
# Gamma spectrometric measurements of nuclear materials

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# Sponsors

- The Swedish Radiation Safety Authority
- Department of Defence
- The Swedish Armed Forces

# Agenda

- Nuclear materials?
- Why measure nuclear materials?
- Theory of isotopic measurements and age determination of nuclear materials
- Results from measurements of:
  - -Plutonium
  - -Uranium
  - -U-233,
  - -Am-241

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# Nuclear materials

- Uranium: DU, Nat. U, LEU, HEU (WGU)
- Plutonium: WGPu, FGPu, RGPu
- U-233
- Am-241
- (Np-237)

# Why?

- An important method for verification of nuclear materials in nuclear Safeguard.
- Method for characterization of nuclear materials in e.g. illicit trafficking scenarios.
- Using high resolution gamma spectrometry, such a characterization can be done already at e.g. an incident place.

# Gamma spectrometric measurement of nuclear materials

- NOT a sensitive method!!!
- However, a non-destructive method
- Isotopic composition and age can be measured on any sample, having any arbitrary shape and matrix
- The same for the age (time since last chemical separation)
- Commercial software availible, but deeper knowledge about the method generates research

# Theory

The efficiency: $$\Psi_{Ej} = \frac{C_{i,Ej}}{I_{i,Ej}\lambda_i N_i}$$ ← $A_i$

Isotope ratio: $$R_i = \frac{N_i}{N_k}$$

Count rate from measurement!

The relative efficiency: $$\Psi_{rel,Ej} = \frac{C_{i,Ej}}{I_{i,Ej}\lambda_i R_i}$$

Isotope ratio(s) as measurand(s)!

Empirical response fcn: $$\Psi_{rel}(E) = e^{c_1 + c_2/E^2 + c_3\cdot(\ln E)^2 + c_4\cdot(\ln E)^3 + c_5/E}$$

**Fit $R_i$ och $c_1$-$c_5$ by means of a least square method**

# Theory

- So, an internal response function is established on the very sample that is measured
- Has to be done on each sample (if shape and matrix differs)
- ➔The method is independant of shape and matrix due to this internal ”calibration”
- The sample has to be isotopic homogeneous

# Theory

**Examples of measurands (Ratios):**

**-Uranium:**

Composition: $R_{U238/U235}$, $R_{U234/U235}$, $R_{Th228/U235}$

Age: $R_{Bi214/U234}$

**-Plutonium:**

Composition: $R_{Pu238/Pu239}$, $R_{Pu240/Pu239}$, $R_{Pu241/Pu239}$

Age: $R_{Am241/Pu241}$ (from $R_{Am241/Pu239}$ and $R_{Pu241/Pu239}$)

**-U233:**

Composition: $R_{U232/U233}$

Age: $R_{Th229/U233}$

The composition can be calculated from ratios:

$$f_i = \frac{R_i}{\sum R_i}$$

*Example*:

$$f_{Pu238} = \frac{R_{Pu238/Pu239}}{R_{Pu238/Pu239} + R_{Pu239/Pu239} + R_{Pu240/Pu239} + R_{Pu241/Pu239}}$$

$$R_{Pu239/Pu239} = 1$$

# Theory: Age determination…

…or the time since the last separation, i.e. separation of daughter radionuclides.

For plutonium and U-233:

-Pu: 1=Pu241, 2=Am241

-U233: 1=U233, 2=Th229

$$t = \frac{1}{\lambda_1 - \lambda_2} \cdot \ln\left[1 - \left(\frac{\lambda_2}{\lambda_1} - 1\right)\frac{N_2}{N_1}\right]$$

$R_{2/1}$

# Theory: Age determination

For uranium the age is calculated from the ratio Bi-214/U-234:

$$^{234}U \xrightarrow{\alpha} {}^{230}Th \xrightarrow{\alpha} {}^{226}Ra \xrightarrow{\alpha} {}^{222}Rn \xrightarrow{\alpha} {}^{218}Po \xrightarrow{\alpha} {}^{214}Pb \xrightarrow{\beta^-} {}^{214}Bi... \longrightarrow {}^{206}Pb$$

