national standards.**
  - Certify the purity of various grades of metals; advanced actinide fuels; oxides; nitrides; carbides; solutions; ^{238}Pu, nuclear or residue, and nuclear forensics samples

— **Minor** – from percent to part-per-thousand level

  - Ability to measure minor components critical to material performance, corrections for interferences and mass balance.

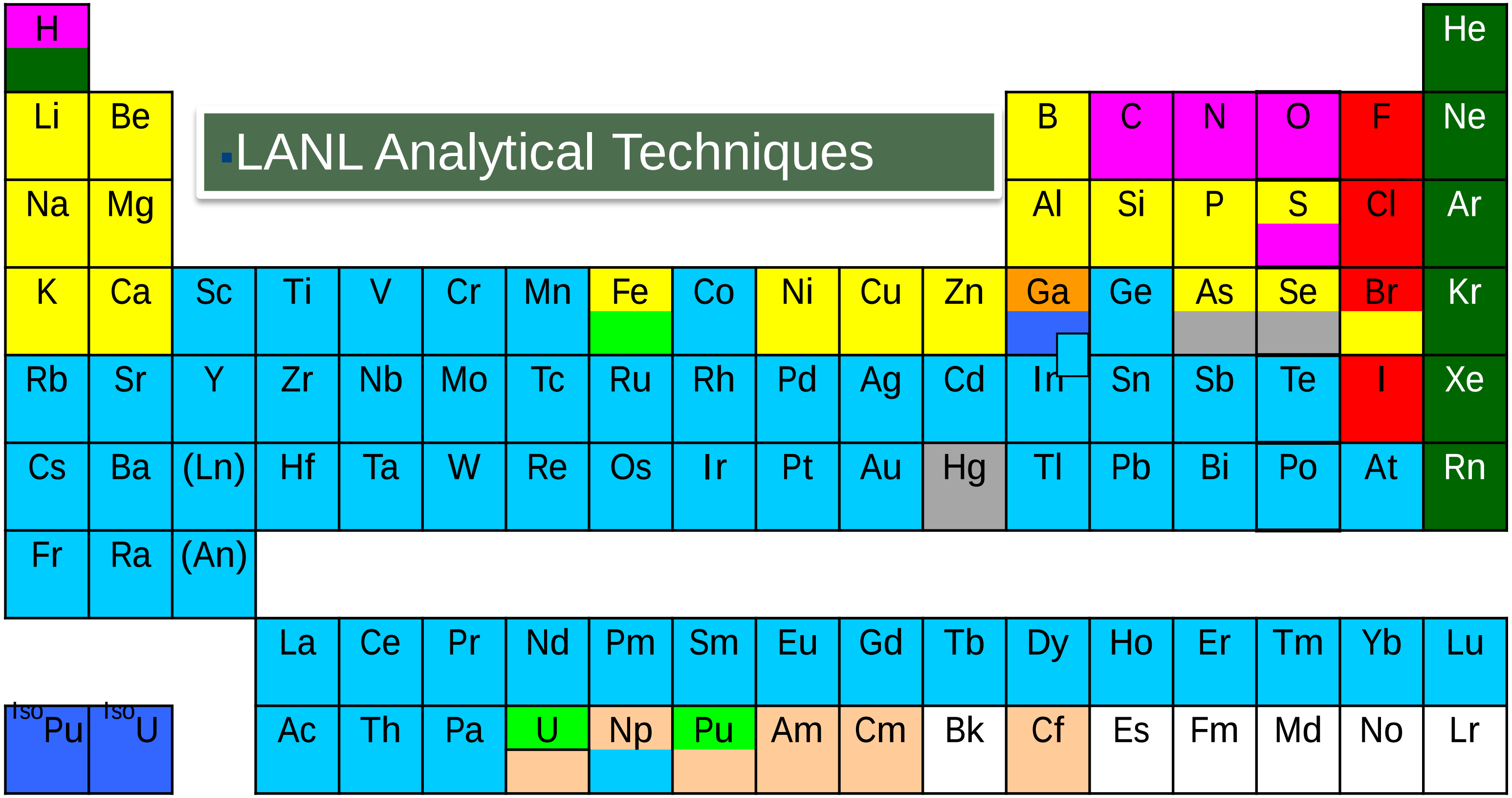
▪Both assay and minor component analyses provide measurements used in defense, nonproliferation, nuclear accountancy/safeguards, counter-proliferation, nuclear materials technologies, basic science and for material control and accountability (MC&A)

Analysis of Trace Impurities

- **Beyond measuring major and minor components, measurement of trace (part-per-thousand to part-per-billion) impurities is a significant capability.**
- **Maintaining these capabilities requires methods which can measure virtually any element from the Periodic Chart within an actinide matrix.**
- **Techniques include**
  - ICP-MS
  - ICP-AES
  - Isotope Dilution Mass Spectrometry
  - Spectrophotometry
  - Radiochemistry
  - Non-metal analyses

