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## Nuclear Forensics Introduction and Laboratory Best Practices

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## Outline



- Introduction to nuclear forensics
- National nuclear forensics libraries
- Laboratory suggestions
- Nuclear forensics example case
- Conclusions
- Questions

## What is Nuclear Forensics?



- Nuclear and radiological material presents a risk if it is unsecured
  - Two tasks arise when material is determined to be out of regulatory control
    - Determination of the material's location and ensuring it remains secure
    - Identifying where the material originated to address nuclear security vulnerabilities and support enforcement of national laws that prohibit such acts
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## What is Nuclear Forensics?



- The U.S. NTNFC defines nuclear forensics as:
  - "Nuclear forensics is the thorough collection, analysis and evaluation of radiological and nuclear material in a pre-detonation state and post-detonation radiological or nuclear materials, devices and debris, as well as the immediate effects created by a nuclear detonation."<sup>1</sup>
- Nuclear forensics conclusions, along with law enforcement and intelligence information, may support the attribution process in suggesting or excluding possible sources of origin for an unknown sample

1. National Technical Nuclear Forensics Center, U.S. Department of Homeland Security, <http://www.dhs.gov/national-technical-nuclear-forensics-center>, 2013.

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# Nuclear Forensics Applications



- Law enforcement and counterterrorism
  - Combatting the illicit trafficking of nuclear materials
  - Pre- and post-detonation forensics
- Nonproliferation
  - NPT verification (IAEA safeguards)
  - CTBT verification (radionuclide component of IMS, OSIs)
  - FMCT verification (determination of reactor's lifetime Pu production)
- Arms control and disarmament
  - Verification of bilateral arms control treaties by NTMs
  - Nuclear intelligence
- Nuclear safety

## Post-Detonation Scenario Analysis



- The table shows approximate times at which different types of information may be obtained in a post-detonation nuclear forensics scenario<sup>1</sup>

Information	Time Scale	Methods
Was radioactivity involved?	An hour or less	Dosimetry at scene
Detonation was nuclear	An hour or less	Visual, seismic, radiation
Uranium or Plutonium fuel	Hours to days	Field measurements
Nuclear device design	Weeks to months	Reconstructive analysis
Age, production, history	A week to months	Iterative analysis

1. M. May and D. Barr, "Nuclear forensics role, state of the art, and program needs," Technical Report, Joint Working Group of the American Physical Society and the American Association of the Advancement of Science, Washington, DC, February 2008.

## National Nuclear Forensics Libraries

- NNFL – a domestic database composed of characteristics of all nuclear and radiological material within a state
  - If material is interdicted within a state, measurements are performed on materials and then compared to the library
  - A NNFL enables the capability to more simply determine if the material originated domestically
  - Can be shared cooperatively under bi- or multilateral agreements or using confidence building measures
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## Nuclear Forensics Laboratory

- It is uncommon for a single laboratory to be utilized for forensics purposes alone
    - Due to the infrequency of usage and cost required typically
    - A network of capabilities is more often utilized in the time of need
  - A network of laboratories can be domestic or regional
  - Very accurate and precise measurements are often required to achieve optimal analysis results
    - With minimal uncertainty
  - Measurements will be discussed in a later talk
-

## Example Scenario: Plutonium at PNNL<sup>1</sup>



- December 2004: remediation activities were being performed on a waste site at the Hanford Site, Washington State, USA
- A sealed safe was uncovered and contents examined



1. A. Luksic and J. Schwantes, "Nuclear Forensics at PNNL: A Case Study", Pacific Northwest National Laboratory, Unclassified Unlimited Release PNNL-SA-86979 and PNNL-SA-66580. (for slides 9-14)

## Scenario 1 (continued)



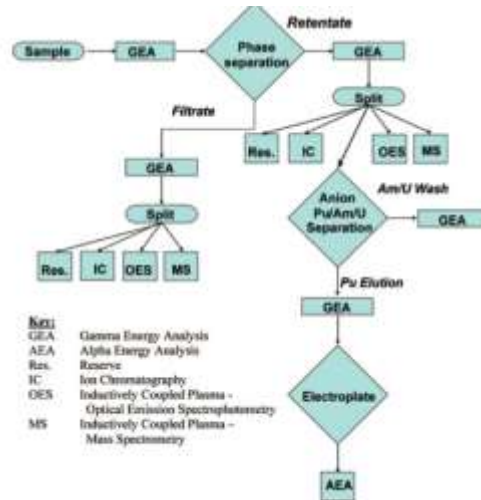
- LaF<sub>3</sub> written on bottle
- Preliminary field gamma analysis detected only <sup>239</sup>Pu
- Paperwork was limited
  - Safe sealed in 1945 after contamination
  - Disposed/buried in early 1950s as waste
- Pu content was estimated to be <1g



