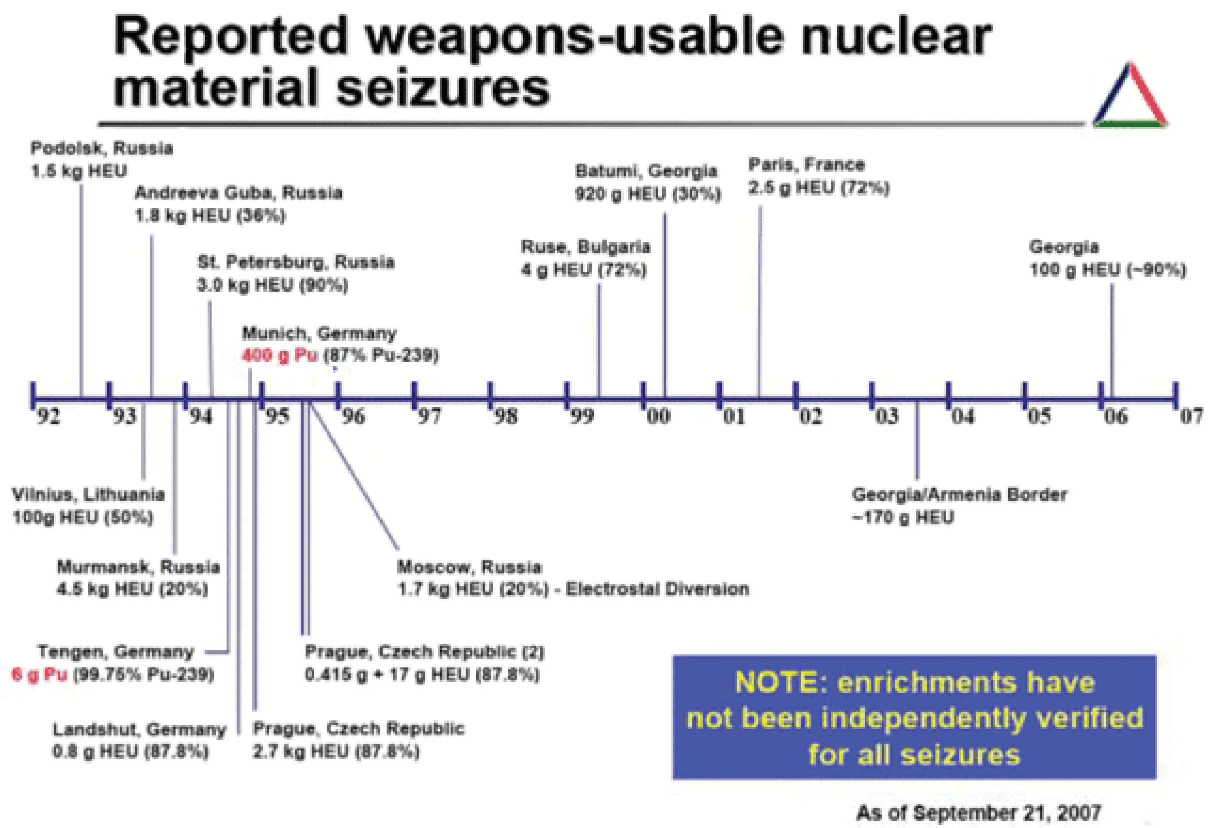
RDCH 702: Lecture 12 Forensics in Nuclear Applications

- From Nuclear Forensics Analysis
  - Moody, Grant, Hutcheon
- Readings
  - Glasstone: Effects of Nuclear Weapon

