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Study on gamma radioactivity analysis from point sources using HPGe spectrometer by Full Spectrum Analysis method combined with Gold unfolding algorithm

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Abstract: In this work, the methodology of estimating the radioactivity of nuclides from point gamma source using method of full spectrum analysis combined with Gold unfolding algorithm are studied. The reference sources with known activities as Ba-133, Cd-109, Co-57, Co-60, Cs-137, Mn-54, Na-22 and Zn-65 are counted in front of the HPGe detector. The source which is mixed by these radioisotopes with known weighting factors C(i) are counted. Its spectra are unfolded to have the relative radioactivity of these isotopes from analyzed source, $C_{unf}(i)$. The difference of less than 0.2% between calculated ratios of $C_{unf}(i)$ from the unfolded spectra and known weighting factors of C(i) opens a possibility to improve the accuracy for the analysis method with low background HPGe detector.

Keywords: *Full spectra analysis, unfolding.*

I. INTRODUCTION

The common approach to analyse the activity from the gamma spectrometer is based on peak counts in each individual energy, also known as Window Analysis method (WA). It is limited for analyzing the energy overlapped peak, subtracting non-linear background under peaks, summing coincidence effect from multigamma source… To reduce these effects, instead of determining the activity based on gamma counting of the individual peaks, an alternative method to determine the activity which are based on recording gamma counts of the full spectrum, which is so called Full Spectrum Analysis (FSA) can be used. It helps us eliminate summing coincidence effect, errors from background subtracting, statistical fluctuations and errors from measurement system. Besides, the computational time can be reduced.

In the typical FSA method, the radioactivity of radionuclides in samples are estimated by fitting techniques for the sample spectrum based on the reference spectra. Maphoto in 2004 has developed methods to determine the activity of U-238, Th-232 and K-40 in the sand, soil and ore samples [1]. The most recent work of the FSA's approach is belong to R.T. Newman and colleagues in 2008 [2]. For the survey, the authors also gained the differences of less than 10% between FSA and WA method. In this work, we developed FSA method combined with Gold unfolding algorithm [3], [4] to evaluate radioactivity from point gamma sources. The reference sources with known activity as Ba-133, Cd-109, Co-57, Co-60, Cs-137, Mn-54, Na-22 and Zn-65 are counted in front of the HPGe detector. The mixed spectra which are combined from these reference spectra with

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given weighting factors are unfolded. Then the radioactivity from the mixed spectra are evaluated through the ratio of counts from each bin of unfolded spectra and total counts from reference spectra.

II. FULL SPECTRA ANALYSIS METHOD AND UNFOLDING ALGORITHM FOR RADIOACTIVITY ANALYSIS USING GAMMA SPECTROMETER

A. Full Spectra Analysis Method (FSA)

Under FSA method, the spectrum of analyzed sample is considered as a weighted superposition from the spectra of reference radionuclides. The spectra of reference radionuclides can be measured experimentally or using Monte Carlo simulation method. Reference spectrum corresponds to the response of the detector for each sample containing 1Bq/kg radionuclides of interest. In the environment measurements, some of the long-lived naturally occurring radionuclides as U-238, Th-232 and series and K-40 are of particular interest. It is noted that samples containing radionuclides of U-238, Th-232 and K-40 seperately for calibration are under the same configuration as analyzed sample. Therefore the measured spectrum can be mathematically expressed in equation as follow:

$$S(i)=C_U\times S_U(i)+C_{Th}\times S_{Th}(i)+C_K\times S_K(i)+C_B\times S_B(i) \quad (1)$$

where S_U , S_{Th} , S_K and S_B are measured spectra from 1Bq/kg of U-238, Th-232, K-40 reference materials and background respectively. C_U, C_{Th}, and C_K are respectively activities of the radionuclides in the analyzed samples, C_B is the background contribution to the analyzed spectrum S. In practice C_B is selected by the ratio between the measuring time for S and one for S_B.

In the general case, the values of C_i radioactivities from analyzed sample could be estimated by fitting the spectra data measured from the analyzed sample with the one from reference sources. When there are several radionuclides taking part into the interested sample, it becomes more difficult for fitting. The unfolding algorithm is an alternative method which has more advantages to estimate radioactivities from analyzed samples.

