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Comparison of Experimental and Monte Carlo Efficiencies of 0.5g/cc Epoxy Matrix Marinelli Source with Multiple Radioactive Nuclides

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Keywords	Abstract
Epoxy Marinelli Source Efficiency Monte Carlo	Using the gamma spectroscopy system, it can be determined whether environmental samples or standard radioactive sources are radioactive, and from which elements their radioactivity originates. The purpose of Monte Carlo (MC) simulation is to model a real-life system with its inputs and evaluate the outputs with real results. This study calculates the experimental efficiency of a p-type HPGe detector using a 0.5 g/cc Epoxy Matrix Marinelli beaker and compares these results with GESPECOR and PHITS MC Simulation programs. Thus, the thickness of the dead layer, which thickens over time and affects the detector efficiency, was determined from the most compatible result of the MC calculations made repeatedly at various alternative thicknesses to the experimental results. For 1.5 mm dead layer thickness, less than 2 % error was found between the test and MC results, especially at energies above 165 keV. As a result, it was determined that the dead layer thickness of the detector reached 1.5 mm with an increase of 114 % after its production. The current value of the dead layer thickness of each detector should be checked, as the efficiency affects the determination of the activity.

Cite

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1. INTRODUCTION

Marinelli beakers are sample containers that fit into the end cap of the detector, designed in such a way that the sample material is close to both the top and sides of the detector crystal for the highest sensitive counting of large sample volumes (Gilmore, 2008). In gamma spectrometry, when the radioactivity detected is low, Marinelli beakers can be used. These beakers maximize the counting efficiency of the detector because of their geometry that almost surrounds the detector (Ahmed et al., 2009). Note that to make quantitative measurements, it is necessary to obtain the full energy peak efficiency (FEPE) curve of the detection systems. The FEPE curve is affected the sample geometry, the sample matrix, and the density of the matrix as well as the detection system (Harb et al., 2008). The FEPE curve can be obtained either by using a known radioactive standard source homogeneously distributed in a Marinelli beaker of the same size and composition (Vasconcelos et al., 2011), or by accurately modeling the source geometry, matrix, the position of the source, and the detection system using MC-based simulations (Azli & Chaoui, 2015; Lépy et al., 2019). Using the Monte Carlo method, the detection process can be simulated and applied to obtain efficiency values for each geometry (Ródenas et al., 2003). It is a convenient tool for situations where it is not always possible to perform experimental calibrations due to expensive standard gamma sources (Bölükdemir et al., 2021), and no radioactive waste is generated in this process (Vasconcelos et al., 2011). This study aims to find the current dead layer (DL) thickness of a p-type HPGe detector with 30 % relative efficiency by using a Marinelli type

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source. DL thickness, which affects the detector efficiency as it changes, was investigated with GESPECOR and PHITS MC codes.

2. MATERIAL AND METHOD

2.1. Experimental Setup

A p-type coaxial HPGe (GCD-30185) detector with a 0.7 mm Al end cap thickness was used for the experiments (Figure 1a). The detector has a relative efficiency of 30 % and a peak to Compton ratio of 58:1 at 1.33 MeV. The resolutions of given energies are 1.85 keV at 1.33 MeV and 0.875 keV at 0.122 MeV. The size information of the detector used in the experiment is given in Table 1. The detector has a connection with a digital signal processing analyzer operating through Gamma Vision spectroscopy software. The experimental efficiency was calculated using a Marinelli beaker with reference multi-radioactive nuclides dispersed in a 0.5 g/cc epoxy matrix. The Marinelli beaker snugged on the detector has a volume of 1 liter that is comprised of radionuclides in the energy range of 46 - 1836 keV (Figure 1b).

