Self-attenuation corrections calculated by LabSOCS Simulations for gamma-spectrometric measurements with HPGe detectors[*]

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Abstract: Simulations from Laboratory Sourceless Object Counting System (LabSOCS) software were used to determine self-attenuation correction factor, which is defined as the efficiency ratio of the sample with the absorbing medium to that of the sample without absorbing medium. The semi-empirical self-attenuation correction formula $F(\mu)$ used to correct self-attenuation of a sample was applied. A comparison of the two methods reveals that formula of sample with $\phi75$ mm$\times25$ mm and $\phi75$ mm$\times10$ mm can be, respectively, used in the self-attenuation correction for μ in the ranges of 0 to 0.5 cm^{-1} and 0.5 cm^{-1} to 2.0 cm^{-1}, indicating that the semi-empirical formula will not be used when μ has exceeded the interval. The semi-empirical formula value is consistent with the experimental value, within 7.9% accuracy. Therefore, this method is correct and effective. Both of our two methods can accurately produce a relative self-attenuation correction factor when the composition of the sample is known. The self-attenuation correction of a sample with unknown composition can only be carried out using a semi-empirical formula method.

Key words: Gamma spectral analysis, self-absorption correction, LabSOCS

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1   Introduction

Gamma-ray spectrometry using high-purity germanium (HPGe) detectors is a widely used procedure in determining the concentrations of natural and artificial radio-nuclides in environmental samples [1]. As a non-destructive technique, this method possesses advantages in multi-element analysis, simplified sample preparation (i.e., does not require any chemical separation process), and applicability for precise quantitative determination of the radioactive content in a sample. When measuring large samples containing γ-ray emitting radioisotopes using an HPGe detector, the calibration of this system is a lengthy process because the counting efficiency depends on both the geometry of the sample and the attenuation coefficient. The most accurate method to determine the activity of each radionuclide is to use an adequate standard source with very similar geometrical dimensions, density, and chemical compositions to the sample under study. However, the preparation of special standards for each case is a time consuming process [1, 2].

To quantify photon attenuation within environmen-

tal samples using gamma-ray spectrometry it is necessary to estimate the relevant self-attenuation corrections. Therefore, there are two main possibilities to perform these estimations, which have been developed to evaluate the necessary self-attenuation correction factor in gamma-ray spectrometry. We can either obtain the self-attenuation correction factor for sample geometry using semi-empirical method or simulate the absolute full energy peak efficiency by Monte Carlo simulation. The former case consists of a numerical integration [3] of the sample-detector geometry, which is calibrated using point sources that are normally located at different sample-detector distances. Some authors have also used a Monte Carlo simulation to calculate photon transmittance and photo peak efficiency for energies above 10 keV [2] but this method does not work well for low energies. In the latter case, a clear advantage of such Monte Carlo code is that it allows one to quickly calculate the efficiency values for changes of the measuring geometry. Some of these simulation codes have been specifically applied to the study of self-attenuation corrections factor in gamma-ray spectrometry [4]. But the accuracy of

efficiency is strongly dependent on an adequate knowledge of the elementary chemical composition of the source matrix and the detailed characteristics of the detector [5]. In this paper, we introduce the LabSOCS method. The NIST (National Institution of Standards and Technology) sources characterize the Ge crystal parameters used in LabSOCS.

The main advantage of the LabSOCS method is its relative rapidity and simplicity. Notably, LabSOCS is only valid from 45 keV to 7000 keV [6]. This method yields considerably accurate results for some special geometries, while powerful Monte Carlo techniques allow simulations for highly complex geometries. We simulate the full-energy peak efficiencies of the sample with a typical cylindrical geometry using the LabSOCS software and obtain the corresponding self-attenuation correction factors. We perform the study with gamma-ray energies ranging from 60 keV to 1500 keV for natural or environmental materials.

The accuracy of self-attenuation correction factor is essentially independent of the detailed characteristics of the detector. Therefore, the simulation results from the BE3830 HPGe detector can be used for other types of HPGe detectors. We can use the correction method for wider range of applications, such as in environmental radioactive analysis.

