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Monte Carlo Determination of Full Energy Peak Efficiency for HPGe Detector and Self-Absorption Correction of Environmental Samples

M.R. Abdi[1,3], M. Mostajaboddavati[1], H. Faghihian[*2,3]
1- Department of Physics, University of Isfahan, P.O. Box: 81747-73441, Isfahan - Iran
2- Department of Chemistry, University of Isfahan, P.O. Box: 81747-73441, Isfahan - Iran
3- Azad University of Shahreza, P.O. Box: 86145-311, Isfahan - Iran

Abstract: The Monte Carlo method was used to determine full energy efficiency of a high-purity germanium (HPGe) coaxial detector within an energy range of 53.2-2614 keV. Also, measurement was carried out for a standard Marinelli beaker of 600cm^3, which was placed into the reference material of mixed gamma. The plot of the experimentally derived efficiency versus the incident gamma radiation energy showed variations at certain energies which is attributed to the detector's characteristics. The results obtained by the Monte Carlo method and those of the experimental measurements resulted in a standard deviation of 2.3 to 7%. The method was also applied to determine self-absorption correction of the environmental radioactive materials which gave rise to a deviation of 3–12%.

Keywords: ***Monte Carlo Method, Self-Absorption, High Purity Ge Detector, Energy Efficiency***

1-Introduction

Currently gamma ray spectrometry with Ge detectors is widely employed to determine the radionuclide concentration of a great variety of samples. The calculation of the activity from the peak areas requires the knowledge of the full-energy peak efficiency at the energy of the γ-ray emissions for the measuring condition. The efficiency depends on several factors that can be classified into two groups: those related to the intrinsic characteristics of the detector, such as its active volume geometry and surrounding materials, and those related to the measured sample, such as geometrical setup and physicochemical characteristics of the matrix. The last two factors imply that an efficiency calibration of the detector needs to be carried out for each source configuration in order to obtain accurate emission rates [1]. In particular, for intended sources, self absorption effects make the detector efficienecy, ε, very sensitive to changes on the type of matrix in the low-energy range causing one to perform a determination of the efficiency for each variation in the matrix composition.

Calibration sources require long counting time and a great deal of work in order to obtain a measured efficiency calibration curve for each setup. Monte Carlo methods are practical tool, that can be applied to calculate new efficiency value conditions. Nevertheless, the accuracy of such simulation techniques must be evaluated before incorporating them in the laboratory. Several studies of the response of high resolution γ ray spectrometers with Monte Carlo calculation have been published. Most authors reported agreement with the experimentally obtained efficiency within 10% [1, 2], and in the recent years the simulation techniques have been improved, with a low error of about 3% [3], except in the low energy range.

It should be emphasized that Monte Carlo studies are seldom discussed for energies of 50 to 3000 keV [4]. However, this region is very interesting in environmental studies since several radionuclide of the ^{238}U and ^{232}Th series emit γ–rays at these energies. For the purpose of our study, the low–energy region is useful for the determination of the main sources of error in the simulation since ε varies rapidly in this interval. In fact, it is the boundaries of the best areas to test the validity of the detector geometrical parameters.

This work determines the influence of detector and the source characteristics in the Monte Carlo simulations of the Ge detector

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*email: h.faghih@sci.ui.ac.ir

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response by comparing the experimental and the calculated efficiencies for environmental sample located in the range 50-3000 keV. Various sources like detector geometrics, several matrices such as soil, sediment and mixed gamma are required to determine the sensitivity of the efficiency of the detector parameters and the sources characteristics.

2-Experimental Efficiency Calculation

Experimental efficiency calibration curves of a Ge detector were determined for a 600 ml Marinelli breaker. The measurement was performed with an ORTEC HPGe detector, with relative photo peak efficiency of 10% at 1332.5 keV and the resolution was 1.8 keV for the 1332.5 keV gamma-ray transition of ^{60}Co. The experimental efficiencies were obtained by measuring calibration sources produced by spiking with a definite volume of reference materials.

The radionuclide employed for calibration were ^{132}Ba, ^{241}Am, ^{152}Eu, ^{24}Na, ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs. whose emissions cover the range of 53.2–2614 keV. The radionuclide activities were low enough to neglect the random–summing effects, but coincidence–summing and self-absorption corrections were made. These correction factors were derived by Monte Carlo experiment and once corrected, the experimental data were fitted to a spline function by a least–square analysis in order to find the optimum parameters of the function. With this procedure the efficiency uncertainty was less that 1% in the whole energy range for environmental samples in Marinelli beakers [5, 6].

3-Monte Carlo

The Monte Carlo method was applied, using the MCNP(1) 4C Code to estimate the HPGe detector efficiency and calculating detector efficiencies [7]. The history of each photon is followed until its energy is dissipated, taking in to account all the secondary particles created by the interaction processes. These interaction processes that were considered for photons are: photoelectric effect, compton scattering and pair production. For the electrons generated in such processes, multiple scattering, ionization and bremsstralung due to difficulties to evaluate the propagation of other uncertainties in the calculated efficiencies. We have only considered statistical uncertainties due to the restricted number of primary photons. A part from this, factors limiting the accuracy of the Monte Carlo simulated efficiencies are as follow:

- The uncertainties in the interaction parameters, such as cross sections and ranges.
- The simplifying assumptions made to simulate interaction processes and
- The errors in the detector and source description due to the lack of a precise knowledge of their characteristics.

With respect to the third factor, whose analysis is the main objective of this work, in all simulation programs the detector is represented by a variable number of geometrical volumes.

