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Performances of different efficiency calibration methods of high-purity-germanium gamma-ray spectrometry in an inter-comparison exercise

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Abstract This study reports the performances of efficiency calibrations for high-purity-germanium gamma-ray spectrometry using the source-, Laboratory Sourceless Object Calibration Software (LabSOCS)- and ANGLE-based methods in an inter-comparison exercise. Although the results of LabSOCS and ANGLE for ^{241}Am emitting low-energy gamma rays were not very satisfactory, all of the three efficiency calibration methods passed acceptance criteria. The results confirmed the reliability of the calculation codes ANGLE and LabSOCS as alternative efficiency calibration methods in high-purity-germanium gamma spectrometry. This study is likely to promote the further application of the ANGLE and LabSOCS calculation codes in radioactivity measurements.

Keywords Efficiency calibration · ANGLE · Laboratory Sourceless Object Calibration Software (LabSOCS) · Gamma-ray spectrometry

1 Introduction

Following a radiological and nuclear emergency, a large number of environmental samples may need to quickly be monitored for possible contamination. Gamma-ray spectrometry is commonly used to analyze radionuclide activity concentration. In order to determine the activity concentration of an environmental sample, it is crucial to know the detection efficiencies of the energy region of interest in advance [1]. The first-reported approach to obtain the detection efficiency, referred to as the source-based method, is based on the use of commercially available radioactive standard sources, which have identical shape, matrix, size, and density with those of the analyzed samples [2, 3]. The second approach is the full Monte-Carlo-based calculation approach using input information, such as detector construction, dimensions of the sample, chemical composition, and sample density [4]. The third approach is the semi-empirical method, which combines the experimental calibration curve of detection efficiency with a mathematical simulation of the detector. Although the source-based method is the most commonly used, it is not possible to obtain a standard source for every measuring geometry, particularly in radiological and nuclear emergency situations.

The modeling method provides an alternative for the direct efficiency calibration of a gamma-ray spectrometer. Calculation codes for efficiency calibration have been developed and are commercially available. Laboratory Sourceless Object Calibration Software (LabSOCS) and ANGLE are examples of full Monte-Carlo-based and semi-empirical calculation codes, respectively. However, LabSOCS and ANGLE are not widely applied in the laboratories in China owing to numerous factors affecting their

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accuracies, despite their advantages of time efficiency and applicability to various samples [5].

In order to evaluate the performances of the LabSOCS- and ANGLE-based efficiency calibration methods for measurements of activity concentrations of environmental radionuclides, this study involved an inter-comparison exercise organized by the Division of Radiation Metrology of the China Institute of Atomic Energy (CIAE). In this paper, the performances of the three efficiency calibration methods obtained in the inter-comparison exercise are reported and compared.

2 Materials and methods

2.1 Samples and preparation

In the inter-comparison exercise, the analyzed material was clay soil prepared by the CIAE; its main components are shown in Table 1. The measured radionuclide ^{40}K is the natural background, while ^{60}Co, ^{137}Cs, ^{241}Am, ^{226}Ra, and ^{232}Th were artificially added to achieve the desired activity concentration. The reference source derived from the National Institute of Metrology of China had the same shape, matrix, and size as those of the inter-comparison sample, which was used for efficiency calibration with the source-based method.

2.2 Measurement of radioactivity by gamma-ray spectrometry

A cylindrical container was employed in this analysis, as it is commonly used for laboratory environmental radioactivity measurements. The soil sample (330 g) was packed into an acrylonitrile–butadiene–styrene (ABS) cylindrical sample container with dimensions of 75 mm × 70 mm ($\Phi \times$ height), with thick walls (3 mm) and thick bottoms (1 mm) (ρ = 1.05 g/cm^3); its chemical composition consisted of carbon, hydrogen, and nitrogen concentrations of 85.31%, 8.05%, and 6.64%, respectively. The cylindrical sample container was purchased from the National Institute of Metrology of China. The sample was sealed and left in the container for at least 3 weeks before measurement to ensure secure equilibrium between the radionuclides.

In the laboratory, the inter-comparison sample was measured by two low-background gamma-ray spectrometry systems. One of them was a broad-energy coaxial high-purity-germanium (HPGe) detector (BE5030, Canberra, USA) with a crystal length of 31.3 mm, diameter of 80.5 mm, and 1.5-mm-thick aluminum round end-cap. The relative efficiency of the detector was 50.5%, while its energy resolution was 1.65 keV (full width at half maximum (FWHM)) at 1.33 MeV for ^{60}Co. GENIE-2000 and LabSOCS were used for the spectrometric analysis. The HPGe crystal parameters used in the LabSOCS (version 4.4) have been characterized by the Canberra Company. The HPGe detector (BE5030) was also used for the source-based method.