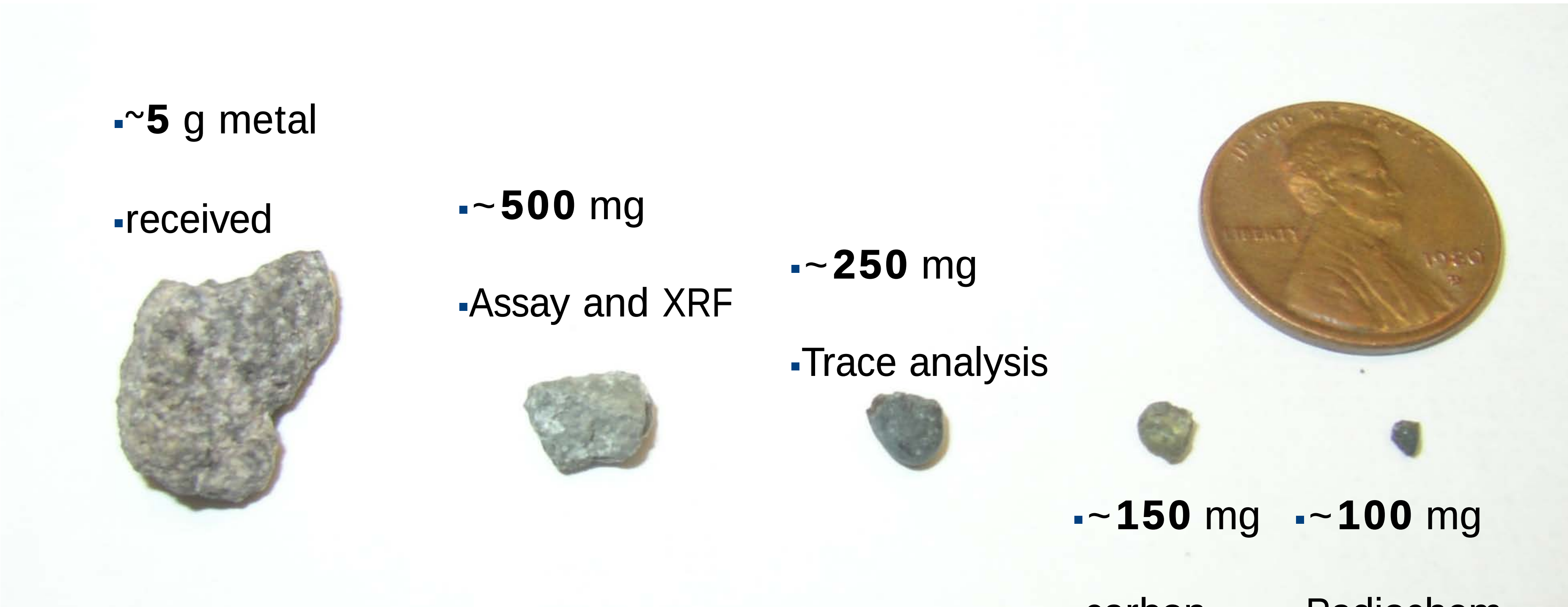
Ability to analyze a large suite of elements with concentrations varying over several orders of magnitude (from % to ppb)

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**

Typical Sample Size for Analysis



- ~**5** g metal
- received
- ~**500** mg
- Assay and XRF
- ~**250** mg
- Trace analysis
- ~**150** mg
- carbon
- ~**100** mg
- Radiochem
- and isotopic

- For most sample types, duplicate cuts are analyzed by each method.

Nuclear Forensic Operations

Nuclear facilities…

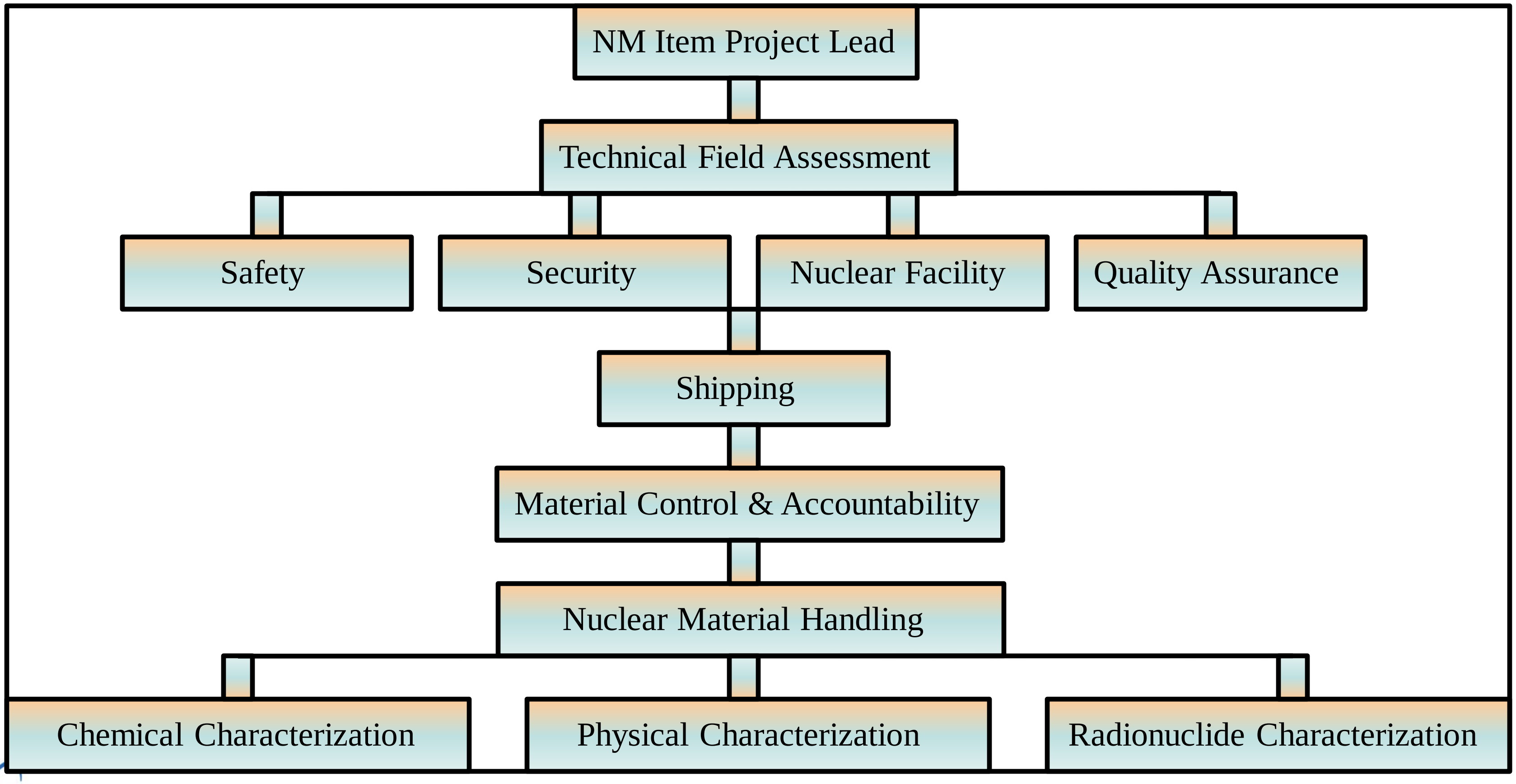
- provide an engineered infrastructure for workers to safely handle significant quantities of nuclear materials