<u>Outline</u>

- Isotopics
  - Variation with reactor
- Signatures from separations
- Devices



Plutonium Isotopics

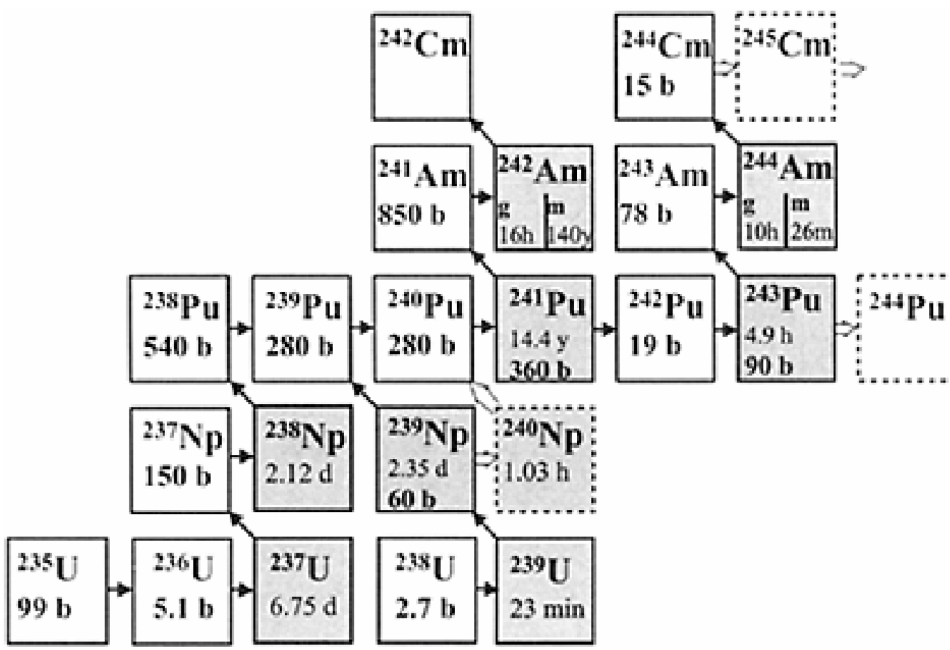
- Reactor types
  - Light water reactor
    - → ^{235}U enrichment
  - CANDU
    - → Natural uranium fuel, D_2O
- Pu formed in core or from blanket
  - U blanket may have a range of isotopic composition
    - → Natural, enriched, and anthropogenic
- Pu formation from neutron capture on ^{238}U
  - ^{239}Pu becomes a target for higher isotope production
    - → (n,γ) up to ^{241}Pu
    - → Also some (n,2n)
      - * E_{min} =5.7 MeV
      - * ^{238}Pu from ^{239}Pu

Plutonium isotopics

- ^{238}Pu can also be produced through successive neutron capture on ^{235}U
  - $^{235}U(n,\gamma)^{236}U$
  - $^{236}U(n,\gamma)^{237}U$
  - ^{237}U beta decay to ^{237}Np
  - $^{237}Np(n,\gamma)^{238}Np$
  - ^{238}Np beta decay to ^{238}Pu
- Mixture of Pu isotopics from fuel or blanket can act as a signature
  - ^{239}Pu dominates at low burnups
    - →Device Pu has 6 % ^{240}Pu

