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# **Technical Note**

# Experimental technique for efficiency transfer along different geometries and volumes

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# ABSTRACT

Efficiency calibration is a fundamental procedure in gamma spectrometric measurement. Experimental technique for efficiency calibration transfer in gamma spectrometer along different geometries and volumes has been developed and validated in this work. The developed technique offers simple and easy procedures to overcome several problems encountered in efficiency calibration of gamma spectrometer such as rate-related correction and different sample volumes. The validation shows that application of the developed technique has a precision of 95%.

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# 1. Introduction

Efficiency calibration (EFFCAL) is a fundamental procedure in gamma spectrometric measurement (GSM). EFFCAL is performed using standard point source (SPS), standard volume sources (SVS) for GSM of point (PS) and volume (VS) samples respectively. In environmental fields, the range of PS and VS activities is very wide. For example, the activity of Naturally Occurring Radioactive Material (NORM) samples ranges from few Bq in ordinary soil sample to thousands Bq in petrol pipes scale. Therefore, EFFCAL is required for various PS and VS at various detector - sample spacing (d). Moreover, it is required for different sample shapes and volumes. Therefore, developing a technique for EFFCAL transfer from specific experimental setup to another is inevitable. Computational techniques were used for EFFCAL transfer in various conditions and cases [1]. Many softwares were used such as Monte Carlo GES-PECOR, MEFFTRAN and ANGLE. The computation results were validated experimentally [2-6]. However, experimental validation has the decisive role. Experimental techniques have been developed for EFFCAL transfer in this work. The EFFCAL obtained by using SPS has been transferred to EFFCAL for VS at different d values. The obtained results were validated using SVS.

# 2. Materials and methods

The measurements were performed using laboratory gamma spectrometer, which consists of: P Type HPGe detector (Bruker Baltic) with high resolution (1.85 keV at 1.33 MeV), relative efficiency of 100% and low background shield; INTERWINNER 0.4: spectroscopy software.

Three SPS (152Eu, 137Cs and 60Co) were measured. They are described in Table 1. Mixed nuclide standard solution RO8 produced by National Physical Laboratory, UK, was measured as liquid SVS. The RGU, RGTH and RGK standard radioactive materials, produced by IAEA [7–9] were measured as powder SVS. The measured SVS are described [10,11] in Table 2.

# 2.1. Calibration

The activity of radionuclide (i) in a cylindrical volume sample, measured at a position (d) along the detector axis is determined using the acquainted count rate of its gamma line (j) by Ref. [12-14]:

$$A_{ijd} = \frac{F_{rlijd}F_{aij}}{\varepsilon_{jd}B_{ij}}cps_{ijd}$$
(1)

Where:  $\varepsilon_{jd}$  –total efficiency for j-th gamma line at (d);  $F_{rlijd}$  and  $F_{aij}$  - are rate-related and self-attenuation corrections factors for j-th

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#### Table 1

The standard point sources used in this work and their activities (uncertainty < 1%).

Source	152Eu	137Cs	60Co
A, Bq	64,211	20,274	743

#### Table 2

The standard volume sources used in this work and their description.

Sample	Ref. Date	Radionuclide	E, keV	A, Bq/Sa	Un%	w, g	Туре
RGU	-	<sup>234m</sup> Pa ( <sup>238</sup> U)	1001.0	795.2	0.6	160	
RGTH	-	<sup>232</sup> Th	911.2	519.5	4.0		powder
RGK	-	<sup>40</sup> K	1460.8	2096.1	0.3	153	
RO-8	1/1/2011	<sup>137</sup> Cs	661.6	79.4	0.7	120	liquid
		<sup>60</sup> Co	1173.2, 1332.5	90.4	0.3	120	nquia

gamma line respectively;  $B_{ij}$ - branching ratio;  $cps_{ijd}$  –j-th measured count rate.

