Reference-free measurement of photon emission intensities in the decay of standard radionuclides

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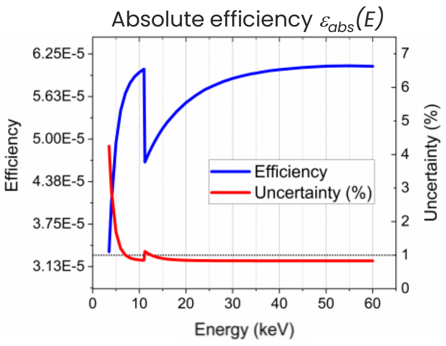
INTRODUCTION

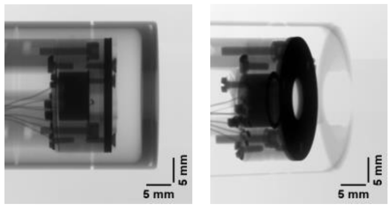
Reliable X-ray emission intensities are essential for radionuclide quantification and the efficiency calibration of energy-dispersive spectrometers. Conventional calibration methods rely on standard radionuclides whose emission intensities are derived from prior decay data, based on tabulated fluorescence yields and theoretical values for internal conversion coefficients or electron capture probabilities. As a result, they are fully correlated, leading to a dependence on previously known intensities when measuring new ones. To overcome this limitation, we applied a calibration method independent of prior decay data for a high-purity germanium (HPGe) detector and used it to directly measure X-ray and selected γ-ray emission intensities, along with their uncertainties, for a set of standard radionuclides in the 5.4 keV to 53.13 keV range.

EFFICIENCY WITHOUT REFERENCE TO DECAY DATA

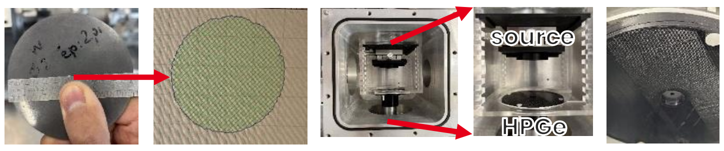
Absolute efficiency of the detector, $\varepsilon_{abs}(E)$, is calculated as $\varepsilon_{abs}(E) = \varepsilon_{int}(E) \cdot \varepsilon_{geom} = \varepsilon_{int}(E) \cdot \Omega / 4\pi$

- $\varepsilon_{int}(E)$: Intrinsic efficiency, depending on the energy E, is measured using a monochromatic photon beam calibrated with an electrically substituted cryogenic radiometer and transfer photodiodes [Elvira, 2024].
- ε_{geom} : Geometrical efficiency is determined from the solid angle Ω which is derived, with 0.83% unc., using radiographically determined detector geometry and an external tungsten collimator, characterised with a micrometer for its thickness and a calibrated vision machine for its diameter.





HPGe detector	Manufacturer data	Radiography data
Crystal thickness	6 mm	6.116(43) mm
Crystal diameter	10 mm	9.786(43) mm
Dead layer thickness	-	1 µm [Elvira, 2024]
Beryllium window thickness	125 µm	131(7) µm
Internal collimator diameter (0.4/1/1 mm thick W/Cu/Al)	8 mm	7.7(9) mm 11.68% unc.
Crystal-to-window distance	10 mm	12.46(10) mm
Window-to-collimator rear distance	-	10.409(22) mm 0.21% unc.



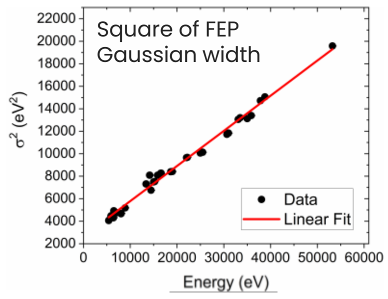
SPECTRAL PROCESSING

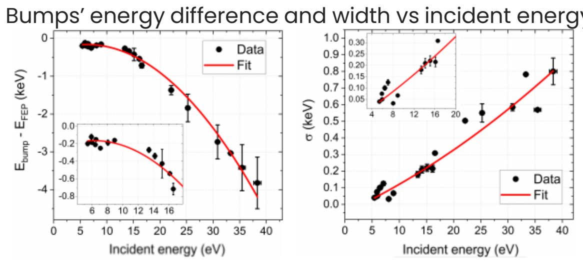
X-ray spectrometry is performed on point sources prepared at LNE-LNHB, at 6.4 cm distance. Measurement durations ranged from 3 to 11 days, depending on the source activity, to accumulate sufficient statistics.