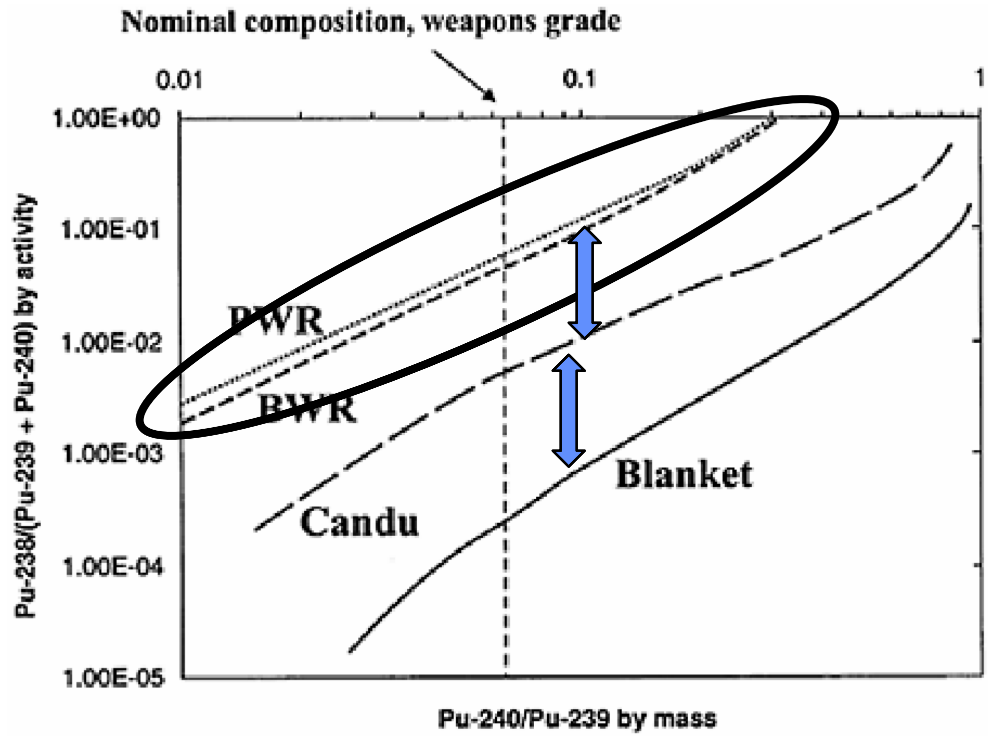
Plutonium isotopics: Neutron Fluence

- Pu isotopics influence by neutron fluence and energy
- Neutron energy effected by
  - reactor operating temperature
  - Moderator
- Fuel size influences distance between neutron generation and moderator
- Fuel composition can influence neutron spectrum
  - Depletion of neutron in ^{235}U resonance region due to higher interactions
- ^{240}Pu production
  - Capture on ^{239}Np produces ^{240}Np, which decays into ^{240}Pu
  - Competition between capture and decay



Plutonium Isotopics

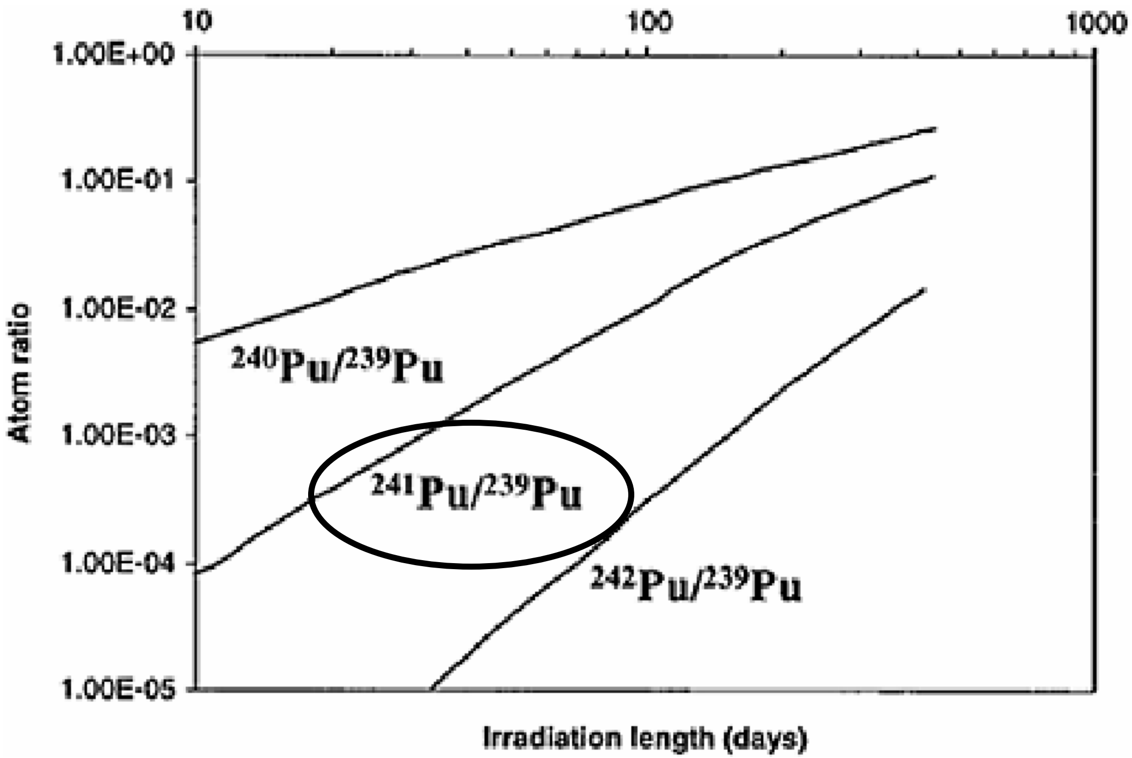
- **Evaluate ratios with ^{240}Pu**
  - **Mass 240:239 (MS)**
  - **Activity 238/(239+240) (alpha spectroscopy)**
- **Varied reactor types, 37.5 MW/ton**



