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Development of coincidence summing and resolution enhancement algorithms for digital gamma ray spectroscopy

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This paper discusses the correction of some of the main problems of digital gamma ray spectroscopy. These problems are the coincidence summing and energy resolution. The coincidence summing effects are evaluated using analytical techniques. Correction is made at different energies for both ^{137}Cs and ^{60}Co radioisotopes. A simple relation is derived between the coincidence summing correction factor and the energies under the conditions of the system configuration. This relation is deduced using the least square approximation method. Consequently, correction can be done at different energies of radiation sources under the constraints of the measured conditions. Furthermore, the coincidence summing algorithm is validated through comparison with published experimental results in the literature and good agreement is found. Coincidence summing correction factors are used to correct the values of the Full Energy Peak (FEP) efficiencies. Correction factors were calculated for predominant gamma emissions significantly affected by coincidence summing effects for both ^{137}Cs and ^{60}Co point sources. Also, a correction algorithm for the resolution–degradation in scintillation (NaI (Tl)) gamma ray detectors using derivative methodology is presented. The derivative methodology is implemented by weighted sum of the original signal, the negative of its second derivative and the positive of its fourth derivative. We noticed that using both derivatives in combination improves the energy resolution, which is enhanced by 24.03%. From the obtained results, the FEP efficiency was enhanced by 1.58% due to coincidence summing correction. Consequently, accurate determination for the source activity can be achieved.

1. Introduction

The coincidence summing correction of gamma spectra has been an important research topic since the 1970's.[1,2] This interesting field has become a focus again in recent years.[3] However, many classical methods are widely used for high efficiency detectors with some good results being obtained. Coincidence summing can occur when a radionuclide emits two or more photons simultaneously in cascade.[4,5] These emitted photons are within the resolving time of the spectrometer. Consequently, correction of the full-energy peak areas of the emissions in the spectra is required. The effect takes two forms: summing out and summing in. Summing out occurs when a gamma ray that should have been recorded in the full-energy peak is recorded elsewhere. This is due to the simultaneous detection and summing with another photon. Summing in is the reverse.[6,7] However, a greater concern is that a full energy event from one transition will sum with a Compton event in another transition, thereby effectively decreasing the efficiency for the detection of an isotope.[8] The aim of gamma ray spectrometric analysis is to determine the activity concentration of gamma ray emitting radionuclides[9] and the associated coincidence summing of the results. To derive the uncorrected apparent activity for summing effects, it must be multiplied by a correction factor. For point sources, the corrections can be calculated by analytical formula. However, for extended sources the computation becomes more complex, since the contribution of each volume element to the efficiency depends on its position in the source. Several methods of a very different nature have been proposed to deal with the problem from purely experimental to Monte Carlo simulations.[10] A realistic evaluation of the coincidence summing effects is a difficult task, especially in the case of nuclides with complex decay schemes. It implies an intricate combination of decay scheme parameters with peak and total efficiencies specific to the measurement conditions.[4]

Furthermore, the requirements for using higher resolutions in the field of nuclear experiments and data analysis are continuously increasing. High resolution gamma ray spectroscopy at relativistic beam energies is an experimental challenge.[11] Signals coming from various sources are overlapped due to the limited resolution of the equipment. For instance gamma ray spectra often contain overlapping photopeaks which make it difficult to estimate correctly their positions, areas and the corresponding radionuclide activities. The accuracy and reliability of the analysis depends on the treatment in order to resolve strong overlapping

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peaks.[12] Over the last few years much effort has been devoted to developing methods for analysis of data from large gamma ray detector arrays.[13] Moreover, high-resolution spectroscopy is used routinely in passive gamma ray assays of nuclear materials and radioactive waste. It provides the capability to identify radionuclides from complex gamma ray spectra. Also, it accurately determines the full-energy response of selected gamma rays using continuum subtraction techniques.[14] The gamma ray for nuclear spectroscopy with higher resolution provides exciting possibilities to detect the gamma rays emitted from complex nuclear reactions with a much higher efficiency. Consequently, some of the general features of gamma ray spectroscopy were studied, along with the performance characteristics of a NaI (Tl) scintillation detection system.

