THE DIRECT ACTIVITY MEASUREMENT OF ^{133}Ba BY USING HPGe SPECTROMETER

by

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The direct measurement of ^{133}Ba source activity by the application of the theoretical count rate equations has been recently developed by Novković *et al*. The analytical and Monte Carlo calculation of activity measurement uncertainties were described in previously published papers for the case of one recorded spectra. The procedure of uncertainty calculation for a sequence of successively recorded spectra, enabling the determination of the correlation between peak count rates, is presented in this paper. The uncertainty of activity obtained by the described method is caused by the uncertainty of the experimentally obtained input data and uncertainties of the used nuclear decay data.

Key words: ^{133}Ba, coincidence summing, measurement uncertainty, HPGe spectrometer

INTRODUCTION

The calibration of a semiconductor HPGe spectrometer could be performed by using a mixture of radionuclides. The most appropriate mixture contains radionuclides whose decays are followed by the emission of one or two photons with energies covering a wide energy range. Radionuclides emitting a number of photons, like ^{133}Ba, are not convenient due to the appearance of a numerous sum peaks. This problem has been observed from the very beginning of using the germanium spectrometer, and many papers refer to this topic [1-4]. In the recently published papers [5, 6], we have developed a new approach which could completely solve the problem, at least at the mathematical level. This method enables:

(1) identification of all possible decay paths,
(2) identification of possible decay path outcomes,
(3) calculation of probabilities of particular path outcomes and their deposited energy in the detector, and
(4) determination of theoretical expressions for count rates for each peak (excluding escape peaks) and also for the total count rate. The unknown quantities in these expressions are the total and peak detection efficiencies. The coefficients of the unknowns are constants which characterize the measured radionuclide and their values are taken from the literature. The formula for the total count rate is of crucial importance for a direct activity measurement.

In the case of a radionuclide with the simplest decay scheme, like ^{139}Ce, the explicit formula for activity could be derived from the count rate expressions. This formula contains only count rates for the single and sum peaks and also the total count rate, and enables the absolute activity measurement of these radionuclides. The application of this method to radionuclides ^{57}Co and ^{133}Ba is explained in detail in references [7, 8].

In the experimentally obtained spectra of ^{57}Co and ^{133}Ba, the number of peaks is larger than the number of unknowns. This could be used for the determination of detection efficiency and also for source activity calculation. Analytical and Monte Carlo methods for the calculation of activity uncertainty determination in the case of only one recorded spectrum are given in detail in reference [9]. In this paper, the procedure of analyzing the uncertainty of a sequence of successively recorded spectra will be elaborated, enabling the determination of the correlation coefficients between certain peak count rates. The nuclear and atomic data are taken from the literature and the influence of their uncertainties could only be determined by using Monte Carlo simulation.

COUNT RATES EQUATIONS

The derivation procedure for theoretical expressions for each single and sum peak count rates and also the total count rates is given in reference [9]. The formulae for the count rates of the most pronounced peaks and adequate formulae for detection or non-detection probabilities calculations are presented in details in reference [6]. The decay of ^{133}Ba is followed by

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the emission of K_α and K_β photons, generated by electron capture and internal conversion processes, and also by gamma photons at nine different energies [10]. The two of these gamma energies are very close (75.91 keV and 81.0 keV) and could be considered as one. The decay scheme data characteristics for ^{133}Ba, as well as the peak and total detection efficiencies (depending on detector characteristics and measurement conditions) are included in the theoretical expressions. Detection efficiencies could be considered as unknown quantities which could be determined by solving the equation system. On the other hand, if the detector is already calibrated, detection efficiencies could be considered as known quantities. Thus, the single and sum peak count rates could be calculated by using the theoretical formulae and compared with experimental values. However, in this paper, detection efficiencies and source activity are considered as unknown values. If the theoretical expressions for count rates were equalized with experimental values, a non-linear equation system would be obtained. In this system, the unknown values are peak and total efficiencies and source activity. The total number of unknown values amounts to 19, *i. e*., 9 peak efficiencies, the same number of total efficiencies and source activity. The number of unknown values could be reduced to 12 if the following function is introduced

