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Passive Nondestructive Assay of Nuclear Materials

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Workshop on Nuclear Material Accounting and Control:
Current Practices and Future Prospectives
Homi Bhabha National Institute
Aushaktinagar, Mumbai, India

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Importance of Non-Destructive Assay Measurements

- NDA measurements involve the detection and analysis of radiation emitted from Special Nuclear Materials (SNM) to detect, identify, locate, and often quantify the composition and/or mass of material.
- NDA measurements have several advantages over DA measurements:
  - produce faster results
  - can be performed with the material in situ
  - produce no waste
- Results from NDA measurements are used for a wide range of facility operations including material control and accounting (MC&A), nuclear security, criticality safety, and waste management.

Spill at Processing Facility

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Role of NDA Measurements - Safeguards

- Most frequently implemented measurement techniques to reliably determine the characteristics of SNM (e.g. 65-75% of all Pu inventory measurements at LANL performed using NDA).

- NDA measurements for MC&A purposes include:
  - Accountability Measurements
  - Confirmatory Measurements
  - Shipper/Receiver Measurements
  - Process control measurements

- Impact of poor quality or erroneous NDA measurement results include possible safety impacts (criticality safety), security impacts (loss of nuclear material) and economic impacts (cost of re-measuring items).

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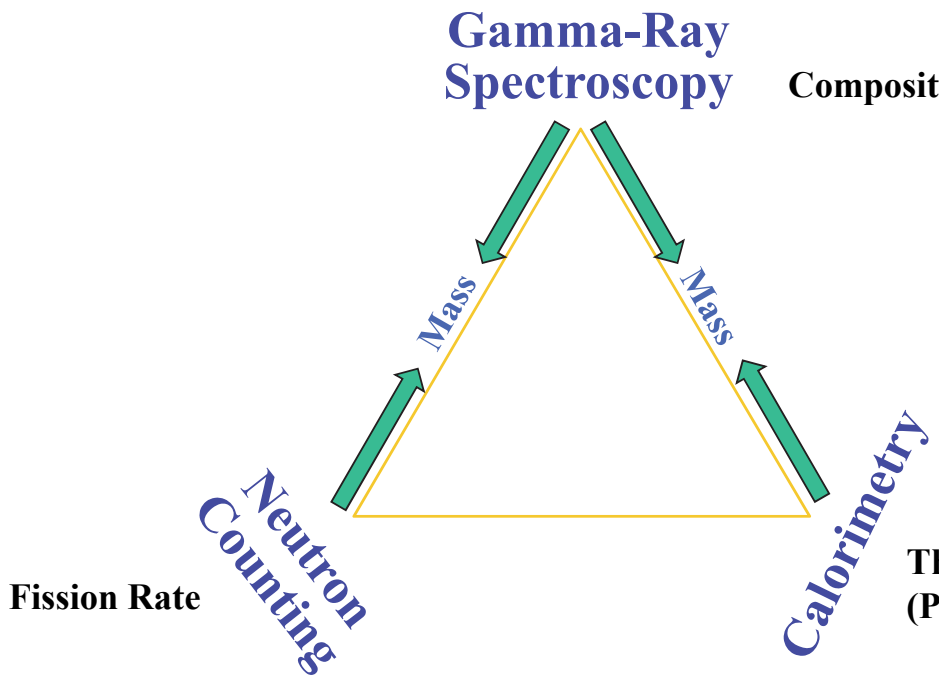
NDA Measurement Techniques



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Gamma-Ray NDA

Enrichment or Isotopic Measurements

- ^{235}U enrichment (NaI, HPGe)
- Pu isotopic composition (HPGe)

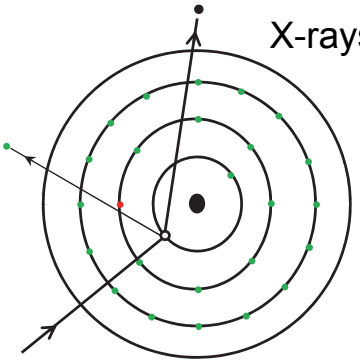
Measuring SNM mass in Nuclear Waste

- Segmented Gamma-Ray Scanner
- Tomographic Gamma-Ray Scanner

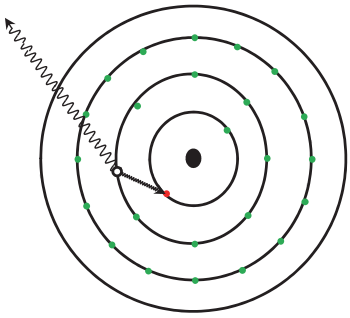
Hold-up measurements (Quantifying amount of material in pipes/ductwork/etc)

Origin of X-rays and Gamma-rays

X-rays



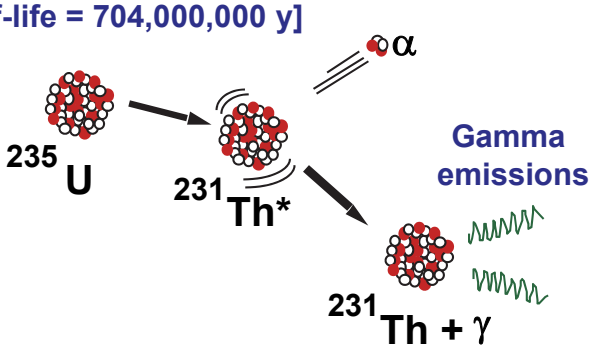
PARTICLE KNOCKS OUT AN ATOMIC ELECTRON AND LEAVES A VACANCY



