INVESTIGATING THE EFFECT OF MATRICES AND DENSITIES ON THE EFFICIENCY OF HPGE GAMMA SPECTROSCOPY USING MCNP

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Abstract. *When determining radioactivities in environmental samples using low-level gamma spectroscopy, in order to raise detection limit, voluminous samples are used. It takes in account for the self-absorption (self-attenuation) of gamma rays in samples. The self-absorption effect is small or large depend on the sample shapes, matrices and densities. In this paper, we investigated the effect of some regular matrices such as water, soil, epoxy resin on the detector efficiency. Some analytical formulas for the correction of matrix and densities for soil sample was established and applied to calculate some activities from standard sample of IAEA-375.*

I. INTRODUCTION

One of the most important problems of radioactivity measurement is investigating the detection efficiency. There are lots of factors can affect the efficiency such as: incident gamma ray energy, measuring geometry, electronic system, detector itself, other effects like coincidence summing or self-absorption...Among them, self-absorption is the most interesting effect when investigating activities of environmental samples because of their large volumes.

One of the most regular geometries used in investigating activities of environmental samples is Marinelli beaker geometry, which has 3π measuring geometry, so the efficiency is very high. Usually, Marinelli beaker samples have large volumes so the self-absorption effect of these samples is significant.

With the MCNP4C2 code [1], by simulating the measuring processes of environmental samples using the HPGe spectroscopy in Nuclear Physics Laboratory, we investigated the effect of matrices and densities on the efficiency. Based on that, a correction method was presented to calculate detection efficiencies for environmental samples.

II. CONFIGURATION OF SPECTROSCOPY - SAMPLE USED IN SIMULATION AND EXPERIMENT

II.1. HPGe spectroscopy

The HPGe detector in Department of Nuclear Physics, model GC2018, is a coaxial detector with configuration showed in Fig.1, including a germanium cylinder crystal with 52 mm outer diameter, 49.5 mm height. Inside the crystal, there is a hole with 7 mm

diameter, 35 mm depth. There are outer n-type contact layer (lithium layer), inner p-type contact layer (boron layer) of the crystal. The detector is hold in an aluminium box with 1.5 mm thickness [3].

There is a lead shield outside detector to absorb gamma rays from environment and suppress spectrum background. The interactions between gamma rays and lead shield layer produce X-rays with energies in the range 7388 keV. These X-rays can be detected by detector and effect on the gamma spectrum. To limit this problem, the copper and tin liners were lined covering the lead shield with the thickness of 1.6 mm and 1 mm respectively. The X-rays emitted by lead will be absorbed by the tin, and X-rays from the tin (about 2530 keV) will be absorbed by cooper. Finally, the cooper emits low energy X-rays (about 8 keV) which does not present on the spectrum.
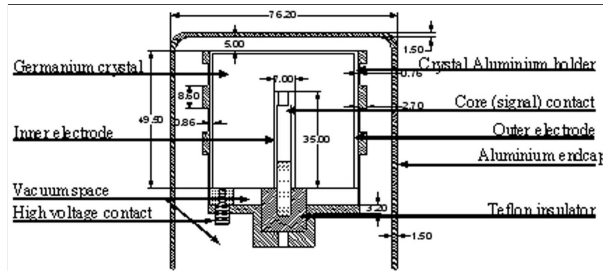

Fig. 1. The configuration of HPGe detector (in milimeter)

II.2. Samples

The samples were contained in Marinelli beakers, which sizes were shown in Fig. 2. These beakers were put on detector to make the 3π measuring geometry.
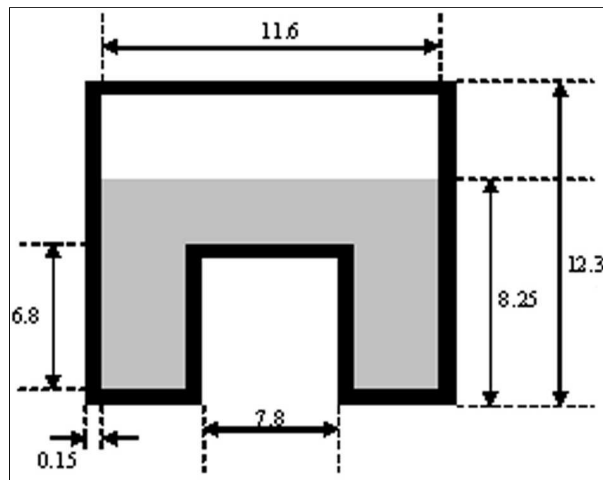

Fig. 2. The configuration of Marinelli sample (in centimeter)

III. SIMULATION OF PEAK EFFICIENCY CURVES OF HPGE DETECTOR WITH MATRICES AND DENSITIES

III.1. Matrices used in simulation

To investigate the effect of matrices on detection efficiency, we need to simulate the efficiencies with and without matrices. There were three types of matrices to simulate: soil, water and epoxy resin. The simulated volumes were the same with all types, the simulated densities were 0.5 g/cm^3, 1.0 g/cm^3 and 2.0 g/cm^3.