$$f(E) \quad \frac{\varepsilon(E)}{t(E)} \quad \frac{1}{1 \quad kE} \tag{1}$$

where $\varepsilon(E)$ and $t(E)$ denote the peak and total efficiencies for energy E, respectively, and k is the unknown parameter. The number of pronounced peaks significantly exceeds the number of unknowns. This is the reason that the equation system could be formed only with equations for the single peaks, one sum peak and total count rates. The single peaks are chosen because the expression for the count rate in the first approximation is proportional to the peak efficiency for that energy. The sum peaks at 437.01 keV and 333.65 keV are very pronounced and they could be used alternatively in the equation system. The system consists of 12 selected equations

$$\begin{aligned}
&N_t \quad n_t,\\
&N(30.8) \quad n(30.8),\\
&N(35.3) \quad n(35.3),\\
&N(53.16) \quad n(53.16),\\
&N(79.61) \quad N(81) \quad n(81),\\
&N(160.61) \quad n(160.61),\\
&N(223.24) \quad n(223.24),\\
&N(276.4) \quad n(276.4),\\
&N(302.85) \quad n(302.85),\\
&N(356.01) \quad N(357.4) \quad n(356.01),\\
&N(383.85) \quad n(383.85),\\
&N(437.01) \quad n(437.01)
\end{aligned} \tag{2}$$

where $N(E_i)$ denotes the theoretical expression for the peak count rate at energy E_i, whereas $n(E_i)$ denotes the experimentally obtained count rate value. In the equation system, decay scheme data for ^{133}Ba are known, so the only unknown variables are detection efficiencies and source activity. In the system (2), $n(81)$ denotes the count rate for merged peaks at energies of 79.61 keV and 81 keV. These peaks are not separated in the calculations. The net area of the sum peak at energy of 357.4 keV amounts to approximately 1% of the net area at the energy of 356.01 keV. This is the reason why these two peaks were taken together.

Since the accuracy of the system solution depends on the accuracy of the decay scheme data taken from the literature, the described method for activity measurement belongs to direct measurement, but not absolute. However, the procedure for the determination of the measurement uncertainty, which depends on the experimental as well as literature input data accuracy, should be defined. The source activity, parameter k, and detection efficiencies for 10 energies could be obtained by solving non-linear equation system (2) numerically. The values of detection efficiencies obtained by the presented method could be compared with the values obtained directly from the peak net area. Thus, the summation effects could be evaluated.

MEASUREMENT UNCERTAINTIES

The measurement uncertainty of source activity obtained by this method is due to the uncertainties of the input data obtained from the measured spectra and the uncertainties of the decay scheme data taken from literature. In reference [9], the analytical procedure for the calculation of uncertainties due to count rate uncertainties, validated by Monte Carlo method, is elaborated. Standard variance of the source activity is given by

$$s^2(R) \quad \sum_{j \ 1}^{12} ([\mathrm{M}^{\ 1}]_{12\,j})^2 s^2(n_j) \quad \sum_{j \ 1 k \ j \ 1}^{11 \quad 12} [\mathrm{M}^{\ 1}]_{12\,j} [\mathrm{M}^{\ 1}]_{12\,k} r_{jk} s(n_j) s(n_k) \tag{3}$$

where $[\mathrm{M}^{-1}]$ denotes the inverse matrix of the linearized system (2), $s(n_j)$ – the standard deviation of j-th peak count rate, and r_{jk} – the correlation coefficient between j-th and k-th peak count rates. The correlation coefficients have to be experimentally determined from the sequence of successively recorded spectra because there is no explicit formula for the source activity calculation. The expression for the correlation coefficients is given by

$$\mathbf{r}_{ij} \quad \frac{(\mathbf{n}_i \quad n_{ia}\mathbf{u})(\mathbf{n}_j \quad n_{ja}\mathbf{u})}{(m \quad 1)s(n_i)s(n_j)} \tag{4}$$

where $\mathbf{n}_i$ denotes the vector consisting of m experimental values for i-th peak count rate, n_{ia} – the mean

value for that count rate, $s(n_i)$ – the standard deviation of the i-th peak count rate, and **u** denotes the unit vector.