AN OUTER ELECTRON "FALLS" INTO THE "HOLE," AND GIVES UP ENERGY IN THE FORM OF AN X-RAY

Gamma-rays

[Half-life = 704,000,000 y]



Gamma Rays:

185.7 keV	57 %
143.8 keV	11 %
163.4 keV	5 %
205.3 keV	5 %

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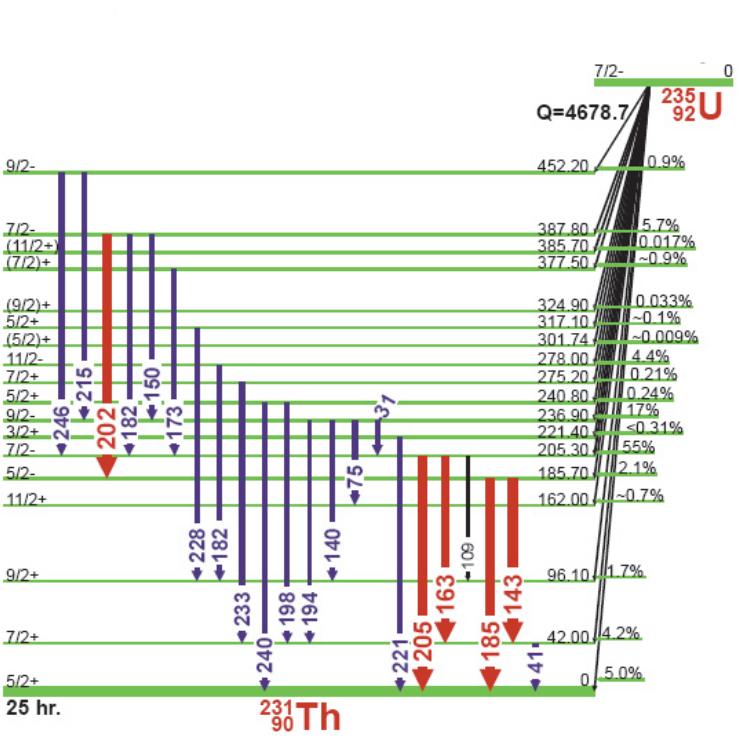
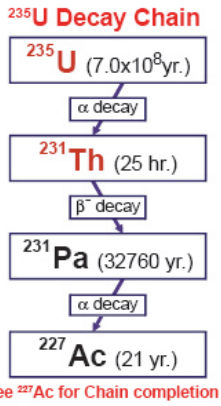
^{235}U Decay Scheme and Branching Ratios





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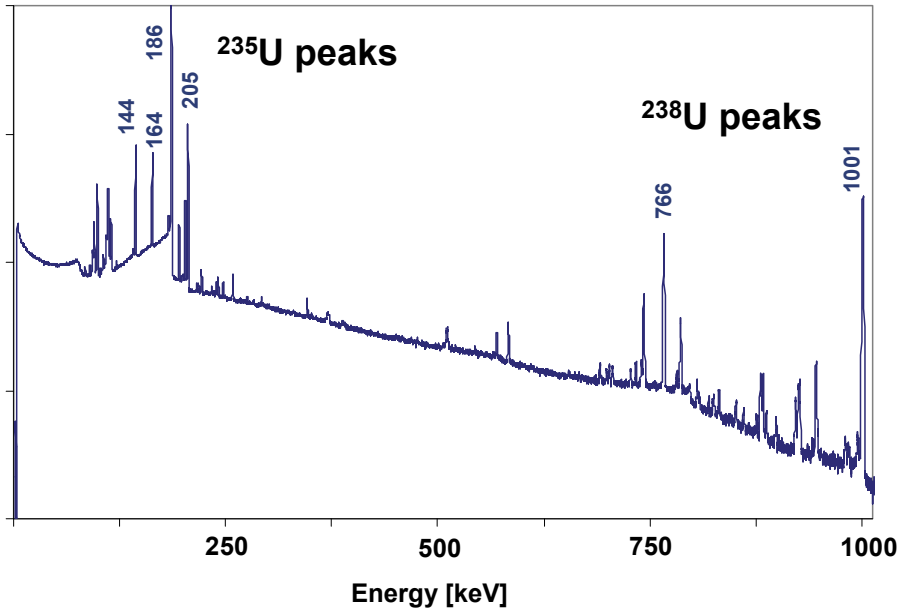
Uranium Gamma-Ray Spectrum (HPGe)

50.5% enrichment (HEU)



