PHOTON EMISSION INTENSITIES IN THE DECAY OF U-235

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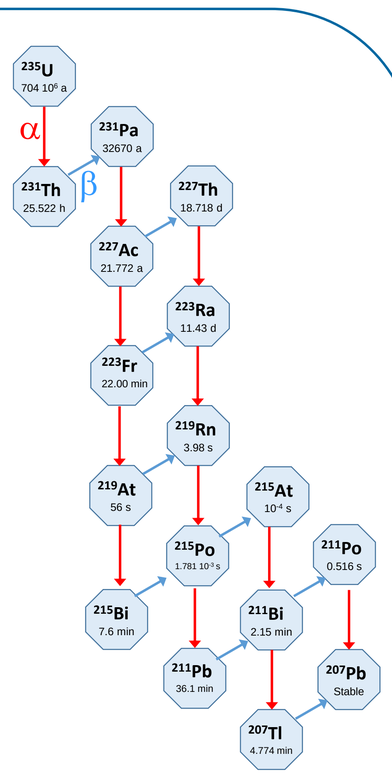
Introduction

- Uranium-235 is the parent of one of the natural radioactive decay series and appears in the background of any spectrometer and is also classed as a NORM (Naturally Occurring Radioactive Material).
- Uranium-235 decay is characterized by about fifty gamma-rays with E < 450 keV, most of them with weak emission intensities (< 1%).
- Probably due to the weak specific activity of U-235 and its presence in any background measurement, only a few experiments have been conducted to measure the photon emission probabilities associated to the U-235 decay.
- Recommended values are based on the intensity of the 185.72-keV gamma ray, used to normalize relative measurements.



235U decay chain

Source characteristics and standardization

Source prepared at JRC-IRMM by deposition of a U-235 solution on glass plate.
Impurities quantified by isotope dilution mass spectrometry (U-234: 5.3%, U-236: 0.110% and U-238: 0.00005%)

Standardization carried out by alpha counting in a defined solid angle (DSA) geometry, using a partially depleted PIPS detector (Canberra) in a vacuum chamber.

Source activity, *A*, is defined as: $A = \frac{N \cdot F_C}{t \cdot G}$

- N: net counts recorded by the detector
- t: acquisition live time
- Fc=0.9458 (7): corrective factor for impurities
- Geometrical factor, $G=\Omega/4\pi$
  (Ω being the solid angle), derived from:
  source-to-collimator distance: 166.29 (21) mm,
  collimator radius: 20.255 (2) mm,
  source radius: 7.8 (20) mm,

- G is defined with 0.26% relative combined standard uncertainty.

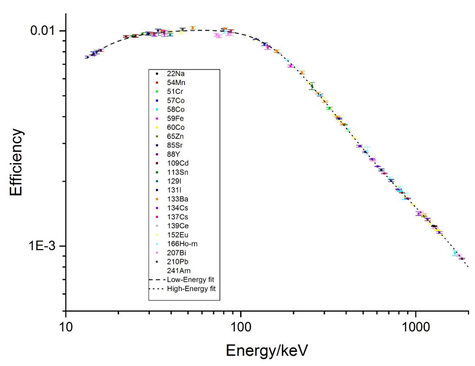
Source activity: A = 48.41 (34) Bq

Contribution	Relative standard uncertainty
Statistics	4.13 e-3
Extrapolation	3.0 e-3
Geometrical factor	2.6 e-3
Backscattering (chamber)	1.0 e-3
Backscattering (window)	1.0 e-5
Counting time	1.0 e-4
Temperature	6.0 e-5
Impurities	7.4 e-4
Reproducibility	3.5 e-3
Total	**6.8 e-3**