In this paper, an algorithm for coincidence summing correction is presented. Moreover, an algorithm for resolution enhancement is investigated. The performance of NaI (TI) scintillation detectors used in nuclear spectroscopy is demonstrated. The current technique will allow further characterization of gamma rays than what is possible with current detector systems. Such features will assist in isotope identification. This paper is organized as follows: Section 2 presents the system component. The coincidence summing correction algorithm, geometry efficiency and FEP efficiency measurements are presented in Section 3. Resolution enhancement algorithm is discussed and summarized in Section 4, and we terminate our study with a brief discussion stating some important conclusions that we note from our obtained results in Section 5.

2. System components

The components of the system for evaluation of coincidence summing and resolution enhancement algorithms contain the following elements; ^{137}Cs/^{60}Co point sources, scintillation detector, coaxial cable, amplifier and acquisition system. Cylindrical scintillation detector with (1.5×7.5) inches NaI (TI) crystal coupled with photomultiplier tube is used to detect the radiation signal from ^{137}Cs and ^{60}Co point sources. The true activity of both isotopes is 1 μCi. This detector is connected to an amplifier through a coaxial cable connected to a PC through ADC with 16 MS s^{-1}.

3. Coincidence summing correction algorithm

Different nuclear states are characterized by the following quantities: energy, total angular momentum, electric quadrupole momentum, parity, magnetic moment, electric quadrupole moment, partial level widths, and total transition probability.[15] All these properties are as well defined for excited levels as for stable and unstable ground states. For radioactive nuclei, and especially for short-lived excited nuclear states, most of these well known methods fail completely and new ways have to be developed. The easiest is to determine the energy difference to the ground state using highly developed gamma spectroscopy.[15] The total transition probability can in most cases also be ascertained by following the radioactive decay over a suitable period or by measuring delayed coincidences.[15]

Angular correlation between two gamma rays emitted in a cascade is defined as the relative yield of γ_2 about the 0° direction defined by the detector position, given that γ_1 emitted in the same direction. Angular correlations arise because the direction of emission of the first gamma ray is related to the orientation of the angular momentum of the intermediate state.[16] If the life time of the intermediate state is short, the orientation of the angular momentum will persist. The direction of the second gamma ray will be related to the angular momentum of the intermediate state, and hence to the direction of the first gamma ray. Angular correlation effects in general are not very significant when correcting for cascade summing effects.[16] Therefore, the influence of angular correlation is assumed to be negligible. Since γ_1 and γ_2 are emitted and detected in true coincidence, the energy deposited in the detector may be the sum of the full energy from γ_1 and only part of the energy (up to full energy) from γ_2. This results in events being lost from the full energy of γ_1 since any type of interaction involving γ_2 will result in a loss of count from the full energy of γ_1. The total detection efficiency of γ_2 is used in determining the cascade summing loss.[16]

The correction of the coincidence summing effect can be made by direct measurement of a single nuclide based on the decay scheme of the radionuclides of concern, geometry of sample shape, the geometry of sample-to-detector and the accurate data of detector shapes.[5]

An efficient algorithm for overcoming and correcting the coincidence summing problem is presented as shown in Fig. 1. The main strength of the proposed algorithm is the derivation of a simple and a general equation that relate the sample energy with the correction factor. Furthermore, the effect of coincidence summing corrections on different forms of efficiency is investigated.

In this work in order to evaluate the coincidence summing corrections we applied a mathematical procedure because point sources are used. Radiation signal from ^{137}Cs/^{60}Co was analyzed. Then, the spectrum of ^{137}Cs/^{60}Co was drawn. It represents the relation between the channel numbers against channel content. The total number of counts within the spectrum was counted. Then, the total efficiency that represents the ratio between the number of events recorded in the entire spectrum and the number of photons emitted by the source[17] was computed. Subsequently, a simple decay scheme with two excited levels for ^{137}Cs is shown in Fig. 2(a). However, a decay scheme with three excited levels for ^{60}Co is depicted in Fig. 2(b).