2   Mathematical viewpoint

In γ-ray spectrometry, the strength, A(Bq), of a given radionuclide in a sample is

$$A = \frac{n}{\varepsilon \cdot F_r \cdot P_\gamma}, \qquad (1)$$

where n is the net count rate (in cps) under the full-energy peak corresponding to the photon energy E_γ emitted by the radionuclide of interest with an emission probability P_γ; ε is the full-energy peak efficiency corresponding to E_γ; and, F_r is the relative self-attenuation correction factor. We express the relative correction factor as the ratio of the absolute self-attenuation correction factor of the sample to that of the standard source [4]:

$$F_r = \frac{F_{sa}}{F_{st}}, \qquad (2)$$

where F_{st} and F_{sa} are the absolute self-attenuation correction factors of the standard source and the sample, respectively. The absolute self-attenuation correction factor is the self-attenuation correction factor, which is mentioned in the following sections.

In this study, we determine the self-attenuation correction factor F for a cylindrical sample using an HPGe detector expressing the correction factor as the efficiency

ratio, as follows [7]:

$$F = \frac{\varepsilon_v}{\varepsilon_{v,0}}, \qquad (3)$$

where, ε_v denotes the efficiency of sample with the absorbing medium and $\varepsilon_{v,0}$ denotes the efficiency of the empty sample without an absorbing medium.

However, it is difficult to obtain the efficiency $\varepsilon_{v,0}$ for a given sample. On the contrary, the LabSOCS software can calculate efficiencies ε_v and $\varepsilon_{v,0}$, and evaluate F as the efficiency ratio between the sample of different materials and the "air sample".

For a given geometrical configuration, μ is the linear attenuation coefficient (in cm^{-1}) of the sample and F depends on μ. Thus, another method is to use a semi-empirical formula $F(\mu)$ to correct the self-attenuation of the sample. The reason to use $F(\mu)$ is that it will be easy to obtain a correction factor for most of the required energy transitions by using the fixed semi-empirical formula when μ is available. Therefore, this method is also essentially independent of the density and matrix of the sample when μ is available. So, this method overcomes the uncertainties produced from the difference in matrices and densities of the samples. Other HPGe detectors can also use the semi-empirical formula $F(\mu)$ that was established by the LabSOCS software for the same sample geometry. So, it is not necessary to do much experimental work or make assumptions for the mathematical approaches.

3   Simulation method

The system consists of a CANBERRA Ultra-Low Background BE3830 HPGe detector (ϕ70 mm$\times$30 mm) of 35.6% relative efficiency. We perform the gamma spectrum analysis using Genie-2000 [8] γ-ray spectroscopy with the LabSOCS software by Canberra.

For samples with known compositions, we can obtain μ theoretically. We first obtain the mass attenuation coefficients of the basic material [9] (see Table 1).

Table 1.   The mass attenuation coefficient.

material	the mass attenuation coefficient/(cm^2/g)		
	60 keV	300 keV	1500 keV
O	0.190	0.107	0.0519
standard source	0.279	0.107	0.0515
Al_2O_3	0.236	0.104	0.0509
Al	0.276	0.101	0.0501
soil	0.439	0.107	0.0509
SiO_2	0.250	0.108	0.0518
Si	0.319	0.108	0.0518
Fe_2O_3	0.893	0.109	0.0497
Fe	1.20	0.110	0.0488

In Table 1, the mass percentage of standard source is: SiO_2 (70%), Al_2O_3(25%), and Fe_2O_3 (5%), the mass

percentage of soil is: SiO_2 (40%), Al_2O_3 (30%), and Fe_2O_3 (30%). We calculate the mass attenuation coefficients of the compound or mixture in Table 1 using Eq. (4), as follows:

$$(\mu/\rho)_{comp} = \sum (\mu_i/\rho_i \cdot W_i), \qquad (4)$$

where ρ denotes the density of the compound; μ_i/ρ_i is the mass attenuation coefficient of one element in the compound; and, W_i is the mass percentage of one element in the compound.