This equation gives the  $A_{ijd}$  value in the general case. The  $F_{rlijd}$  factor includes all rate related corrections, i.e., random and true coincidence corrections. The total efficiency for j-th gamma line at the position (d)  $\varepsilon_{jd}$  for cylindrical volume sample is expressed as a product of three factors [15]:

$$\varepsilon_{jd} = \varepsilon_{intj} \varepsilon_d \varepsilon_{vd} \tag{2}$$

Where:  $\varepsilon_{intj}$  - detection unit (detector and its housing) intrinsic efficiency for the j-th gamma line;  $\varepsilon_{d}$ - geometric efficiency factor for standard PS at the position d;  $\varepsilon_{vd}$  - volume efficiency factor for volume cylindrical samples.

The  $\varepsilon_{intj}$  is the probability that a gamma ray that enters the detector unit will interact and give a pulse in the full-energy peak. The  $\varepsilon_d$  is the fraction of emitted photons emitted by PS that are intercepted by the detector. Therefore,  $\varepsilon_d$  depends on (d). The  $\varepsilon_{vd}$  value accounts for the differences between VS and PS.

Each standard source of a specified radionuclide (i) gives a specified total count rate value ( $CR_{id}$ ) when measured at (d). The  $F_{rlijd}$  value for a specified detector depends on j, d and i. Therefore,  $F_{rliid}$  can be expressed by a function as:

$$F_{rlijd} = f(CR_{ijd}) \tag{3}$$

As the  $CR_{ijd}$  value decreases the  $F_{r1 ijd}$  value decreases. However, when  $CR_{ijd}$  decreases to a threshold value ( $CR_{ijt}$ ) and lower,  $F_{r1 ijd}$ tends to unity, i.e.:

$$CR_{iid} \le CR_{iit} \Rightarrow F_{rliid} = 1$$
 (4)

This equation defines the condition for free-rate related correction position. Therefore, the index "t" indicates the position, which satisfy Eq. (4).

The  $\varepsilon_{\rm vd}$  value depends on d only for constant sample volume. The PS has neither volume nor attenuation, therefore, ( $\varepsilon_{\rm v} = 1$ ,  $F_{aj} = 1$ ) for PS and consequently, Eq. (2) becomes:

$$\varepsilon_{pjd} = \varepsilon_{intj}\varepsilon_d \tag{5}$$

Substituting Eq. (5) in Eq. (1) gives the PS activity by:

$$A_{pijd} = \frac{F_{prljd}}{\varepsilon_{pid}B_{ij}} cps_{pjd}$$
(6)

Where: the index p – indicate the point sample.

# 2.2. PS efficiency transfer for different geometries

A point standard source (i ref) is selected to satisfy Eq. (4) regardless of j and d, i.e.:

$$F_{rliref} = 1 \tag{7}$$

The source (i ref) is measured in the positions (d) and (t). The measured activities in both positions must be equal. Therefore, applying Eq. (4) gives:

$$A_{p iref jd} = A_{p iref jt} \tag{8}$$

Substituting Eq. (6) in Eq. (8) and rearranging gives

$$\varepsilon_d = \frac{cps_{pjd}}{cps_{pjt}}\varepsilon_t \tag{9}$$

This equation gives  $\varepsilon_d$  as a function of  $\varepsilon_t$  experimentally using (iref) source. This means that this equation transfer  $\varepsilon_d$  value from position (t) to position (d). The transfer factor is defined by:

$$T_d = \frac{cps_{pjd}}{cps_{pit}} \tag{10}$$

Substituting this equation in Eq. (9) gives  $\varepsilon_d$  transfer equation as:

$$\varepsilon_d = T_d \ \varepsilon_t \tag{11}$$

EFFCAL at position (t) using (iref) source is performed by applying Eq. (6) and rearranging to get:

$$\epsilon_{pjt} = \frac{cps_{pjt}}{A_{pij}B_{ij}} \tag{12}$$

Noting Eq. (5) gives the efficiency value at the position (t) by:

$$\varepsilon_{pjt} = \varepsilon_{intj}\varepsilon_t \tag{13}$$

Noting Eq. (11)and Eq. (5) gives the efficiency value at the position d by:

$$\varepsilon_{pjd} = \varepsilon_{intj}\varepsilon_d$$
 (14)

Substituting Eq. (11) in Eq. (14) and reducing equal terms, gives:

$$\varepsilon_{pjd} = \varepsilon_{intj} \, \varepsilon_t T_d \tag{15}$$

This equation allows transferring GS EFFCAL from position (t) to any position (d) for PS.