- **Large Pu isotopic variation**
- **Obvious variation for blanket and CANDU**

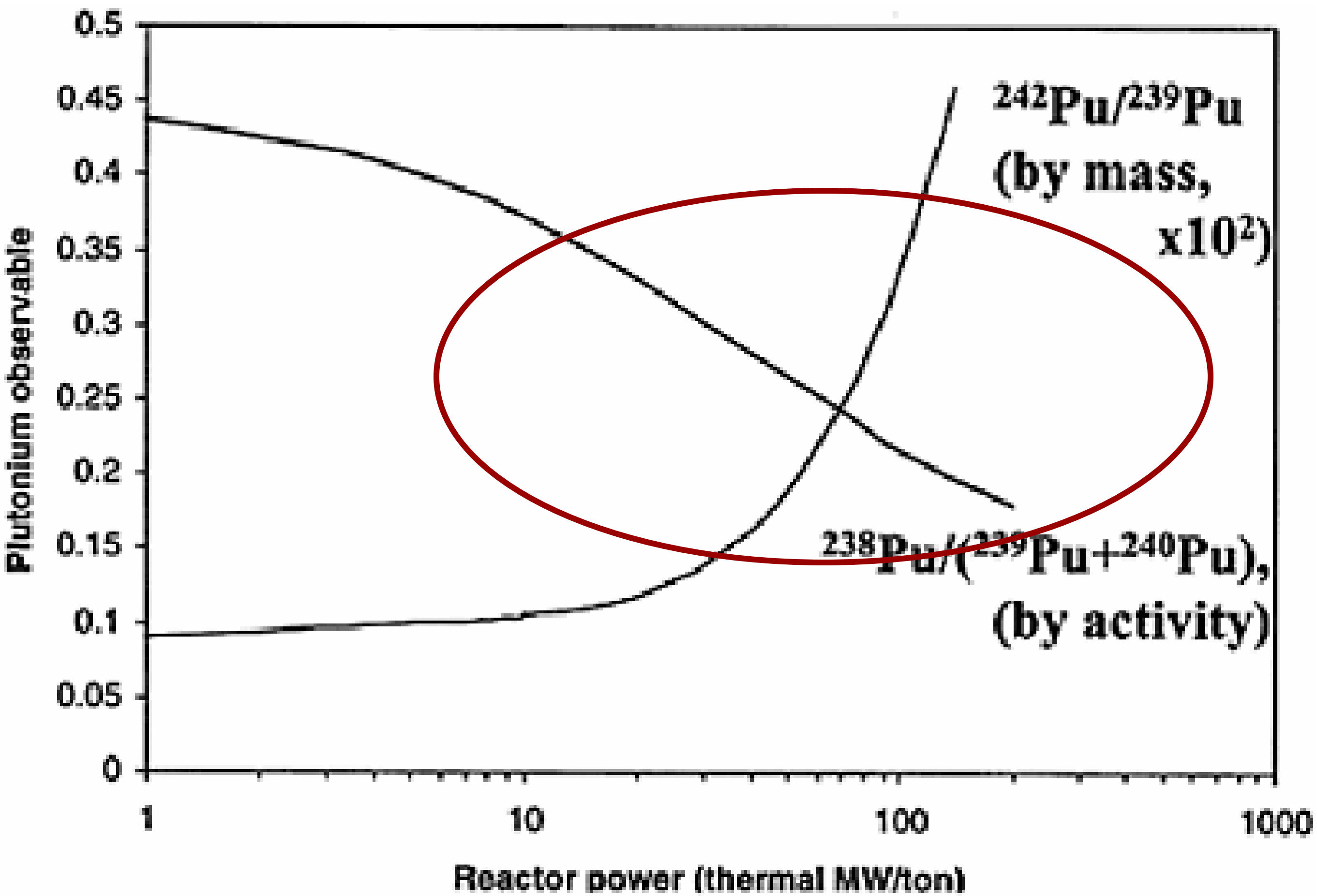
Plutonium Isotopics: Irradiation time

- Comparison to ^{239}Pu concentration
  - Can provide time since discharge