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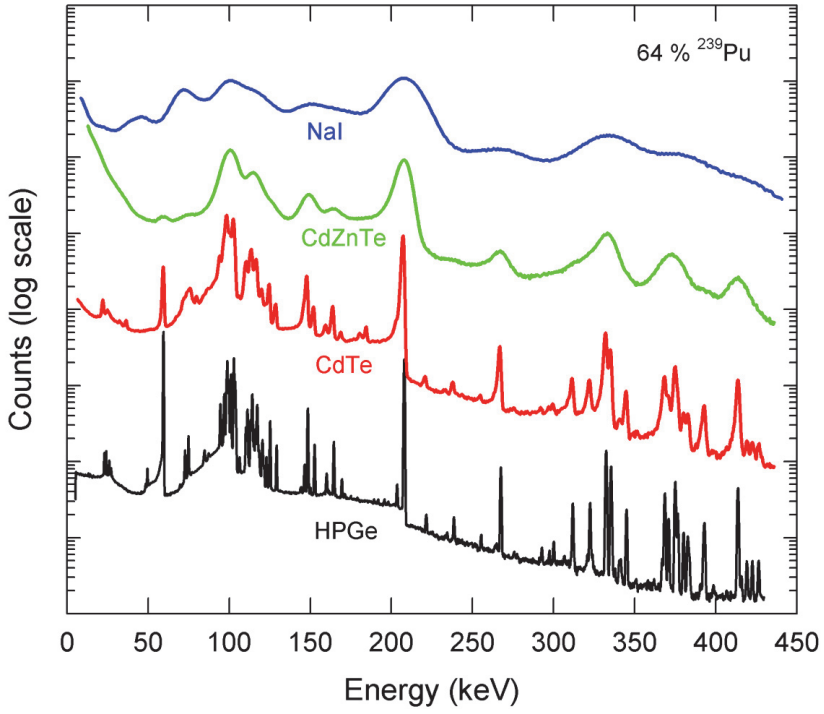
Various Gamma-Ray Detector Resolutions



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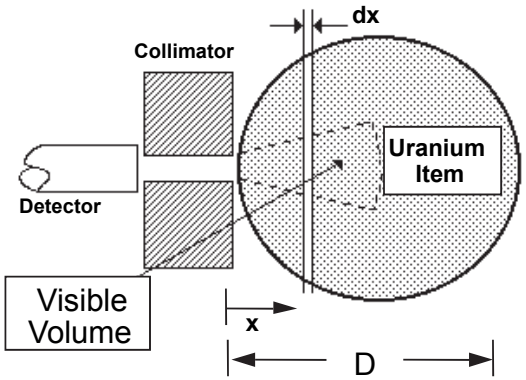
^{235}U Enrichment Measurement with NaI Detector

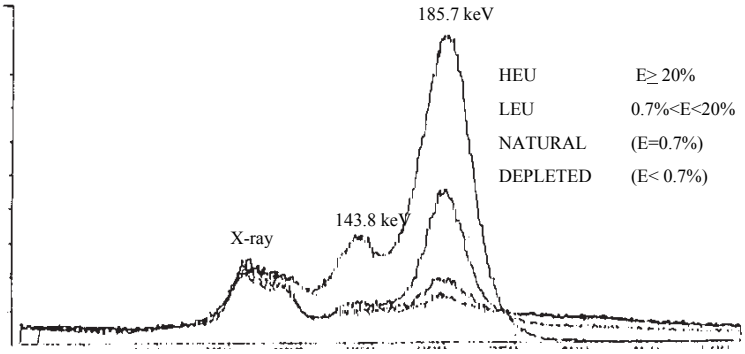


Infinite Thickness Method

Detector views the item through a collimator.

Visible volume determined by collimator size and the absorption coefficient of U.

The measured 186-keV intensity is proportional to the ^{235}U in the visible volume. For an enrichment measurement on an infinite item thickness, the detector views a portion of the item volume through a collimator. Isotopic uniformity is a requirement of this type of measurement.



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Accuracy of the Infinite Thickness Method

- Method requires one or two NM standards to calibrate the system
- Corrections required for container wall thickness and differences in chemical composition between item being assayed and NM standards
- 1-5% typically, depending on the item
- NaI and HPGe can provide similar accuracy
- In special applications involving installed systems, 0.1-0.2% accuracy is possible.

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Ratio-Based Isotopic Measurements

$$\frac{N(E_1)}{N(E_2)} = \frac{C(E_1)}{C(E_2)} \cdot \frac{T_{1/2}(1)}{T_{1/2}(2)} \cdot \frac{BR(E_2)}{BR(E_1)} \cdot \frac{RE(E_2)}{RE(E_1)}$$

$N(E_i)$	Atom Number for Isotope i
$C(E_i)$	Photo-peak Area at Energy E_i
$T_{1/2}(i)$	Half Life of Isotope i
$BR(E_i)$	Branching Ratio at Energy E_i
$RE(E_i)$	Relative Efficiency at Energy E_i