Table 1. Geometric dimensions of the detector (mm)

Basic Detector dimensions		
Detector diameter	57.3	
Detector length	57.3	
Hole diameter	7	
Hole depth	40.8	
Detector dimensions and materials		
Description	Dimension	Material
End cap to crystal cap	7.5	N.A.
End cap diameter	83	Aluminum
End cap window	0.6	Aluminum
Insulator/shield	0.01	Aluminized/ Mylar
Outside contact layer	0.7	Lithium
Hole contact layer	0.003	Boron
Mount cap wall	1.5	Aluminum
End cap wall	1.5	Aluminum



Figure 1. a) HPGe Detector, b) Marinelli beaker with 0.5 g/cc density used in the experiment

$$\varepsilon(E) = \frac{N_p(E)/t}{A \cdot f_\gamma(E)} F_{coi} \quad (1)$$

Where $N_p(E)$ is the full energy peak net area, t is the gamma acquisition live time in seconds, A is the source activity in Bequerrels at the measurement date, f_γ , and F_{coi} are the gamma-ray emission probability of the interested energy (%) and TCS correction factor, respectively. To obtain the coincidence summing factors, GESPECOR, a program that can also calculate coincidence-summing effects with cascade gamma photons, coincidence losses, K_α , K_β , and multiple X-rays, was used.

2.2. PHITS

In this study, HPGe detector was modeled using PHITS version 3.26 (Particle and Heavy Ion Transport Code System) and Marinelli beaker that snugs on the detector. PHITS, which has recently gained increasing popularity, is a Monte Carlo particle transport simulation code and was developed by the Japanese Atomic Energy Agency in collaboration with various institutions in Japan and Europe (Sato et al., 2018). PHITS can transport most types of particles with an energy of up to 1 TeV using several nuclear reaction models and data libraries. While modeling the experimental setup with this simulation, first the given geometric parameters of the detector and its surroundings, such as crystal dimensions, the thickness of the end cap window, the DL thickness, the structure of the detector components, etc. are defined (Ordóñez et al., 2019; Zamzamian et al., 2020). Marinelli beaker with its dimensions, its wall thickness, and the material (EPOXY) inside it was also modeled in the simulation (Figure 2). T-deposit tally reveals the deposited energy distribution in a specified location by ionization of charged particles. It is used to calculate FEPE values for the energy of interest. The history number in this simulation is 10^6 for each energy value, a sufficient amount for the required counting statistics to obtain an uncertainty of around 1 %.

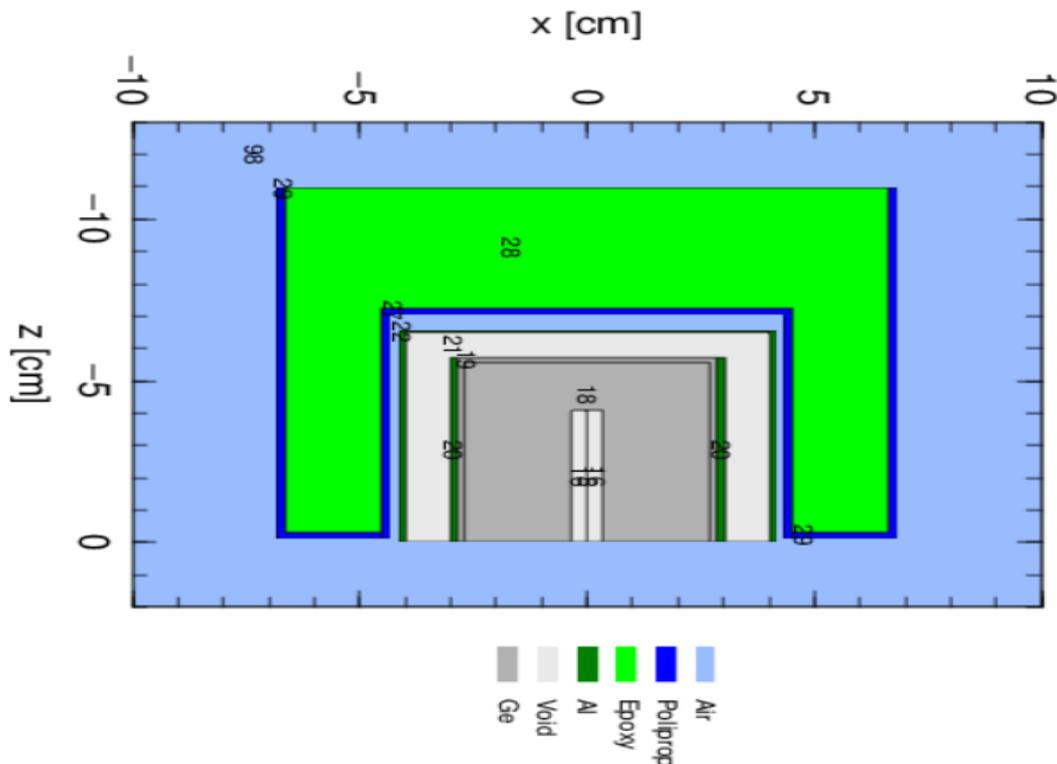


Figure 2. Two-dimensional view of the detector and Marinelli beaker modeled in PHITS

