The direct measurement of ^{133}Ba activity by the sum-peak method

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ABSTRACT

The subject is a direct measurement of the activities of ^{133}Ba sources by application of the theoretical count rate expressions recently developed by Novković et al. The total uncertainties of the activity of the measured sources, obtained in two manners, are described in this paper. The presented method is successfully tested using a germanium spectrometer with efficiencies of 50%. The accuracy of this method is proved to be comparable with the present methods of the activity measurement of ^{133}Ba.

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1. Introduction

In gamma spectrometry, coincidence summing of X-ray and gamma-ray is used for the absolute activity measurement of radionuclides: ^{24}Na, ^{60}Co, ^{88}Y, ^{207}Bi [1], ^{139}Ce [2] and ^{125}I [3,4]. These radionuclides have simple decay schemes, therefore the explicit expressions for activity are obtained, which contain only the peak and the total count rates. Since these expressions contain only count rates determined from the spectra, these measurements can be classified as absolute. In previously published papers [5,6], a new method for creating theoretical expressions for all peak count rates (single- and sum-peaks) in the spectra as well as for the total count rate was developed. The unknown quantities in those expressions are the source activity, and the peak and total efficiencies for all emitted photon energies. The coefficients in the expressions contain the nuclear and atomic parameters. The system of equations is obtained by equalization of the theoretical count rate expressions with the corresponding experimental values. By solving this system of equations, the activity and efficiencies can be obtained. For the simplest radionuclides, like ^{139}Ce, the solution of the system of equations gives the analytical formula for the activity, which does not contain any nuclear or atomic parameter [2,5]. However, for nuclides with somewhat more complex decay scheme, the system of equations cannot be solved analytically. The activity, as well as the peak efficiencies, is obtained by using numerical values for the nuclear and atomic parameters and by the numerical solving of the system of nonlinear equations [5]. Due to numerous nuclear and atomic

parameters existing in the system of equations, this method could not be classified as an absolute measurement, but as a direct activity measurement. The decay scheme of ^{133}Ba is more complex than that of ^{57}Co, but despite of its complexity, the activity could be directly measured by this method [6]. The main problem of the activity measurement using this method is determination of the measurement uncertainty and this paper is focused on it.

2. The system of count rate equations of ^{133}Ba

The theoretical number of ^{133}Ba peaks amounts to 167 [6], but many of them cannot be recorded due to their small probabilities. The number of the well-defined peaks in the spectrum recorded by a 50% efficiency detector was 47 [6]. The 10 single energy peaks, 1 sum peak and total count rate were used for the activity measurement of ^{133}Ba. Usage of other peaks in the system gave a less precise result. The system of 12 nonlinear equations (1) was obtained by equalizing the theoretical expressions for count rates, N, with the corresponding experimental values, n. The equations were published in ref. [6] except for the energy of 357.4 keV, which is contributed in Appendix A. The unknowns in Eq. (1) are 10 peak efficiencies, 10 total efficiencies and the activity (21 unknown quantities). If the total efficiencies are approximated by the formula $t(E) = (1+kE)\varepsilon(E)$ (k is also an unknown parameter to be determined), the number of unknowns can be reduced to 12 (activity, 10 peak efficiencies and parameter k) [6].

Function $f(E) = \varepsilon(E)/t(E) = 1/(1+kE)$ represents peak to total efficiency ratio.

$$N_t = n_t,$$
$$N(30.8) = n(30.8),$$

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$$N(35.3) = n(35.3),$$
$$N(53.16) = n(53.16),$$
$$N(79.61) + N(81) = n(81),$$
$$N(160.61) = n(160.61),$$
$$N(223.24) = n(223.24),$$
$$N(276.4) = n(276.4),$$
$$N(302.85) = n(302.85),$$
$$N(356.01) + N(357.4) = n(356.01),$$
$$N(383.85) = n(383.85),$$
$$N(437.01) = n(437.01). \tag{1}$$

The left-hand sides of the above system are the functions of the peak and total efficiencies, the source activity and decay scheme data. The decay scheme data for ^{133}Ba are known [6,7], and unknown quantities are only efficiencies and the activity. The system of equations is nonlinear, but it can be numerically solved by searching the roots of all unknown variables simultaneously with the program package Mathematica 5.2, Wolfram Research Company[1].

3. Uncertainties

The accuracy of ^{133}Ba activity, achieved by this method [6], could be better than 1%, which allows its application to standardization of ^{133}Ba sources. The reliability of the obtained results requires establishment of an exact procedure for determination of the measurement uncertainty. The activity uncertainties can be divided into two groups.

Those of the first group are caused by the peak count rates and gross count uncertainties of the spectrum. These uncertainties are mainly determined by the quality of applied spectrometer.

Those of the second group are caused by the nuclear and atomic data uncertainties taken from the literature.

