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Consistency of photon emission intensities for efficiency calibration of gamma-ray spectrometers in the energy range from 20 keV to 80 keV

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HIGHLIGHTS

- Efficiency calibration of four HPGe detectors using standard radioactive sources.
- Accurate processing of the full-energy peaks and Monte Carlo simulation.
- Absolute photon emission intensities derived for Ba-133.
- Absolute K X-ray emission measured for Sr-85, Y-88, Cd-109, Eu-152 and Ho-166 m.

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ABSTRACT

The efficiency calibration for different high-purity germanium detectors in the low-energy range was established by the conventional method, using standard radioactive sources. The peak shapes were carefully analysed taking account of natural linewidth, full-energy width at half maximum and scattering. Complementary information was obtained by Monte Carlo simulation using the PENELOPE code, after optimization of the geometrical parameters. This was used to measure photon emission intensities of some low-energy emitting radionuclides, including ^{133}Ba, and compared to the tabulated values.

1. Introduction

Efficiency calibration of X-ray spectrometers in the low-energy range has been a topic of concern for many years (*e.g.,* Campbell and McGhee, 1986; Debertin and Helmer, 1988). Two kinds of energy-dispersive detectors are generally used in this energy range, both based on semi-conductor materials (silicon and germanium). Lithium-drifted silicon detectors (Si(Li)), with thickness up to 5 mm, were preferably used in the low-energy range, with the drawback that they require liquid nitrogen cooling. Today, the technology of silicon drift detectors (SDD) makes it possible to benefit from electrical Peltier cooling, but the energy range is still restricted to energies below 50 keV, because of the thinness of the silicon wafer (500 µm). High-purity germanium (HPGe) detectors are very commonly used and planar or semi-planar crystal shapes have been proposed to extend the traditional energy range of coaxial detectors down to 5 keV (Martin and Burns, 1992). Conventional efficiency calibration of the spectrometers relies on experimental measurement of standard radionuclides. Complementary approaches based on Monte Carlo simulation have also been developed (see *e.g.* Helmer et al., 2003; Peyres and García-Toraño, 2007; Sima and Arnold, 2009). Maidana et al. (2013) performed a mixed study utilizing an analytical expression as proposed by Seltzer (1981) which takes into account the different interactions in the detector. Plagnard et al. (2007) used a tunable monochromatic X-ray source and a reference proportional counter in an innovative experiment to calibrate the intrinsic efficiency of an HPGe detector, without any radioactive source. Any alternative approach nevertheless requires validation with standard sources to check the consistency of calculation with experiment and/or to determine the effective detection solid angle. Barium-133 and americium-241 are frequently used in the low-energy range, where only a few gamma-emitting radionuclides are available for this purpose. But we observed systematic deviations, with relative differences of about 2–3%, between the full-energy peak (FEP) efficiencies experimentally obtained for the peaks at 53 keV (^{133}Ba) and 59.6 keV (^{241}Am), despite the small energy gap between them. It was then decided to collect further information and to examine in detail the experimental FEP calibration in the low-energy range for HPGe detectors with different sizes to improve the consistency of the photon emission intensities in the 20–80 keV energy region.

2. Experimental conditions

At the Laboratoire National Henri Becquerel (LNHB), X-ray

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Table 1
Main characteristics of the HPGe detectors.

HPGe detector	G1	G2	G3	G4
Crystal geometry	Coaxial	Coaxial	Planar	Planar
Crystal diameter	49.5 mm	48.7 mm	15.1 mm	2 mm
Crystal thickness	47.8 mm	55.4 mm	6.7 mm	1 mm
Dead layer thickness	0.3 µm	0.3 µm	0.8 µm	0.7 µm
Beryllium window thickness	500 µm	500 µm	127 µm	10 µm
Energy resolution (Full Width at Half Maximum at 5,9 keV)	–	–	225 eV	140 eV
Energy resolution (Full Width at Half Maximum at 59 keV)	670 eV	690 eV	370 eV	330 eV
Calibration distance (source-to-detector window)	10.7 cm	10.3 cm	7.8 cm	3.7 cm (In vacuum)

spectrometry is performed with N-type HPGe detectors. Four detectors are available for the present study and their main characteristics are reported in Table 1. Each spectrometer is calibrated with standard point sources installed at the reference distance. For each energy, E, the full-energy peak (FEP) efficiency, $\varepsilon(\boldsymbol{E})$, is derived from the net counting, N (E) according to:

$$\varepsilon(E) = \frac{N(E)}{A \cdot I(E) \cdot t} \prod_i C_i \quad (1)$$

where:

A is the activity of the standard radionuclide (Bq),
I(E) is the emission intensity of the line with energy *E,*
t is the acquisition duration (live time) (seconds),
C_i stands for different correction factors (radioactive decay, coincidence summing, etc.).