2.3. GESPECOR

Another MC simulation program used in this study is GESPECOR version 4.2, which is a practical and useful code commonly used in Gamma Measurement Laboratories. The efficiency computation, self-absorption effects, and coincidence summing can be calculated by just entering the parameters into its user-friendly interface. GESPECOR is easily applicable to various sample geometries such as point, cylindrical, and Marinelli measured using well-type, closed-end coaxial HPGe detectors (Sima et al., 2001; Murphy et al., 2020). While creating the model of the experiment using GESPECOR, first the detector was modeled by entering the relevant parameters in the interface, then the source geometry was selected as Marinelli and the experimental setup was simulated by entering the dimensional parameters of the Marinelli beaker, the components and density of the epoxy material in the container (Figure 3). The number of photons in this simulation was again taken 10^6 . The DL thickness was increased to 1.7 mm in 0.1 mm steps from the 0.7 mm provided by the manufacturer to determine the optimum DL thickness that was consistent with the experimental efficiency. As the DL thickness increased, the efficiency values decreased and the agreement with the experiment was observed.

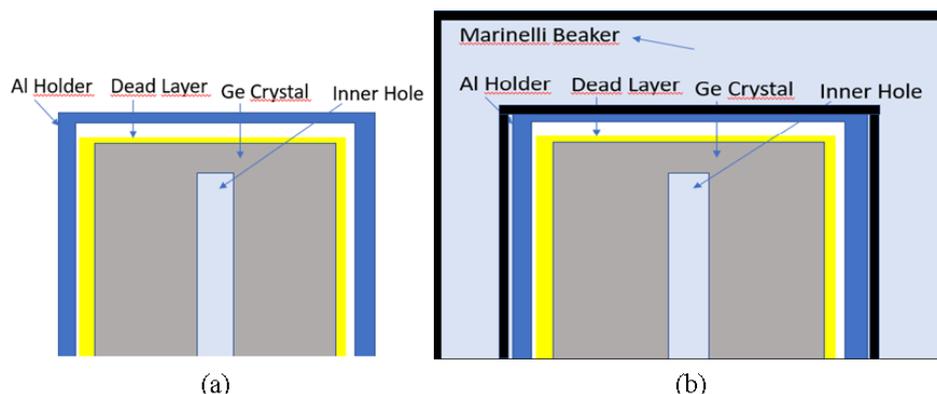


Figure 3. Two-dimensional view of the detector and Marinelli beaker modeled in GESPECOR

3. RESULTS AND DISCUSSION

The experiment was carried out with a Marinelli beaker containing a multi-energy point radioactive source in an epoxy matrix with a density of 0.5 g/cc attached to the detector. The elemental composition of the epoxy used was 62.040 % C, 27.547 % O, and 10.413 % H by weight. The radionuclides used in the experiment, the energy values of the gamma rays emitted from these nuclides, the source activity, the error percentage of the source activity, the half-life (days) of the radioactive sources, the gamma density, the experimental efficiency, the total count rate, and absolute error uncertainties are given in Table 2.

Table 2. The energy (keV) values, activities (μCi), half-lives, gamma emission probabilities, net count rate, experimental efficiency values, and percentage error of the multiple radioactive Marinelli sources used in the experiment