- provide protection to the population from exposure to unsecured nuclear materials
  - inadvertent -
  - advertent – weapon of mass destruction (WMD), improvised nuclear device (IND), radiological dispersal device (RDD), radiological exposure device (RED)

- demand formalized conduct-of-operations for all routine and non-routine nuclear material handling, processing, and measurement activities

Nuclear Forensic Operations

Conduct-of-Operations Teams



Material Processing

- **Material Handling**

- Clean nuclear material enclosures
- Collect swipes of “clean” enclosures to monitor contamination levels
- Handle each material in a new glove-bag
- Collect swipes on target surfaces for ultra-trace signatures

Nuclear Forensic Operations

Chemical Characterization Capabilities

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

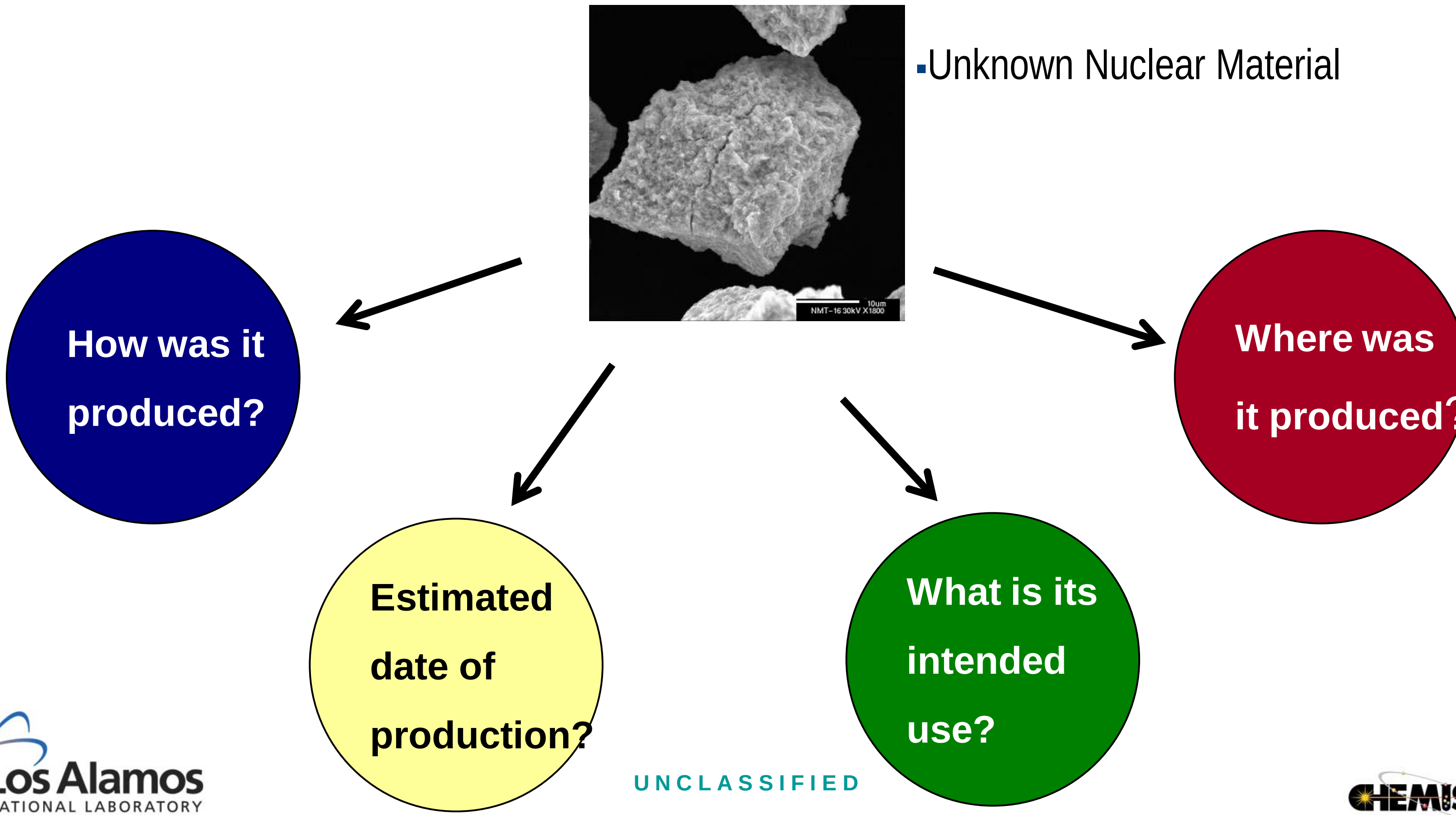
Nuclear Forensic Operations

Physical Characterization Capabilities

- gravimetry
- optical microscopy
- particle size analysis
- photography
- pycnometry
- secondary electron microscopy
- x - ray diffractometry
- x - ray radiography