Furthermore, to evaluate the coincidence summing corrections, a mathematical procedure was applied because point sources are used. The corrections for the peak areas corresponding to energies E_1, E_2 and E_3 are, respectively:[1,2,18,19]

$$C_1 = \frac{1}{1 - P_{12}\varepsilon_{T_2}} \quad (1)$$

$$C_2 = \frac{1}{1 - P_{21}\varepsilon_{T_1}} \quad (2)$$

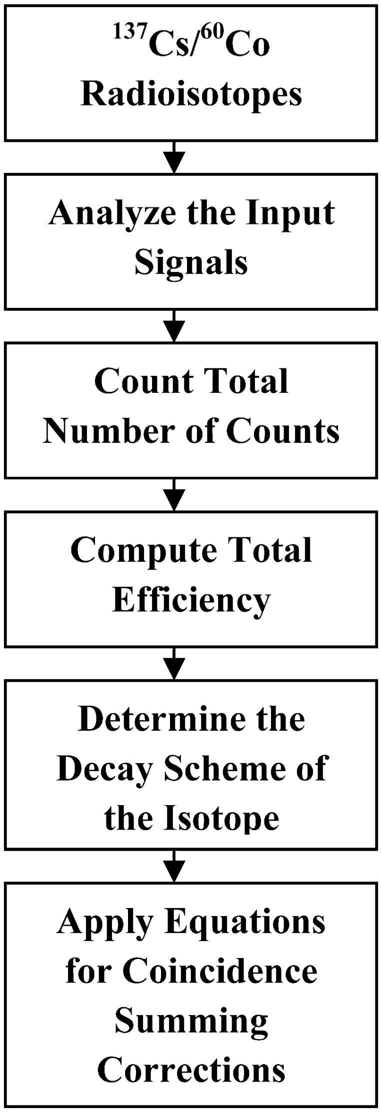

Fig. 1 Coincidence summing correction algorithm.

$$C_3 = \frac{1}{1 + \frac{I_{\gamma_1}}{I_{\gamma_3}} \frac{\varepsilon_{p_1}\varepsilon_{p_2}}{\varepsilon_{p_3}} P_{12}} \tag{3}$$

where P_{ij}, ε_{pi}, ε_{T_i}, and I_{γ_i} denote the conditional probability for emitting γ_i simultaneously with γ_j, the FEP efficiency for energy E_i, the total efficiency for energy E_i, and the photon emission intensities for energy E_i, respectively.

Moreover, the coincidence summing factor (C) can be calculated using the following equation[20]

$$C = \left[1 - \frac{\sum_i P_{t_{ij}} P_i P_j \varepsilon_{t_j}}{I_{\gamma_i}}\right] \left[1 + \frac{\sum_{k,m} P_{t_{km}} P_k P_m \varepsilon_k \varepsilon_m}{I_{\gamma_i} \varepsilon_i}\right] \tag{4}$$

where I_{γ_i}, ε_i, ε_{t_j}, $P_{t_{ij}}$, P_i, α_t, ε_k, ε_m, and P_m denote photon emission probability, FEP efficiency, total efficiency, probability of coincident transitions (i, j), probability of photon emission in transition i, the total internal conversion coefficient, the FEP efficiency for energy E_k, FEP efficiency for energy E_m, and probability of photon emission in transition m, respectively. The photon emission probability is specified by[20]

$$I_{\gamma_i} = P_i T_i \tag{5}$$

where, T_i denotes the transition probability. However, the total efficiency is illustrated as[20]

$$\varepsilon_{t_j} = \frac{\varepsilon_j}{K R_\sigma E_\gamma} \tag{6}$$

where, ε_j, K and R_σ denote the FEP efficiency for energy E_j, a constant and the cross section ratio that is given by[20]

$$R_\sigma = \frac{R_\varepsilon}{R} = \frac{R_\varepsilon}{K E_\gamma} \tag{7}$$

where, R denotes the relative efficiency. The probability of photon emission in transition k is presented as[20]

$$P_k = \frac{1}{1 + \alpha_t} \tag{8}$$

Therefore, the coincidence summing correction factor (C_i) is given by[20]

$$C_i = 1/C \tag{9}$$

However, the FEP count rate, n_i, is specified as[20]

$$n_i = A I_{\gamma_i} \varepsilon_i C_i \tag{10}$$

where A denotes the current activity that was computed by

$$A = A_0\, e^{-\lambda t} \tag{11}$$

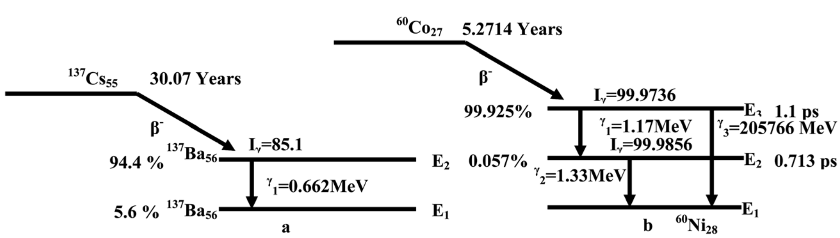

Fig. 2 Simple decay scheme with (a) two excited levels for ^{137}Cs (b) three excited levels for ^{60}Co.