Three types of matrices [3]: Soil (% mass of atom in molecular): hydrogen 2.2%, oxygen 57.5%, aluminium 8.5%, silicon 26.2%, iron 5.6%; Epoxy resin (% mass of atom in molecular): hydrogen 6.0%, oxygen 21.9%, carbon 72.1%; Water (% mass of atom in molecular): hydrogen 11.11%, oxygen 88.89% .

To obtain the efficiency without matrix, simulated sample was chosen is air sample with density 0.00129 g/cm^3, includes 79% nitrogen and 21% oxygen. The size and volume of this sample is the same as soil, water and resin samples.

The simulated results of air matrix (efficiencies without self-absorption) were presented in Table 1.

Table 1. Detection efficiencies with air matrix (ε_0)

Radionuclide	Energy (keV)	Detection efficiency (ε_0)
^{241}Am	59.6	0.0186080
^{238}U	63.3	0.0225394
^{109}Cd	88.2	0.0423534
^{228}Ac	93.3	0.0446783
^{57}Co	122.0	0.0508223
^{214}Pb	295.0	0.0316577
	352.0	0.0268676
^{137}Cs	661.6	0.0151516
^{54}Mn	834.8	0.0124248
^{60}Co	1173.3	0.0094232
	1332.5	0.0085116

By presenting the dependence of efficiency on energy as a logarithmic function [3] by fitting, we have:

$$\ln(\varepsilon) = 0.0221(\ln E)^5 - 0.7226(\ln E)^4 + 9.4711(\ln E)^3$$
$$- 62.158(\ln E)^2 + 203.16\ln E - 266.2 \tag{1}$$

Fig. 3, Fig. 4, and Fig. 5 presented the simulated efficiencies with different matrices and densities.

There are some comments based on the above results:

- The difference between soil, water and epoxy resin in compare with air samples increases when densities of matrices increase. This can be explained when we know that if
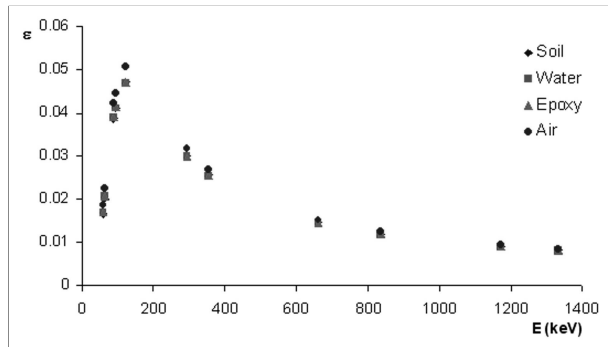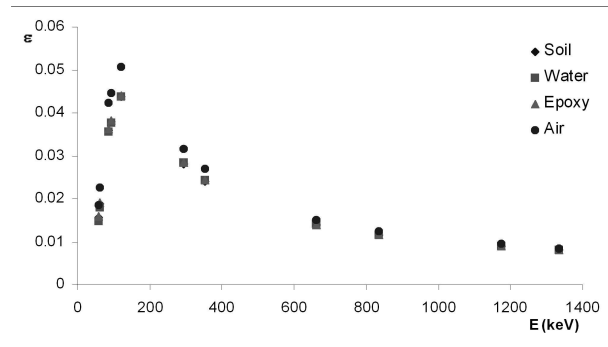
Fig. 3. Efficiencies at density 0.5 g/cm^3
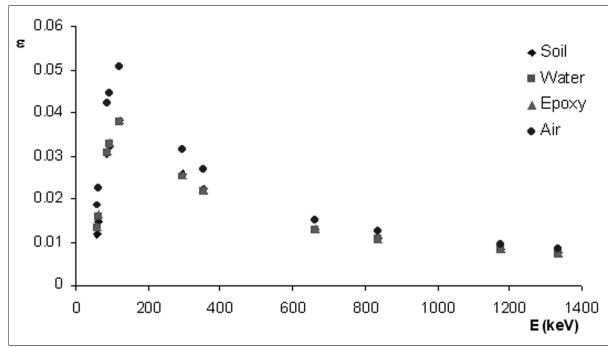

Fig. 4. Efficiencies at density 1.0 g/cm^3

Fig. 5. Efficiencies at density 2.0 g/cm^3

the density increases, the number of gamma rays can reach detector will decrease (because of losing more energy by interacting with matrix), so the efficiency will decrease.

