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ARTICLE

Low-energy Gamma Unfolding Using NaI(Tl) Geant4 Detector Model and GRAVEL Code

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Abstract: In the field of gamma spectrometry with calibrated NaI(Tl) 3" × 3" γ-ray spectrometer, the correction of the photo-peak region of the original spectrum was discussed. In the present paper, GRAVEL algorithm based on Bayes theorem was used to test the unfolding process at low energetic interval, where Compton continium and background contributions are dominant. The simulated response matrix was constructed from mono-energetic γ-ray using validated Geant4 code. The calculated spectrum was compared with experimental ^{133}Ba gamma spectrum. The compared activities were found in good agreement in 4.10 % order.

Keywords: Spectrum unfolding, Gamma-ray spectra, Geant4 simulation, NaI(Tl) detector.
PACS: NaI(Tl): Sodium iodide (NaI) detector activated by thallium (Tl), Geant4: GEometry ANd Traking, FWHM: Full Width at Half Maximum.

1. Introduction

Up to now, the unfolding concept was successfully presented as a powerful method for spectral readability, especially for poor-resolution detectors, complex spectra and raw data. The unfolding process for neutron and gamma spectra was carried out by computer programs [1-7], where the instrument response function and pulse height distribution are needed. At this end, the instrument response function was constructed by numerical simulation using validated Geant4 code.

In this study, the interesting part is to check the validity test of unfolding GRAVEL code at the low-energetic range. Hence, the iterative GRAVEL validity was carried out for ^{133}Ba gamma-ray spectrum using an NaI(Tl) detector. The unfolded spectrum was compared with certified source activity to validate the followed approach and an acceptable increment of 4.10 % was found.

2. Materials And Methods

2.1 Running the GRAVEL Code

Let $Z(E)$ be the original spectrum emitted by a parallel γ calibration source located at X distance from the front surface of the detector, being the unknown and registered by an NaI(Tl) scintillation counter. $\Phi(E)$ is the experimentally observed spectra. $R(E, E_0)$ is the detector response function, recorded at energy E for emitting gamma ray at E_0.

The unfolding mathematical equation concerning $Z(E)$, $\Phi(E)$ and $R(E, E_0)$ is generally described as [4, 8-10]:

$$\phi(E) = \int_0^\infty Z(E) \times R(E, E_0) dE \qquad (1)$$

To solve this equation, the matrix detection system $R(E, E_0)$ and the measured pulse height

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spectrum $\Phi(E)$ can be expressed as in the following matrix equation [9]:

$$\phi(E) = Z(E) \times R(E, E_0) \quad (2)$$

For the unfolding process and inverting the instrument matrix $R^{-1}(E, E_0)$ in few channels by a successful way [11-12], a modified version of SAND-II method based on an iteration algorithm [6, 11] to a new method called GRAVEL [10] is used. The counts' vectors $\Phi(E)$, $Z(E)$ and the response matrix $R(E, E_0)$ can be rewritten in a matrix form as [7, 11]:

$$\phi(E) = \sum_l R_{kl} \times Z_l \quad (3)$$

The next gamma iteration record obtained after the unfolding process can be exploited discretely as [14-15]:

$$Z_i^{\alpha+1} = Z_i^{\alpha} \times \exp\left(\frac{\sum_k^n k \times A_{ik}^{\alpha} \times \ln\left(\frac{\phi_k}{R_k}\right)}{\sum_{k=1}^n k \times A_{ik}^{\alpha}} \right) \quad (4)$$

where $Z_i^{\alpha+1}$ is the new spectrum, Z_i^k is the γ emitted after α iteration, A_{ik}^{α} is the weighting factor [12] and R_k^{α} is the numeric pulse corresponding to γ convolution fluence [11-12].

The GRAVEL iteration algorithm stops running when the χ^2 is minimum [9]. It should be noted that the χ^2 value per degree of freedom n (Eq. 5) describes the goodness of the deconvolved spectrum. Also, it is used as a criterion for stopping the iteration procedure [9,11]:

$$\frac{\chi^2}{n} = \frac{1}{n} \sum_i \frac{\left(\sum_j R_{ij} Z_j - Z_i \right)^2}{\sigma_i^2} \quad (5)$$

From Eqs. (5) and (3):

$$\frac{\chi^2}{n} = \frac{1}{n} \sum_i \frac{\left(\phi_i - Z_i \right)^2}{\sigma_i^2} \quad (6)$$

Z_i and Φ_i are the unfolded spectrum and the measured spectrum, respectively, after i iterations. σ_i is the uncertainty of the measured spectrum.