The other gamma-ray spectrometer was equipped with a Detective DX-100T HPGe detector (ORTEC, USA). The relative efficiency of the detector was 46%, while its resolution (FWHM) was 2.18 keV at 1.33 MeV for ^{60}Co. A coaxial *p*-type HPGe crystal was employed with nominal diameter of 50 mm and depth of 30 mm. The ANGLE software (version 3.0) was used to obtain efficiency calibration curves, which is compatible with the ORTEC's Gamma Vision. Technical characteristics of the employed detector, such as type and dimensions, were input into the software, while the reference efficiency curve was obtained experimentally using a reference source.

The energy calibrations of both spectrometers were performed using a reference source derived from the National Institute of Metrology of China. The analyzed radionuclides and associated decay data are summarized in Table 2 [6]. The counting time was set to a value larger than 86,400 s. Furthermore, a container with the same geometrical dimensions was used to obtain the background gamma-ray spectrum. The activity concentration of radionuclides in the sample was calculated by:

$$A = \frac{(n_s/T_s - n_b/T_b)}{I_\gamma \times \varepsilon \times m \times F_1 \times F_2}, \tag{1}$$

where A is the activity concentration of the radionuclide, n_s is the net count of the sample, n_b is the net count of the background, T_s is the live time of the sample measurement, T_b is the live time of the background measurement, I_γ is the gamma-ray emission probability, ε is the detection efficiency, m is the dry weight of the sample, F_1 is the density correction factor, and F_2 is the geometry correction factor. In this study, F_1 and F_2 were both 1.0.

The expanded uncertainty U_{total} of the activity concentration can be calculated using the following equation according to the International Atomic Energy Agency (IAEA) technical documents [7]:

Table 1 Chemical composition of the inter-comparison soil material

Components	Si	Al	Fe	MgO	CaO	Na	K	MnO	Ti	P
Mass percentage (%)	68.57	13.22	4.8	1.68	1.42	1.74	2.5	0.091	0.703	0.116

Table 2 Radionuclide and associated decay data

Radionuclide	Half-life (years)	Corresponding daughters	γ-ray energy (keV)	Emission probability
^{241}Am	432.6		59.5409	0.3592
^{226}Ra	1600	^{214}Pb	351.932	0.356
		^{214}Bi	609.312	0.4549
^{232}Th	1.402×10^{10}	^{208}Tl	583.187	0.85
		^{228}Ac	911.196	0.262
^{137}Cs	30.05		661.657	0.8499
^{60}Co	5.2711		1173.228	0.9985
			1332.492	0.999826
^{40}K	1.2504×10^{9}		1460.822	0.1055

$$U_{\text{total}} = k\sqrt{U_1^2 + U_2^2 + U_3^2 + U_4^2} \tag{2}$$

where U_1 is the efficiency calibration for the HPGe detector uncertainty, which was estimated from a fitted calibration curve using the commercial software, k is equal to 2, U_2 is the uncertainty of the statistical counts, U_3 is the uncertainty of the sample weights, and U_4 is the uncertainty of the sample geometry.

2.3 Performance evaluation and scoring

An evaluation of the measured results obtained from the different efficiency calibration methods was performed according to the proficiency test method of the IAEA [8–10], which considers the accuracies and precisions of the expected and measured data together with their uncertainties.

2.3.1 *Relative bias*

The relative bias was calculated using the equation:

$$\text{Relative bias} = \frac{\text{Value}_{\text{Analyst}} - \text{Value}_{\text{Ref.}}}{\text{Value}_{\text{Ref.}}} \times 100\% \tag{3}$$

where $\text{Value}_{\text{Analyst}}$ is the measured activity concentration and $\text{Value}_{\text{Ref.}}$ is the reference activity concentration.

2.3.2 *Z-score value*

The *Z*-score was calculated as:

$$Z_{\text{score}} = \frac{\text{Value}_{\text{Analyst}} - \text{Value}_{\text{Ref.}}}{\sigma} \tag{4}$$

Considering the "fitness for purpose" principle, the standard deviation (σ) of the reference value is $0.10 \times \text{Value}_{\text{Ref}}$. According to the evaluation report of the IAEA [8], $|Z_{\text{score}}| < 2$ indicates that the performance is satisfactory, for $2 < |Z_{\text{score}}| < 3$ the performance is considered questionable, and if $|Z_{\text{score}}| \geq 3$, the performance is unsatisfactory.