## Scenario 1 (continued)



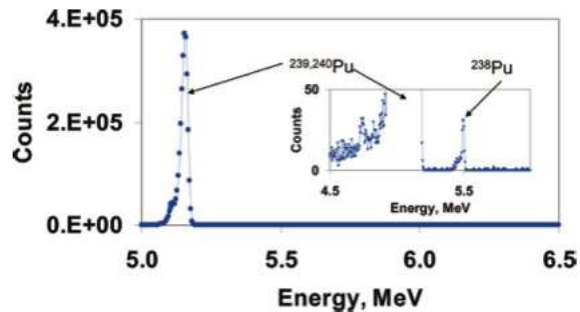
- A radiochemical analysis was performed on a sample to determine the isotopic concentrations



## Scenario 1 (continued)



- Alpha spec results



## Scenario 1 (continued)



- ICP-MS results

**Table 3. ICPMS Results of Filtrate and Retentate of the Pu Sample as of July 2007**

isotope	filtrate (mg)	2 $\sigma$ (mg)	retentate (mg)	2 $\sigma$ (mg)	total (mg)	2 $\sigma$ (mg)
[ <sup>238</sup> Pu] <sup>a</sup>	nm <sup>c</sup>	nm <sup>c</sup>	1.48 × 10 <sup>-1</sup>	2.20 × 10 <sup>-1</sup>	1.48 × 10 <sup>-1</sup>	2.20 × 10 <sup>-1</sup>
[ <sup>239</sup> Pu]	1.51 × 10 <sup>-2</sup>	2.33 × 10 <sup>-2</sup>	4.86 × 10 <sup>-2</sup>	1.72 × 10 <sup>-1</sup>	4.86 × 10 <sup>-2</sup>	1.72 × 10 <sup>-1</sup>
[ <sup>240</sup> Pu]	5.76 × 10 <sup>-2</sup>	8.00 × 10 <sup>-2</sup>	1.87 × 10 <sup>-1</sup>	4.24 × 10 <sup>-1</sup>	1.87 × 10 <sup>-1</sup>	4.24 × 10 <sup>-1</sup>
[ <sup>241</sup> Pu] <sup>b</sup>	bd <sup>d</sup>	bd <sup>d</sup>	9.29 × 10 <sup>-2</sup>	1.85 × 10 <sup>-1</sup>	9.29 × 10 <sup>-2</sup>	1.85 × 10 <sup>-1</sup>
[ <sup>242</sup> U]	4.03 × 10 <sup>-1</sup>	1.69 × 10 <sup>-1</sup>	8.25 × 10 <sup>-1</sup>	9.64 × 10 <sup>-1</sup>	1.23 × 10 <sup>-1</sup>	1.30 × 10 <sup>-1</sup>
[ <sup>238</sup> U]	2.76 × 10 <sup>-1</sup>	6.92 × 10 <sup>-1</sup>	5.83 × 10 <sup>-1</sup>	1.10 × 10 <sup>-1</sup>	8.59 × 10 <sup>-1</sup>	1.74 × 10 <sup>-1</sup>
[ <sup>235</sup> U]	3.97 × 10 <sup>-1</sup>	7.41 × 10 <sup>-1</sup>	8.38 × 10 <sup>-1</sup>	1.94 × 10 <sup>-1</sup>	1.24 × 10 <sup>-1</sup>	2.74 × 10 <sup>-1</sup>
[ <sup>238</sup> U]	1.86 × 10 <sup>-1</sup>	6.84 × 10 <sup>-1</sup>	3.64 × 10 <sup>-1</sup>	4.66 × 10 <sup>-1</sup>	5.50 × 10 <sup>-1</sup>	1.15 × 10 <sup>-1</sup>
isotope ratio	filtrate	2 $\sigma$	retentate	2 $\sigma$		
<sup>238</sup> Pu/ <sup>239</sup> Pu <sup>(1)</sup>	nm <sup>c</sup>	nm <sup>c</sup>	3.17 × 10 <sup>-1</sup>	3.54 × 10 <sup>-1</sup>		
<sup>239</sup> Pu/ <sup>240</sup> Pu	3.63 × 10 <sup>-1</sup>	2.12 × 10 <sup>-1</sup>	3.84 × 10 <sup>-1</sup>	2.68 × 10 <sup>-1</sup>		
<sup>241</sup> Pu/ <sup>240</sup> Pu <sup>(2)</sup>	bd <sup>d</sup>	bd <sup>d</sup>	1.89 × 10 <sup>-1</sup>	3.77 × 10 <sup>-1</sup>		
<sup>242</sup> U/ <sup>238</sup> U	2.17 × 10 <sup>-1</sup>	9.58 × 10 <sup>-1</sup>	2.38 × 10 <sup>-1</sup>	4.06 × 10 <sup>-1</sup>		
<sup>242</sup> U/ <sup>235</sup> U	1.50 × 10 <sup>0</sup>	1.71 × 10 <sup>-1</sup>	1.69 × 10 <sup>0</sup>	2.23 × 10 <sup>-1</sup>		
<sup>242</sup> U/ <sup>238</sup> U	2.15 × 10 <sup>-1</sup>	7.36 × 10 <sup>-1</sup>	2.41 × 10 <sup>-1</sup>	3.08 × 10 <sup>-1</sup>		