3.1 The self-attenuation factor simulation for $\phi75$ mm$\times$10 mm sample

The following part is carried out for the 60 keV γ-ray's self-attenuation correction. The self-attenuation factor simulation for a sample with $\phi75$ mm$\times$10 mm is implemented using the above theory. We only use pure aluminum material to conduct the self-attenuation calculation for 60 keV γ-ray and eventually obtain different values of μ by changing the density of aluminum. Meanwhile, we use the LabSOCS software to calculate F of the samples. Table 2 shows the simulated efficiencies ε_v of various samples using this software. In this simulation, we should input some parameters of the sample, including sample diameter, thickness, material of the sample box, sample material, density, γ-ray energy, and the distance between the sample and the detector. Secondly, we refer to the efficiency of the air sample as $\varepsilon_{v,0}$ and to the other efficiencies of sample as ε_v. We can then calculate the correction factors F using Eq. (3). The results are given in Table 2 and Fig. 1.

Table 2. F of aluminum sample calculated using LabSOCS for 60 keV γ-ray.

$\rho/(\text{g/cm}^3)$	μ/cm^{-1}	ε_v	F
1.80	0.497	0.127	0.764
2.60	0.718	0.114	0.686
3.40	0.939	0.103	0.619
4.20	1.16	0.0931	0.562
5.00	1.38	0.0849	0.512
5.80	1.60	0.0776	0.468
6.60	1.82	0.0713	0.430
7.40	2.05	0.0659	0.398
0.0012(air)	—	0.166	1

Figure 1 shows that the relationship between the F and μ is given by:

$$F(\mu) = 0.926 \cdot e^{-0.421\mu}. \qquad (5)$$

The correlation coefficient is $R^2 \approx 0.997$. Using this semi-empirical formula, we can easily obtain a correction factor for μ in the 0.5–2.0 cm^{-1} interval.

The semi-empirical formula $F(\mu)$ method is performed by fixing the linear attenuation coefficient by theoretical calculation or a source collimation experiment.

The correction formula is then selected according to μ, and used to obtain F' for the most required energy. F' denotes the self-attenuation correction factor obtained from Eq. (5). We calculate the μ of different samples in Table 3 using Eq. (4) and data from Table 1, the μ values are shown in Table 3, and then we calculate F' of some samples using Eq. (5). The results are shown in Table 3.
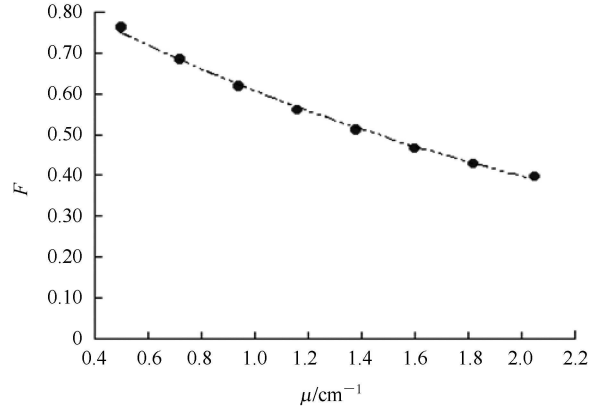

Fig. 1. Fitting curve for F of a sample with $\phi75$ mm$\times$10 mm for a 60 keV γ-ray.

Table 3. Self-attenuation factors of $\phi75$ mm$\times$ 10 mm sample for 60 keV γ-ray.

sample	μ/cm^{-1}	F'	ε_v	F
Fe_2O_3*	0.625	0.711	0.118	0.710
Al*	1.27	0.542	0.0889	0.536
Soil	0.702	0.689	0.114	0.686
Si*	0.574	0.727	0.123	0.740
Fe_2O_3	1.88	0.420	0.0684	0.413
Si	0.743	0.677	0.113	0.683
Al	0.746	0.676	0.112	0.677
Fe	9.41	0.0175	0.0162	0.0980
Air	—	—	0.166	1

Meanwhile, we use LabSOCS software to calculate the samples' F in Table 3. We simulate the efficiencies ε_v of different samples with this software, and then we refer to the efficiency of air sample as $\varepsilon_{v,0}$, and all of the other efficiencies of sample as ε_v. We calculate the correction factors F using Eq. (3). The results are given in Table 3, where we refer to the F' of Al* as F'_{st}, and for all other samples' F' as F_{sa}. Then, we calculate F_r using Eq. (2). The results are shown in Table 4. We refer to the F of Al* as F_{st}, and for all other samples' F as F_{sa}. Then, we calculate F_r using Eq. (2). The results are shown in Table 4.