Objective

- To perform forensic analysis on nuclear materials by identifying key elements for forensic investigations



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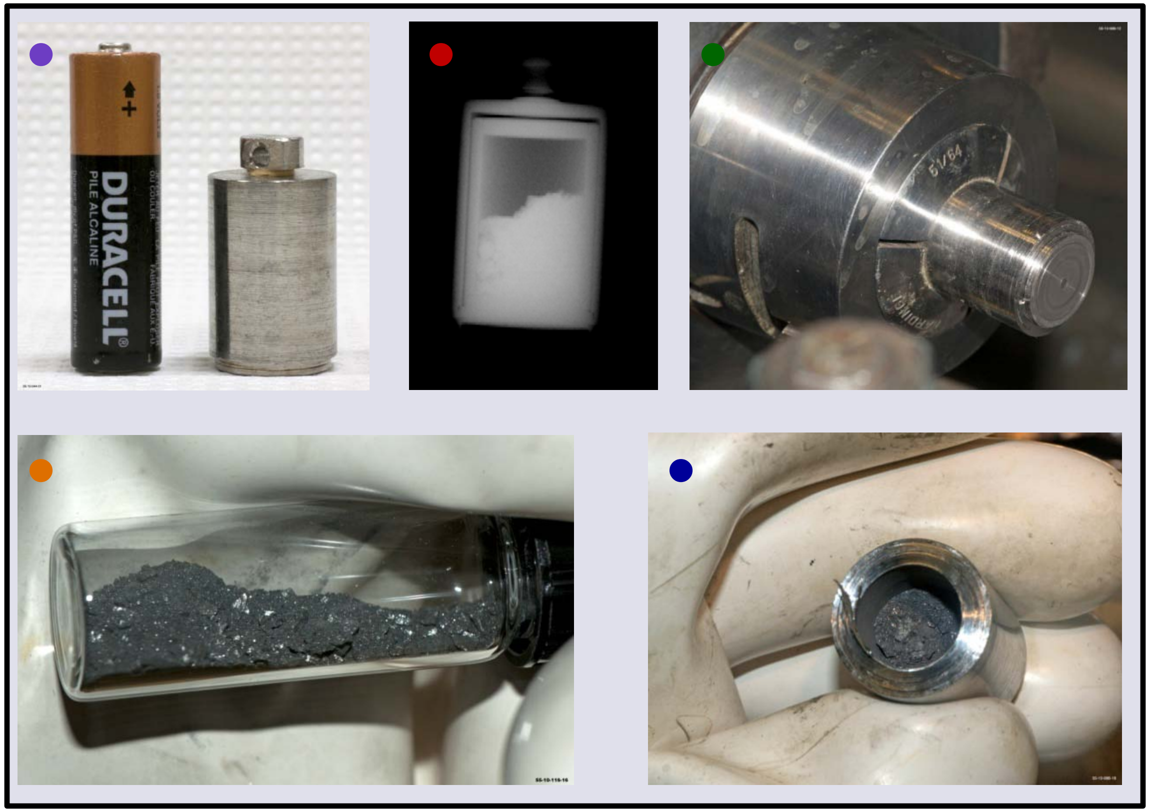
Nuclear Forensic Operations

- **What ?** measured nuclear material characteristics
- **When ?** nuclear material processing timeline
- **Why ?** intended use of nuclear material
- **How ?** processing methods to produce the nuclear material
- **Who?** infer from "How"
- **Where?** infer from "Who"

Nuclear Forensics – Study I

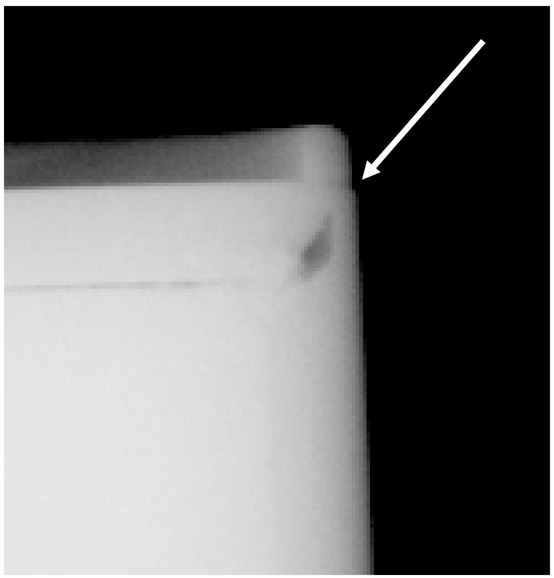
Sealed neutron source: plutonium-beryllium

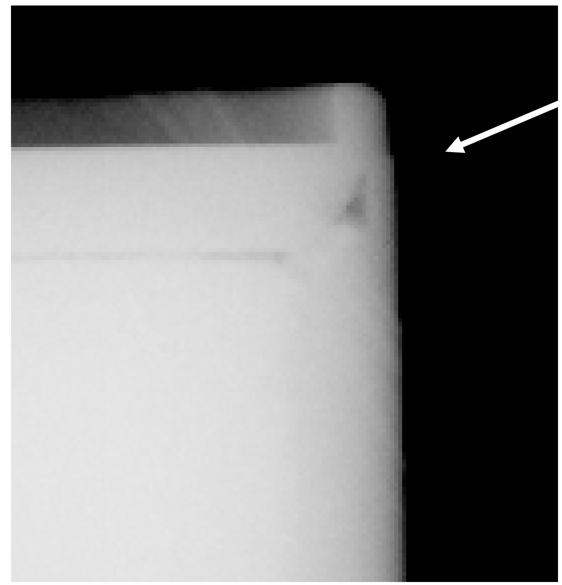
- photography
- radiography
- lathe machining
- opening
- extraction