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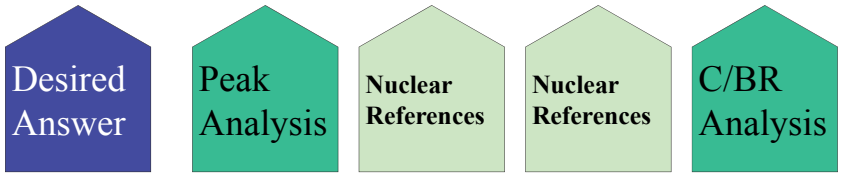
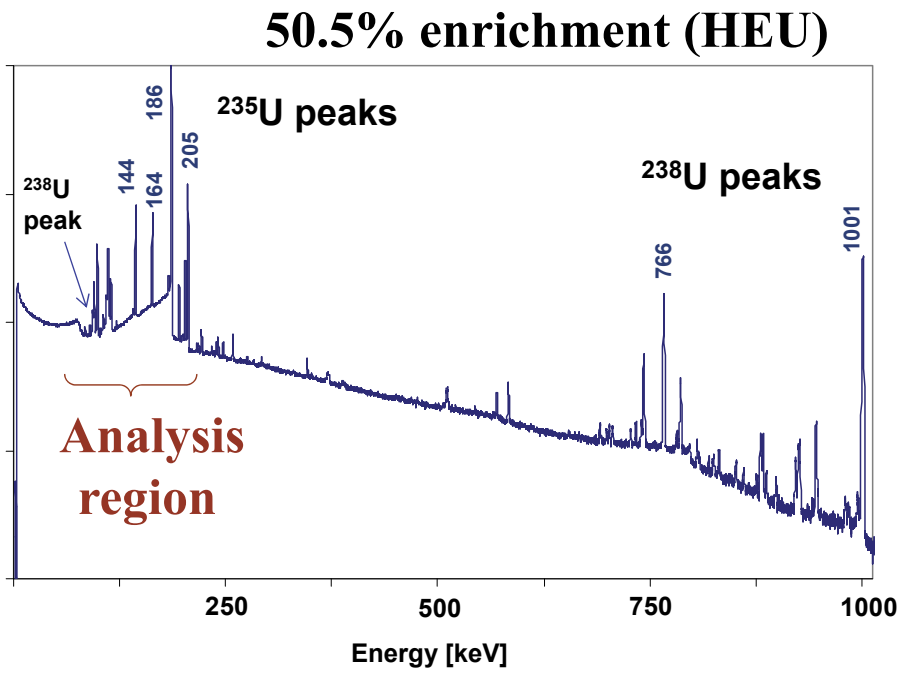
The Isotopic Ratio Formula

$$\frac{N(E_1)}{N(E_2)} = \frac{C(E_1)}{C(E_2)} \cdot \frac{T_{1/2}(1)}{T_{1/2}(2)} \cdot \frac{BR(E_2)}{BR(E_1)} \cdot \frac{RE(E_2)}{RE(E_1)}$$



All these values are obtained from the spectrum itself or from standard nuclear references; no calibration is necessary!

Uranium Gamma-Ray Spectrum (HPGe)



Calculating Isotopic Fractions

Based on measuring relative intensities of gamma-ray lines, we can get:

$$\frac{^{234}U}{^{238}U} \qquad \frac{^{235}U}{^{238}U}$$

And we know that:

$$^{234}U + ^{235}U + ^{238}U = 100\%$$

So

$$\frac{^{234}U}{^{238}U} + \frac{^{235}U}{^{238}U} + \frac{^{238}U}{^{238}U} = \frac{1}{^{238}U}$$

We can solve the above equation for ^{238}U which allows us to calculate ^{234}U and ^{235}U from the original ratios:

$$\frac{^{234}U}{^{238}U} \times {}^{238}U = {}^{234}U \qquad \frac{^{235}U}{^{238}U} \times {}^{238}U = {}^{235}U$$

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Independence of Analysis

Using **relative** efficiency and isotope **ratios** makes analysis independent of:

- **Item Characteristics**
  - Size
  - Shape
  - Physical and Chemical Composition
  - Packaging
  - Filtering
- **Data Acquisition Limitations**
  - Pulse Pile-up
  - Deadtime

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Two commercially available analysis programs

- MGA/MGAU (Multi Group Analysis) (R. Gunnick, LLNL)
  - Can analyze low energy region (94-104 keV) or high energy region (<1 MeV)
  - Rigid analysis routine
  - User friendly
- FRAM (Fixed energy Response function Analysis with Multiple efficiencies) (T. Sampson and D. Vo, LANL)
  - Can analyze gamma-rays up to more than 1 MeV
  - User configurable analysis (flexible)
  - Learning curve associated with initial use of code

Why Measure Pu-Isotopic Composition?

- **Neutron Coincidence/Multiplicity Counting**
  - $^{240}Pu_{eff} = (2.52 \times ^{238}Pu) + (1.00 \times ^{240}Pu) + (1.68 \times ^{242}Pu)$

- **Calorimetry**

$$P_{eff} = \sum_{i=1}^{n} R_i P_i$$

  - n = number of isotopes in the item
  - R_i = mass fraction ($m_i/M_{total\ Pu}$) of the i^{th} isotope
  - P_i = specific power of the i^{th} isotope

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Relative Isotopic Error

Isotopic Precision varies by isotope:

Isotope	% RSD
^{238}Pu	<1 - 10
^{239}Pu	0.1 - 0.5
^{240}Pu	0.5 - 4
^{241}Pu	0.2 - 0.8
^{241}Am	0.2 - 10
P_{eff}	0.2 - 2.0
$^{240}Pu_{eff}$	0.5 - 4.0