Table 1 Parameter values of coincidence summing algorithm

$K = 0.0102207$ (2 cm)	$P_3 = 0.0180/100$ (^{60}Co)	$I_\gamma = 0.851 \pm 0.00002$ (^{137}Cs)	$T_{1/2} = 5.2$ years (^{60}Co)
$P_1 = 99.925/100$ (^{60}Co)	$P_{21} = (99.925 - 0.057)/100$ (^{60}Co)	$I_{\gamma_1} = 0.99857 \pm 0.00022$ (^{60}Co)	$T_{1/2} = 30.1$ years (^{137}Cs)
$P_2 = 0.057/100$ (^{60}Co)	$P_{12} = (94.4 - 5.6)/100$ (^{137}Cs)	$I_{\gamma_2} = 0.99983 \pm 0.00006$ (^{60}Co)	$\alpha_t = 0.004 \pm 0.0002$

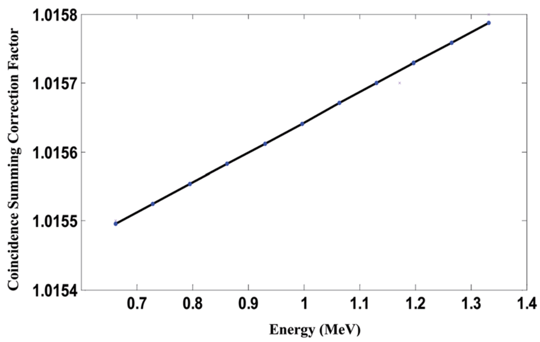

Fig. 3 Coincidence summing correction factor against energy.

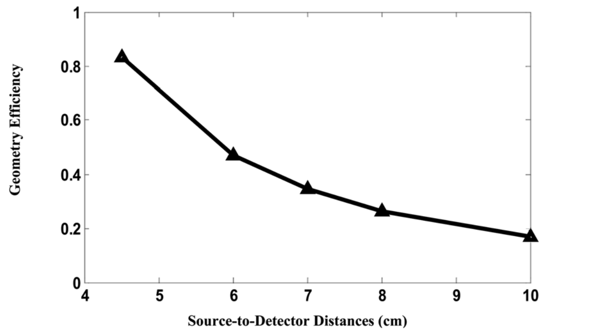

Fig. 5 Geometry efficiency against different source-to-detector distances.

Table 2 Coincidence summing correction factors for ^{137}Cs and ^{60}Co at 1.7 cm obtained from the analytical and experimental[22] methods for point source geometry

Isotope	Energy (MeV)	Coincidence summing correction Factor	Experimental results
^{137}Cs	0.662	1.0155	1.02
^{60}Co	1.173	1.0157	1.10
^{60}Co	1.332	1.0158	1.10

Table 3 Obtained values of least square approximation method

Parameters	P_1 (MeV^{-1})	P_2 (MeV^{-1})	Sum Square Error
Values	1.015	0.000434 MeV^{-1}	5.066×10^{-10}

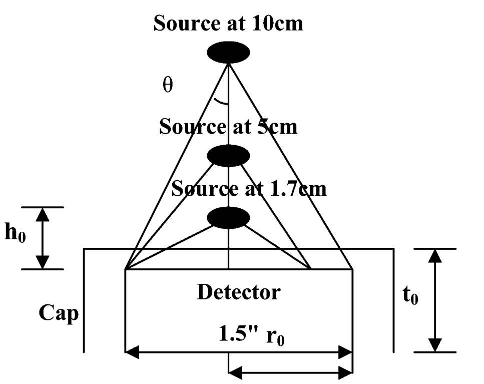

Fig. 4 Geometrical arrangement using NaI crystal and ^{137}Cs/^{60}Co isotopes where r_0 is the detector radius, t_0 is the detector length and h_0 is the distance between the source and detector.