## 2.3. $\varepsilon_{vd}$ determination

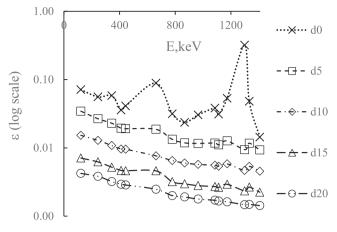
To validate the obtained results of PS EFFCAL transfer for VS, the activities of the above-described SVS of the same shape and volume were measured using the obtained  $\varepsilon_{jd}$  values. The volume efficiency factor for volume cylindrical samples  $\varepsilon_{vd}$  value is determined to get  $\varepsilon_{jd}$  as follows. Substituting Eq. (14) in Eq. (2), gives:

$$\varepsilon_{jd} = \varepsilon_{pjd} \varepsilon_{vd} \tag{16}$$

Rearranging this equation gives

$$\epsilon_{vd} = \epsilon_{jd} / \epsilon_{pjd} \tag{17}$$

This equation is used to get  $\varepsilon_{vd}$  experimentally.



**Fig. 1.** The obtained efficiency calibration curves of 152Eu and points of 137Cs and 60Co at different d values (uncertainty values < 1%).

# 3. Results and discussion

# 3.1. CR<sub>iit</sub> Determination

The first experimental step is to define  $CR_{ijt}$  for the used GS. This step were performed by GS EFFCAL at different (d) values using the above-described SPS (152Eu, 137Cs and 60Co). The 152Eu rich spectrum cover wide range of gamma lines. This is a great advantage of this source. However, its activity is high and it cannot be used for GS EFFCAL in a close geometry. Because the count rate would be too high and would raise  $F_{rl}$  to too high value. However in the position (t), which satisfies Eq. (4) these sources can be used for EFFCAL. EFFCAL were performed using the three standard sources at different positions correspond to d = 0, 5, 10, 15, 20 cm. The obtained EFFCAL curves at these positions were plotted for the sources (Fig. 1). The position (t), which correspond to  $CR_{ijt}$ , was defined as the position at which the calibration curve of 152Eu pass the calibration points of 137Cs and 60Co sources.

Analyzing this figure shows the following:

- Calibration points of 137Cs and 60Co deflect clearly at calibration curve of 152Eu in the positions correspond to d = 0, 5, 10, 15 cm. The worst deflection appears at d = 0 cm due to the highest CR value;
- Calibration curve of 152Eu passes the calibration points of 137Cs and 60Co in the positions correspond to d = 20 cm. This means that, the position d = 20 is the free-rate related correction position (t);
- The corresponding *CR<sub>ijt</sub>* value was 814 cps of 152Eu source. Therefore, 152Eu rich spectrum can be used to calibrate efficiency along wide range of gamma lines in this position the.

## 3.2. Efficiency transfer for PS

The possibility of EFFCAL transfer for PS from the position (t) to

closer positions (d < 20 cm) was investigated experimentally. The methodology described in section 2.1 was applied. The 60Co PS described in Table 1 was selected as (i ref) source. Because its count rate satisfy Eq. (4) in close geometry positions. The  $\varepsilon_d$  values for close positions (d < 20 cm) were obtained using Eq. (9). Then, the  $\varepsilon_{pjd}$  values were obtained using Eq. (14). Thus, the obtained EFFCAL at the position (t) was transferred to the EFFCAL at position (d). The obtained  $\varepsilon_{pjd}$  values were used to measure the activities of 137Cs and 60Co SPS at different (d) values. The obtained values were compared to the SPS reference values corrected to the measurement date (Table 3).

Analyzing Table 3 shows that the measured values agree with the reference one in all cases, which satisfies Eq. (4). The difference does not exceeds 5%. These values should be converged to the nominal value at the position "t". However, the standard deviation of the obtained values is less than the measurement uncertainty; therefore, the converging is not visible. Therefore, the applied methodology for efficiency transfer for PS has succeeded. The CR value for 137Cs at d = 0 was about 4500 cps, which is much higher than  $CR_{ijt}$ , therefore, the corresponding measurement was excluded.