- In the energy range below 100 keV, the effect of matrix is more significant than in the energy range above 100 keV.

- With the same density and measuring condition (geometry, volume, energy, ...), the efficiencies with different matrices are nearly the same.

With above comments, we can deduce some conclusions: with environmental samples matrices like soil, water and resin, the role of matrix is not important if we just need suitable accuracy (no need high accuracy). Therefore, when measuring with high energy above 100 keV, the matrix correction between measured and standard samples can be neglected. So, the standard sample preparation will be easier, saving time and cost to obtain the acceptable results.

III.2. Self-absorption correction

The self-absorption correction factor is determined by the ratio of efficiencies with and without self-absorption effect:

$$f = \frac{\varepsilon}{\varepsilon_0} \tag{2}$$

where f is self-absorption correction factor, ε is efficiency with self-absorption effect, ε_0 is efficiency without self-absorption effect.

Different environmental samples usually have different matrices. This will be the obstacle for measuring with large number of samples. In this part, the investigation of f by simulation of soil matrix with different densities in the range from 0.5 to 2.0 g/cm^3 was carried out to figure out the dependence of detection efficiency on density and energy with the same measuring geometry. Based on that, when measuring the activity of sample with any density in the investigated range, we use this correction factor to calculate detection efficiency.

Table 2 presented the calculated results obtained from simulation of self-absorption correction factor f of soil sample with energy E and density ρ.

Based on the dependence of f on E as in Fig. 6, we can approximate f for energy E as follow:

$$f(E, \rho) = ax^2 + bx + c, \quad x = \ln E \tag{3}$$

With different densities, fitting values of f for E, we got the parameters a, b and c.

With a, b, c obtained from different densities in the investigated range, we realized that a, b and c depend linearly on ρ, so the fitting of a, b, c to ρ was carried out [2]. The obtained results were:

$$a(\rho) = -0.0071\rho - 0.0054 \quad (R = 0.9773) \tag{4}$$

$$b(\rho = 0.1144\rho + 0.0710 \quad (R = 0.9842) \tag{5}$$

$$c(\rho = -0.5067\rho + 0.7622 (R = 0.9907) \tag{6}$$

III.3. Testing and applying into calculating activities of radionuclides in IAEA-375

The standard sample IAEA-375 in Laboratory of Nuclear Physics Department supplied by International Atomic Energy Agency (IAEA) is a sample collected from a farm in Novozybkov, Brjansk, Russia in July, 1990 [4]. The weight of sample is 760g, contained in

Table 2. Self-absorption correction factor of soil sample

E (keV)	Self-absorption correction factors f at densities ρ				
	0.5 g/cm³	0.8 g/cm³	1.0 g/cm³	1.2 g/cm³	2.0 g/cm³
59.6	0.89	0.83	0.79	0.76	0.64
63.3	0.89	0.83	0.80	0.77	0.65
88.2	0.92	0.87	0.84	0.81	0.72
93.3	0.92	0.87	0.85	0.82	0.72
122.0	0.93	0.89	0.86	0.84	0.75
295.0	0.95	0.92	0.90	0.88	0.81
352.0	0.95	0.92	0.91	0.89	0.83
661.2	0.96	0.94	0.93	0.91	0.86
834.8	0.97	0.95	0.93	0.92	0.88
1173.3	0.97	0.95	0.94	0.93	0.89
1332.5	0.97	0.96	0.95	0.94	0.90
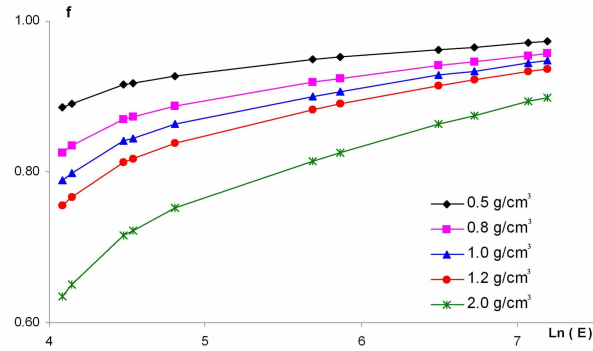
Fig. 6. The dependence of factor f on energy and density of soil matrix

Marinelli beaker with the same geometry as the simulation (Fig. 2). The sample density $\rho = 1.503$ g/cm³, sample was measured for 3 days with HPGe detector.