Nuclear Forensics – Study I

- Radiographic Interpretation

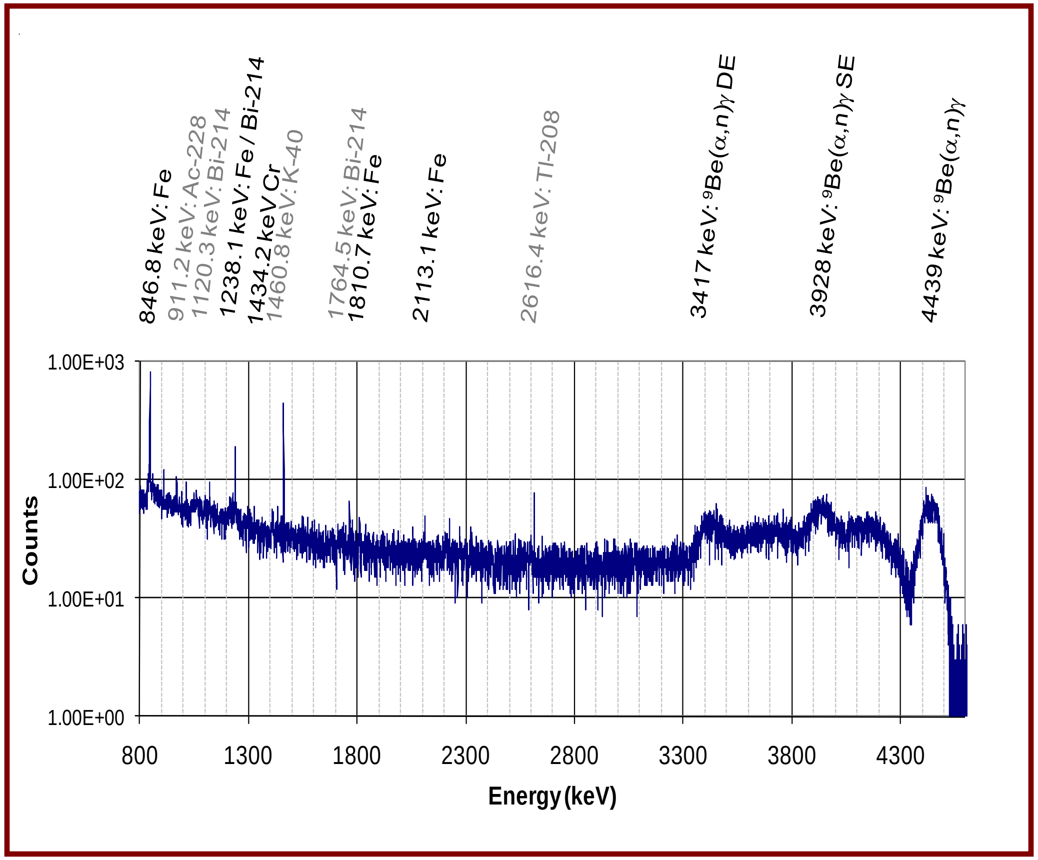




- Press fit? No evidence of weld

- possibly all powered/granules or surfaces are powdered

Source characterization



- Gamma Spectrometry on a PuBe source

Nuclear Forensics – Study I

Plutonium-beryllium metal

- **Plutonium** and **beryllium** assay results on dissolved plutonium-beryllium metal

Analytical Method	Element	Result
Controlled potential coulometry	Plutonium	(61.57 ± 0.15) wt%
ICP – atomic emission spectroscopy	Beryllium	(38.4 ± 0.8) wt%

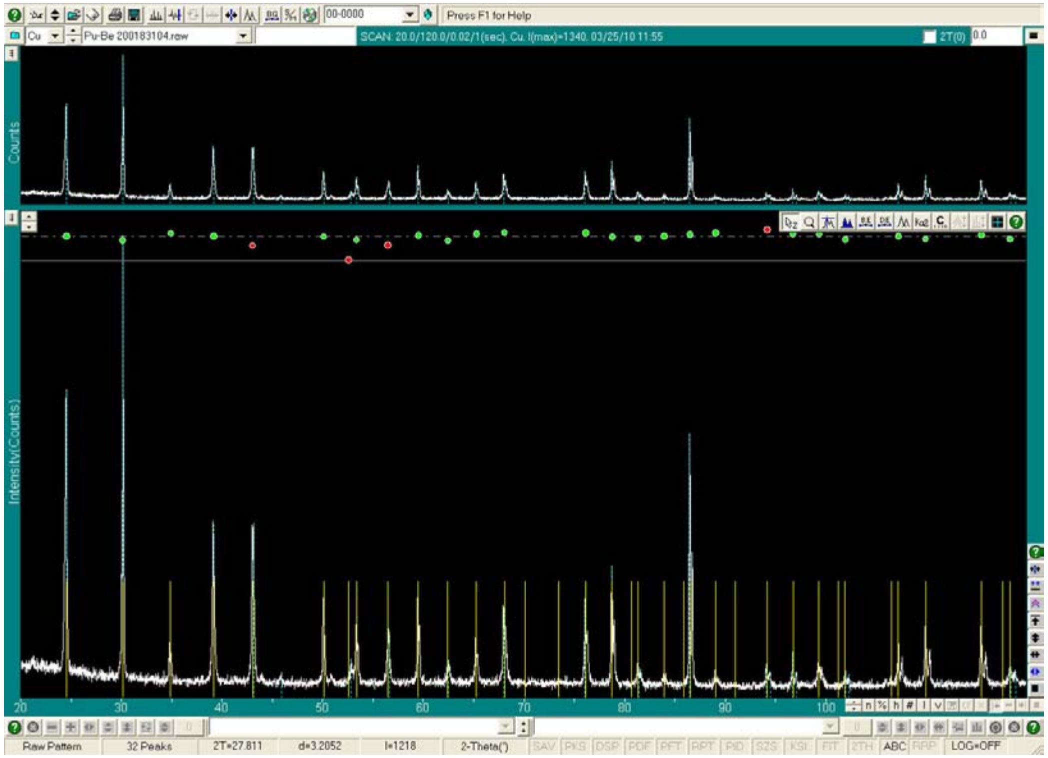
- Plutonium-beryllium **chemical form** inferred from assay measurements