As a consequence, if χ^2 per degree of freedom is almost 1, the iteration is convergent [11]. In this work, we have used 10000 iterations.

2.2 Detector Modelling

A cylindrical scintillation detector is coupled directly to a photomultiplier tube, hermetically sealed on the back of the crystal, housed by aluminium around it with a density of 2.7 $g.cm^{-3}$ and surrounded by MgO powder. The MgO and NaI(Tl) crystal densities are 2.0 $g.cm^{-3}$ and 3.667 $g.cm^{-3}$, respectively [13-14]. The SiO_2 back part was not considered during Geant4 (package 4-9.6.4 versions) simulation. In this study, the physics list used is G4EmStandardPhysics.

In Fig. 1, a screenshot of the NaI(Tl) 3"×3" detector model is shown.

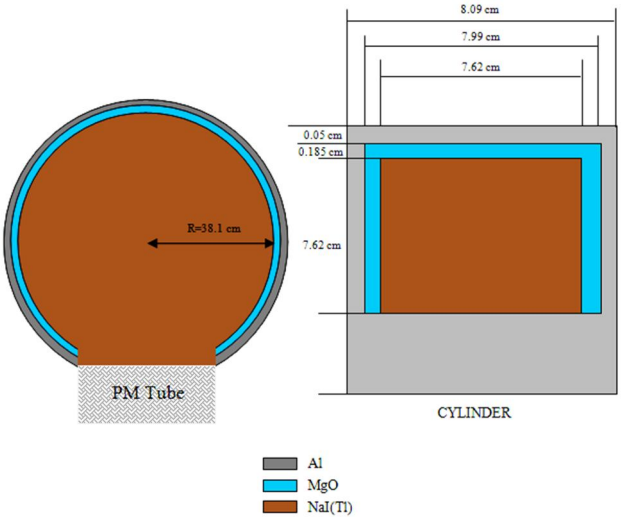

FIG. 1. Representation of the scintillator detector model.

The Gaussian energy broadening function was accounted for [15] by using three photopeaks of ^{137}Cs (661.6 keV) and ^{60}Co (1173.2 and 1332.5 keV) sources. Then, a non-linear fitting of the measured FWHM is used. In Eq. 7, the measured FWHM as an energy function can be expressed as:

$$FWHM = a + b\sqrt{E + cE^2} \quad (7)$$

where E is the incident particle energy, a, b and c are the energy broadening parameters determined by non-linear fitting (see Table 1).

TABLE 1. Adjusting FWHM parameters.

a (keV)	*b (keV^{-1})*	*c (keV^{-1})*
-88.7888	8.57634	0.000558254

The simulated geometry will be used to validate the Monte Carlo code by a punctual ^{137}Cs source fixed at 1 cm from the central axis of the detector. Experimentally, the acquisition time was kept as long as possible to get 1% statistical uncertainty.

During the Monte Carlo simulation, 10^6 events were generated.

2.3 NaI(Tl) Response Function

Due to the lack of mono-energetic sources, experimental measurements of the response function *R(E, E$_0$)* are not possible. As known, the spectral response function can be calculated using Monte Carlo methods [18-19]. In the case of a certified Bariym-133 source, gamma emission at 80.99 keV is chosen.

The simulated low-energetic boundary varied from 20 to 110 keV. The simulation series was carried out at an increment of 1 keV. The structure of each single response matrix for E_0 should be arranged in ascending order [11]. For good statistical results of each centroid peak, the history numbers were chosen by Geant4 code to achieve 1% uncertainly.

3. Results and Discussion

3.1 Validation of NaI(Tl) Detector Model

In Fig. 2, a 3D visualisation of the 3"×3" NaI(Tl) detector model considered in Geant4 simulation is presented. On the right side, we see the vertical section and on the left side, the NaI(Tl) detector structure is shown. In the same figure, the inner structure as crystal and the surrounding materials can be easily distinguished. Fig. 3 and Table 2 show a direct comparison between experimental and simulated pulse height distributions as the energy function of ^{137}Cs calibration source counted by the NaI(Tl) detector.