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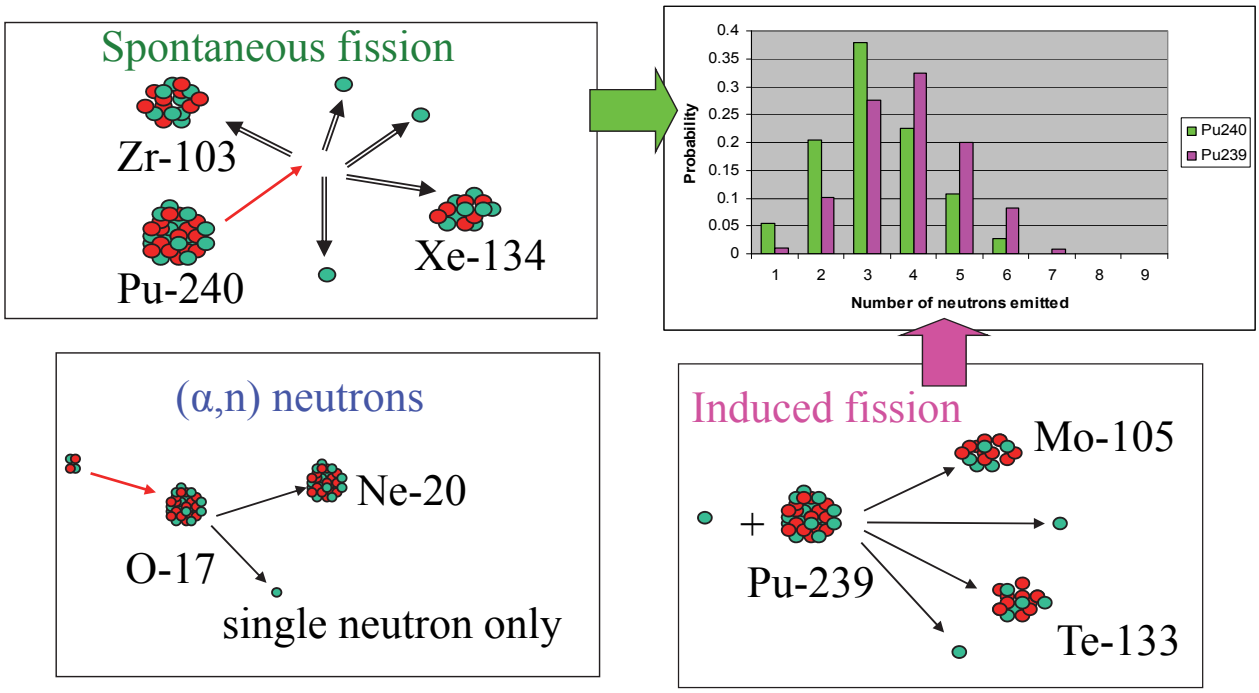
Neutron Signatures for Passive NDA



Spontaneous fission

Nuclide	Specific Intensity [n/(g.s)]
^{234}U	0.005
^{235}U	0.0003
^{236}U	0.0055
^{238}U	0.0136
^{238}Pu	2590.
^{239}Pu	0.022
^{240}Pu	1020.
^{241}Pu	~0.05
^{242}Pu	1720.
^{241}Am	1.18
^{242}Cm	2.1×10^{7}
^{244}Cm	1.08×10^{7}
^{252}Cf	2.34×10^{12}

$$^{240}Pu_{eff} = (2.52 \times ^{238}Pu) + (1.00 \times ^{240}Pu) + (1.68 \times ^{242}Pu)$$

N. Ensslin, et al., LA-13422-M (1998)

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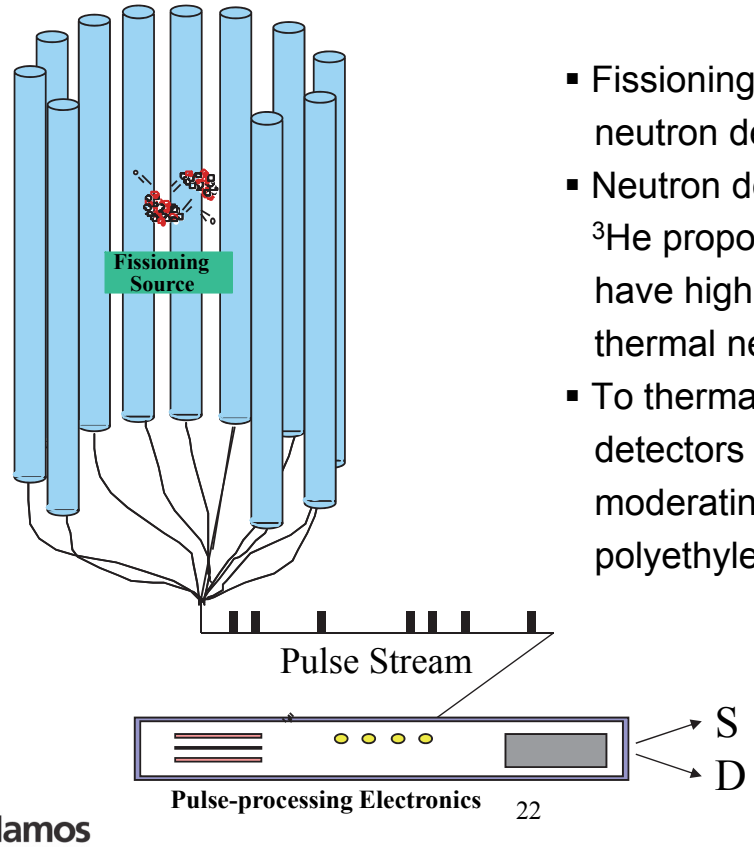
Passive Neutron Counter