2.3.3 *U-test value*

The *U*-test value was calculated by:

$$U_{\text{test}} = \frac{\left|\text{Value}_{\text{Analyst}} - \text{Value}_{\text{Ref.}}\right|}{\sqrt{\text{Unc}_{\text{Ref.}}^2 + \text{Unc}_{\text{Analyst}}^2}}. \tag{5}$$

In this study, the limiting value of the *U*-test was set to 2.58 at a probability level of 99% to determine whether the result is satisfactory ($U < 2.58$) [8–10].

2.4 Evaluation criteria of the inter-comparison exercise

2.4.1 *Trueness*

The measured result is assigned as "Acceptable" if $A_1 \leq A_2$ [9], where A_1 and A_2 can be calculated as:

$$A_1 = \left|\text{Value}_{\text{Ref.}} - \text{Value}_{\text{Analyst}}\right|, \tag{6}$$

$$A_2 = 2.58 \times \sqrt{\text{Unc}_{\text{Ref.}}^2 + \text{Unc}_{\text{Analyst}}^2}. \tag{7}$$

2.4.2 *Precision*

The precision *P* value was calculated by [9]:

$$P = \sqrt{\left(\frac{\text{Unc}_{\text{Ref.}}}{\text{Value}_{\text{Ref.}}}\right)^2 + \left(\frac{\text{Unc}_{\text{Analyst}}}{\text{Value}_{\text{Analyst}}}\right)^2} \times 100\% \tag{8}$$

The limit of accepted precision for the ^{241}Am, ^{226}Ra, ^{232}Th, ^{137}Cs, ^{60}Co, and ^{40}K comparison exercise was set at 10%. The result was scored as "Acceptable" for a precision $P \leq 10\%$.

A result is assigned as "Acceptable" if it satisfies both criteria. However, if the accuracy or precision tests fail, the result is assigned as "Not acceptable," and then a further check is performed. The maximum acceptable level of the relative bias was set at 15% for this exercise. The performance was assigned as "Warning" when the relative bias was smaller than 15%; otherwise “Not acceptable” was the final score.

3 Results and discussion

The measured results obtained by the source-, LabSOCS-, and ANGLE-based methods are presented in Table 3. The above evaluation parameters were calculated and are summarized in Tables 4, 5 and 6, respectively. Tables 4, 5 and 6 reveal that the measured results of ^{241}Am, ^{226}Ra, ^{232}Th, ^{137}Cs, ^{60}Co, and ^{40}K by the three methods satisfied the trueness and precision criteria. This confirms the good agreement of the source-, LabSOCS-, and ANGLE-based methods, which implies that instead of using standard sources for efficiency calibration, one can utilize any available standard with similar geometry and mass and calculate the efficiency with LabSOCS and ANGLE.

It is worth noting that the relative bias for ^{241}Am obtained by LabSOCS is up to 25.8%, and its *Z*-score is larger than 2, but smaller than 3, making these data questionable. Regarding ANGLE, the relative bias of ^{241}Am is up to 14.3%, while its *Z*-score of 1.4 is lower than 2. Compared with that of the source-based method, the performances of LabSOCS and ANGLE for ^{241}Am are lower. This may be attributed to an inaccurate efficiency calibration of gamma-ray spectrometry at energies lower than 100 keV, which are challenging to obtain owing to the more significant self-absorption and relatively high background level recorded by the detector in this low-energy range.

It is challenging to achieve an accurate efficiency calibration in the energy range below 100 keV for gamma-ray spectrometry. The accuracy and precision of the detector calibration efficiency were affected by many factors, such as background interference, source geometries, and matrix. ANGLE requires the used detector parameters, measurement configuration, and experimental efficiency calibration data. The LabSOCS calculation also requires information about the detector configuration, source characteristics, matrix, etc. Therefore, it is essential to have accurate information regarding the input parameters used by LabSOCS and ANGLE to ensure accuracy of the efficiency calibration for the low-energy range.

According to the performed exercise, in order to improve the accuracy and precision of gamma-ray analysis, the laboratory should use almost identical sample shapes and sizes to minimize the deviations in geometry. It would be useful to establish a chemical composition database of the main sample types including soil and food. If a matrix sample composition is unknown, materials with similar chemical compositions to those of the targeted samples analyzed with the LabSOCS technique can be used. In addition, it is important to create a protocol for quality control and assurance including regular detector calibration and measurement of backgrounds.

The reliabilities of ANGLE and LabSOCS for efficiency calibrations have been reported [11–14]. In this study, the effectiveness of the two calculation codes to determine activities in soil samples was demonstrated with the laboratory proficiency test scoring method of the IAEA [8], which considers the accuracy and precision. The acceptability of ANGLE and LabSOCS was more comprehensively verified in this study.