Nuclide	Energy (keV)	A_0 (μCi)	A_0 (μCi)		$t_{1/2}$ (day)	$f(\gamma)$	Net Count	ϵ -Exp	$U_{\text{exp}}\%$
			A_0 (μCi)	$U_{\text{Total}}\%$					
^{210}Pb	46.54	0.30320	4.0	8140	4.252	1.1	0.00229	4.4	
^{241}Am	59.54	0.03014	3.0	157740	35.920	4.0	0.01008	3.4	
^{109}Cd	88.03	0.42430	3.0	462.6	3.660	14.8	0.02750	3.1	
^{57}Co	122.06	0.01661	3.0	271.79	85.490	16.1	0.03431	3.1	
^{139}Ce	165.86	0.02051	3.0	137.64	79.900	16.1	0.03335	3.1	
^{203}Hg	279.20	0.06153	3.0	46.595	81.480	23.3	0.02418	3.1	
^{113}Sn	391.70	0.07979	3.0	115.09	64.970	27.5	0.01868	3.0	
^{85}Sr	514.00	0.10000	3.0	64.849	98.500	33.7	0.01481	3.1	
^{137}Cs	661.66	0.07143	3.0	11012	84.990	27.3	0.01218	3.0	
^{88}Y	898.04	0.16840	3.0	106.63	93.700	40.5	0.00969	3.0	
^{60}Co	1173.23	0.08579	3.0	1924.3	99.850	23.2	0.00786	3.0	
^{60}Co	1332.49	0.08579	3.0	1924.3	99.983	21.0	0.00710	3.1	
^{88}Y	1836.07	0.16840	3.0	106.603	99.346	24.3	0.00553	3.0	

The experimental setup was modeled using GESPECOR and PHITS MC programs. The DL thickness was increased from 0.7 mm given by the manufacturer to 1.7 mm in 0.1 mm steps. As the DL thickness was increased, a decrease was observed in the efficiency values. Except for 46.5 keV, 59 keV, and 88 keV energies, the photopic efficiency closest to the experimental efficiency was obtained at 1.5 mm thickness in both GESPECOR (Figure 4) and PHITS MC (Figure 5) simulations. This mismatch in the low-energy region is due to the DL thickness in p-type HPGe detectors (Jeřkovský et al., 2019). The DL thickness is due to lithium

intrusiveness on the outer surface of the Germanium crystal in high-purity germanium detectors (Modarresi et al., 2018). Even at room temperature, lithium atoms are aggressive to the crystal surface (Huy et al., 2007). The reason is that the manufacturer does not explain the DL thickness with precise measurements and that it reaches this thickness randomly during the lifetime of the detector. Even the millimeter variation in DL thickness must be carefully considered to arrive at an accurate calculation of the experimental efficiency. Discrepancies in the low energy region have also been reported for both Marinelli geometries and point sources in the literature (Ródenas et al., 2003; Huy, 2011; Britton et al., 2013).

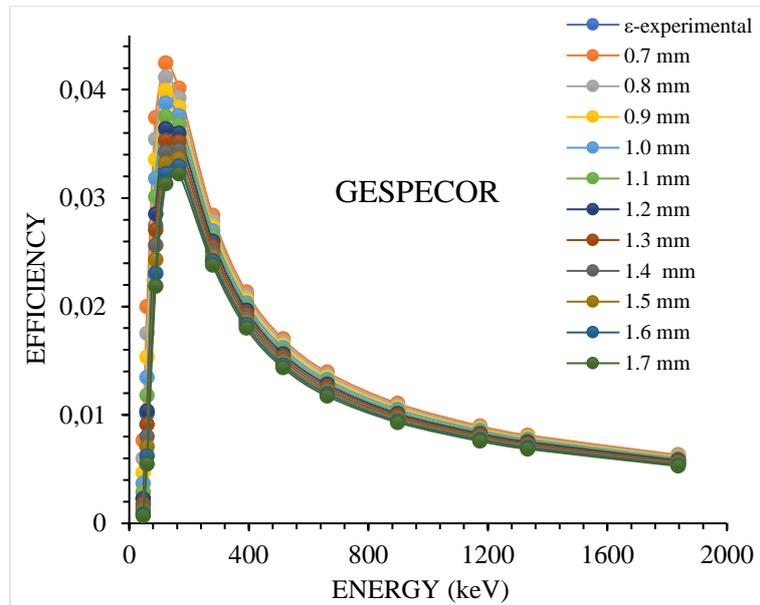


Figure 4. Energy dependence of efficiency from GESPECOR for DL thicknesses between 0.7 - 1.7 mm