    - → ^{241}Pu is time sensitive
- Change in measure and expect ratio can be used to determine time since discharge



Plutonium Isotopics

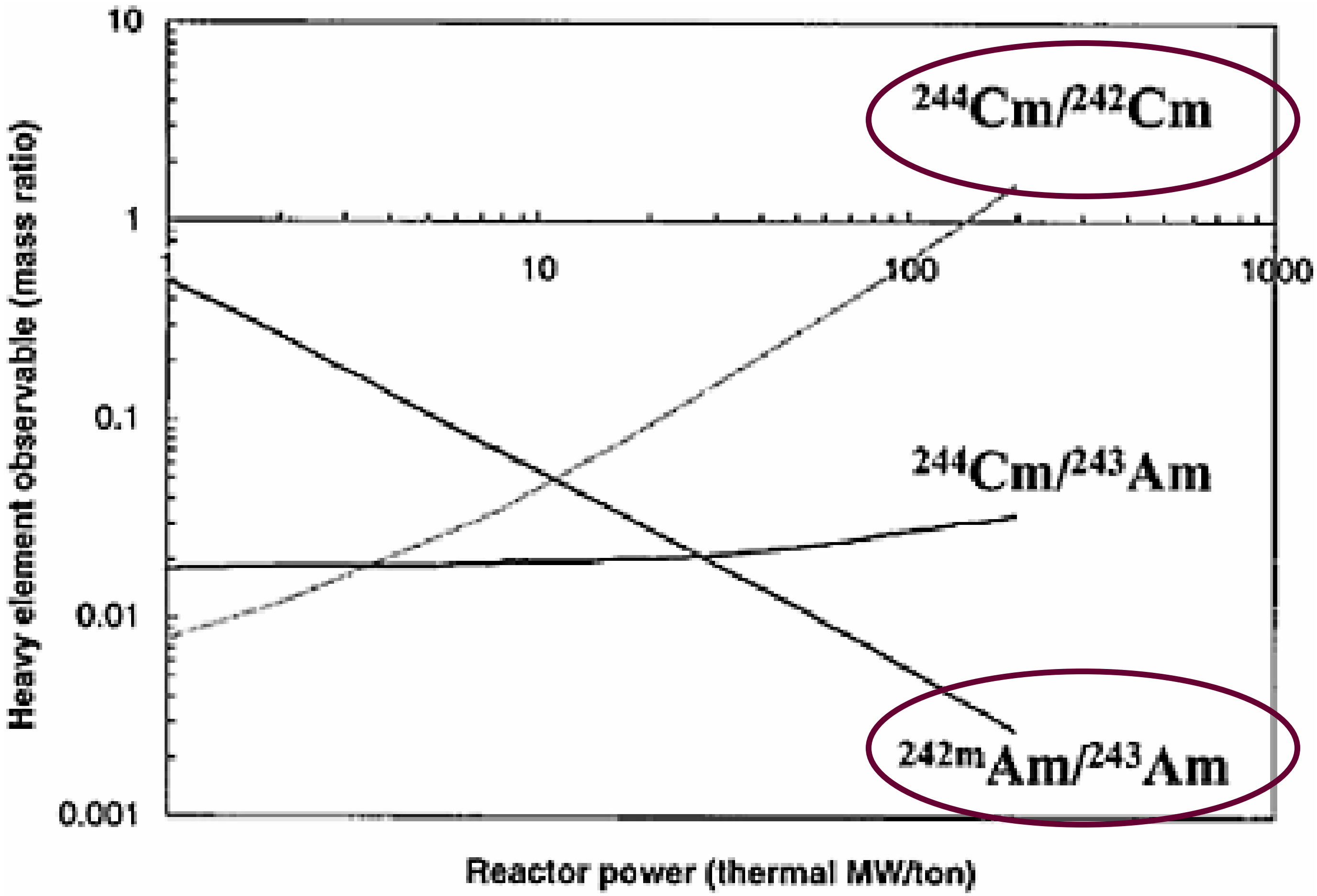
- Use for evaluating reactor power
  - Mass 242:239 ratio
  - Activity 238/(239 +240)