…

$$\frac{A_{214Bi}(t)}{A_{234U}(t)} = \lambda_{230} \cdot \lambda_{226} \cdot \left[ \frac{e^{-\lambda_{234} \cdot t}}{(\lambda_{230} - \lambda_{234}) \cdot (\lambda_{226} - \lambda_{234})} + \frac{e^{-\lambda_{230} \cdot t}}{(\lambda_{234} - \lambda_{230}) \cdot (\lambda_{226} - \lambda_{230})} + \frac{e^{-\lambda_{226} \cdot t}}{(\lambda_{234} - \lambda_{226}) \cdot (\lambda_{230} - \lambda_{226})} \right]$$

After Taylor expansion:

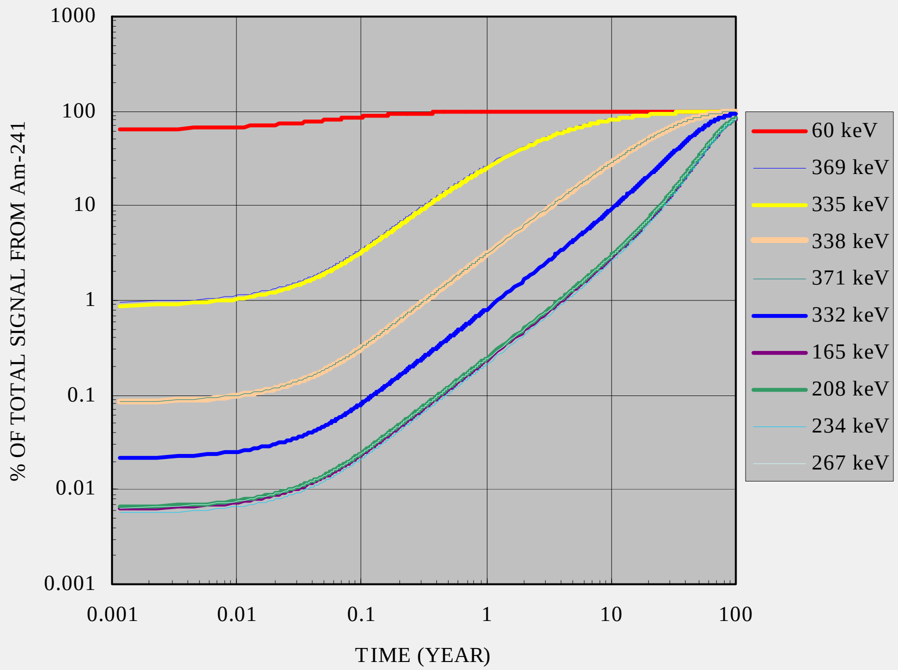
All other terms "small" (for t<100 y)

$$\frac{A_{214Bi}(t)}{A_{234U}(t)} = \frac{1}{24} \cdot t^2 \cdot \lambda_{230} \cdot \lambda_{226} \cdot \left[ t^2 \cdot \lambda_{234}^2 + t^2 \cdot \lambda_{234} \cdot \lambda_{230} + t^2 \cdot \lambda_{234} \cdot \lambda_{226} + t^2 \cdot \lambda_{230}^2 + t^2 \cdot \lambda_{230} \cdot \lambda_{226} + t^2 \cdot \lambda_{226}^2 - 4 \cdot t \cdot \lambda_{234} - 4 \cdot t \cdot \lambda_{230} - 4 \cdot t \cdot \lambda_{226} + 12 \right]$$

$$\Rightarrow \quad \frac{A_{214Bi}(t)}{A_{234U}(t)} = \frac{1}{2} \cdot t^2 \cdot \lambda_{230} \cdot \lambda_{226}$$

# Am-241 in Pu materials

- Pu-241 decays to Am-241, but also to U-237 (0.0025% BR).
  Both Am-241 and U-237 decay to Np-237 and populates in some cases the same energy levels, i.e. yields the same gamma ray energies:




However, e.g. 146.55 and 169.56 keV can be used for measurement of Am-241 (no interference from U-237, altough 146.55 keV has to be corrected for Pu-239)!