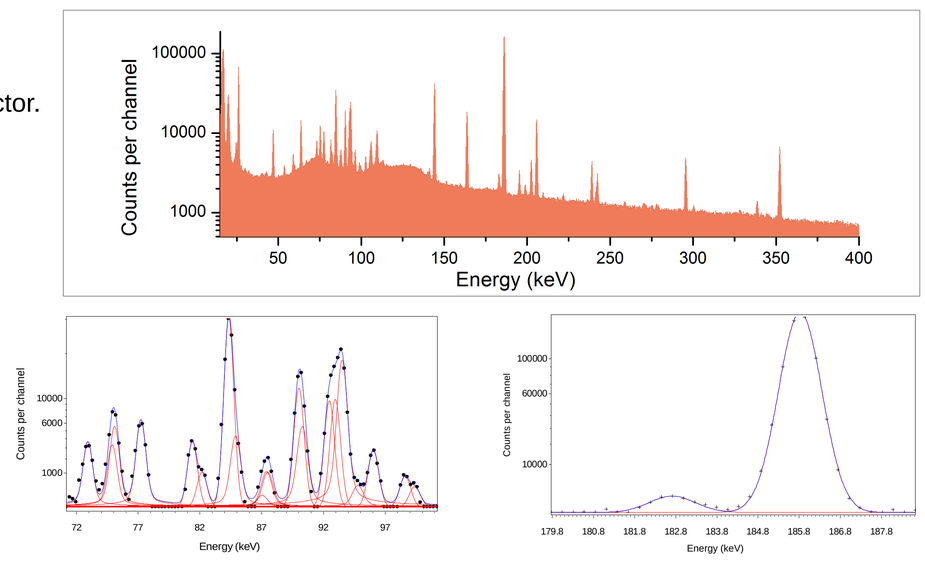
Uncertainty budget for the source activity

Gamma-ray spectrometry

Measurements performed using high-purity germanium detector.
Source at 10 cm from the detector window.



Full-energy peak efficiency calibration



235U spectrum (top) and processing of X-rays (left) and 185.72-keV (right) and regions of interest

Contribution	Relative standard uncertainty
Source activity	7.0 e-3
Statistics	1.3 e-3
Peak area determination	2.0 e-3
Efficiency	1.0 e-2
Counting time and radionuclide decay	negligible
Geometry	1.0 e-3
Coincidence summing	1.0 e-4
Reproducibility	2.8 e-3
Total	**1.28 e-2**

Uncertainty budget for the absolute photon emission intensity of the reference peak (185.72 keV)

Results

For each peak with energy E_i, the absolute intensity, I_i is computed as:

$$I_i = \frac{N_i \cdot C_{Gi} \cdot C_{Ci}}{A \cdot \varepsilon_i \cdot t}$$

- N_i: net counts of the full-energy peak, obtained using GAMMAVISION® for well isolated peaks or COLEGRAM for the complex regions,
- C_{Gi}: correction for source geometry,
- C_{Ci}: correction for coincidence summing effects,
- A: source activity (Bq),
- ε_i: full-energy peak efficiency,
- t: acquisition live time (2.8 10^6 seconds).

Reference	Experimental value (%)	Comment
Vaninbroukx and Deneke (1982)	57.5 (9)	Enriched material - Activity: mass spectrometry isotope dilution and alpha counting
Olson (1983)	56.1 (8)	Enriched material (97.66%) – NBS standard
Helmer and Reich (1984)	57.2 (5)	NBS standard
Lin and Harbottle (1991)	56.8 (13)	Enriched material (92.8%)
Chatani (1999)	*58 (2)*	*Indirect measurement - U-Al alloy and purified ^{231}Th source*
Al-Saleh et al. (2006)	*80.19 (4)*	*Uranyle nitrate hexahydrate – powder sample*
Present study	**57.6 (7)**	**Enriched material (94.58%)**

Comparison of experimental photon emission intensities for the reference peak (185.72 keV)

The relative photon emission intensities, I_{iR}, are computed using the 185.72-keV peak (M) intensity as normalization factor:

$$I_{iR} = \frac{\frac{N_i C_{Gi} \cdot C_{Ci}}{\varepsilon_i \cdot A}}{\frac{N_M C_{GM} \cdot C_{CM}}{\varepsilon_M \cdot A}} \cdot 100 = \frac{N_i C_{Gi} \cdot C_{Ci}}{N_M C_{GM} \cdot C_{CM}} \cdot \frac{\varepsilon_M}{\varepsilon_i} \cdot 100$$