Correlation matrix $[\mathbf{r}_{ij}]$ becomes unit matrix, and formula (3) is reduced to the first addend under the root, if there is no correlation between peaks.

The influence of uncertainty of any atomic or nuclear parameter P is determined by Monte Carlo method. Every parameter should be varied according to the formula

$$P \quad \overline{P} \quad s\sqrt{2\ln w_i}\cos(2\pi w_2) \qquad (5)$$

where s denotes the standard deviation for the parameter, whereas w_1 and w_2 denote the generated random numbers. The equation system could be solved with parameters varied in that way. The uncertainty caused by uncertainty budget for atomic and nuclear data could be defined by the set of m activity solutions.

EXPERIMENT

The applicability of activity measurement by coincidence summation, as well as the definition of measurement uncertainty, has been experimentally verified by using two HPGe spectrometers.

– Spectrometer I: Schlumberger HPGe detector with the relative efficiency of 15%, amplifier Ortec 672 and multichannel analyzer Canberra Multiport II, and
– Spectrometer II: AMETEK-AMT (Ortec) GEM 30-70 with the relative efficiency of 37%, amplifier Canberra 2020 and multichannel analyzer Canberra Multiport II.

^{133}Ba source was measured twice by spectrometer I. The first series consisted of 12 measurements, each lasting 40 000 s, and the spectra were processed by program Anges [11]. The second series consisted of 17 measurements, each lasting 20 000 s, and the spectra were processed by program Genie 2k [12]. The same Ba source was measured once by spectrometer II. The series consisted of 10 measurements, each lasting 20 000 s, and the spectra were processed by program Anges. The spectra recorded by both spectrometers are presented in figs. 1 and 2.

DISCUSSION

The results obtained by the analysis of 12 spectra using program Anges are presented in tab. 1. The characteristic of this series is: if the correlation term, second term in eq. (3), is taken in account, the obtained uncertainty is smaller. The processed results obtained in the analysis of the spectra recorded by the spectrometer II are presented in tab. 2. Spectrometer II has better performances than spectrometer I, and the measurement uncertainties are several times smaller. The

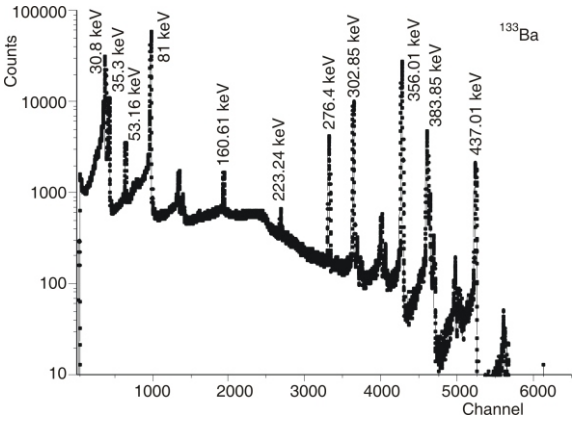

Figure 1. The recorded spectrum of ^{133}Ba point source by spectrometer I

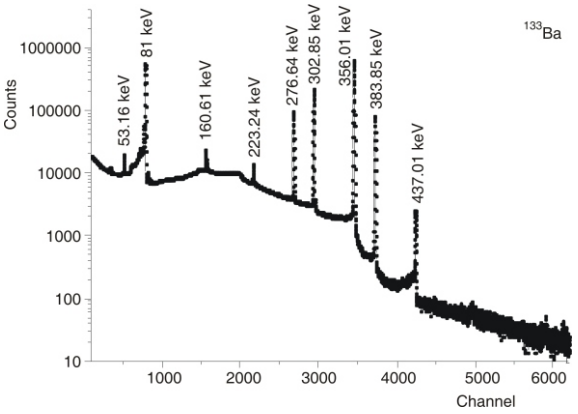

Figure 2. The recorded spectrum of ^{133}Ba point source by spectrometer II

comparative values for the activity and activity uncertainty are given in tab. 3. The spectra in the second series, on spectrometer I (third row in tab. 3), were processed by Canberra's program Genie 2k. The obtained activity uncertainty is 1 Bq and it is unrealistically small, because this software gives unrealistically small count rate uncertainties for the peaks taken into account.