Transplutonium Isotopics

- Am (242m, 243) and Cm (242, 244) sensitive to reactor power
    - Due to relative decay of ^{241}Pu and formation of ^{241}Am
        - → More ^{241}Am is available for reactions from longer times with low flux
            - * Weapons grade Pu from 10 day to 2 year irradiation
- From neutron reaction and beta decay of heavier Pu
- ^{243}Pu beta decay to ^{243}Am
    - Capture and decay to ^{244}Cm
- Neutron capture on ^{241}Am
    - ^{242}Am states
    - Ground state decay to ^{242}Cm
- Strong flux dependence on ratios

6% ^{240}Pu



- ^{244}Cm and ^{243}Am arise from multiple neutron capture on ^{241}Pu
- ^{242}Cm ($t_{1/2}$=163 d) can determine time since discharge
  - Change from expected value compared to other ratios

Reprocessing

- 510 gU/g Pu for used fuel
- Majority of U remains, enrichment level to 0.62%
  - Depends upon the level of burnup
- Reprocessing limitations
  - Remote handling
  - Criticality issues
  - Limit impurities
- Range of reprocessing techniques
  - Precipitation
  - Molten salt
  - Ion exchange
  - Fluoride volatility
  - Solvent extraction

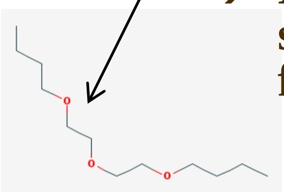
Table 3.1 Long-Lived Fission Products Present at Discharge in Reactor Fuel Used to Produce 1g of Pu Containing 6% ^{240}Pu by Mass

Nuclide	Half-Life (years)	Mass (mg)	Activity (dpm)
^{90}Sr	28.5	11	3.4×10^{12}
^{93}Zr	1.5×10^{6}	14[a]	8.0×10^{7}
^{99}Tc	2.1×10^{5}	16	6.1×10^{8}
^{106}Ru	1.01	4.1	3.0×10^{13}
^{125}Sb	2.77	0.45	1.0×10^{12}
^{129}I	1.6×10^{7}	3.4	1.3×10^{6}
^{135}Cs	2.0×10^{6}	2.4	7.1×10^{6}
^{137}Cs	30.2	22	4.2×10^{12}
^{143}Nd	Stable	20	0
^{155}Eu	4.96	0.23	2.4×10^{11}

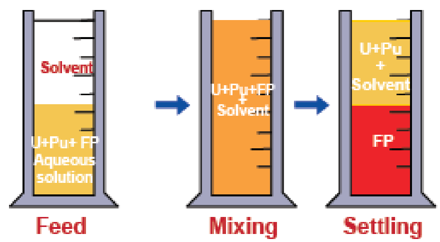
Note: ORIGEN2 calculation, natural-U-fueled, graphite-moderated reactor, 37.5 MW_t/ton.
[a]Lower limit; ^{93}Zr is also produced in claddings.