# 3.3. Efficiency transfer for volume source

The CR values of standard RO8 volume samples satisfy Eq. (4) for all positions. The  $F_{aij}$  values in these samples are unity because they are aqueous solutions, where the self-attenuation is negligible for the considered gamma emissions. Therefore, Eq. (1) is simplified to:

$$A_{ijd} = \frac{cps_{ijd}}{\varepsilon_{id}B_{ij}}$$
(18)

Rearranging this equation gives  $\varepsilon_{id}$  as:

$$\epsilon_{jd} = \frac{cps_{ijd}}{B_{ij}A_{iid}} \tag{19}$$

The obtained results were used to calculate  $\varepsilon_{vd}$  using Eq. (17). The obtained results were plotted versus d in Fig. 2.

– The  $\varepsilon_{vd}$  values increases as (d) increases. It attains it maximum value ( $\varepsilon_{v} = 1$ ) at (d = 20 cm) for this specific sample geometry, because  $\varepsilon_{vd}$  depends generally on sample volume and measurement geometry. This means that at this position the used VS can be considered as PS. Therefore at this position PS EFFCAL can be used for VS;

- The  $\varepsilon_{vd} = f(d)$  curve accept fitting of the form:

$$\varepsilon_{vd} = -0.0007 \, d^2 + 0.041 \, d + 0.4413, \ R^2 = 0.9963$$
 (20)

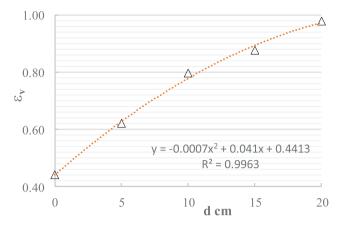
- The obtained  $\varepsilon_{vd}$  values allow getting  $\varepsilon_{jd}$  for VS using Eq. (2). Therefore, the PS EFFCAL has been transferred to VS.

The activities of the above-described SVS were measured using the obtained  $\varepsilon_{id}$  values. The 1001, 911and 1460 gamma lines were

#### Table 3

The reference, measured activities values of 137Cs, 60Co standard sources at different d values and the deviation between the nominal and the measured values.

Point Source	A, Bq; Deviation %										
	Ref. Value	d, cm									
		0		5		10		15		20	
137Cs	20,274	a		21,342	5	19,288	5	21,267	5	21,290	5
60Co	743	700	6	715	4	714	4	705	5	709	5



**Fig. 2.** The volume efficiency factor for volume cylindrical samples ( $\varepsilon_{vd}$ ) value variation versus detector - sample spacing (d) values (uncertainty values < 1%).

#### Table 4

The measured activities of standard volume samples using the developed efficiency transfer technique and their difference from the reference values.

Nuclide	E, keV	A, Bq	Dif. %
<sup>238</sup> U	1001.0	850	6.7
<sup>232</sup> Th	911.2	498	4.3
<sup>40</sup> K	1460.0	2163	1.3

used to get the activities of 238U, 232Th and 40K respectively. The 1001 keV, 911 keV gamma lines are related to 234 mPa (238U daughter) and 228Ac (232Th daughter) respectively. The daughter gamma line was used to measure the activity of the parent nuclide, because the parent nuclide and its daughter are in secular equilibrium in both 238U and 232Th cases. The 911 keV (26.2%) was used to determine the activity, because it has no coincidence summing practically (less than 3%). These gamma lines have high energies; therefore, the  $F_{aij}$  value for each of them is practically unity. Moreover, the CR values for them satisfy Eq. (4). So, Eq. (18) was used to get their activities. Table 4 lists the obtained activities along with difference from the reference values. Analyzing this table shows that the difference does not exceed 7%. This means that the PS EFFCAL to VS EFFCAL has succeeded.

# 4. Conclusions

Experimental technique for efficiency calibration transfer in gamma spectrometry along different geometries and volumes has been developed and validated in this work. The developed technique offers simple and easy procedures to overcome several problems encountered in efficiency calibration of gamma spectrometry such as rate-related correction and different sample volumes. The validation shows that application of the developed technique has a precision of 95%.