Furthermore, our laboratory will continue to participate in proficiency testing, inter-comparison studies, and emergency exercises to obtain accurate, precise, and meaningful measurement results for multiple radionuclides in soil, food, water, and other types of samples by the three different methods of efficiency calibration.

4 Conclusion

In this study, we demonstrated that the performances of the three methods of efficiency calibration were acceptable. As in other analysis techniques, the source-based method is the most accurate quantitative analysis method of gamma-ray spectrometry, particularly for low-energy gamma-ray radionuclides. Nowadays, owing to the large number of samples to be measured for radioactive contamination in a short time in case of a nuclear emergency, mathematical simulation is a very useful method for efficiency calibration of gamma-ray spectrometry. ANGLE, as a semi-empirical method, has the advantage of applicability to different detectors. Conversely, “characterized” detector is

Table 3 Activity concentrations of radionuclides in the inter-comparison sample obtained using the three different efficiency calibration methods

Efficiency calibration method	Activity concentration (Bq/kg) ± expanded uncertainty					
	^{241}Am	^{226}Ra	^{232}Th	^{137}Cs	^{60}Co	^{40}K
Reference value	207 ± 8	253 ± 8	236 ± 7	203 ± 4	331 ± 10	653 ± 26
Source-based	216 ± 17	251 ± 19	231 ± 18	193 ± 15	308 ± 24	645 ± 50
LabSOCS	260 ± 20	245 ± 19	234 ± 18	210 ± 16	314 ± 24	665 ± 52
ANGLE	177 ± 14	223 ± 17	218 ± 17	200 ± 15	308 ± 24	647 ± 50

Table 4 Evaluation of the results for the source-based method

Analyte	Relative bias (%)	*Z*-score	*U*-test	A_1	A_2	Trueness	*P* (%)	Precision	Final score
^{241}Am	4.40	0.44	0.49	9.11	47.57	Acceptable	8.60	Acceptable	Acceptable
^{226}Ra	– 0.70	– 0.07	0.08	1.77	53.90	Acceptable	8.31	Acceptable	Acceptable
^{232}Th	– 1.93	– 0.19	0.23	4.55	50.09	Acceptable	8.37	Acceptable	Acceptable
^{137}Cs	– 5.16	– 0.52	0.68	10.47	39.45	Acceptable	7.92	Acceptable	Acceptable
^{60}Co	– 7.01	– 0.70	0.91	23.21	66.01	Acceptable	8.23	Acceptable	Acceptable
^{40}K	– 1.29	– 0.13	0.15	8.45	146.38	Acceptable	8.78	Acceptable	Acceptable

Table 5 Evaluation of the results for the LabSOCS method

Analyte	Relative bias (%)	*Z*-score	*U*-test	A_1	A_2	Trueness	*P* (%)	Precision	Final score
^{241}Am	25.81	2.58	2.48	53.43	55.62	Acceptable	8.60	Acceptable	Acceptable
^{226}Ra	– 2.89	– 0.29	0.36	7.32	52.90	Acceptable	8.31	Acceptable	Acceptable
^{232}Th	– 0.80	– 0.08	0.10	1.89	50.60	Acceptable	8.37	Acceptable	Acceptable
^{137}Cs	3.45	0.35	0.42	7.01	42.80	Acceptable	7.92	Acceptable	Acceptable
^{60}Co	– 5.28	– 0.53	0.67	17.49	67.06	Acceptable	8.23	Acceptable	Acceptable
^{40}K	1.90	0.19	0.21	12.41	150.16	Acceptable	8.78	Acceptable	Acceptable

Table 6 Evaluation of the results for the ANGLE method

Analyte	Relative bias (%)	*Z*-score	*U*-test	A_1	A_2	Trueness	*P* (%)	Precision	Final score
^{241}Am	– 14.31	– 1.43	1.80	29.62	42.46	Acceptable	8.98	Acceptable	Acceptable
^{226}Ra	– 11.77	– 1.18	1.57	29.79	48.82	Acceptable	8.31	Acceptable	Acceptable
^{232}Th	– 7.54	– 0.75	0.97	17.79	47.24	Acceptable	8.30	Acceptable	Acceptable
^{137}Cs	– 1.64	– 0.16	0.21	3.32	40.80	Acceptable	7.91	Acceptable	Acceptable
^{60}Co	– 6.83	– 0.68	0.88	22.62	66.06	Acceptable	8.22	Acceptable	Acceptable
^{40}K	– 0.85	– 0.08	0.10	5.53	144.19	Acceptable	8.62	Acceptable	Acceptable

a prerequisite for the use of LabSOCS. Although the performances of LabSOCS and ANGLE for ^{241}Am emitting low-energy gamma rays were not very satisfactory, they are still promising for further development and improvement with the progress of efficient calculation techniques.

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