# Gamma rays from some Pu isotopes

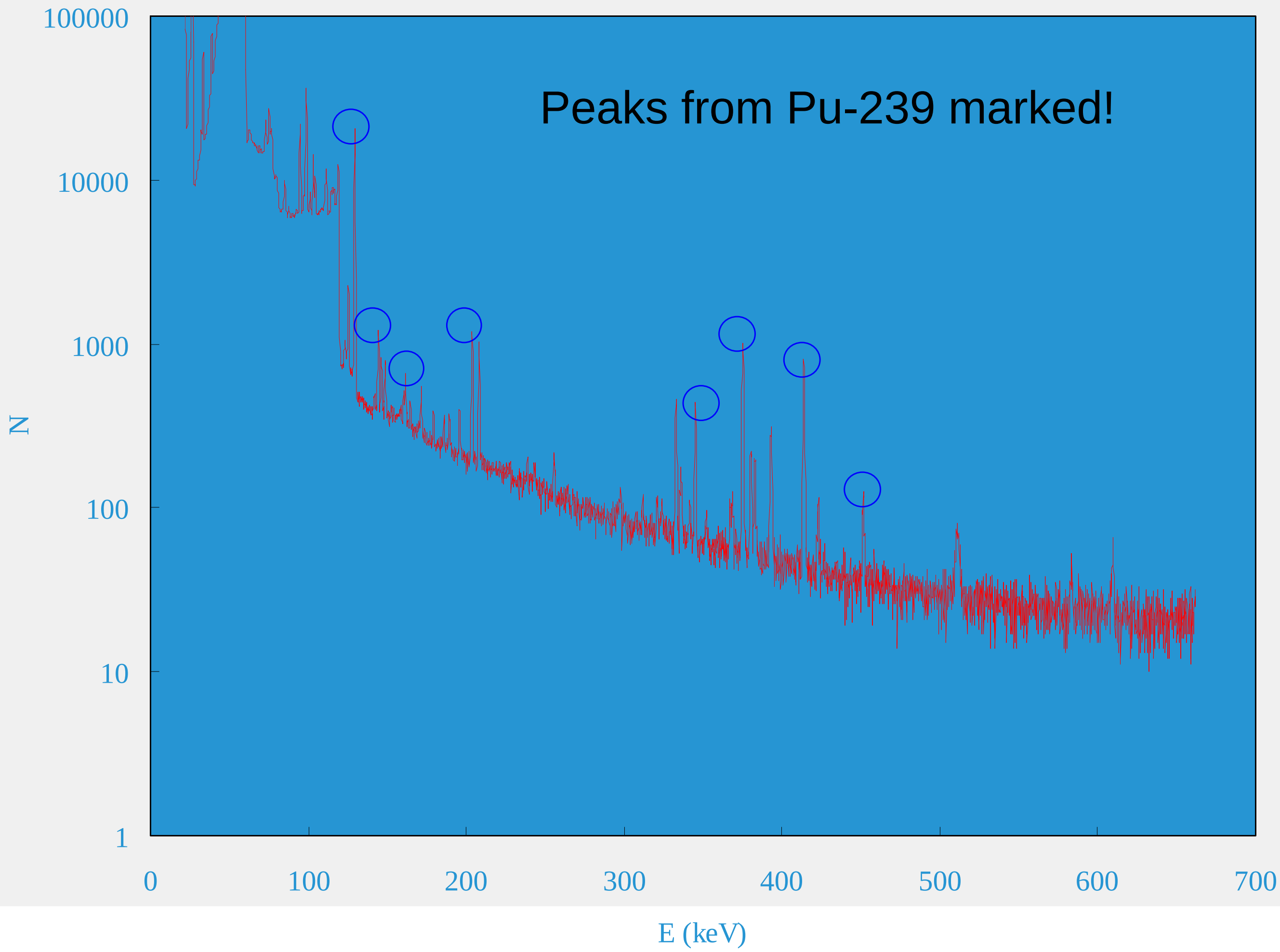
| A | Energy | BR (γ/decay) |
|---|---|---|
| 239 | 129.296 | 0.0000631 |
| 239 | 144.201 | 0.00000283 |
| 239 | 146.094 | 0.00000119 |
| 241 | 148.57 | 0.000001855 |
| 238 | 152.72 | 0.00000937 |
| 241 | 159.955 | 6.54E-08 |
| 240 | 160.308 | 0.00000402 |
| 239 | 161.45 | 0.00000123 |
| 241 | 169.56 | 0.00000173 |
| 239 | 195.679 | 0.00000107 |
| 239 | 203.55 | 0.00000569 |
| 239 | 345.013 | 0.00000556 |
| 239 | 375.054 | 0.00001554 |
| 239 | 413.713 | 0.00001466 |
| 239 | 451.481 | 0.00000189 |
| 240 | 642.35 | 0.00000013 |
| 239 | 645.94 | 0.000000152 |
| 238 | 766.39 | 0.00000022 |