B. Gold unfolding algorithm

The unfolding algorithm is based on the integral equation [3]:

$$Y(\mu)=\int_{\nu_1}^{\nu_2} A(\nu,\mu)X(\nu)d\nu \qquad (2)$$

where X (ν) is the distribution of radiations at energy ν, Y (μ) is the radiation level recorded at energy μ from the recording device, A (ν, μ) is the response function of detector, it is the probability that a energy radiation ν is recorded in the energy bin μ. In equation (2), μ and ν are continuous variables on the range of energy which are studied. In practice, the design of the spectrometer only allows them to show signal distribution into discrete channels, so equation (2) is transformed into:

$$Y = A.X \qquad (3)$$

where Y is the measured spectrum data vector, also known as the output spectrum vector containing the discrete values corresponding to counts at each channel of the spectrometer, X is the input spectrum vector, which is representing the energy distribution of the source, A is the detector response matrix, which is reflecting the way that the original radiation spectrum was recorded in the detector. Thus, the y_i elements of vector Y will be determined by:

$$y_i=\sum_{j=1}^{n} a_{j,i}x_j \qquad (4)$$

with x_j are elements of the vector X, $a_{j,i}$ are elements of the response matrix A, n is the level of response matrix A. Equation (3) can be solved by multiplying both sides of the equation with the inverse matrix A^{-1}. In practice, Gold [3] used iterative methods to find approximate solution of equation (3) as follows:

$$x_i^{(k+1)} = \frac{z_i x_i^{(k)}}{d_i} \qquad (5)$$

where $z = A^T y$

and $d = A^T A.x^{(k)}$

A^T is transposition matrix from A.

An unfolding code for finding the solution of the equation (3) by the iterative formula (5) was built by the authors of this work. The values of C_i relative radioactivities of analyzed sample in equation (1) can be estimated from the solution vector X, where Y is the measured spectrum data vector, the response matrix A is built from spectra of reference radionuclides which have expected for existence in the analyzed samples.

III. RESULTS AND DISCUSSION

In this work, FSA method combined with Gold unfolding algorithm as above was developed to evaluate radioactivity from point gamma sources. The reference sources with known activities as Ba-133, Cd-109, Co-57, Co-60, Cs-137, Mn-54, Na-22 and Zn-65 are counted in front of the HPGe detector. The analyzed spectra which are sum of spectra from eight radionuclide above with known weighting factors are unfolded and used for evaluation of the activities. The symbol of each reference spectrum, radionuclide, and the total count of the spectrum are shown in Table I. Symbols of mixed spectra, factors of involved components of radionuclides C(i) are shown in Table II.

Table I. Symbols of reference spectra, radionuclides, and counts of the total spectra

Reference spectra	Radionuclide	Total spectrum count S(i)
1	Ba-133	439690
2	Cd-109	25384
3	Co-57	149712
4	Co-60	374507
5	Cs-137	186059
6	Mn-54	56421
7	Na-22	417428
8	Zn-65	37457

The response matrix for unfolding is illustrated in Figure 1. It includes eight measured reference spectra from Ba-133, Cd-109, Co-57, Co-60, Cs-137, Mn-54, Na-22 and Zn-65 radionuclides.

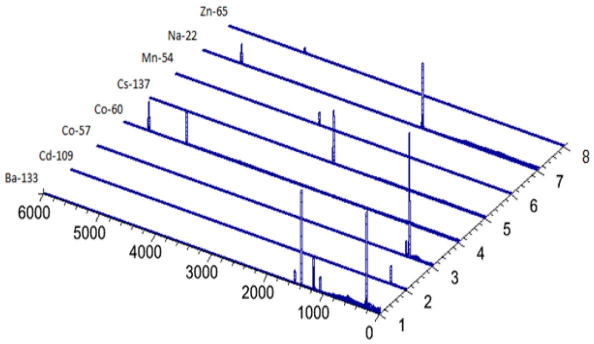

Fig. 1. Response matrix of reference spectra

Table II. The involved components in the analyzed source

Radionuclide	Weighting factor
Ba-133	1
Cd-109	3
Co-57	1
Co-60	5
Cs-137	2
Mn-54	2
Na-22	1
Zn-65	4

The spectrum of analyzed source before and after unfolding are illustrated in Figure 2 and Figure 3, respectively.

It is noted that the unfolded spectrum includes bins every which contains the total counts of every radionuclide involved in the analyzed sample. The ratios $C_{unf}(i)$ of the counts of each bin from the unfolded spectrum and the total counts from corresponding reference spectra were evaluated and compared with the correspondingly weighting factors C(i) and shown in Table III.