Chemical Form
$PuBe_{16.6}$

Nuclear Forensics – Study I

Plutonium-beryllium metal

- Plutonium-beryllium **chemical form** from x-ray diffraction measurement



Chemical Form
$PuBe_{13}$

Nuclear Forensics – Study I

Plutonium-beryllium metal

- Measured elemental composition on dissolved plutonium-beryllium metal

Analytical Method	Element	Result
Gamma spectrometry	Americium	(1550 ± 4) μg/g
Isotope dilution mass spectrometry	Uranium	(935 ± 1) μg/g
Alpha / gamma spectrometry	Neptunium	(108 ± 5) μg/g
ICP – mass spectrometry	Chromium	(114 ± 2) μg/g
ICP – mass spectrometry	Magnesium	(33 ± 4) μg/g
All methods	∑Elements	101 wt%

Nuclear Forensics – Study I

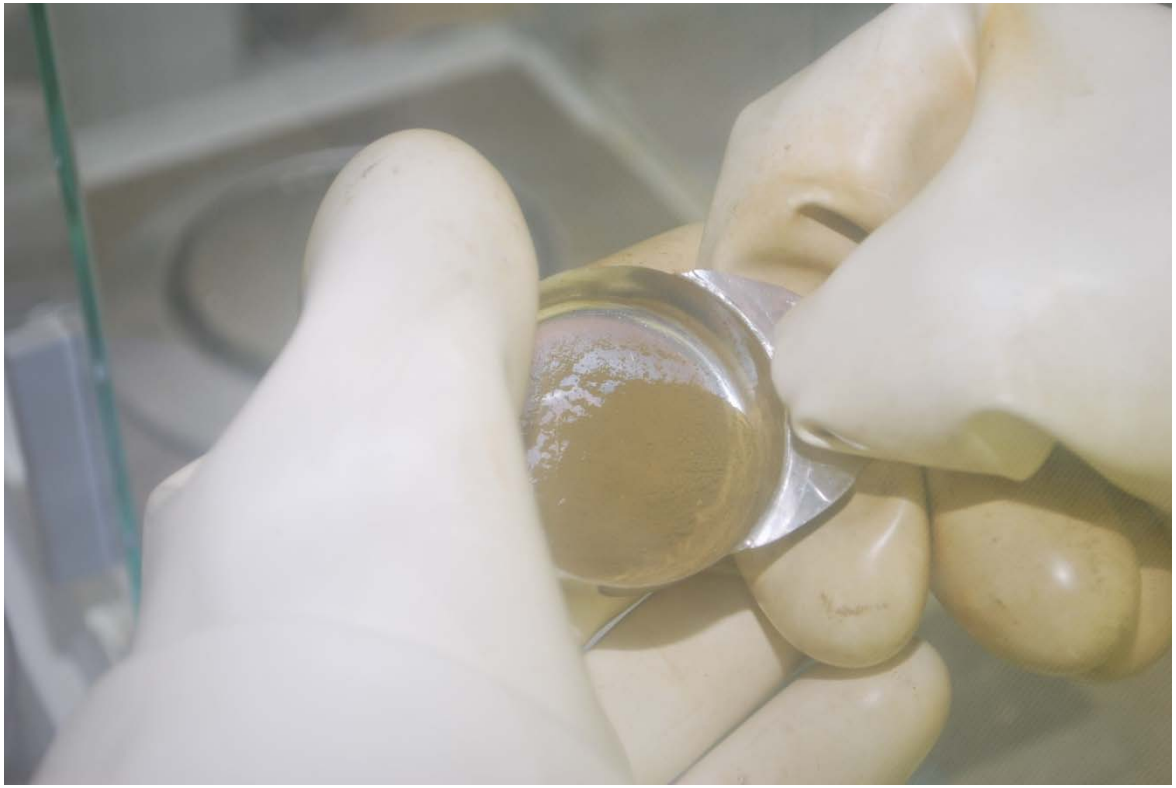
Plutonium-beryllium metal

Radionuclide Measurements			Ages
• ^{238}Pu	→ ↑ $^{4}He^{2+}$	• ^{234}U	47 years
• ^{239}Pu	→	• ^{235}U	45 years
• ^{240}Pu	→	• ^{236}U	45 years
• ^{241}Pu	→ • ^{241}Am →	• ^{237}Np	46, 46 years
• ^{242}Pu	→	• ^{238}U	---

- Radiochemistry
- Thermal ionization mass spectrometry

Nuclear Forensics – Study II

Neptunium-oxide powder



Nuclear Forensics – Study II

Neptunium-oxide powder

- **Neptunium** assay results on dissolved neptunium powder

Analytical Method	Assay Result	Replicates
Alpha Counting/Spectrometry	(88.7 ± 2.5) wt%	8
Controlled Potential Coulometry	(88.14 ± 0.40) wt%	4
Gamma Spectrometry	(87.7 ± 3.5) wt%	4

Nuclear Forensics – Study II

Neptunium-oxide powder

- Trace **plutonium** concentration result on dissolved neptunium powder

Analytical Method	Plutonium Result	Replicates
Isotope Dilution Mass Spectrometry	(44.9 ± 0.4) μg/g	4