In Table 4, the deviations are within 3%, except for the Fe sample. These data indicate that the semi-empirical formula value is consistent with the LabSOCS software calculation. So, the semi-empirical Eq. (5) is suitable for these samples with $\phi75$ mm$\times$10 mm. It is clearly noticed that considerable deviations are encountered in the process for the Fe sample.

Table 4. Comparison of relative self-attenuation factors of samples with $\phi75$ mm$\times10$ mm for 60 keV γ-ray.

sample	F_{r}	F_{r}'	$\Delta\%$
Fe_2O_3*	1.33	1.31	-1.5
Al*	1.00	0.999	-0.10
Soil	1.28	1.27	-0.78
Si*	1.38	1.34	-2.9
Fe_2O_3	0.770	0.774	0.52
Si	1.27	1.25	-1.6
Al	1.26	1.25	-0.79
Fe	0.183	0.0322	-82

Table 5. Linear attenuation coefficient (cm^{-1}) of different samples under different gamma-ray energies.

sample	$\rho/(\mathrm{g/cm^3})$	$\mu/\mathrm{cm^{-1}}$		
		60 keV	300 keV	1500 keV
Fe_2O_3*	0.70	0.625	0.0763	0.0348
standard source	1.44	0.401	0.153	0.0740
Al*	1.50	0.414	0.152	0.0751
Soil	1.60	0.702	0.171	0.0815
Si*	1.80	0.574	0.195	0.0933
Fe_2O_3	2.10	1.88	0.229	0.104
Si	2.33	0.743	0.252	0.121
Al	2.70	0.746	0.273	0.135
Fe	7.86	9.41	0.863	0.384
Air	0.0012	—	—	—

We have discovered some rules. For serious self-attenuation, we cannot use the semi-empirical formula Eq. (5), such as Fe sample, because the rays emitted from the The upper part of the sample is absorbed completely, making the equation unsuitable for this sample. Therefore, we should establish a new $F(\mu)$ formula for a thinner sample, where this equation can only be used for a certain range of μ. The results will be inaccurate when μ is beyond this range or when the sample shape is changed.

In this study, if the composition of the sample is unknown, then the LabSOCS software calculation cannot be used and the established semi-empirical formula $F(\mu)$ can be used instead. The most important step is to obtain the μ for a sample. We perform a simple source collimation experiment [10] instead of a chemical analysis procedure to obtain μ, where μ depends not only on the γ-ray energy but also on the density and matrix of the sample.

3.2 Self-attenuation factor simulation for $\phi75$ mm$\times25$ mm sample

The correction process of F for cylindrical samples with $\phi75$ mm$\times25$ mm different for gamma ray energies of 60 keV, 300 keV and 1500 keV are presented.

In the semi-empirical formula $F(\mu)$ method, we can obtain μ using Eq. (4) (Table 5).

We calculate $F(\mu)$ using Eq. (6), as follows [10]:

$$F(\mu)=0.965\mathrm{e}^{-1.20\mu}. \qquad (6)$$

The correlation coefficient is $R^2 \approx 0.997$ obtained from Monte Carlo simulations for the cylindrical sample, which is consistent with the disk source experimental result of $F(\mu)=0.966\mathrm{e}^{-1.18\mu}$ [10]. We then calculate F_{r}' and F_{r} for different samples. The values are listed in Table 6.