In the next simulation, the detector consist of absorbing layers (aluminum end cap, inactive germanium) as specified by the manufacturers, were all included in the geometry and the detectors response was calculated for the same energy range again. The F8 (pulse height) tally used for photons in all cases was investigated without array variance reduction. The statistical behavior of the results and the assurance of the valid confidence intervals for each tally bin were assessed for each run by checking the associated tables in the tally fluctuation chart bin. A homogeneous source with a shape of a Marinelli beaker was modelled. The influence of the plastic composition of the beaker, as well as the reduction of the volume filled with Material were analyzed by Rodenas [8]. The number of total histories considered in each run, must be large enough to obtain tally results with the minimum uncertainty associated with the calculated efficiency. The rule given in manual is to keep the relative error (S_x/x) below 0.10 [7]. Nevertheless, when 1,000,000 source particles are started, we generally obtain a relative error of no more than 0.02, with an observed minimum of approximately 0.001. Consequently, this number of total histories was adopted for all runs.

4- Results and discussion

The effect of the detector window has been assumed to be negligible because of its low atomic number (Z=4) and its small thickness. The front dead layer was not considered, because the n-type Ge crystal configuration has a dead layer thickness of less than a few micrometers. As expected for its geometry, the Marinelli beaker is the most sensitive setup to the thickness of the aluminum. In this research, the experimental calibration curve and the Monte Carlo efficiencies for the geometrics Marinelli beaker, are as those shown in Fig. 1. The agreement was good, the differences being less than 7% (Table 1). When extended sources were analyzed, based on measured efficiency data, differences in density and chemical composition between the matrices of the samples and those used in the calibration process, can lead to errors in the deduced radionuclide activities due to differences in the self-absorption. In order to reduce errors for the soil samples that currently measured in our laboratory seven samples were prepared with the density between 1.1-1.6 gr/cm^3. For each fraction, the experimental efficiency were derived from seven samples at the energies of the calibration standard gamma-emission, and obtained different estimations for different samples. The experimental efficiency curve was calculated by a least-square fit of these values to a spline function. This fitting method smoothes the effect of experimental errors, such as a bad homogenization of the calibration standard. However, the variability in the efficiency values for the seven samples could be caused not only by experimental errors, but also by differences in the self-absorption (Table 2) [6].

5- Conclusions

The full-energy peak efficiencies for three different standards have been successfully simulated by MCNP in the energy range of 50-3000 keV. MCNP efficiency values have been presented even for energies below 88 keV, with the relative errors less than 4%. For volume sources, an accurate determination of the activity requires one to know not only the apparent density, but also the chemical composition of the matrix. It has been shown that even for environmental samples with very similar mineralogical, differences between their compositions density, provoke considerable changes on the efficiency value.

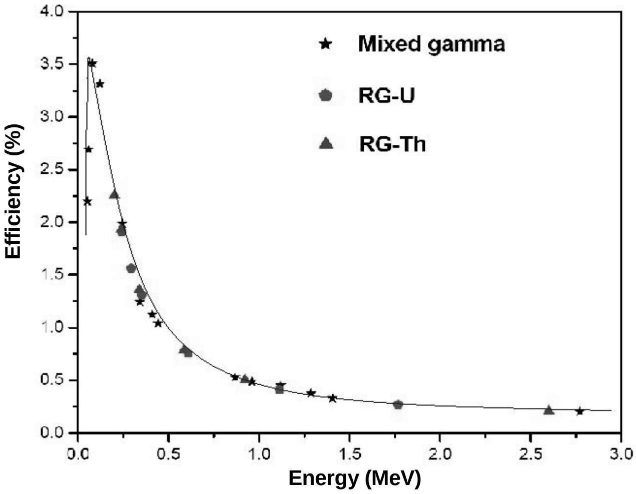

Figure 1- Comparison of the experimental efficiency calibration curve with the calculated efficiencies (solid-line) in the energy range 50-3000 keV for the Marinelli beaker.

Table 1- Comparison of experimental and simulated full-energy peak efficiencies for geometrics Marinelli beaker (with density of 1.207 gr/cm^3).

Energy(keV)	$\varepsilon_{\exp}$	ε_{MC}	$\delta(\%)$
53.2	2.2144	2.1435	- 3.2
59.54	2.662	2.72	2.2
81.0	3.5048	3.547	1.2
121.8	3.324	3.284	- 1.2
202.39	2.2437	2.172	- 3.2
238.63	1.923	2.021	5.1
295.21	1.569	1.6312	4.0
344.3	1.334	1.25	- 6.3
609.32	0.758	0.74	- 2.3
661.6	0.6936	0.6832	- 1.5
778.9	0.58824	0.622	5.8
911.16	0.5062	0.528	4.5
1120.28	0.41074	0.404	- 1.5
1460.8	0.31695	0.325	2.8
1764.52	0.26725	0.26	- 2.6
2614.7	0.20943	0.224	7.1
2754	0.20552	0.199	- 3.4

Table 2- Comparison of experimental and simulated self-absorption [6].

Energy(keV)	S_{exp}	S_{MC}	$\delta(\%)$
53.2	0.966	0.9196	- 4.8
59.54	0.968	1.0057	3.9
81.0	0.9734	0.9967	2.4
121.8	0.9779	0.9603	- 1.8
202.39	0.9708	0.9397	- 3.2
238.63	0.9696	1.040	7.3
295.21	0.9619	1.0388	8.0
344.3	0.9546	0.8524	- 10.7
609.32	0.929	0.8863	- 4.6
661.6	0.926	0.898	- 3
778.9	0.925	1.02675	11.0
911.16	0.928	1.005	8.3
1120.28	0.942	0.9232	- 2
1460.8	0.9885	1.0389	5.1
1764.52	1.0506	0.9949	- 5.3
2614.7	1.307	1.4664	12.2
2754	1.372	1.2869	- 6.2

Foot Note:

1– MCNP: Monte Carlo N-Particles Transport

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