- Trace **uranium** concentration result on dissolved neptunium powder

Analytical Method	Uranium Result	Replicates
Isotope Dilution Mass Spectrometry	(0.9 ± 0.08) μg/g	4

Nuclear Forensics – Study II

Neptunium-oxide powder

- **Plutonium isotopic** composition results on dissolved neptunium powder

Plutonium Isotopic Ratio	TIMS Ratio Result	Replicates
Pu-238 / Pu-239	(2.767 ± 0.008)	2
Pu-240 / Pu-239	(0.138 ± 0.001)	2
Pu-241 / Pu-239	(0.009 ± 0.0001)	2
Pu-242 / Pu-239	(0.105 ± 0.001)	2

Nuclear Forensics – Study II

Neptunium-oxide powder

- **Uranium isotopic** composition results on dissolved neptunium powder

Uranium Isotopic Ratio	TIMS Ratio Result	Replicates
U-233 / U-238	(0.95 ± 0.05)	2
U-234 / U-238	(0.98 ± 0.05)	2
U-235 / U-238	(1.47 ± 0.08)	2
U-236 / U-238	(0.52 ± 0.03)	2

Nuclear Forensics – Study II

Neptunium-oxide powder

- Calculated **age**-since-separation estimates for neptunium powder

Daughter / Parent Chronometers	Age Result	Replicates
U-233 / Np-237	(7.7 ± 0.3) months	4
U-234 / Pu-238	(7.9 ± 0.3) months	4

Nuclear Forensics – Study II

Neptunium-oxide powder

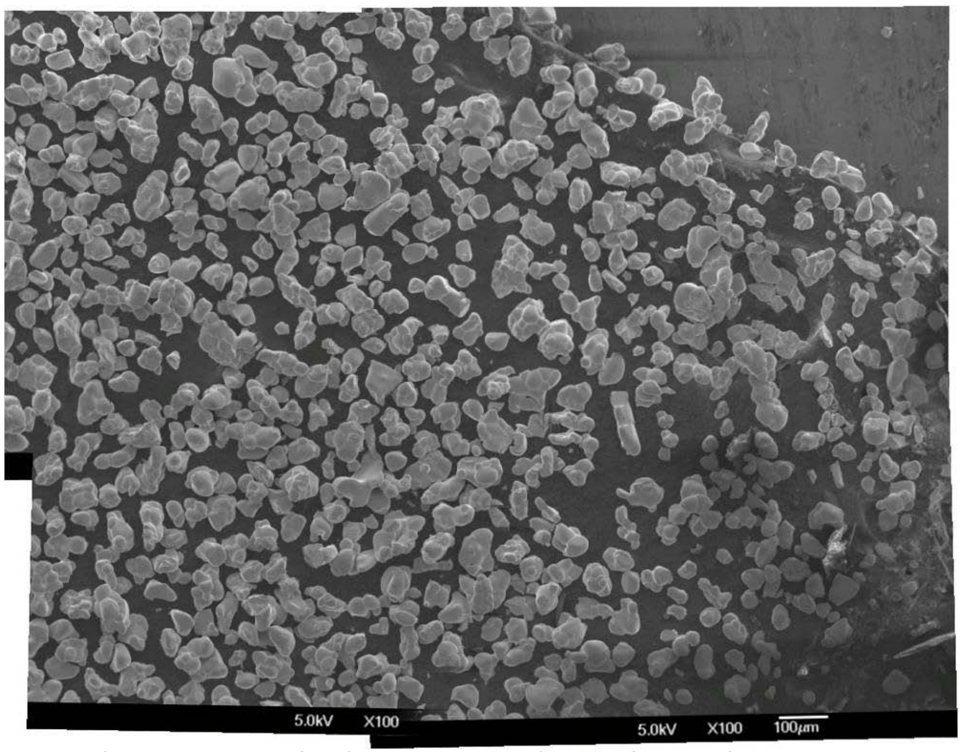
- Metal contaminants in dissolved neptunium powder by ICP-AES/MS

Metal Contaminant	Result	Replicates
Thorium	(2550 ± 510) μg/g	6
Cerium	(73 ± 15) μg/g	6
Phosphorus	(72 ± 14) μg/g	6
Chromium	(12 ± 2) μg/g	6
Tin	(3.5 ± 0.7) μg/g	6
Silver	(1.4 ± 0.3) μg/g	6
Cadmium	(0.73 ± 0.15) μg/g	6
Lanthanum	(0.51 ± 0.10) μg/g	6