- Fissioning source surrounded by neutron detectors
- Neutron detectors are nominally ^{3}He proportional counters which have high detection efficiency for thermal neutrons
- To thermalize the neutrons, ^{3}He detectors are embedded in moderating matrix such as polyethylene.

Neutron Multiplicity Counting

Standard Point Model Multiplicity Equations for Pu:

$$S = m_{eff} F_0 \varepsilon \nu_{s1} M (1+\alpha)$$

$$D = \frac{1}{2} m_{eff} F_0 \varepsilon^2 f_d M^2 \left\{ \nu_{s2} + \left(\frac{M-1}{\nu_{i1}-1} \right) \nu_{s1} (1+\alpha) \nu_{i2} \right\}$$

$$T = \frac{1}{6} m_{eff} F_0 \varepsilon^3 f_t M^3 \left\{ \nu_{s3} + \left(\frac{M-1}{\nu_{i1}-1} \right) [3\nu_{s2}\nu_{i2} + \nu_{s1}(1+\alpha)\nu_{i3}] + 3\left(\frac{M-1}{\nu_{i1}-1} \right)^2 \nu_{s1}(1+\alpha)\nu_{i2}^2 \right\}$$

Converts S, D, T counting rates into properties of plutonium bearing item:

- effective mass of ^{240}Pu in item (m_{eff})
- ratio of (α,n) to spontaneous fission (SF) neutrons (α)
- multiplication (M)

Total Pu mass: $m = \frac{m_{eff}}{^{240}Pu_{eff}}$

Note: Measuring S and D known as coincidence counting
Measuring S, D, and T known as multiplicity counting

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Comparison of Coincidence and Multiplicity Counting

Coincidence Counting

- Pure samples
- Results dependent on knowledge or assumptions concerning α and M
- In general, shorter measurement times than multiplicity counter measurements
- Coincidence counters less expensive than multiplicity counters

Multiplicity Counting

- Both pure and impure samples ($\alpha < 10$)
- Information on item α and M determined from measurement
- In general, longer measurement times than coincidence counting measurements
- Multiplicity counters more expensive than coincidence counters.
- Multiplicity counter can be used as coincidence counter

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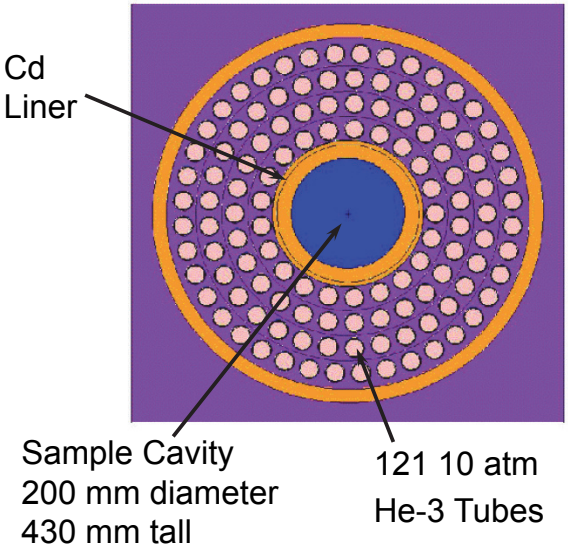
Epithermal Neutron Multiplicity Counter (ENMC)

H.O. Menlove and C.D. Rael, LA-UR-05-3041 (2005)



ε = 64% (Pu energy)
Die-away time = 19.1 μs

Neutron Multiplicity Uncertainty

N. Ensslin, et al., LA-13422-M (1998)

Table 7.1. Summary of past or expected multiplicity counter performance on various nuclear material categories. A well-designed multiplicity counter with roughly 50% to 55% detection efficiency is used, unless otherwise specified in the text.