**All weak!**

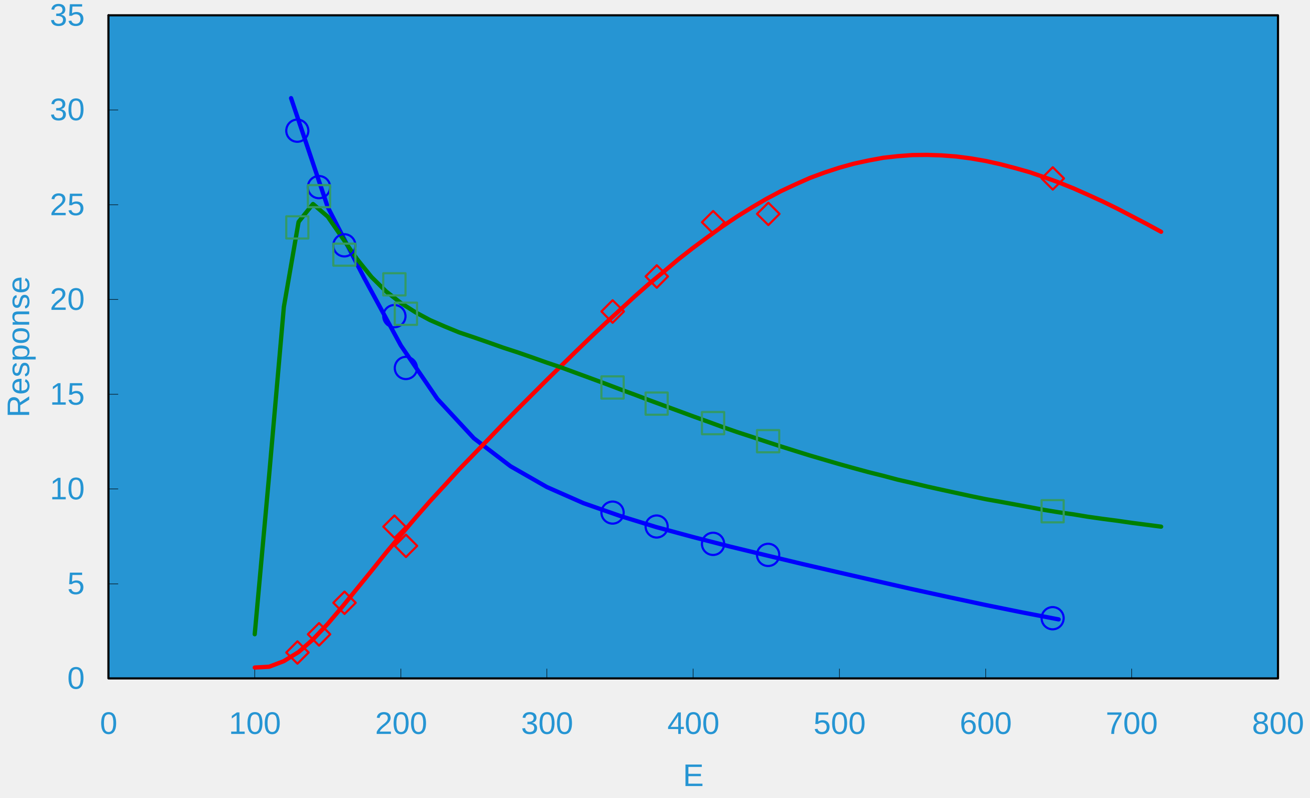
**Comparison:**
-1 µg Cs-137 emits 66 Mgammas per second@662 keV
-1 µg Pu-239 emits 3.5 gammas per second@129.3 keV

➔NOT a sensitive method!!!