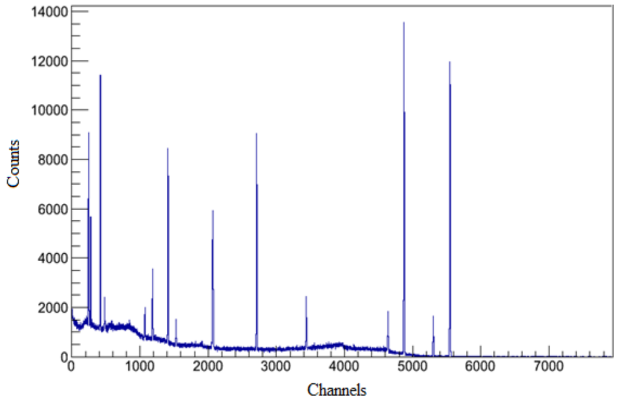

Fig. 2. The spectrum of analyzed source before unfolding

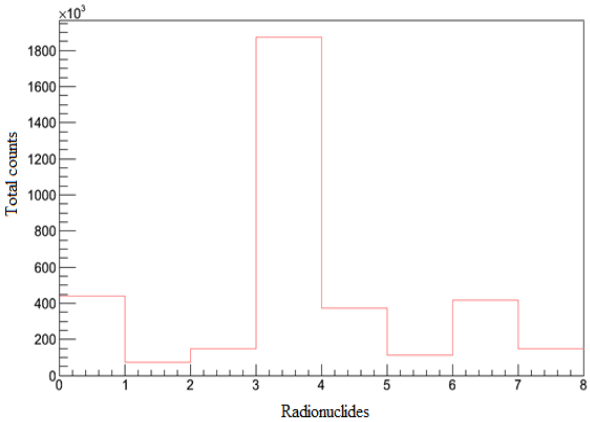

Fig. 3. The spectrum of analyzed source after unfolding

Table III. The ratios $C_{unf}(i)$ of the counts of each bin from the unfolded spectrum and the total counts from corresponding reference spectra and deviation δ (i) between the C_{unf} (i) and C(i)

i	S(i)	C(i)	N(i)	$N_{unf}(i)$	C_{unf} (i)	δ (i) (%)
1	439690	1	439690	439349	0.999	0.08
2	25384	3	76152	76083	2.997	0.09
3	149712	1	149712	149517	0.999	0.13
4	374507	5	1872535	1872170	4.99	0.02
5	186059	2	372118	372032	2.000	0.02
6	56421	2	112842	112804	1.999	0.03
7	417428	1	417428	417292	1.000	0.03
8	37457	4	149828	149740	3.998	0.06
Sum			3590305	3588987		

where the symbol i is i^th^ radionuclide, S(i) is the total counts of the i^th^ reference spectrum, C(i) is weighting factor of the i^th^ isotope in the analyzed spectrum, N(i) is the total counts of i^th^ reference spectrum; $N_{unf}(i)$ the total counts of i^th^ bin in the unfolded spectrum; $C_{unf}(i)$ is ratio of the counts of each bin from the unfolded spectrum and the total counts from corresponding reference spectra and also being relative radioactivity of ith radioisotope in the analyzed source, δ (i) is deviation (%) between the value of C_{unf} (i) and C (i).

It is shown that the difference between $C_{unf}(i)$ from the unfolded spectrum and known C(i) is lower of 0.2%. It is noted that there is neither effects of mono-energy nor multi-energy from radioisotopes included in the mixed spectra influencing on the results of analysis. It also shows that effects of summing coincidences when analyzing the radioactivity of gamma emitters using the FSA method are negligible. This is a major advantage of the technique developed.

IV. CONCLUSION

In this work we developed determination the activity of point source of mono-gamma and multi-gamma using the FSA method combined with Gold unfolding algorithm. The reference sources with known activities as Ba-133, Cd-109, Co-57, Co-60, Cs-137, Mn-54, Na-22 and Zn-65 are counted in front of the HPGe detector. The mixed spectra which are combined from these reference spectra by given weighting factors are unfolded. It is showed from the results of unfolding that the difference of less than 0.2% between calculated weighting factors of radionuclides from the unfolded spectra and mixed weighting factors may be neglected. The evaluation of radioactivity from the mixed spectra through the ratio of counts from each bin of unfolded spectra and total counts from reference spectra can reduce propagation of errors from nuclear data as well as summing coincidence, subtracting Compton continuum under the surveyed peaks which are often happened for WA method. This approach opens a possibility to improve the accuracy for the analysis method with low background HPGe detector which is often used in measurements of environmental samples.

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