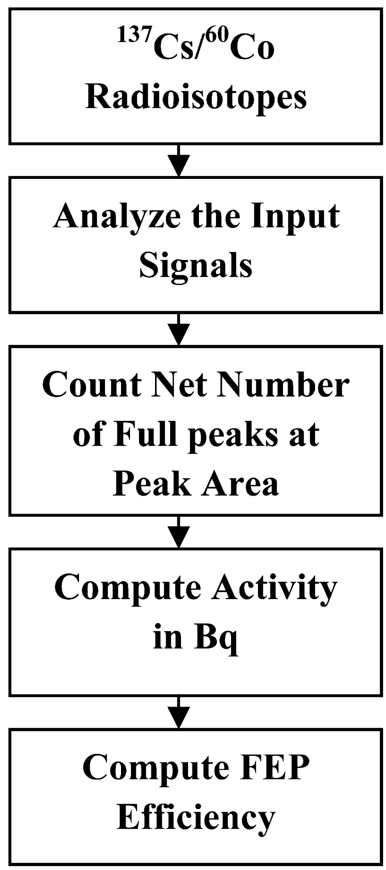

Fig. 6 FEP efficiency algorithm.

where, A_0, t, $\lambda = 0.693/T_{1/2}$, and $T_{1/2}$ denote the true activity, the age of the radioisotope, the decay constant, and the half life time of the isotope, respectively. The reference values of the parameters used in coincidence summing algorithm are shown in Table 1.[20,21]

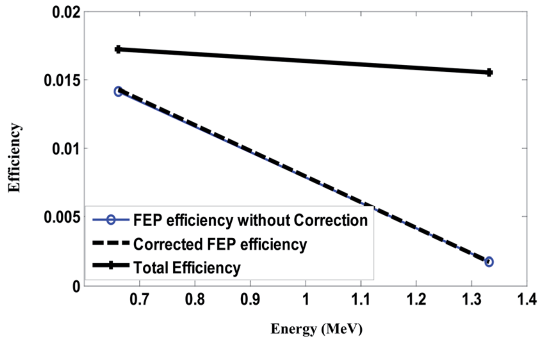

Fig. 7 FEP efficiency and total efficiency *vs.* energy.

Table 4 Effect of coincidence summing correction factor on different FEP efficiencies

Isotope	Energy (MeV)	ε_p (FEP efficiency)	ε_t (total efficiency)	Corrected FEP Efficiency
^{137}Cs	0.662	0.0141	0.0172	0.0143
^{60}Co	1.173	0.0016	0.0156	0.00157552
^{60}Co	1.332	0.0017	0.0155	0.00172686

The coincidence correction factors for both ^{137}Cs and ^{60}Co at different energies are shown in Table 2. These values are computed as illustrated in Fig. 1. Comparisons between our obtained analytical results and published results in the literature are given in Table 2. As illustrated in this table, the obtained analytical results are in good agreement with the experimental results in Ref. 22. However, the coincidence correction factor against energy is illustrated in Fig. 3. From this figure, the required correction factor increases with energy. We noticed that the number of simultaneously emitted photons increases with energy. Consequently, a higher correction value is required.

By applying the least square approximation method to the data of Table 2, a general relation between the correction factor and energy is obtained. Therefore, the coincidence summing correction factor is introduced as follows:

$$\mathrm{CF} = P_1 + P_2E \tag{12}$$

where P_1 and P_2 are the adjustment parameters, and E denotes the energy in MeV. The values of these constants are depicted in Table 3.

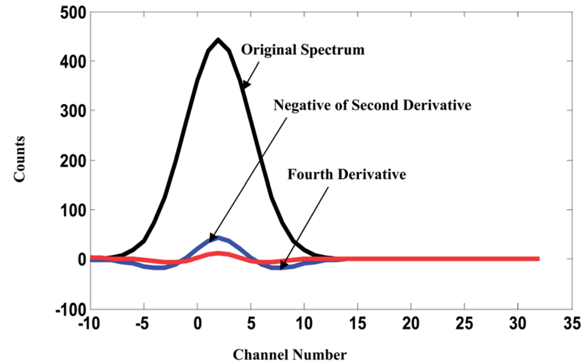

Fig. 9 Original spectrum in combination with negative second and positive fourth derivative.