# Pu spectrum


Peaks from Pu-239 marked!
100000
10000
1000
100
10
1
N
0
100
200
300
400
500
600
700
E (keV)

# Response functions for plutonium (Different detectors and materials)



# Result: Plutonium

Composition:

| Isotope | WGPu<br>γ-spectrometry | WGPu<br>ICP-SFMS+α-spectrometry<br>(REFERENCE METHOD!!!) |
|---|---|---|
| 238 | 0.0087±0.0039 | 0.00668±0.00020 |
| 239 | 94.68±0.21 | 94.483±0.021 |
| 240 | 5.26±0.50 | 5.541±0.021 |
| 241 | 0.055±0.003 | 0.05952±0.00060 |

**AGE:**

Gamma spectrometry: (36.8±2.1) y

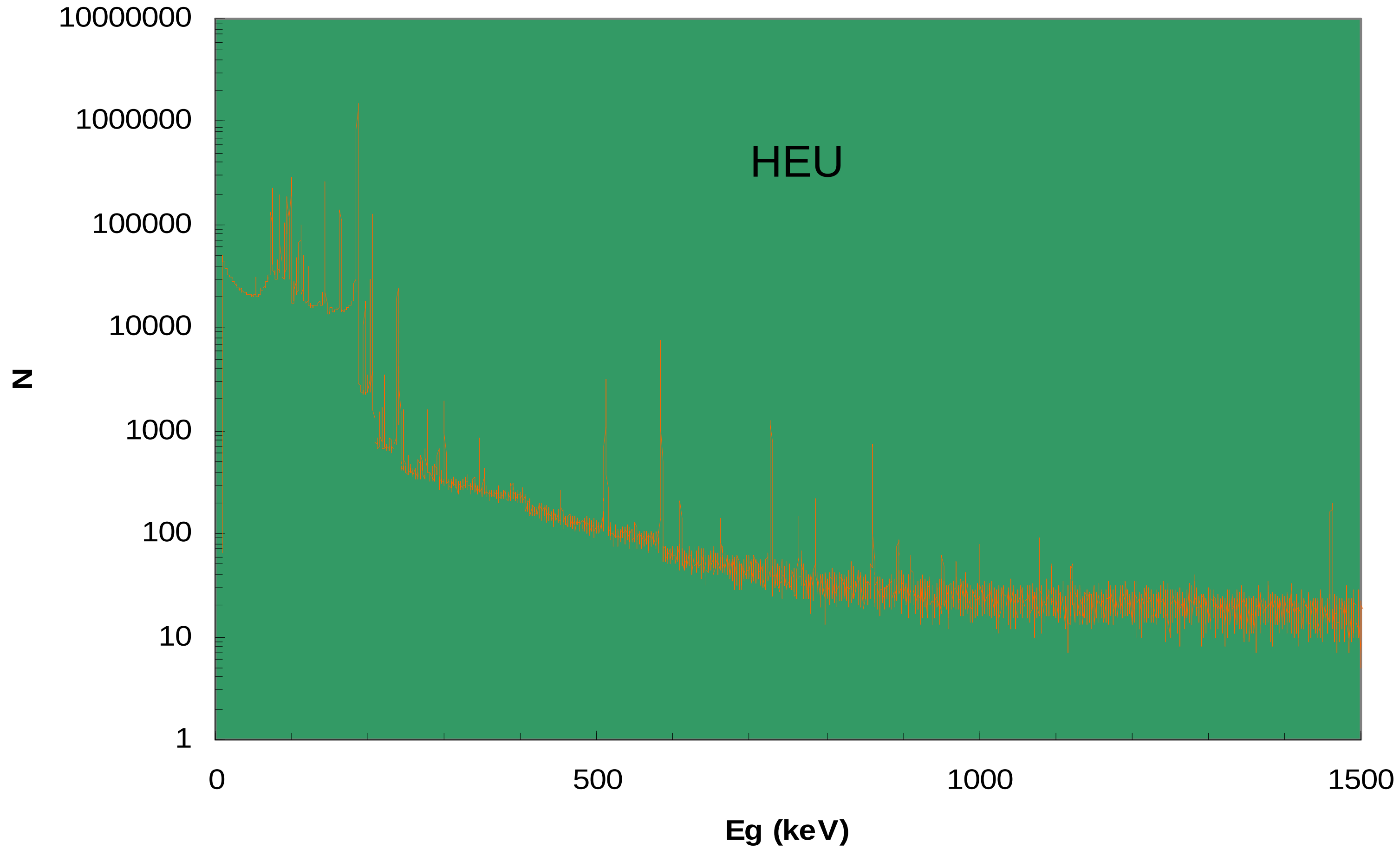
ICP-SFMS: (35.0±0.30) y

Separated around 1980!

# Gamma rays for uranium measurements

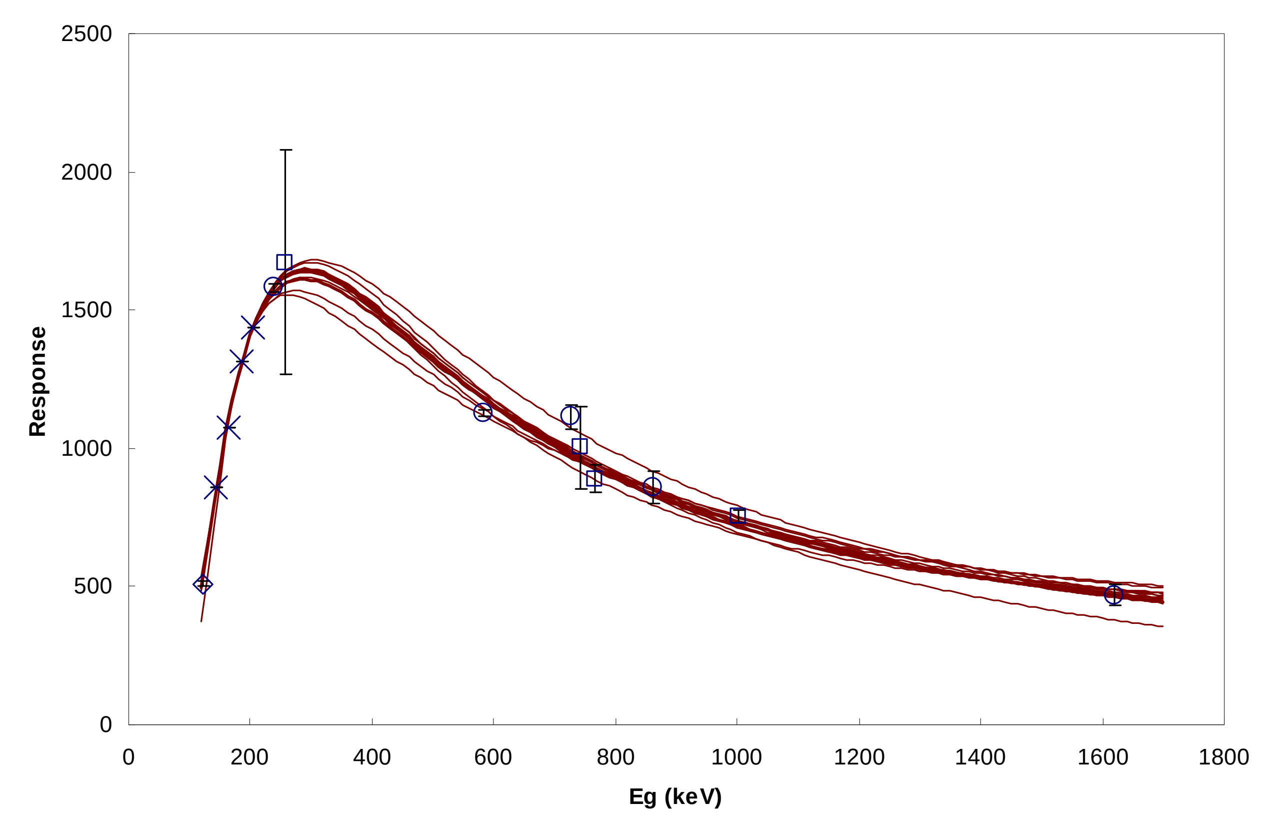
- U-234: 120.9 keV
- U-235: 143.8, 163.3, 185.7 och 205.3 keV
- U-238/Pa-234m: 258.3, 742.8, 766.4 och 1001 keV
- Th-288 (daughters): 238.6, 727.3, 1078.6, 1620.5, 583.2, 860.5 keV [Signature for U-232]
- Bi-241 (Age): 609 keV