Activities of long-lived radionuclides were calculated by absolute method:

$$A = \frac{S}{\varepsilon(E).\theta.m.t_m} \tag{7}$$

A is the source activity at the time of acquisition (Bq/kg), S is the net peak area of the concerned peak,

$\varepsilon(E)$ is the efficiency at energy E, m is sample weight (kg), θ is the branching ratio of the observed nuclide at this energy $E(\%)$, t_m: the live time of the measurement (s).

Using formulas (4), (5), and (6) to calculate three parameters a, b, and c:

$$a = -0.0071 \times 1.503 - 0.0054 = -0.01607$$
$$b = 0.1144 \times 1.503 + 0.0710 = 0.24294$$
$$c = -0.5067 \times 1.503 + 0.7622 = 0.00063$$

After that, using formula (3) to obtain self-absorption correction factor f.

Applying formula (1) to calculate the detection efficiencies without self-absorption ε_0. The actual efficiencies were calculated by formula (2).

Calculated results were presented in Table 3.

Table 3. Detection efficiencies at some investigated energies of standard sample IAEA-375

Radionuclide	E (keV)	Correction factor f	Detecting efficiency	
			ε_0	ε
^{137}Cs	661.7	0.900569	0.013684	0.012323
^{212}Pb (^{232}Th)	238.6	0.848982	0.036071	0.030624
	338.3	0.870405	0.026255	0.022853
	583.2	0.895998	0.015454	0.013847
	911.6	0.909874	0.010112	0.009201
^{214}Pb (^{226}Ra)	295.2	0.862510	0.029861	0.025755
	351.9	0.872578	0.025279	0.022058
	609.3	0.897642	0.014813	0.013297
^{40}K	1460.8	0.917563	0.006670	0.006120

Using formula (7) to calculate activities of radionuclides after background subtraction, results are presented in Table 4:

Table 4. Activities of investigated radionuclides

Radionuclide	E (keV)	Peak area S	Emission probability (%)	Activity A (Bq/kg)
^{137}Cs	661.667	7,308,484 (0.04)	0.8499	5.190 ± 260
^{212}Pb (^{232}Th)	238.632	52,289 (2.70)	0.436	20 ± 1
	338.320	10,554 (7.52)	0.1127	21 ±2
	583.187	16,239 (2.64)	0.845	19.6 ± 1.1
	911.204	11,134 (1.16)	0.258	23.9 ± 1.2
	Mean activity of ^{232}Th : A = 21 ± 1 Bq/kg			
^{214}Pb (^{226}Ra)	295.224	19,546 (4.23)	0.18414	21.0 ± 1.4
	351.932	34,019 (2.36)	0.356	22.1 ± 1.2
	609.316	22,391 (2.80)	0.4642	18.5 ± 1.0
	Mean activity of ^{226}Ra : A = 20.2 ± 0.7 Bq/kg			
^{40}K	1460.822	54,922 (0.42)	0.1066	429.4 ± 21.6

Note: The number in parentheses is the relative standard deviation (%) due to counting statistics.

Finally, comparing calculated results with values of IAEA:

Table 5. Activity comparison of investigated radionuclides of standard sample IAEA-375

Radionuclide	Activity A (Bq/kg) (95% Confidence Interval)	
	Our results	IAEA [4]
^{137}Cs	4680 – 5700	5200 – 5360
^{226}Ra	17.2 – 23.2	18 – 22
^{232}Th	17.8 – 24.2	19.2 – 21.9
^{40}K	387 – 472	417 – 432

From Table 5, the calculated activities of three radionuclides ^{137}Cs, ^{226}Ra, ^{232}Th and ^{40}K agreed with given values of IAEA-375. In brief, we can accept this calculation method in calculating detection efficiency of environmental samples by using self-absorption correction factor with varied density.

IV. CONCLUSION

In this paper, the MCNP4C2 code was used to investigate the effect of matrices on detection efficiency of HPGe detector of Nuclear Physics Department, University of Natural Sciences, HCMC. The results showed that with regular densities (from 0.5 to 2.0 g/cm^3), the effect of matrices can be neglected when investigating gamma rays with energies higher than 100 keV. Then the MCNP4C2 code was continued to establish the relation between self-absorption correction factor and sample density. The simulation results showed that the correction factor changes linear with the change of sample density, and we also established the analytic formulas for correction factor. With the obtained analytic formulas, we carried out correcting detection efficiency with standard sample IAEA-375. The agreement between calculated activities with self-absorption correction and values from IAEA showed that the correction is quite exact. Therefore, the simulation method with MCNP4C2 code can help us in investigating and correcting the effect of matrices and density on detection efficiency of gamma spectroscopy.

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