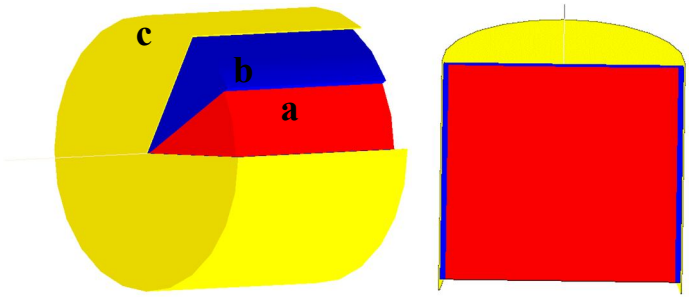

FIG. 2. Representation of scintillator detector model, (a): NaI(Tl) crystal, (b): MgO powder and (c): Aluminium cover.

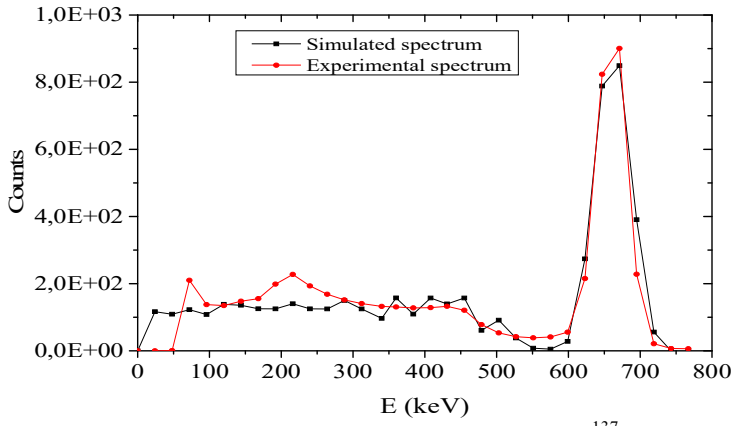

FIG. 3. Experimental and simulated pulse height distributions of ^{137}Cs punctual source.

TABLE 2. Measured and simulated data of ^{137}Cs source.

Energy (keV)	*Experimental*	*Geant4*
0	0	0
24	0	117
48	1	109
72	210	122
96	136	106
120	133	137
144	147	135
168	155	124
192	198	124
216	227	139
240	139	124
264	168	124
288	151	150
312	140	124
340	132	97
360	130	157
384	127	109
408	128	157
431	132	139
455	121	157
479	78	61
503	53	91
527	41	38
551	39	8
575	41	5
599	56	27
623	215	278
647	823	778
671	901	849
695	228	390
719	21	56
743	7	0
767	6	0

The obtained pulse height distribution was normalized by source activity [14]. The background radiation was subtracted from the original spectra and an acceptable accuracy between simulated and experimental data even in energy resolution at 661.6 keV is observed.

A similar test configuration using Monte Carlo codes was carried out with NaI(Tl) detector [14] and the same spectra form, approximately, was observed.

Below 300 keV, the Compton edges are lower than the measured spectrum, probably because of the scattered photons in the unclosed cylindrical shielding system around the detector [16], noting that the K-X-ray peaks of ^{137m}Ba source [17] are not considered in Geant4 simulation.

3.2 Unfolding Experimental Distribution by GRAVEL Algorithm

After background removal from ^{133}Ba experimental spectrum, $\Phi(E)$, GRAVEL iteration algorithm is used now for a few channel energies. The simulated response function $R(E, E_0)$ and pulse height vector $\Phi(E)$ are used. In Fig. 4, the unfolding spectrum (continuous line) shows a pulse height lower than in the measured spectra in the energy range 60 to 90 keV. It is due to the Compton and background removal. Consequently, the deconvolved area will be used to estimate the source activity. These results will be presented as histogram data. This type of figure is commonly used to represent similar results (e.g., Matzek (2002) and Cheminet (2003)).

Table 3 presents a comparison between the certified activity and the estimated activity.