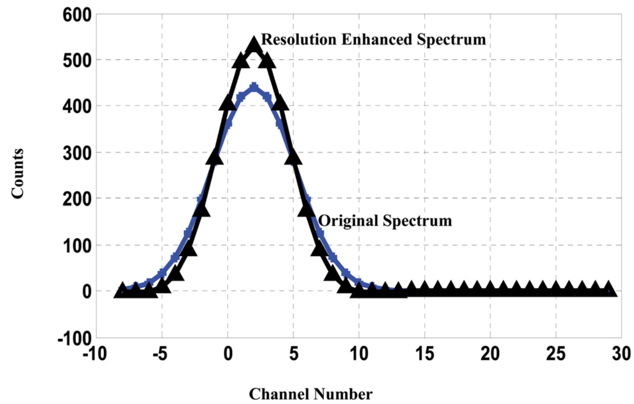

Fig. 10 Resolution enhanced spectrum using the negative second derivative weighting factors.

3.1 Geometry efficiency measurement

Geometry efficiency is the fraction of gamma rays that are emitted by the source and arrive at the detector surface.[19,23] In other words, it is the fraction of emitted photons that are intercepted by the detector.[23] The geometry efficiency is independent of the source. However, it depends on both the source-to-detector distance and the detector geometry[23,24] as depicted in Fig. 4. Therefore, we are interested in the calculation of the geometry efficiency of NaI detectors in detecting gamma radiation from ^{137}Cs and ^{60}Co point source. For a point source, the geometry efficiency is given by:

$$\varepsilon_{\mathrm{geom}} = \frac{a}{4\pi d^2} \tag{13}$$

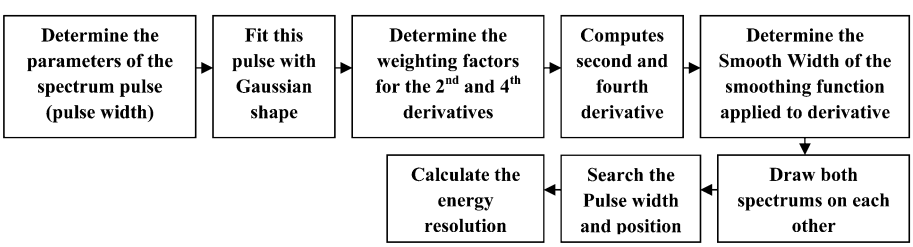

Fig. 8 Resolution enhancement algorithm of gamma ray spectroscopy.

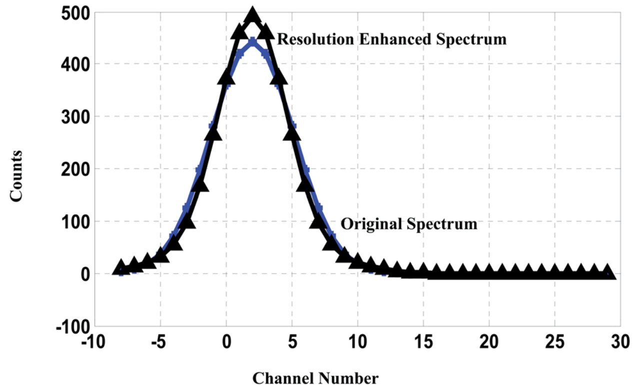

Fig. 11 Resolution enhanced spectrum using the positive fourth derivative weighting factors.

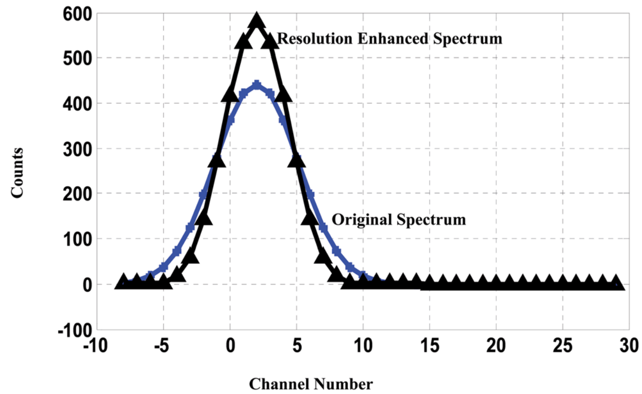

Fig. 12 Resolution enhanced spectrum using the negative second and positive fourth derivative weighting factors.

where a and d denote the cross-sectional area of the detector and the source-to-detector distance, respectively. This factor is essentially independent of photon energy. It manifests the well-known inverse-square law for counting rates as a function of source-to-detector distance. Geometric correction against source-to-detector distance is depicted in Fig. 5. We noticed that the correction factor becomes acceptable at a specific distance and suddenly drops with distance. Also, there is a specific correction for each individual source detector distance.