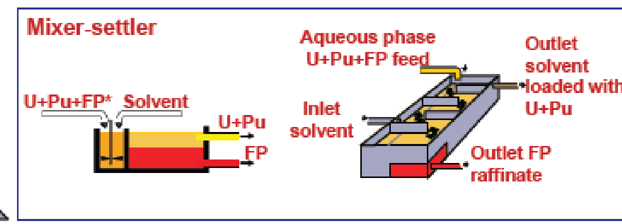
Reprocessing

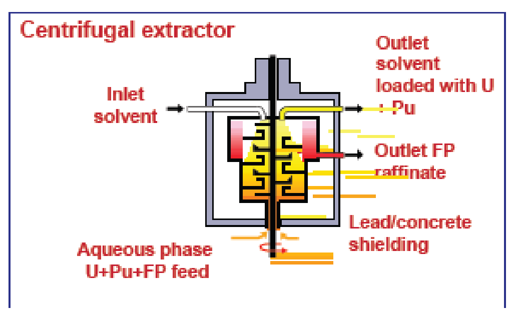
- **BUTEX**
  - **Dibutyl carbitol solvent**



    → **Relatively poor separation from Ru**
      * **1E3 rather than 1E6**
    → **1 g Pu has 1E7 dpm ^{106}Ru**
- **HEXONE**
  - **Methylisobutylketone solvent**
  - **Decontamination factors**
    → **1E4 Ru**
    → **1E5 for Zr, Nb, and Ce**
      * **1 g Pu has 1000 dpm ^{93}Zr**
- **PUREX**
  - **Zr, Tc, and Ru observed**
  - **Ln, Np, and Th non-negligible**
  - **Nd is a key isotope, nature levels in reprocessing materials**
    → **Natural and reactor Nd ratios are plant signatures**







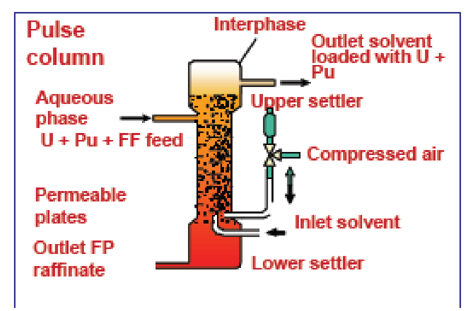


Table 3.3 Production of Selected Heavy-Element Nuclides in Reactor Fuel (at Discharge) Used to Produce 1g of Pu Containing 6% ^{240}Pu by Mass

Nuclide	Half-Life (years)	Mass (g)	Activity (dpm)
^{237}Np	2.14×10^{6}	5.6×10^{-3}	8.8×10^{6}
^{241}Am[a]	432	9.7×10^{-6}	7.4×10^{7}
^{242m}Am	141	5.3×10^{-8}	1.2×10^{6}
^{243}Am	7370	3.9×10^{-6}	1.7×10^{6}
^{242}Cm	0.45	4.4×10^{-7}	3.2×10^{9}
^{244}Cm	18.1	7.6×10^{-8}	1.4×10^{7}

Note: From the same ORIGEN2 calculation used to generate Table 3.1 data.

[a] Assumes no in-growth from post-discharge decay of ^{241}Pu.

- **Isotopes in Pu from incomplete separation and decay**
  - **Small masses, high activity**

Reprocessing

- Organic analysis
  - Radiation effects
    - → Polymerization
    - → TBP degradation
      - * Di, mono, and phosphoric acid
    - → N-butonal and nitrobutane
- Changes extraction behavior, limits extraction efficiency
  - Formation of carboxylic acid, esters, ketones, nitroorganic compounds
- Signatures from processing
  - Range of sample states, including gas
  - GC-MS methods

Table 3.2 Neodymium Isotope Ratios (by Mass) and the Production of ^{143}Nd vs. Total Pu Production (6% ^{240}Pu) as a Function of the Power Level of a Natural-U-Fueled, Graphite-Moderated Reactor

	Reactor Power Level (MW_t/ton)			Natural Nd
Mass Ratio	1.25	12.5	125	
^{142}Nd/143Nd	0.00172	0.00081	0.00019	2.212
^{144}Nd/143Nd[a]	0.964	0.951	0.949	1.973
^{145}Nd/143Nd	0.704	0.697	0.699	0.692
^{146}Nd/143Nd	0.571	0.567	0.572	1.478
^{148}Nd/143Nd	0.343	0.342	0.351	0.487
^{150}Nd/143Nd	0.160	0.159	0.160	0.484
^{143}Nd/totPu	0.0202	0.0202	0.0194	

Note: Natural Nd isotopic composition is included for comparison.
[a] Assumes complete decay of 285-day 144Ce.