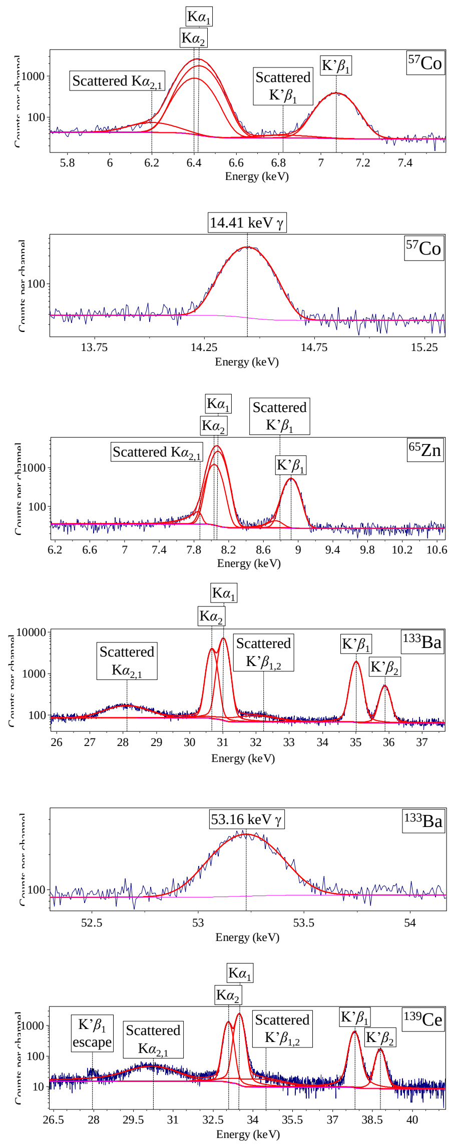
Spectrum analysis is carried out using COLEGRAM software with the following fitting functions:

- Voigt function for the K X-ray full-energy peak (FEP) accounting for
  - Gaussian instrumental broadening
  - Lorentzian natural X-ray linewidth
- Gaussian with a left-sided tail for "bumps" associated with Compton scattered photons
- Simple Gaussian for γ-ray peaks

Low-energy X-ray FEP (< 10 keV) are more affected by bump disturbances, introducing up to 0.5% differences in net peak areas, which are included as additional uncertainty contributions.







RESULTS

Absolute emission intensities, $I(E)$, of X-rays or γ-rays are derived as

$$I(E) = \frac{N(E) \cdot \prod_{i=1}^{n} C_i}{\varepsilon_{abs}(E) \cdot A \cdot t}$$

where

- $N(E)$: Net counts in the FEP corresponding to photons of interest with energy E,
- A : Source activity, standardised by various methods available at LNE-LNHB,
- t : Acquisition live time,
- $\varepsilon_{abs}(E)$: Absolute detection efficiency for FEP with energy E,
- C_i : Correction factors associated with
  - decay between the measurement and reference dates,
  - decay during the measurement,
  - transmission through Mylar foil and air.
- ✓ Contributions of individual relative standard uncertainties to the combined values are derived following the uncertainty propagation procedure described in the Guide to the Expression of Uncertainty in Measurement (GUM) [JCGM, 2008].

Uncertainty budget for photon emission intensity

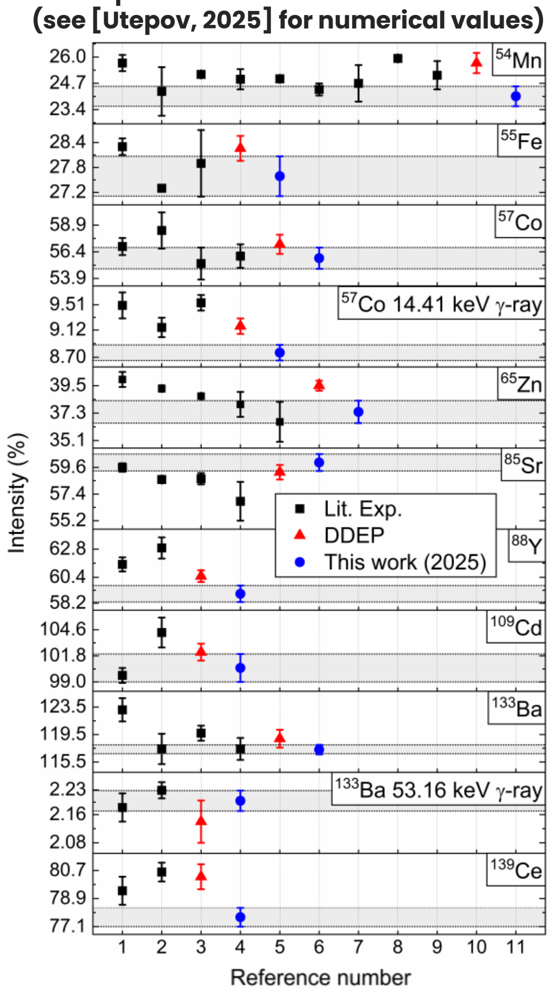
Origin	Relative standard uncertainty		
	$XK\alpha$	$XK\beta$	γ-ray
Activity	0.33% – 1.6%		≤ 0.43%
Counting statistics & Spectral deconvolution	< 1.0% – 2.5%	< 1.5% – 3.3%	< 1.0%
Decay during measurement	< 0.01%		
Decay correction to a reference date	< 0.10%		
Geometrical efficiency	0.83%		
Intrinsic efficiency	< 1.2%		< 0.40%
Transmission through Mylar foil and air	≤ 1.4%		≤ 0.17%
Combined uncertainty	**≤ 2.7%**	**≤ 3.4%**	**≤ 1.4%**