3.2 FEP efficiency measurement

Absolute full energy peak (FEP) efficiency relates the peak area in the spectrum to the number of gamma rays emitted by the source. It depends on the geometrical arrangement of the source and detector. An algorithm for FEP calculation is illustrated in Fig. 6. The acquired ^{137}Cs/^{60}Co radiation signal was analyzed and the spectrum is drawn. Then, the number of peaks corresponding to the position of maximum peak is computed. The activity was converted into disintegration per second. This efficiency is of most significance in practical gamma-spectroscopy and is given by applying the following formula:[19]

$$\varepsilon_{\mathrm{p}} = \frac{R}{SI_{\gamma}} \tag{14}$$

where R and S denote the full energy peak count rate in counts per second, and the source strength in disintegrations per second (Bq), respectively. However, absolute total efficiency relates the number of gamma rays emitted from the source to the number of counts detected anywhere in the spectrum taking into account the full energy peak and all incomplete absorptions represented by the Compton continuum.

The origin of the deviation of the efficiency data from smooth curves as a function of energy is due to the presence of important coincidence summing effects in the case of ^{137}Cs and ^{60}Co isotopes. The coincidence summing effects are peak and nuclide specific, therefore it is necessary to consider them in order to obtain a generally useful efficiency curve.

The efficiency calibration represents the efficiency of the semiconductor detector system as a function of energy. It

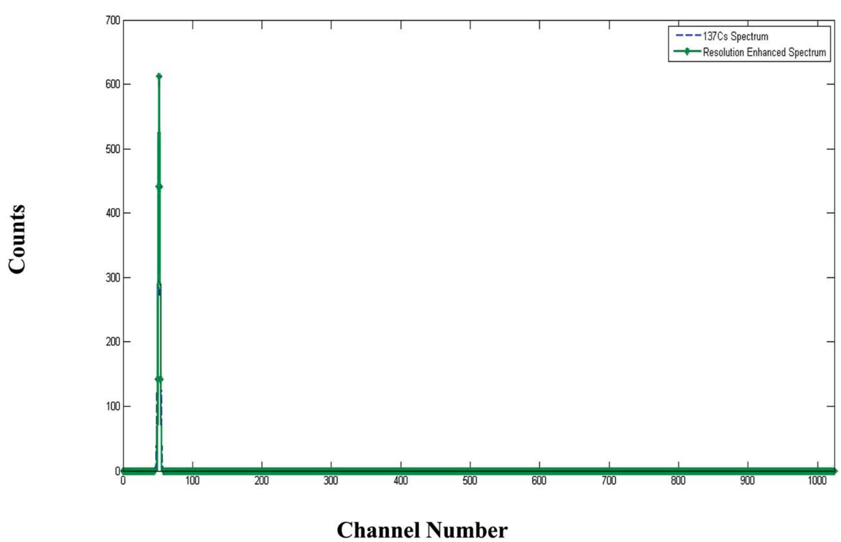

Fig. 13 Resolution enhanced spectrum of ^{137}Cs using the negative second and positive fourth derivative weighting factors.

Table 5 Actual and measured resolution obtained values for negative second and positive fourth derivative weighting factors

	Actual resolution	Measured enhancement resolution	Resolution enhancement factor	Execution time (ms)
Negative of 2nd derivative only	3.7054	3.1193	0.1582	172
Positive 4th derivative only	3.7054	3.2796	0.1149	78
Both 2nd and 4th derivative	3.7054	2.8149	0.2403	203

includes various effects from the intrinsic detector crystal, the detector and source geometry, the materials surrounding the detector, and absorption in the source matrix.[9] It is needed for each source-to-detector combination. Therefore, total efficiency and FEP efficiency before and after coincidence summing correction against the energy for $^{137}Cs/^{60}Co$ isotopes are depicted in Fig. 7. The corrected FEP efficiency is comparable to that without correction as indicated in Table 4. This is due to smaller source-to-detector distance. Also, the counting rate in this experiment is small. Consequently, smaller numbers of photons are emitted simultaneously and fewer enhancements occur. Moreover, the total efficiency at different energies for these isotopes were measured as indicated in Table 4.