Nuclear Material Category	SNM Mass (g)	(α,n)/sf rate α	Counting Time (s)	Assay Precision (% RSD)	Assay Bias (%)	References
Plutonium Metal	2000 g	0 to 0.2	1000 s	7.1%	-10.6%[a]	Langner 91b
	2000 g	0 to 0.2	3000 s	5.1%	-4.7%[a]	Krick 92b
	4000 g	0 to 0.2	1800 s	2.0%	0.0%	Langner 93b
	200-4000 g	0 to 1.3	3600 s	3.3%	-1.8%	Ensslin 98
Plutonium Oxide	2000 g	1	5000 s	0.7%	0.6%	Langner 91b
	1000 g	1	3000 s	0.8%	-2.7%	Krick 92b
	1000 g	1	1800 s	2.2%	-0.1%	Langner 93b
	4000 g	1-4	1800 s	3.0%	2.4%	Stewart 95
	1000 g	1-4	600 s	1-3%	0.9%	Stewart 98
Plutonium Scrap	100 g	5	1000 s	12%	2-5%	Langner 92
	100-1200 g	1-6	3600 s	4.5%	1.5%	Ensslin 98
Plutonium Residues	120 g	13-29	3000 s	20%	2-10%	Krick 92b
	300 g	7-34	3600 s	18.9%	-4.0%	Ensslin 98
	20-100 g	8-30	3600 s	7%	7%	Langner 98
	100 g	5-9	3600 s	8.7%	3.2%	Langner P.C.
Plutonium Waste (estimated)	1 g	1	1000 s	2%	1-2%	Ensslin 95
	1 g	5	1000 s	10%	2-5%	Ensslin 95
	1 g	20	1000 s	50%	5-10%	Ensslin 95
Plutonium Oxide in Excess Weapons Materials	1000 g	1 - 10	1500 s	6.0%	0.02%	Stewart 96
	1000 g	1 - 8	1000 s	5.0%	1.0%	Stewart 97(PC)
	4000 g	1 - 6	1800 s	4.2%[b]	0.8%	Langner 96b
	4000 g	1 - 6	1800 s	5.8%[c]	-1.0%	Langner 97b
Mixed Uranium/ Plutonium Oxide	300 g	1 - 2	1000 s	1-2%	1-3%	Menlove 93
Large Drum Inventory Verification	1 - 4000 g	1 - 6	6 - 12 h	10.2%	-0.5%	Rinard 97
	1 - 4000 g	7 - 50	6 - 12 h	N/A	N/A	Rinard 97

[a] Assay bias quoted without multiplication correction curve for metal.
[b] Assay precision based on counting statistics, gamma-ray isotopics, and scatter relative to calorimetry.
[c] Assay precision based on counting statistics and scatter relative to destructive analysis.

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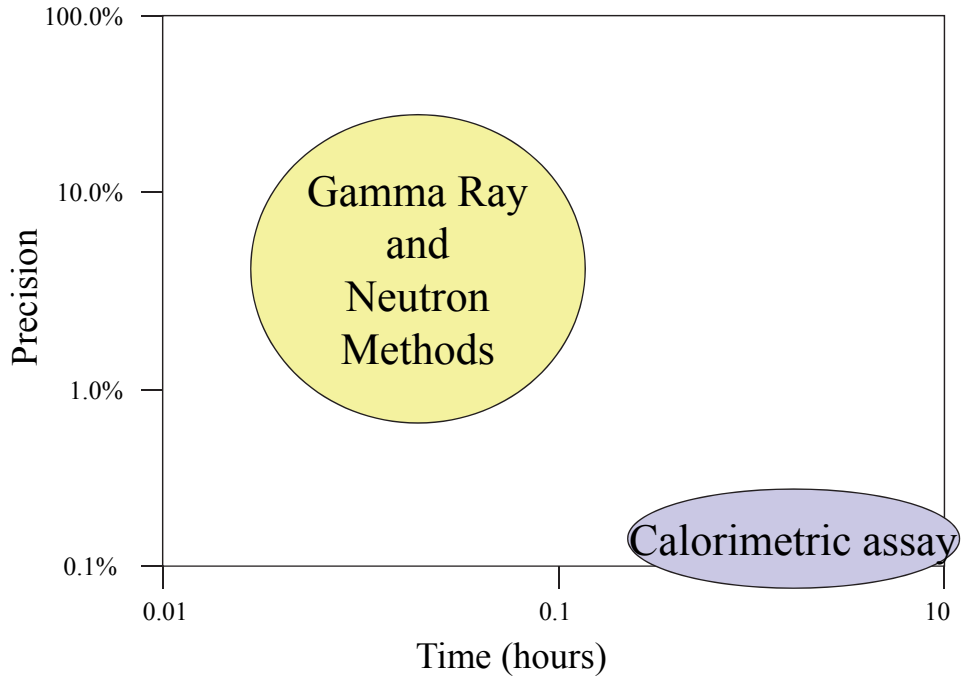
Calorimetric assay compliments other NDA techniques



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Calorimetry

- The most accurate and precise NDA measurement of Pu mass. The precision nominally ranges from:
  - ~0.5% for low power items (≤ 0.2 W)
  - ~0.1% for higher power items (≥ 1 W)
- You can't shield the heat of the material
- Inherently matrix independent
- Answer is bias free
- Integrates over total sample volume
- No representative physical standards needed

Calorimetric assay is the foundation for NDA of Pu in the US DOE.