Radionuclide point source	Relative standard uncertainty for activity
^{54}Mn	0.33%
^{55}Fe	0.40%
^{57}Co	0.32%
^{65}Zn	0.34%
^{85}Sr	0.75%
^{88}Y	0.29%
^{109}Cd	1.60%
^{133}Ba	0.43%
^{139}Ce	0.56%

Partial K X-ray and selected γ-ray emission intensities measured in this work

Radionuclide	Photon type	Intensity (%)	Relative standard uncertainty
^{54}Mn	$XK\alpha_{2,1}$ (Cr)	21.23(49)	**2.3%**
	$XK'\beta_1$ (Cr)	2.83(6)	**2.3%**
^{55}Fe	$XK\alpha_{2,1}$ (Mn)	24.25(47)	**1.9%**
	$XK'\beta_1$ (Mn)	3.34(7)	**2.0%**
^{57}Co	$XK\alpha_{2,1}$ (Fe)	49.1(10)	**1.9%**
	$XK'\beta_1$ (Fe)	6.72(15)	**2.2%**
	14.41 keV γ-ray (^{57}Fe)	8.77(12)	**1.4%**
^{65}Zn	$XK\alpha_{2,1}$ (Cu)	32.9(9)	**2.7%**
	$XK'\beta_1$ (Cu)	4.47(15)	**3.4%**
^{85}Sr	$XK\alpha_{2,1}$ (Rb)	51.0(7)	**1.4%**
	$XK'\beta_1$ (Rb)	8.07(12)	**1.5%**
	$XK'\beta_2$ (Rb)	0.898(18)	**2.0%**
^{88}Y	$XK\alpha_{2,1}$ (Sr)	49.9(7)	**1.4%**
	$XK'\beta_1$ (Sr)	8.08(12)	**1.5%**
	$XK'\beta_2$ (Sr)	1.006(20)	**2.0%**
^{109}Cd	$XK\alpha_{2,1}$ (Ag)	82.8(15)	**1.9%**
	$XK'\beta_1$ (Ag)	14.90(29)	**1.9%**
	$XK'\beta_2$ (Ag)	2.78(6)	**2.2%**
^{133}Ba	$XK\alpha_2$ (Cs)	33.26(35)	**1.0%**
	$XK\alpha_1$ (Cs)	61.4(6)	**1.0%**
	$XK'\beta_1$ (Cs)	18.15(18)	**1.0%**
	$XK'\beta_2$ (Cs)	4.53(5)	**1.1%**
	53.16 keV γ-ray (^{133}Cs)	2.199(29)	**1.3%**
^{139}Ce	$XK\alpha_2$ (La)	21.63(29)	**1.4%**
	$XK\alpha_1$ (La)	40.6(5)	**1.3%**
	$XK'\beta_1$ (La)	12.32(16)	**1.3%**
	$XK'\beta_2$ (La)	3.14(5)	**1.6%**

Total K X-ray and the indicated γ-ray emission intensities measured in this work compared with those in the literature (see [Utepov, 2025] for numerical values)



CONCLUSION

K X-ray emission intensities were determined for nine radionuclides, along with γ-ray emission intensities for the 14.41 keV transition of ^{57}Co and the 53.16 keV transition of ^{133}Ba. The results agree well with existing data, although some differences are observed, likely due to uncertainties in decay data and the use of different standard radionuclides. The reference-free method used in this work avoids these limitations and is, to our knowledge, the first time that such a method has been used for measuring absolute emission intensities of low-energy photons in radioactive decay. The uncertainty for the total K X-ray intensity of ^{133}Ba was improved to 0.6%, while uncertainties for other radionuclides remain comparable. For all investigated intensities, a fully traceable uncertainty budget, derived according to the GUM, has been provided.

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