The standard sources are prepared at LNHB from radioactive solutions calibrated by primary methods, thus their activity is characterized with standard relative uncertainty of 0.5% or less. For the practical use, to derive the efficiency for any energy, the set of raw data (energies and efficiencies) is fitted using a log-log polynomial function, taking into account the correlations between input data. An example of the experimental data and the resulting calibration curve in the range from 12 keV to 120 keV for detector G1 is plotted in Fig. 1. Relative standard combined uncertainties are about 1–2% in the energy range of interest. The fitted calibration curves versus the energy for the four detectors are compared in Fig. 2. Whereas, the efficiency of the two coaxial detectors decreases strongly in the low-energy part, it keeps a rather stable value in the range from 40 keV to 60 keV. On the other hand, the efficiency of the smaller detector is quite constant between 20 keV and 40 keV and then decreases due to its small active thickness. The present study focusses on two topics: 1) The ^{133}Ba absolute gamma-ray intensities based on data recorded with the two large coaxial detectors (G1 and G2) and experimental efficiency calibration; and 2) the KX-ray emission intensity ratios of selected nuclides based on the information derived from measurements and Monte Carlo simulations with the smaller detectors (G3 and G4).

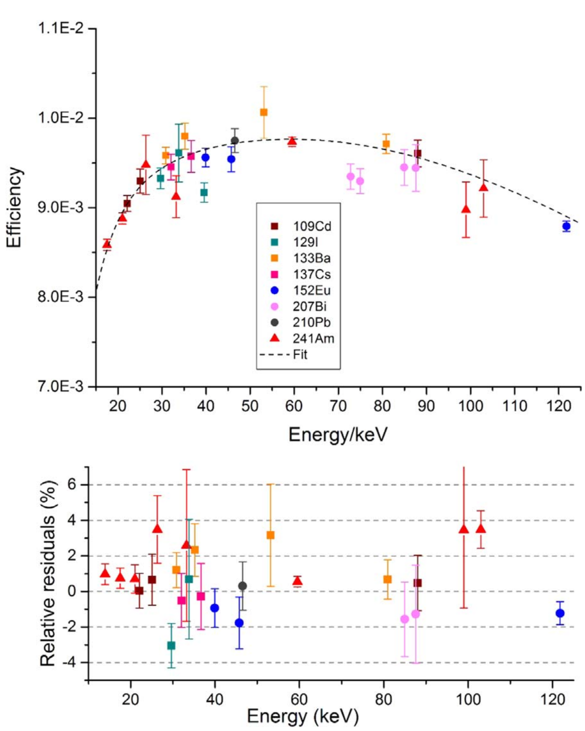

Fig. 1. Efficiency calibration of detector G1 for point sources – Experimental values and fitted curve. The relative residuals are plotted in the lower panel.

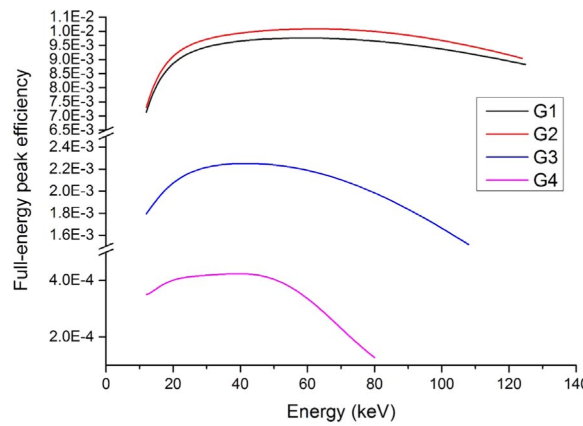

Fig. 2. Comparison of the efficiency calibration for the 4 HPGe detectors for point sources.

3. Peak processing

The net areas of the full-energy peaks were obtained with the COLEGRAM software (Ruellan et al., 1996), which proposes different mathematical functions to fit the experimental points by the least-squares minimization method, and paying particular attention to the following:

1. Natural linewidth of the incident photons;
2. Full-width at half maximum versus energy;
3. Scattering effects.

As discussed early by Debertin and Pessara (1981), one has to distinguish between peaks shapes resulting from interactions of X-rays and the ones induced by gamma-rays. The observed peak width results from the convolution of the so-called natural line width, due to the intrinsic nature of the incident photons, by the Gaussian broadening induced by the charge pair creation statistics. The energy distribution of a gamma emission, due to transitions between nuclear levels is well defined and has a Lorentzian shape whose width is a few meV. The X-ray emission is linked to atomic relaxation and the intrinsic Lorentzian width ranges from some eV to some tens of eV, depending on the atomic number of the element, Z, and on the electronic (sub)-shells concerned in the atomic rearrangement. Consequently, according to the energy range and the detector energy resolution, the X-ray linewidth cannot always be considered as negligible and must be taken into account in the fitting procedure used. Thus a Voigt function (convolution of a Lorentzian function by a Gaussian shape) is used:

$$V(E) = \int_{-\infty}^{+\infty} L(E') \cdot G(E - E') \cdot dE' \quad (2)$$