Energy (keV)	Relative intensity (%)	Absolute intensity (%)	
	This study	This study	NUCLÉIDE
Th Ll X-rays	1.036 (26)	0.599 (15)	
Th Lα X-rays	35.1 (8)	20.26 (44)	
Th Lβ X-rays	30.1 (6)	17.42 (38)	
Th Lγ X-rays	4.20 (9)	2.43 (5)	
Th total L X-rays	70.4 (17)	40.7 (10)	40 (22)
72.7	0.506 (10)	0.292 (6)	0.116 (20)
74.94	0.755 (14)	0.436 (8)	0.051 (6)
Th Kα2 X-ray	3.62 (5)	2.087 (29)	3.56 (7)
Th Kα1 X-ray	7.78 (10)	4.49 (6)	5.75 (11)
96.09	1.045 (17)	0.603 (10)	0.091 (11)
Th Kβ'1 X-rays	2.96 (5)	1.706 (28)	2.05 (5)
Th Kβ'2 X-rays	0.947 (12)	0.546 (7)	0.690 (19)
109.19	2.55 (6)	1.469 (33)	1.66 (13)
115.45	0.0441 (5)	0.0255 (3)	0.03 (1)
136.55	0.124 (10)	0.071 (6)	0.012
140.76	0.257 (22)	0.148 (13)	0.20 (1)
143.767	18.81 (24)	10.84 (14)	10.94 (6)
150.936	0.095 (23)	0.055 (13)	0.09 (3)
163.356	8.57 (12)	4.94 (7)	5.080 (3)
182.62	0.750 (25)	0.432 (14)	0.39 (5)
185.72	100.0 (12)	57.6 (7)	57.1 (3)
194.94	1.19 (3)	0.685 (18)	0.63 (1)
202.12	2.09 (4)	1.204 (25)	1.08 (2)
205.316	8.90 (12)	5.12 (7)	5.02 (3)
221.386	0.176 (34)	0.101 (20)	0.118 (5)
240.88	0.14 (4)	0.082 (23)	0.074 (4)
275.35 275.49	0.10 (7)	0.059 (43)	0.051 (6) 0.032
291.65	0.12 (1)	0.0677 (45)	0.040 (6)
343.54	0.024 (3)	0.0139 (16)	0.0032
345.92	0.054 (5)	0.0309 (26)	0.040 (6)
387.84	0.052 (5)	0.0299 (29)	0.040 (6)

Relative and absolute photon emission intensities in the decay of U-235, and comparison with NUCLÉIDE database

Due to the equilibrium between U-235 and Th-231 it was also possible to determine some photon emission intensities associated to the decay of Th-231.

Energy (keV)	Relative intensity (%)	Absolute intensity (%)	
	This study	This study	NUCLÉIDE
Pa Ll X-rays	2.88 (7)	1.66 (4)	
Pa Lα X-rays	50.9 (11)	29.4 (6)	
Pa Lβ X-rays	49.9 (11)	28.8 (6)	
Pa Lγ X-rays	8.53 (18)	4.93 (11)	
Pa total L X-rays	112.2 (24)	64.8 (14)	65 (3)
25.64	24.14 (31)	13.95 (18)	13.9 (7)
58.57	0.76 (9)	0.44 (5)	0.480 (16)
81.228	1.268 (41)	0.731 (24)	0.905 (23)
82.087	0.349 (21)	0.201 (12)	0.418 (13)
84.214	11.61 (16)	6.70 (10)	6.70 (7)
Pa Kα2 X-ray	3.56 (5)	2.05 (39)	0.37 (4)
99.278	0.134 (5)	0.077 (3)	0.137 (6)
102.27	0.750 (14)	0.433 (8)	0.441 (11)
Pa Kβ'1 X-ray	0.595 (12)	0.343 (7)	0.21 (2)
163.101	0.272 (7)	0.157 (4)	0.156 (5)

Relative and absolute photon emission intensities in the decay of Th-231, and comparison with NUCLÉIDE database

Conclusion

The reference photon emission intensity (185.72 keV) is obtained with 1.28% relative combined uncertainty. Most of the present results agree with the tabulated data.
Some discrepancies are noticed in the 70-keV and 90-keV energy regions: the complex spectrum structure with gamma- and K X-rays from different nuclides, together with the strong influence of the background and uranium self-fluorescence make it difficult to unambiguously attribute the counting to individual lines.
It is expected that these new experimental values will provide helpful information for further evaluation of the decay schemes of U-235 and Th-231.

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