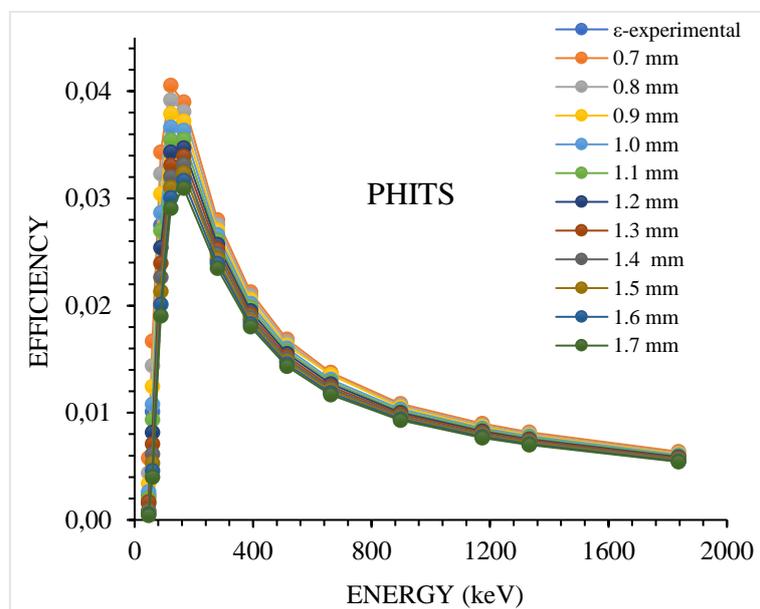


Figure 5. Energy dependence of efficiency from PHITS for DL thicknesses between 0.7 - 1.7 mm

The photon energy of 59.54 keV is insufficient to reach the crystal just through the detector's outer top dead layer, but the photon energy of 662 keV is sufficient to reach both the outer lateral and inner dead layers of the crystal (Azli & Chaoui, 2015). At energies above 122.06 keV, the results of both GESPECOR (Table 3) and PHITS MC (Table 4) codes are in good agreement with experimental efficiencies with a maximum deviation of 2 % and 2.8 % respectively, at 1.5 mm dead layer. These DL thicknesses can be determined effortlessly using simulation programs.

Table 3. The percentage difference in the efficiency values calculated using GESPECOR according to the dead layer change in the experimental efficiency

Energy (keV)	ϵ_{exp}	0.7 mm	0.8 mm	0.9 mm	1.0 mm	1.1 mm	1.2 mm	1.3 mm	1.4 mm	1.5 mm	1.6 mm	1.7 mm
46.54	0.00229	233.8	160.6	103.9	59.8	25.4	1.4	22.4	38.8	51.7	61.9	69.9
59.54	0.01008	98.3	73.6	52.0	33.3	17.0	2.8	9.7	20.6	30.1	38.5	45.8
88.03	0.02750	36.1	28.8	22.0	15.6	9.6	3.8	1.7	6.8	11.6	16.2	20.5
122.06	0.03431	23.7	19.9	16.3	12.9	9.4	6.0	2.8	0.2	3.2	6.0	8.7
165.86	0.03334	20.3	17.7	15.2	12.8	10.3	7.9	5.5	3.2	1.0	1.1	3.3
279.20	0.02418	17.5	15.4	13.4	11.5	9.5	7.6	5.7	3.8	2.0	0.3	1.6
391.70	0.01868	14.2	12.5	10.6	8.8	6.9	5.2	3.4	1.6	0.1	1.9	3.6
514.00	0.01481	14.9	13.0	11.1	9.3	7.4	5.6	3.8	2.1	0.2	1.4	3.0
661.66	0.01218	14.5	12.6	10.7	8.8	7.1	5.2	3.4	1.6	0.3	2.0	3.7
898.04	0.00968	14.1	12.2	10.4	8.6	6.5	4.6	3.1	1.4	0.2	2.0	3.5
1173.23	0.00786	14.3	12.5	10.8	9.1	7.2	5.5	3.9	2.1	0.6	1.2	3.1
1332.49	0.00710	14.5	12.9	11.0	9.3	7.7	6.0	4.0	2.3	0.6	1.1	2.9
1836.07	0.00552	14.3	12.1	10.4	8.7	7.2	5.4	3.4	1.5	0.3	2.0	3.8

Table 4. The percentage difference in the efficiency values calculated using PHITS according to the dead layer change in the experimental efficiency