# Uranium



HEU
N
Eg (keV)
10000000
1000000
100000
10000
1000
100
10
1
0
500
1000
1500

# Response function(s) for a HEU material



# Result: Uranium

Composition:

| Isotope | Gamma spectrometry | ICP-SFMS |
|---|---|---|
| U-234 | 1.28±0.23 | 1.1654±0.0062 |
| U-235 | 93.28±0.88 | 92.773±0.020 |
| U-238 | 5.44±0.79 | 6.061±0.017 |

**AGE:**
Gamma spectrometry: (47.5±7.5) y
ICP-SFMS: (46.0±0.50) y

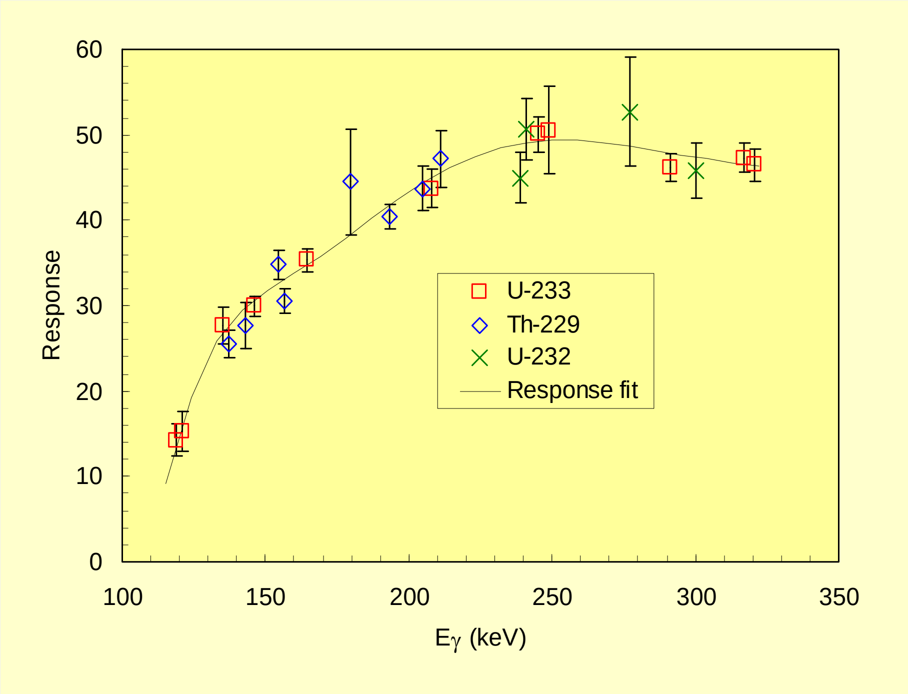
Separated around 1964!

# U-233

- U-233 may be used as e.g. nuclear fuel
- Significant amounts has been produced
- Importance will be increased if/when the Th-fuel cycle start to play a role

# U-233: Response function

gram-quantities of $^{233}U_3O_8$ (AEA-material):



# Results: $^{233}U$

**U-232 content:**

| Measurement | $^{232}U/^{233}U$ (IRMM-040a) | $^{232}U/^{233}U$ (Material 2) |
|---|---|---|
| Gamma spectrometry (LOAX) | $(3.14\pm0.15)\cdot10^{-6}$ | $(1.20\pm0.10)\cdot10^{-6}$ |
| Gamma spectrometry (TF2) | *not measured* | $(1.16\pm0.08)\cdot10^{-6}$ |
| Alpha spectrometry | $(3.06\pm0.14)\cdot10^{-6}$ | $(1.12\pm0.02)\cdot10^{-6}$ |