4. Resolution enhancement algorithm

The first and most important detector parameter to consider is energy resolution. It is a measure of its ability to separate two peaks that are fairly close together in energy.[25] A detector with high resolution usually gives more accurate assays than one with low resolution. It is easier to determine accurately the area of full-energy peaks in a complex spectrum when the peaks do not overlap. Also, the probability of overlap is less with narrower peaks.[26] Moreover, full-energy-peak areas are easier to evaluate in high-resolution spectra because the interference from small-angle Compton scattering in the sample is reduced. Gamma rays that undergo small-angle scattering lose only a small amount of energy.[26] For a peak with a main pulse height (H_0) the resolution is obtained by the following relation:[26]

$$R = \frac{\text{Full Width Half Maximum}}{H_0} \tag{15}$$

An efficient algorithm enhancing the resolution of the resulting gamma ray spectroscopy is introduced. This algorithm is illustrated in Fig. 8. A resolution enhancement algorithm is very important especially with complex spectrum using NaI (TI) crystals. This algorithm is based on the smoothed second derivative and fourth derivative of the original signal. In other words, this algorithm represents the weighted sum of the original signal and the negative of its second derivative. On the other hand, this algorithm is based on subtracting the smoothed second derivative and adding the smoothed fourth derivative to the original signal according to the following relation:[27]

$$\Re = \mathrm{O} - \delta \times \mathrm{O}'' + \delta \times \mathrm{O}'''' \tag{16}$$

where $\Re$, O, O$''$, O$''''$, and δ denote the resolution enhanced signal, the original signal, the second derivative of O, the fourth derivative of O, and a user-selected weighting factor, respectively. The exact value of the weighting factors, δ, for the resolution enhancement algorithm is user dependent.[27] The optimum choice depends upon the width, shape, and digitization interval of the signal.[27] As a starting point, a reasonable value for δ is $\omega^2/25$ for peaks of Gaussian shape, where ω is the number of data points in the half-width of the component peaks.[27] The original signal, negative of second derivative and the fourth derivative signal are depicted in Fig. 9. The most important property of the present algorithm is that it does not change the total peak area.

The resolution enhancement results for negative second derivative, positive fourth derivative and summation of both negative second and positive fourth derivative weighting factors are shown in Fig. 10–12, respectively. Comparison between these derivative methods for actual and measured resolution is depicted in Table 5. Therefore, the accuracy of the proposed algorithm is validated as shown in this table. From the results of this algorithm, we concluded that using the negative second derivative and positive fourth derivative introduces better enhancement of the energy resolution. However, the negative second derivative method gives better resolution enhancement than the positive of the fourth derivative method. Moreover, the effect of resolution enhancement algorithm on real data spectra of ^{137}Cs is illustrated in Fig. 13.

5. Conclusion

This paper presents the development of algorithms that could be used in improving the performance of gamma ray spectrometers. These algorithms are the coincidence summing, FEP efficiency and resolution enhancement. Correction of the coincidence summing effect is based on mathematical equations since radiation point sources are used. Correction factor for both ^{137}Cs and ^{60}Co isotopes were implemented. The relation between the correction factors against different energies is obtained through a simple linear fitting equation. This is accomplished using the least square approximation method with minimum sum square error. FEP efficiency which is the figure-of-merit of gamma ray spectroscopy is taken into consideration. It was calculated for two cases; before and after coincidence correction. We noticed that correction of coincidence summing enhances both FEP efficiency and consequently the total efficiency of gamma ray spectroscopy. The FEP efficiency was enhanced by a factor of 1.58%. Therefore, it guarantees accurate determination of the source activity. Moreover, good agreement was obtained from the comparison of our results of coincidence summing algorithm with experimental results. Additionally, an algorithm for improving the energy resolution is presented. This algorithm is necessary for radioisotopes with complex energy spectra. This algorithm is based on the second and fourth derivative method and their weighting factors. From the obtained results, the energy resolution was enhanced by a factor of 24.03%.

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