Energy (keV)	ϵ_{exp}	0.7 mm	0.8 mm	0.9 mm	1.0 mm	1.1 mm	1.2 mm	1.3 mm	1.4 mm	1.5 mm	1.6 mm	1.7 mm
46.54	0.00229	152.6	91.2	48.7	13.7	11.1	32.3	25.4	60.7	68.1	75.4	81.2
59.54	0.01008	65.3	42.6	23.4	7.1	7.4	19.1	29.6	39.3	47.3	54.4	60.3
88.03	0.02750	24.8	17.5	10.7	4.2	1.9	7.6	12.9	17.7	22.4	26.9	30.8
122.06	0.03431	18.1	14.2	10.4	6.8	3.3	0.0	3.5	6.7	9.6	12.4	15.3
165.86	0.03333	16.9	14.3	11.6	9.1	6.6	4.1	1.8	0.5	2.8	5.0	7.1
279.20	0.02418	15.9	13.9	11.8	10.0	8.1	6.3	4.5	2.6	0.8	1.0	2.9
391.70	0.01868	13.8	11.8	10.0	8.1	6.3	4.5	2.8	1.0	0.7	2.1	3.6
514.00	0.01481	13.9	12.1	10.2	8.3	6.5	4.7	3.0	1.5	0.2	1.7	3.3
661.66	0.01218	13.0	11.2	11.2	7.5	5.8	4.1	2.3	0.7	0.9	2.6	4.3
898.04	0.00968	12.0	10.5	8.6	6.9	5.1	3.6	1.9	0.2	1.3	2.3	3.8
1173.23	0.00785	14.3	12.4	10.7	9.1	7.5	5.7	4.0	2.4	0.9	0.8	2.2
1332.49	0.00709	15.1	13.5	11.6	10.0	8.1	6.5	5.1	3.5	1.9	0.5	1.0
1836.07	0.00552	15.1	12.9	11.1	9.5	8.1	6.3	4.8	3.2	1.5	0.0	1.4

4. CONCLUSION

In this study, the FEPE curve of the detector for this Marinelli geometry was experimentally obtained using a Marinelli source containing an epoxy matrix with a density of 0.5 g/cc. Experimental efficiencies were obtained by counting a multi-energy point radioactive source in the range of 46.5 - 1836 keV in the HPGe detector. The experimental efficiency values of the detector at 0.7 mm dead layer thickness given by the manufacturer were calculated with Monte Carlo. Experimental efficiency results were compared with Monte Carlo efficiency values. In comparison, a difference between the experimental efficiency value of 233.8 % and 14.1 % was observed in the GESPECOR program in the range of 46.5 - 1836 keV. Likewise, a difference of 152.6 % - 12 % was observed in this energy range in the PHITS program. In order to obtain the correct dead layer thickness and to understand the change in the active volume of the crystal after eight years of use, when Modeling was done using Monte Carlo simulation codes GESPECOR and PHITS, the most compatible results with the experiment were obtained at 1.5 mm DL thickness for both GESPECOR and PHITS. In the GESPECOR program, a difference of 51.7 %, 30.1 %, 11.6 %, 122 keV and 3.2 % was obtained at 46.5 keV, 59 keV and 88 keV, and 122 keV, respectively, while this difference was found between 2 % and 0.1 % from 165 keV. In the PHITS program, 46.5 keV, 59 keV, 88 keV and 122 keV are 68.1 %, 47.3 %, 22.4 % and 9.6 %, respectively, while the difference from the experimental efficiency is between 2.8 % and 0.2 % in the 165 - 1836 keV range. In addition, while the simulations are being run, it has been determined that the active volume of the detector crystal decreases as the dead layer thickness increases and thus the efficiency values decrease. Good agreement was obtained at 1.5 mm DL in the high energy region, although inconsistency was observed between simulations and experiments in the low energy region of 46.5 keV and 122 keV. Marinelli beakers are almost completely they cover the detector crystal, are very useful geometries for calculating experimental efficiency against energy values. The compatibility between the detector's efficiency calculation and the Monte Carlo simulation for multi-energy radioactive sources was studied in an epoxy Marinelli beaker with a density of 0.5 g/cc. Inconsistencies in the low energy region such as 46.5, 59, 88 and 122 keV are due to the detector being p-type. This incompatibility is explained by the random and time-time diffusion of lithium on the crystal surface in high purity germanium detectors (Modarresi et al., 2017). It can be caused by errors in the position of the Marinelli beaker on the detector or by the wall thickness of the Marinelli beaker.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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