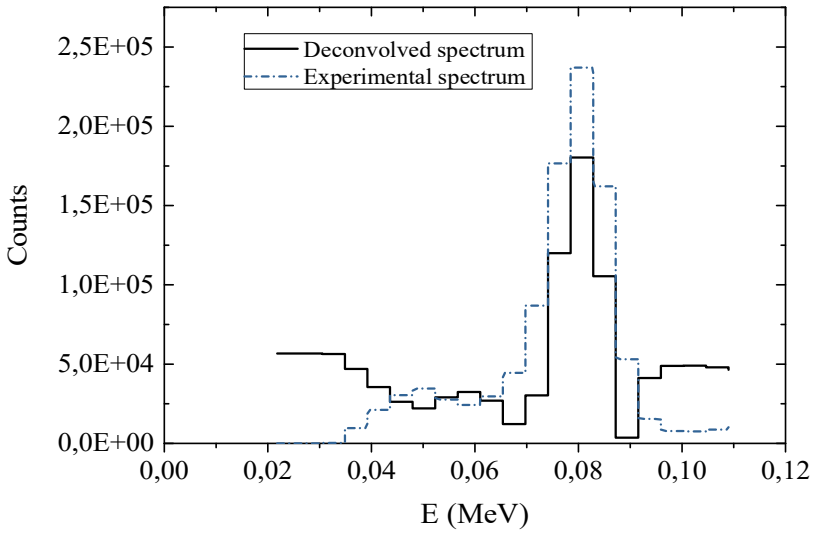

FIG. 4. Original spectrum (dashed line) and unfolded spectrum obtained by GRAVEL algorithm (continuous line) after 10000 iterations using ^{133}Ba at 81 keV energy.

TABLE 3. ^{133}Ba activity, (10^3 Bq).

Isotope	*Reference activity*	*GRAVEL*	*Relative error (%)*
^{133}*Ba*	29.2	28.0	4.10

4. Conclusion

In this work, the NaI(Tl) response function constructed by virtual mono-energetic photon sources using the validated Geant4 code is presented. The Geant4 simulation showed that the Gaussian energy broadening, experimentally measured, must be introduced during detector simulation.

The GRAVEL iterative method has been applied to experimental data in the 20-110 keV energy range.

The GRAVEL results are very encouraging and can be used with a high accuracy at low-energetic ranges. Besides, the unfolding spectra can be quickly analyzed because of Compton and background removal.

References

[1] Hartman, J. and Barzilov, A., Physics Procedia, 66 (2015) 410.

[2] Tripaty, S.P., Sunil, C., Nandy, N., Sarkar, P., Sharma, D. and Mukherjee, B., Nuclear Instruments and Methods in Physics Research-Section A, 583(2007) 421.

[3] Felter, T., "Development of a Neutron Unfolding Code", (2013) 1–9.

[4] Ewa, L. and Katarzyna, MM., Fusion Engineering and Design, 156 (2020) 111593.

[5] Shuhuan, L., Chen, D., Nannan, D., Jingye, A., Wenshou, Z. and Kai, W., Nuclear Science and Techniques, 18 (2007) 77.

[6] Rahman, M.S., Cho, G. and Kang, B.S., Radiation Protection and Dosimetry, 135 (2009) 203.

[7] De, S., Thomas, R.G., Rout, P.C., Suryanarayana, S.V., Nayak, B.K. and Saxena, A., Journal of Instrumentation, 13 (2018) 460.

[8] Tsoulfanidis, N., "Detection of Radiation Fields", Second Edn., (Taylor and Francis, University of Missouri-Rolla, 2005).

[9] Matzke, M., Instruments and Methods in Physics Research-Section A: Accelerators, Spectrometers, Detectors and Associatted Equipment, 476 (2002) 230.

[10] McElory, W.N., Berg, S., Crockett, T. and Hawkins, R.G., SAND II (Report AFWL-TR-67-41). US Weapons Laboratory, United States (1967).

[11] Matzke, M., The HEPROW Program System, (2003).

[12] Cheminet, A., Ph.D Thesis, University de Toulouse, (2013).

[13] Tekin, H.O., Science and Technolgy of Nuclear Installations, 2016 (2016) 7.

[14] Salgado, C.M., Brandão, L.E.B., Schirru, R., Pareira, C.M.N.A. and Conti, C.C., Progress in Nuclear Energy, 59 (2012) 19.

[15] Rahman, M.S. and Cho, G., Journal of Scientific Research, 2 (2010) 221.

[16] Berger, M.J. and Seltzer, S.M., Nuclear Instrumentation and Methods in Physics Research, 104 (1972) 317.

[17] Heath, R.L., "Scintillation Spectrometry: Gamma-ray Spectrum Catalogue", New Version, (1997).

[18] Kovaltchouk, V. and Machrafi, R., Annals of Nuclear Energy, 38 (2011) 788.

[19] Belgya, T. and Szentmiklósi, L., Nuclear Instrumentation and Methods in Physics Research-Section A, 991 (2021) 165018.