<sup>a</sup> By AEA. <sup>b</sup> By GEA. <sup>c</sup> nm, not measured. <sup>d</sup> bd, below detection limit.

## Scenario 1 (continued)



- A radiochemical analysis was performed on a sample to determine the plutonium isotopic concentrations
  - <sup>238</sup>Pu – very low
  - <sup>239</sup>Pu – 99.961%
  - <sup>240</sup>Pu – 0.039%
  - <sup>241</sup>Pu – very very low
  - <sup>242</sup>Pu – very very low
- This ratio represents extremely low burnup Pu
  - Typical reactor plutonium contains significant <sup>238</sup>Pu, <sup>239</sup>Pu, <sup>240</sup>Pu, <sup>241</sup>Pu, and <sup>242</sup>Pu due to additional nuclear interactions over an extended period of exposure to neutrons

## Scenario 1 (continued)



- Age of sample estimated to be ~62a (1944) from U isotopic compositions

Am237 73.0m	Am238 98m	Am239 11.9h	Am240 50.8a	Am241 432.7a	Am242 16.02h/141a	Am243 7.4E3a
237.0499	238.0519	239.0530	240.0552	241.0568	242.0595	243.0613
Pu236 2.858a	Pu237 45.2d	Pu238 87.7a	Pu239 2.41E+4a	Pu240 6.56E+3a	Pu241 14.4a	Pu242 3.75E+5a
236.0460	237.0484	238.0459	239.0521	240.0538	241.0568	242.0587
Np235 1.085a	Np236 22.5h/1.5E+5	Np237 2.14E+6a	Np238 3.83E+5d	Np239 2.3565d	Np240 61.9m/7E+2	Np241 13.9m
235.0440	236.0465	237.0481	238.0509	239.0529	240.0561	241.0582
U234 2.46E+5a	U235 7.04E+8a	U236 2.34E+7a	U237 6.75d	U238 4.47E+9a	U239 23.45m	U240 14.1h
234.0409	235.0439	236.0455	237.0487	238.0507	239.0543	240.0565

## Scenario 1 (continued)



- $^{240}\text{Pu}$  composition too low compared to Hanford reference material of this time frame
  - Meaning sample has lower burnup
- Where could it have come from?



## Scenario 1 (continued)

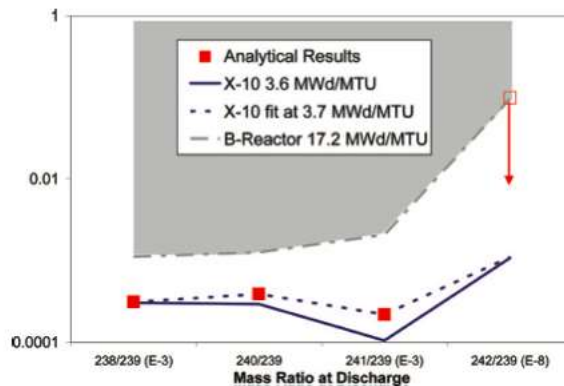


- After more investigation, historic material produced at X-10 reactor in ORNL was shipped to Hanford for testing
  - 96 X-10 spent fuel slugs
  - ~112kg uranium
  - Irradiated to 0.0036 GWD/MTU
  - Shipped to Hanford for experimental separations
  - Produced matching Pu with 0.03%  $^{240}\text{Pu}$

## Scenario 1 (continued)



- A radiochemical analysis was performed on a sample to determine the plutonium isotopic concentrations



## Scenario 1 (continued)



- The shipment to Hanford provided Pu for Handford's T-Plant operations
  - Records indicate the first material was separated on December 9, 1944, which originated from ORNL.
    - All future full scale batches were from B-reactor
  - It is concluded that this material:
    - Is part of the first batch of Pu separated at the world's first industrial-sized reprocessing facility
    - Represents the oldest known collection of  $^{239}\text{Pu}$  in the world
    - And to be the 2<sup>nd</sup> oldest collected plutonium ever!
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## Thanks



- Questions, discussion, and comments?