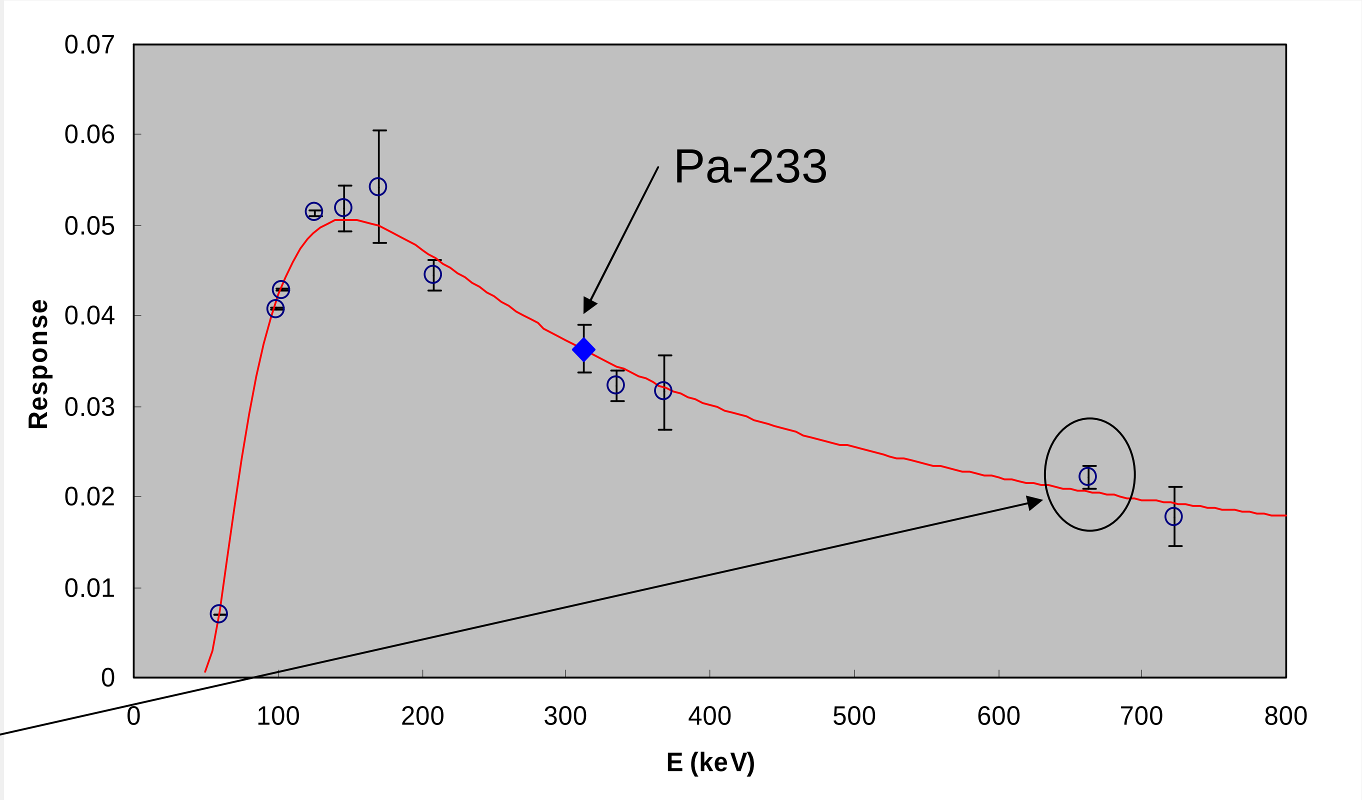
**AGE:**

| Measurement | Age of IRMM-040a | Age of Material 2 |
|---|---|---|
| Gamma spectrometry (LOAX) | (29.8±1.6) y | (41.2±2.6) y |
| Gamma spectrometry (TF2) | *not measured* | (42.7±3.4) y |
| ICP-MS | (29.9±0.4) y | (42.9±0.4) y |

Separated around: 1977 1964

# ”Curiosity”: Am-241

Measurement of a smoke detector:




662 keV not to be mistaken for Cs-137!

**Separation around 1983!**

# Thank you!