Nuclear Forensics – Study III

Uranium-oxide powder

Enriched Uranium Octoxide, U_3O_8



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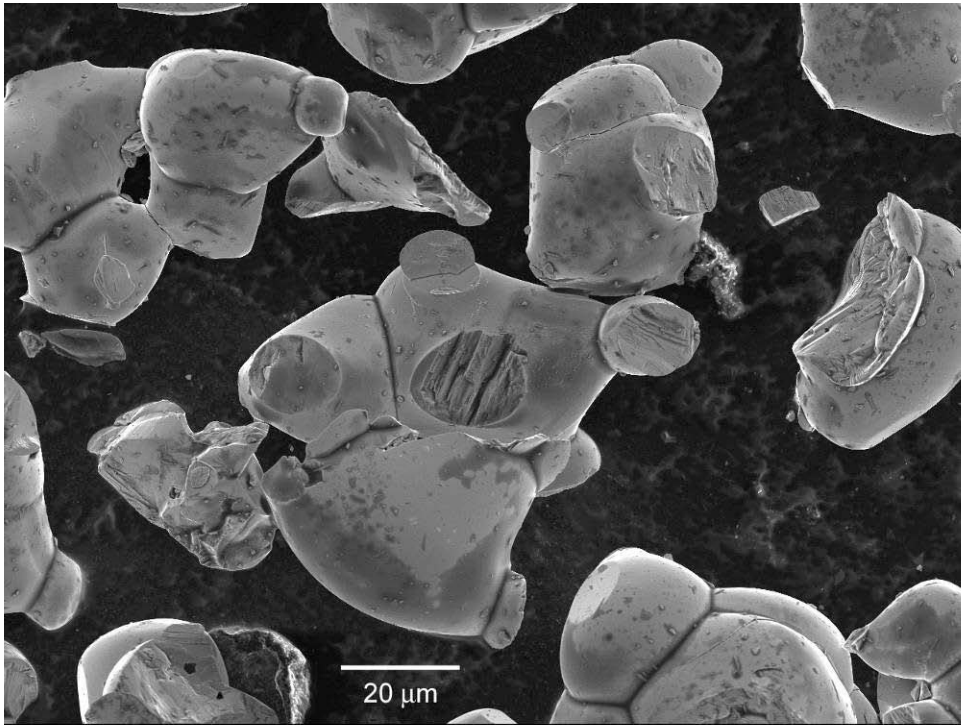
- **Coarse black powder**
- **Homogeneity**
- **Particle Shape**
- **Particle Size**
  - 41 ± 18 µm equivalent circular diameter

Enriched Uranium Octoxide, U_3O_8



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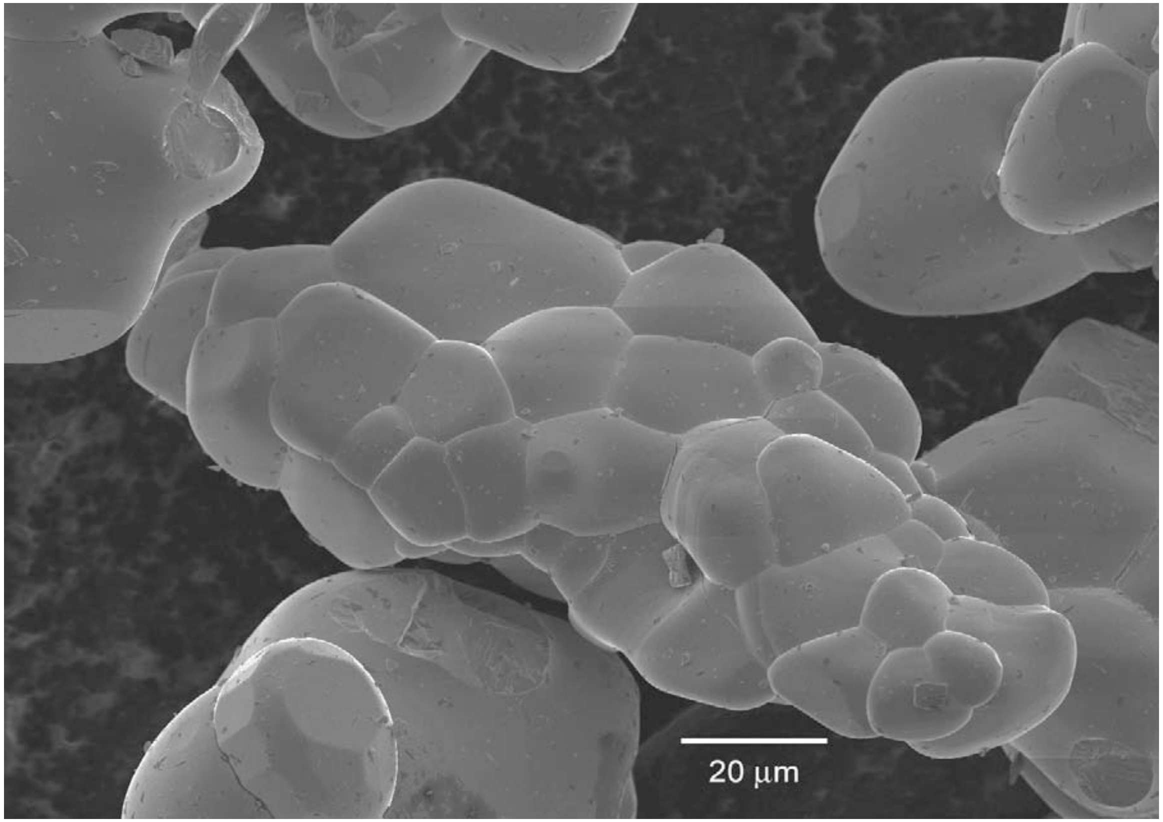
- **Particle structure common to sintered ceramics**
- **Particles composed of smaller, roughly spherical particles**
- **Smaller particles have been sintered at high temperature**
- **Large number of fractured particles suggests a crushing operation**

Nuclear Forensics – Study III

Uranium-oxide powder

- Secondary electron microscopic image of uranium powder



Enriched Uranium Octoxide, U_3O_8

- **Uranium Metal**
  - dissolution and precipitation of uranyl nitrate
- **Oxide Conversion**
  - uranyl nitrate converted to uranium octoxide
- **U_3O_8 Purification**
  - dissolved, precipitated, calcined, sintered
- **U_3O_8 Particle Sizing**
  - rod milled, sized, high fired
- **Intended Use**
  - ceramic grade U_3O_8 for fuel fabrication

Conclusions

Nuclear facilities…

- The actinide analytical chemistry group at Los Alamos National Laboratory sees opportunity to validate and refine formal procedures for the chemical and physical characterization of nuclear materials operated by functional teams identified in our forensic conduct-of-operations model.

- Periodic evaluation is crucial to maintaining operable nuclear forensic procedures that accurately address current site institutional policy, organizational structure, technical capability, and staff training.

Nuclear Forensic Team Acknowledgments

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