3.1. Uncertainties caused by net areas statistics

3.1.1. The analytical procedure for the determination of uncertainties caused by net areas statistics

The system of equations (1) is not suitable for evaluation of uncertainties by an analytical procedure, due to its nonlinearity, and because the count rate data (right-hand side of the equations) are mutually correlated. The simplest analytical way for estimation of the measurement uncertainty is the linearization of the above system in the vicinity of its solution. All equations are expanded in Taylor series with respect to the efficiencies and activity in the vicinity of the system solution, rejecting higher order terms. A system of linear equations is obtained

$$m_{1,1}(\varepsilon_1 - \varepsilon_1^{sol}) + m_{1,2}(\varepsilon_2 - \varepsilon_2^{sol}) + \cdots + m_{1,11}(k - k^{sol}) + m_{1,12}(R - R^{sol}) = n_{tot} - n_{tot}^{exp}$$
$$m_{2,1}(\varepsilon_1 - \varepsilon_1^{sol}) + m_{2,2}(\varepsilon_2 - \varepsilon_2^{sol}) + \cdots + m_{2,11}(k - k^{sol}) + m_{2,12}(R - R^{sol}) = n_2 - n_2^{exp}$$
$$\vdots \qquad \vdots \qquad \vdots \qquad \vdots \qquad \vdots$$
$$m_{11,1}(\varepsilon_1 - \varepsilon_1^{sol}) + m_{11,2}(\varepsilon_2 - \varepsilon_2^{sol}) + \cdots + m_{11,11}(k - k^{sol}) + m_{11,12}(R - R^{sol}) = n_{11} - n_{11}^{exp}$$
$$m_{12,1}(\varepsilon_1 - \varepsilon_1^{sol}) + m_{12,2}(\varepsilon_2 - \varepsilon_2^{sol}) + \cdots + m_{12,11}(k - k^{sol}) + m_{12,12}(R - R^{sol}) = n_{12} - n_{12}^{exp} \tag{2}$$

where n_i^{exp} denotes the experimental (mean) value of the count rate and n_i the possible value (difference $|n_i - n_i^{exp}|$ is less than the experimental uncertainty). Matrix form of the above

system is

$$
\begin{pmatrix}
m_{1,1} & m_{1,2} & \cdots & m_{1,11} & m_{1,12} \\
m_{2,1} & m_{2,2} & \cdots & m_{2,11} & m_{2,12} \\
\vdots & \vdots & \ddots & \vdots & \vdots \\
m_{11,1} & m_{11,2} & \cdots & m_{11,11} & m_{11,12} \\
m_{12,1} & m_{12,2} & \cdots & m_{12,11} & m_{12,12}
\end{pmatrix}
\begin{bmatrix}
\varepsilon_1 - \varepsilon_1^{sol} \\
\varepsilon_2 - \varepsilon_2^{sol} \\
\vdots \\
k - k^{sol} \\
R - R^{sol}
\end{bmatrix}
$$
$$
=
\begin{bmatrix}
n_{tot} - n_{tot}^{exp} \\
n_2 - n_2^{exp} \\
\vdots \\
n_{11} - n_{11}^{exp} \\
n_{12} - n_{12}^{exp}
\end{bmatrix} \tag{3}
$$

where $M = [m_{i,j}] = [\partial N(E_i)/\partial\varepsilon_j], j = 1,2, \ldots, 10; m_{i,11} = \partial N(E_i)/\partial k$ and $m_{i,12} = \partial N(E_i)/\partial R$ denotes a matrix of the expansion of the nonlinear system of equations into linear in the vicinity of the solution $(\varepsilon_1^{sol}, \varepsilon_2^{sol}, \ldots, k^{sol}, R^{sol})$. The system of equations can be formed in different manners combining expressions for different peaks of the recorded spectrum. Some of them can give an unstable system. For instance, a system involving peaks 30.8 keV (K_α), 35.3 keV (K_β), 61.6 keV ($2K_\alpha$), and 70.6 keV ($2K_\beta$) is an unstable system because the ratio of the expressions for the first two energies are equal to the ratio of the expressions for the last two energies. We tested different combinations of the peaks (47 detected peaks) in the spectrum and the quoted system of equations (1) is the stable system and gave solutions with minimum uncertainties for activity and efficiencies.

If system (3) is multiplied from the left-hand side by the inverse matrix $\mathbf{M}^{-1}$, the unknown variables $(\varepsilon_1, \varepsilon_2, \ldots, \varepsilon_{10}, k, R)$ are obtained

$$
\begin{bmatrix}
\varepsilon_1 - \varepsilon_1^{sol} \\
\varepsilon_2 - \varepsilon_2^{sol} \\
\vdots \\
k - k^{sol} \\
R - R^{sol}
\end{bmatrix}
=
\begin{pmatrix}
m_{1,1} & m_{1,2} & \cdots & m_{1,11} & m_{1,12} \\
m_{2,1} & m_{2,2} & \cdots & m_{2,11} & m_{2,12} \\
\vdots & \vdots & \ddots & \vdots & \vdots \\
m_{11,1} & m_{11,2} & \cdots & m_{11,11} & m_{11,12} \\
m_{12,1} & m_{12,2} & \cdots & m_{11,12} & m_{12,12}
\end{pmatrix}^{-1}
$$
$$
\times
\begin{bmatrix}
n_{tot} - n_{tot}^{exp} \\
n_2 - n_2^{exp} \\
\vdots \\
n_{11} - n_{11}^{exp} \\
n_{12} - n_{12}^{exp}
\end{bmatrix}. \tag{4}
$$

i.e.,

$$\varepsilon_1 = \varepsilon_1^{sol} + \sum_{j=1}^{12}(M^{-1})_{1,j}(n_j - n_j^{exp})$$

$$\varepsilon_2 = \varepsilon_2^{sol} + \sum_{j=1}^{12}(M^{-1})_{2,j}(n_j - n_j^{exp})$$
$$\vdots \qquad \vdots$$
$$k = k^{sol} + \sum_{j=1}^{12}(M^{-1})_{11,j}(n_j - n_j^{exp}).$$

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[1] The built-in function *FindRoot*, based on Newton method, is used for simultaneous search of 12 roots of system (1).