The two methods are compared in Table 6 and the semi-empirical formula $F(\mu)$ method is validated by the LabSOCS software calculation. We calculate the discrepancies from the reference data in Table 6 as follows: $\Delta\% = \dfrac{F(\mu)_{\mathrm{r}}-F_{\mathrm{r}}}{F_{\mathrm{r}}}\%$, where $\Delta\%$ is 1% for γ-ray energies of 1500 and 300 keV, except Fe (13%), which means that we can use these two methods to correct the self-attenuation of these samples for more than 300 keV γ-ray energy, except for the Fe samples. Furthermore, Eq. (6) is not applicable in correcting self-attenuation for less than 300 keV γ-ray energy for Fe sample; $\Delta\%$ is in 2% for 60 keV except (-50%), Al (-9.3%), soil (-6.8%), Si (-10%), and Fe (-100%), in which the deviations are larger than those in other samples due to their serious self-attenuation. Deviation values obtained from Table 4 (i.e., 0.52%, -0.79%, -0.78%, -1.6%, and -82% for

Table 6. Relative self-attenuation factors under different gamma-ray energies.

sample	60 keV			300 keV			1500 keV		
	F_{r}	F_{r}'	$\Delta\%$	F_{r}	F_{r}'	$\Delta\%$	F_{r}	F_{r}'	$\Delta\%$
Fe_2O_3*	0.802	0.767	-4.3	1.10	1.10	-0.50	1.05	1.05	-0.20
standard source	1.00	1.00	0.0	1.00	1.00	0.0	1.00	1.00	0.0
Al*	0.991	0.984	-0.70	1.00	1.00	0.50	1.00	1.00	0.0
Soil	0.752	0.701	-6.8	0.976	0.979	0.30	0.987	0.991	0.40
Si*	0.864	0.815	-5.6	0.952	0.952	0.0	0.975	0.978	0.20
Fe_2O_3	0.352	0.175	-50	0.911	0.915	0.40	0.956	0.965	1.0
Si	0.746	0.668	-10	0.888	0.890	0.20	0.940	0.947	0.70
Al	0.734	0.665	-9.3	0.859	0.868	1.1	0.923	0.931	0.80
Fe	0.076	0.000	-100	0.500	0.433	-13	0.694	0.694	0.10

Fe_2O_3, Al, soil, Si, and Fe, respectively) are smaller when compared with that the values in Table 6. So, for serious self-attenuation, we should choose thinner sample for the material's self-attenuation correction. Therefore, Eq. (6) cannot be used to correct their self-attenuation. From the analysis, it is shown that the deviation value increases with the increase of μ. From Table 6, we can see that Eq. (6) is usually suitable for γ-ray energy in the 300–1500 keV interval, and μ in the 0–0.5 cm^{-1} interval. The sample's thickness should decrease with the increase of μ, which leads to the establishment of another semi-empirical formula $F(\mu)$ of thinner ϕ75 mm×10 mm sample when μ is large. From Table 6, we can see that although both of the two correction methods can be used for γ-ray energies ranging from 300 keV to 1500 keV, they are unsuitable for 60 keV γ-ray.

3.3   Experiment validation

Disk source experimental data have been previously used to validate the calculation results. The relationship between the peak count rate $f(h)$ (in cps) of a ^{241}Am disk source and the distance h is [11]:

$$f(h)_{s,0} = 13.8\cdot h^2 - 67.2\cdot h + 132, \tag{7}$$

$$f(h)_s = 114\cdot e^{-0.942\cdot h}, \tag{8}$$

where both equations come from Figs. 4 and 5 of the reference [11]. The efficiencies of volume sources $\varepsilon_{v,0}$ and ε_v are obtained using Eq. (9) [11]

$$\varepsilon_v = \frac{1}{H}\int_0^H \frac{1}{AP_\gamma}f(h)\mathrm{d}h, \tag{9}$$

where h is in the 0–1.0 cm interval for sample with ϕ75 mm×10 mm. We calculate $F_{experiment}$ using Eq. (3), with the $F_{experiment}$ value shown in Table 7. According to the composition of standard source, we can calculate the linear attenuation coefficient of a standard source using Eq. (4) to obtain μ=0.40 $cm^{-1}\cdot F'$ can be obtained from Eq. (5) , the result is also shown in Table 7.