Device general signatures

- Designed to have highest possible k (neutron multiplication factor)
  - Greatest increase of neutrons and fission from one generation to the next
  - Cooling from explosion
    - → k goes to zero
    - → Need to complete reaction in short time
- Designs maximized neutron and fissile material reaction
- Generation time
  - Average time between neutron release and capture
  - Neutron energy 1 MeV
    - → 10 n sec per generation
      - * Shake
  - 50 to 60 shakes to produce 1 kt of fission yield
    - → About 0.5 microseconds
      - * Energy generation exponential
      - * Most energy from last few generations

Device general information

- Fast neutron reflection important for device
  - Better use of neutrons, lower critical mass
- Fission spectrum neutrons drive the n,f reaction
  - Not thermalized
- Cross sections for fast neutrons orders of magnitude lower than thermal reactions
  - n, gamma reaction for non-fissile isotopes also lower
- Limiting non-fissile isotopes results in material signatures
  - Less than 7 % ^{240}Pu
  - More than 20 % ^{235}U
  - Low ^{232}U in ^{233}U (10 ppm)

Device general information

- Low Z material in Pu not desired
  - React with neutrons
  - Lowers density
  - Moderates neutrons
    - →Used as signature
      - * Pu device isotopes in non-metal form indicates storage or starting material
- Alloys
  - Ga in Pu
  - Nb in U

Post-detonation analysis

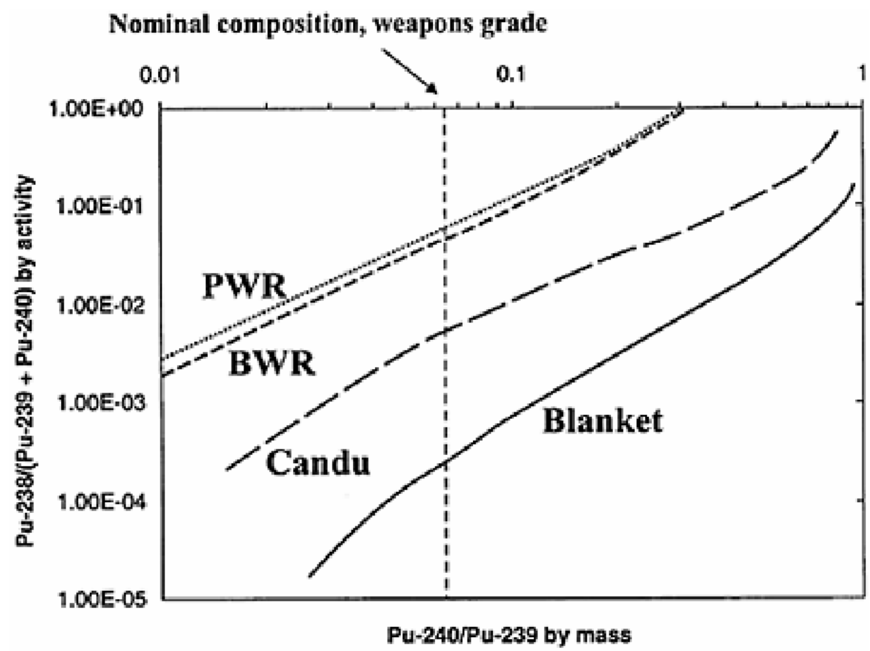
- Debris provides information on
  - Function
  - Source
- Efficiency of yield from fuel and fission products
  - High efficiency indicates sophisticated device
    - → Also with multiple fissile material and (n,2n) reactions
- Indication of initial isotopic compositions
  - Neutron energy influences fission product distribution
    - → Also varies with nuclear fuel

Overview

- Discussed signatures
  - Isotopic ratios
- General device overview

Questions

- What signatures are available from
  - Pu isotopic ratios
- What material can be alloyed with Pu?
- Why do Nd fission product isotopes differ from natural



Questions

- **Respond to blog with questions**
- **PDF Quiz 12**