# **Declaration of competing interest**

All authors have approved the manuscript and agree with its submission and there is no conflict of interest.

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# References

- Jelena Krneta Nikolic, Rajacic Milica, Todorovic Dragana, Vidmar Tim, The first experimental test of the MEFFTRAN software on HPGe detector calibration for environmental samples, J. Environ, Radioact. 165 (2016) 191–196.
- [2] Haluk Yücel, Senem Zümrüt, Recep Narttürk Bora, Gizem Gedik, Efficiency calibration of a coaxial HPGe detector-Marinelli beaker geometry using an 152Eu source prepared in epoxy matrix and its validation by efficiency transfer method, Nuclear Eng. Technol. 51 (2) (2019) 526–532.
- Liye Liu, Jizeng Ma, Didier Franck, Loic de Carlan, Binquan Zhang, Monte Carlo efficiency transfer method for full energy peak efficiency calibration of three type HPGe detectors: a coaxial N-type, a coaxial P-type and four BEGe detectors, Nucl. Instrum. Methods Phys. Res., Sect. A 564 (1) (2006) 608–613.
   T.M. Semkow, CJ. Bradt, S.E. Beach, D.K. Haines, A.J. Khan, A. Bari, M.A. Torres,
- [4] T.M. Semkow, C.J. Bradt, S.E. Beach, D.K. Haines, A.J. Khan, A. Bari, M.A. Torres, J.C. Marrantino, U.-F. Syed, M.E. Kitto, T.J. Hoffman, P. Curtis, Calibration of Ge gamma-ray spectrometers for complex sample geometries and matrices, Nucl. Instrum. Methods Phys. Res., Sect. A 799 (2015) 105–113.
  [5] O. Sima, D. Arnold, Transfer of the efficiency calibration of Germanium
- [5] O. Sima, D. Arnold, Transfer of the efficiency calibration of Germanium gamma-ray detectors using the GESPECOR software, Appl. Radiat. Isot. 56 (1-2) (2002) 71–75.
- [6] M. C Lépy, T. Altzitzoglou, D. Arnold, F. Bronson, R. Capote Noy, M. Décombaz, F. De Corte, R. Edelmaier, E. Peraza Herrera, S. Klemola, M. Korun, M. Kralik, H. Neder, J. Plagnard, S. Pommé, J. de Sanoit, O. Sima, F. Ugletveit, L. Van Velzen, T. Vidmar, Intercomparison of efficiency transfer software for gammaray spectrometry, Appl. Radiat. Isot. 55 (4) (2001) 493–503.
- [7] https://nucleus.iaea.org/rpst/ReferenceProducts/ReferenceMaterials/ Radionuclides/IAEA-RGK-1.htm.
- [8] https://nucleus.iaea.org/rpst/ReferenceProducts/ReferenceMaterials/ Radionuclides/IAEA-RGTh-1.htm.
- [9] https://nucleus.iaea.org/rpst/ReferenceProducts/ReferenceMaterials/ Radionuclides/IAEA-RGU-1.htm.
- [10] https://wwwndc.jaea.go.jp/cgi-bin/nuclinfo2014?19%2C40.
- [11] https://wwwndc.jaea.go.jp/cgi-bin/nuclinfo2014?27%2C60.
- [12] Kh Haddad, True coincidence summing correction determination for 214Bi principal gamma lines in NORM samples, J. Radioanal. Nucl. Chem. 300 (2) (2014) 829–834.
- [13] Kh Haddad, R. Albiyat, Correction factors determination in large samples gamma assay using its own multi gamma lines spectrum, Appl. Radiat. Isot. 67 (10) (2009) 1819–1823.
- [14] Kh Haddad, M. Bouch, Validation of MCNP volume efficiency calculation for gamma spectrometric assay of large NORM samples, J. Radioanal. Nucl. Chem. 289 (1) (2001) 97–101.
- [15] Kh Haddad, R. Albiyat, Volume efficiency correction factor determination for gamma spectrometry using 82Br, J. Radioanal. Nucl. Chem. 292 (1) (2012) 29–32.