The point source experimental data can also be used to validate the calculation results. For the air sample, the peak efficiency function parameters of the point sources are given by [12]:

$$\varepsilon_{v,0} = \frac{1}{V}\int_0^H\int_0^R 2\pi r\cdot\varepsilon_P^P(E_\gamma, r=0, h)$$

$$\times \exp\left[-2.773\frac{r^2}{\Gamma(h)^2}\right]\mathrm{d}r\mathrm{d}h, \tag{10}$$

$$\varepsilon_p^p(E_\gamma, r=0, h) = (a_3 h^2 + a_4 h + a_5)^{-1}, \tag{11}$$

$$\Gamma(h) = a_1 e^{a_2 h}, \tag{12}$$

where a_1, a_2, a_3, a_4 and a_5 represent the fitting parameters given by the linear best-fit curve of the experimental data. The fitted parameters are given by

[12] a_1=6.595, a_2=0.1057, a_3=0.2874, a_4=0.5691, and a_5=3.942 for V=44.16 cm^3, H=1.0 cm, and R=3.75 cm. The volume source efficiency $\varepsilon_{v,0}$=0.159 is then obtained from Eq. (10).

For a standard soil sample, the peak efficiency ε_v can be expressed as Eq. (13) using point sources [13].

$$\varepsilon_v = \frac{1}{H}\int_0^H \varepsilon(h_i)\mathrm{d}h, \tag{13}$$

$$\varepsilon(h_i) = \frac{1}{d + e\cdot h_i + f\cdot h_i^2}, \tag{14}$$

where d, e, and f represent the fitting parameters from the linear best-fit curve of the point source experimental data, and are given by [13] d=6.48, e=−0.075, f=3.41. The volume source efficiency ε_v=0.134 is then obtained from Eq. (13) [13]. Finally, we calculate $F_{experiment}$=0.84 by Eq. (3), which is shown in Table 7.

Table 7.   Comparison with the experimental result for ^{241}Am(59.54 keV)

	disk source experiment	point source experiment	LabSOCS method
$F_{experiment}$	0.72	0.84	—
F'	—	—	0.78
$\Delta\%$	7.9	−7.1	—

We calculate discrepancies from the reference data in Table 7 as follows: $\Delta\% = \dfrac{F' - F_{experiment}}{F_{experiment}}\%$.

The value of the semi-empirical formula F' value is consistent with the experimental value of $F_{experiment}$ in 7.9%. Therefore, the semi-empirical formula $F(\mu)$ method is proven to be both correct and effective. We can also conclude that the two simulation methods can be used in self-attenuation simulation because the semi-empirical formula $F(\mu)$ is also established through LabSOCS software calculation.

4   Conclusions

We can only use the semi-empirical Eq. (6) for sample with ϕ75 mm×25 mm whose μ is in the 0–0.5 cm^{-1}interval, and Eq. (5) for sample with ϕ75 mm×10 mm, μ in the 0.5–2.0 cm^{-1} interval, respectively. We should establish the semi-empirical formula $F(\mu)$ of a thinner sample of the Fe sample. We establish Fig. 1 for an aluminum sample with different densities. However, it can correct the self-attenuation factor of samples with different compositions, thereby neglecting the discrepancies in the chemical composition and density of the reference and other bulk samples.

The chemical composition and the density of the sample must be known to complete this simulation, which limits the area of application of this method. However,

if we cannot obtain the chemical composition and density of the sample, then we can fix the μ of sample by another simple source collimation experiment, so the semi-empirical self-attenuation correction formula $F(\mu)$ method can be more widely used than the LabSOCS software simulation. The results indicate that some system errors can be neglected in the LabSOCS software simulation, producing accurate results for self-attenuation correction. However, this method cannot be used to calculate the efficiencies of the samples for low energies because of the system errors. Although the use of Monte Carlo simulation for the efficiency calibration in gamma-ray spectrometry has become more common, we have to consider the detailed chemical composition of the samples and the detailed characteristics of the detector [14]. If these parameters are accurately obtained, then we can also use the Monte Carlo method instead of the LabSOCS software to complete the self-attenuation correction. The self-attenuation correction will also be studied in a future reactor with the application of a high purity Germanium (HPGe) spectrometer in determining the fuel element burnup [15].

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