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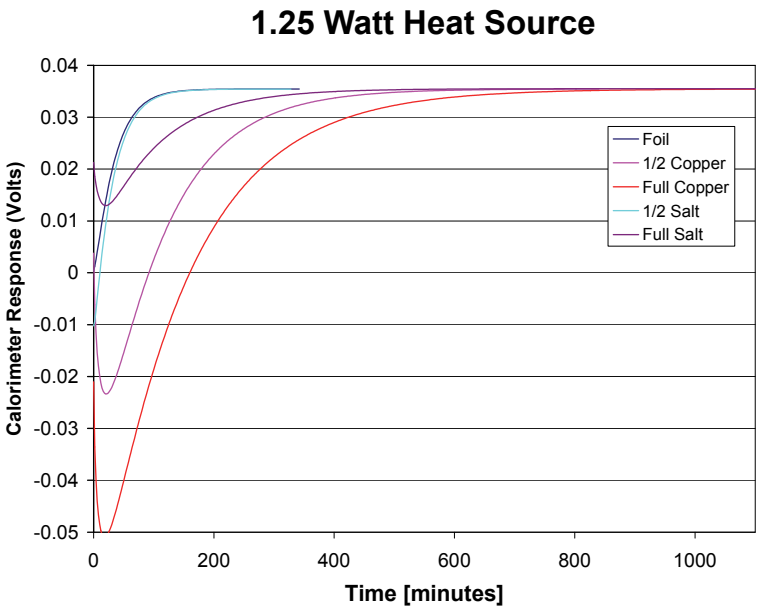
Calorimetry

- Calorimetry – quantitative measurement of heat
- Calorimetric assay – Determination of the mass of radioactive material through the measurement of its thermal power by calorimetry and isotopic composition by gamma-ray or mass spectrometry.
- The approach to thermal equilibrium as a function of time is exponential in nature



D.S. Bracken, et al. "Application Guide to Safeguards Calorimetry", LA-13867-M

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Calorimeter Measurement Uncertainty

Heat standard Power, Watts	Calorimeter diameter, m	Calorimeter Type, operation mode	Number Of Meas.	Precision, % RSD	Bias, %
98.0	0.06	rod, servo	29	0.065	0.02
3.5	0.15	rod, servo	55	0.09	0.00
4.0	0.25	twin, passive[1]	22	0.05	0.03
4.9	0.30	twin, passive[1]	34	0.06	0.05
0.0786	0.04	Solid state, passive[2]	10	0.23	0.001
[1]Pooled results from two calorimeters.					
[2]Measurements made in laboratory.					

D.S. Bracken, et al. “Application Guide to Safeguards Calorimetry”, LA-13867-M

Calorimetric Assay measurement technique

- Measure total *Sample Power* in a calorimeter (Watts)
- Measure isotopic composition of sample, including all significant heat producing isotopes (e.g. ^{241}Am)
- Compute *Effective Specific Power, P_{eff}*

$$P_{eff} = \sum P_i \cdot f_i$$

P_i – Watts/gram for isotope i
f_i – Isotopic fraction in sample relative to total Pu
i – Pu isotopes, ^{241}Am

$$\text{grams Pu} = \frac{\text{Sample Power (Watts)}}{P_{eff}\ \text{(Watts/g Pu)}}$$

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Isotopics and Calorimetry

Heat due primarily to **alpha decay** **(not fission)**, except ^{241}Pu which is due to beta decay.

Isotope	Half-Life (Yr)	Specific Power (W/g)
^{238}Pu	87.7 ± 0.1	0.56757 ± 0.05%
^{239}Pu	24110 ± 30	0.0019288 ± 0.02%
^{240}Pu	6561 ± 7	0.0070824 ± 0.03%
^{241}Pu	14.325 ± 0.006	0.003412 ± 0.06%
^{242}Pu	3.75×10^5	0.0001159 ± 0.22%
^{241}Am	432.6 ± 0.6	0.1142 ± 0.37%
^{233}U	1.592×10^5	0.00028
^{234}U	2.455×10^5	0.00018
^{235}U	7.04×10^8	5.996×10^{-5}
^{238}U	4.468×10^9	8.51×10^{-6}

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Calorimetric Assay Importance to Safeguards at LANL

D.S. Bracken, et al. "Application Guide to Safeguards Calorimetry", LA-13867-M

Table 1. Measurement Methods for LANL Pu Inventory Mass % for Each Material Form[1]				
Material form	Calorimetry + gamma spec	Analytical chemistry	Neutron counter + gamma spec	Segmented gamma scanner
Metal	71%	29%	0.3%	0%
Compounds pure	64%	35%	0.9%	0.3%
Compounds impure	72%	23%	2.6%	2.6%

[1]From LANL Material Accounting and Safeguards System (MASS) database April 1999.

Routine Plutonium Calorimetric Assay

- Shipper receiver measurements
- Outlier Resolution
- Accountability Measurements
- Calibration of NDA standards
- Process Control Measurements

Only Calorimetric Assay

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Summary

- NDA measurements play an important role in managing nuclear material through their ability to detect, identify, locate, and often quantify the composition and/or mass of material within a relatively short amount of time
- The three main passive NDA measurement techniques that are utilized in managing nuclear material are
  - Gamma-ray spectroscopy (composition)
  - Neutron Multiplicity counting (spontaneous fission rate of Pu items)
  - Calorimetry (heat production from decay of Pu bearing items)
- Through combination of Gamma-ray spectroscopy and either neutron multiplicity counting or calorimetry, mass of Pu bearing items can be measured.