Uncertainty in activity determination caused by atomic and nuclear parameter uncertainties calculated by using the Monte Carlo method is given in the forth column of tab. 3. It is interesting to notice that this uncertainty for spectrometer I is five times smaller than for spectrometer II, which has superior performances. We could not explain this phenomenon. Atomic and nuclear parameter uncertainties for both spectrometers have negligibly small influence on the total uncertainty.

The possible source of errors like live time and half-life uncertainty are not included in tabs. 1, 2, and 3 because they are several orders of magnitude smaller [9].

Table 1. The series of twelve measurements on spectrometer I (the spectra were processed by Anges program)

Activity [Bq]	Uncertainty [Bq] (without correlation)	Uncertainty [Bq] (with correlation)
806.8	11.9	9.4
807.1	11.2	10.0
814.2	13.5	11.6
808.3	12.5	10.1
806.1	11.3	10.0
814.1	13.2	11.0
814.6	13.0	10.8
810.4	12.7	10.5
809.9	12.6	10.2
812.7	12.4	10.2
812.4	12.7	10.4
806.0	12.0	9.5
Mean value = 810.2	Standard uncertainty $\sqrt{\frac{\sum_{i=1}^{m} s_{0i}^2}{(m-1)m}}$ 3.75	Standard uncertainty $\sqrt{\frac{\sum_{i=1}^{m} s_i^2}{(m-1)m}}$ 3.11

Table 2. The series of ten measurements on spectrometer II (the spectra were processed by Anges program)

Activity [Bq]	Uncertainty [Bq] (without correlation)	Uncertainty [Bq] (with correlation)
815.6	4.8	5.5
815.5	3.3	4.4
817.9	5.5	7.1
815.1	4.1	4.6
818.4	5.5	6.25
817.0	4.5	5.0
818.1	5.4	6.2
820.6	5.5	6.2
819.1	4.9	5.7
818.9	4.5	5.4
Mean value = 817.6	Standard uncertainty $\sqrt{\frac{\sum_{i=1}^{m} s^2}{(m-1)m}}$ 1.6	Standard uncertainty $\sqrt{\frac{\sum_{i=1}^{m} s^2}{(m-1)m}}$ 1.9

Table 3. The comparison of the results of the three series of measurements

Activity [Bq]	Standard uncertainty [Bq] (without correlation)	Standard uncertainty [Bq] (without correlation)	Standard uncertainty caused by decay scheme data uncertainty [Bq]	Total uncertainty [Bq]	Note*
810.2	3.8	3.1	0.07	3.1	F, A
803.8	1.6	1.9	0.37	1.94	S, A
803.1	1.0	1.03	0.07	1.03	F, C

*Note: F – Spectrometer I, S – Spectrometer II, A – the spectra were processed by Anges, C – the spectra were processed by Genia 2k

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ДИРЕКТНО МЕРЕЊЕ АКТИВНОСТИ ^{133}Ba ПОЛУПРОВОДНИЧКИМ HPGe СПЕКТРОМЕТРОМ

Директно мерење активности извора ^{133}Ba применом теоријски добијених једначина за брзине бројања недавно су развили Новковић и сарадници. Аналитички и Монте Карло прорачуни несигурности мерења активности детаљно су описани у раније објављеном раду. У овом раду приказана је процедура прорачуна несигурности серије узастопно снимљених мерења, при чему се одређује и корелација између брзина бројања у пиковима. Такође је детаљно анализиран утицај несигурности експериментално одређених података, као и несигурности коришћених нуклеарних параметара, на укупну мерну несигурност активности одређене описаном методом.

Кључне речи: ^{133}Ba, коинцидентно сумирање, мерна несигурност, HPGe спектрометар