



Original Article

Efficiency calibration of a coaxial HPGe detector-Marinelli beaker geometry using an ^{152}Eu source prepared in epoxy matrix and its validation by efficiency transfer method

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ABSTRACT

In this study, an in-house ^{152}Eu calibration source was produced from a custom epoxy matrix with a density of $\rho = 1.14 \text{ g cm}^{-3}$, which is chemically stable and durable form after its solidification. The homogeneity of ^{152}Eu in matrix was obtained better than 98%. For a Marinelli beaker geometry, an efficiency calibration procedure was applied to a n-type, coaxial, 78.5% relative efficient HPGe detector in the energy range of 121.7–1408.0 keV by using in-house ^{152}Eu calibration source. Then the measured efficiencies for Marinelli geometry were compared with the results calculated by MEFFTRAN and ANGLE softwares for the validation. Although MEFFTRAN and ANGLE have two different efficiency transfer algorithms to calculate the efficiencies, they usually need to use a reliable and accurate *reference efficiency* values as input data.

Hence, *reference efficiency* values were obtained experimentally from a multinuclide standard source for the same detector- Marinelli geometry. In the present source characterization, the corrections required for self-absorption and true coincidence summing effects for ^{152}Eu gamma-rays were also obtained for a such close counting geometry condition. The experimental results confirmed the validity of efficiency calculations obtained by MEFFTRAN and ANGLE softwares that are calculation tools.

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1. Introduction

In practice, high resolution gamma-ray spectrometry (HGRS) is preferably employed to determine radionuclide activities or concentrations in a variety samples. Because it is quite easy, a less laborious and simple from point of sample preparation. If one aims at obtaining high quality results in terms of higher accuracy and precision, the samples shall be counted under the same measuring conditions (geometric setup, chemical composition, density, etc.) as those under which the gamma-ray spectrometer has been calibrated. However, it is fact that in many cases this is not possible, all above and especially when measured environmental samples [1]. This is mainly due to different chemical compositions and density of the samples. This needs to correct for self-absorptions in different matrix, and true coincidence summing effects for some radionuclides when they measured in the case of close counting

geometry.

For the quantification of the activity for a nuclide in samples by using HRGS, one needs to know accurately the efficiency of the measuring system since it provides the relation between the activity and the measured signal [2]. As calibration depends on the used measurement system itself, density and composition of the sample, sample-detector geometry arrangement and incident photon energy, the efficiency calibration of HGRS is always a key issue for analysts. For instance, many approaches have been used for the efficiency calibration for the case of HPGe detectors to quantify environmental samples [3].

The efficiency calibration of an HPGe detectors can be done in different methods:

- The first method, the efficiency calibration can be performed using *Calibration Sources* which equal to the sample to be measured in matrix, density and its sample geometry (container type, filling height of the sample), and sample to detector configuration. This way is always difficult to realize for all types samples because different calibration sources need to be prepared for different samples.

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- The second method is to have a few number of *Calibration Sources* in the laboratory, e.g., either aqueous solutions or powder forms in different containers. If the sample differs from the chosen *Calibration Source* in terms of matrix, density and filling height, then we need to make some corrections for different self-absorption effects by using special methods. To do this, some software packages such as ANGLE is based on semi empirical approach, whereas GESPECOR and EFFTRAN/MEFFTRAN is based on either Monte Carlo method [4–7]. In this method, the *reference efficiency curve* is usually required and derived from a radioactive *standard source*.
- Third method is to derive a mathematical efficiency using LabSOCS in which the detector has to be characterized by full factory measurements together with Monte Carlo modelling [8]. The LabSOCS allows to the user to establish the efficiency calibration for different samples without the need for calibration sources [9].

The motivation for this study is that low density materials as source matrix are commonly preferred to simulate food, ashed-food and other environmental samples since they have low gamma-ray self-absorption effects. However, these matrix materials are rarely found for the source preparation. Hence, the aim of this study is to prepare an in-house gamma-ray calibration source from custom epoxy resin mixed with ^{152}Eu radioisotope and as the second approach describe above, this new source will be used for characterization of a coaxial HPGe detector-Marinelli beaker geometry. In this study, only one radioisotope, ^{152}Eu that emits many gamma-rays having the useful energy range of 121.7–1408 keV was chosen and mixed with epoxy matrix because it has a long half-life and offers an economical advantage at a relatively low cost compared to a multinuclide source. Low density of an epoxy matrix has an advantage from point of low self-absorption effects for gamma-rays, however, we need to do the corrections for true coincidence summing effects from x-and gamma cascading transitions of ^{152}Eu nuclide because of its complex decay scheme. Especially, it is expected that true coincidence summing corrections for the gamma-rays from ^{152}Eu have much larger for the case of a close counting geometry. Hence, in this study, self-absorption and true coincidence summing correction factors were purposely estimated for the case of a HPGe detector- Marinelli beaker geometry. Finally, the obtained experimental efficiencies were used to determine some radionuclides (^{210}Pb , ^{137}Cs and ^{60}Co) containing in another matrix to validate the results from obtained by an in-house epoxy based ^{152}Eu source with those of the calculated results from efficiency transfer (ET) function methods implemented in MEFFTRAN and ANGLE.

2. Material and methods

2.1. Source preparation procedure

This work covers only use of epoxy resin for use of ^{152}Eu in calibration source since it has a low density (1.14 g cm^{-3}) matrix and has extra good properties. The simple and practical source preparation procedure was developed at the laboratory of Institute of Nuclear Sciences, Ankara University, Turkey. The present procedure was applied to prepare in-house calibration source from epoxy resin. First, suitable epoxy resin was chosen for the intended use by taking into account physical and chemical properties, its durability and inert phase, and low cost. The elemental composition of the presently used custom epoxy was analysed by means of PHI Versaprobe 5000 Scanning X-Ray Photoelectron Spectrometer (XPS). LECO CHNS-932 Elemental Analyser was also used to determine Carbon, Hydrogen, Nitrogen and Sulphur content. The

elemental composition of epoxy is determined to be 68.25% C, 14.67%O, 7.91%H, 4.56%N, 1.11% Ca, 1.48%Cl, 0.456% Zn and 1.56% Si in weight. Its effective atomic number is calculated to be $Z_{\text{eff}} \approx 6.1$. In source preparation procedure an aliquot having a prescribed activity is taken from ^{152}Eu stock solution (EuCl_3 in 0.5 M HCl in a glass ampoule) by micro-pipetting and then pH acidity was adjusted. This solution was gently mixed with epoxy resin by means of mechanical stirrer rod at a given rotation of 70–80 rpm. The epoxy resin mixture is waited for at least 24 h to remove humidity/water content. If required, the beaker is immersed in hot water or cold water bath while adding its hardener to the mixture at a given ratio. The final mixture is continuously stirred until it starts the solidification. Before this step, some aliquots of the whole source matrix are sampled meticulously from the beaker to fill several vials and tubes. For testing the homogeneity of the source matrix, small vials and tubes are always filled at the same height. The density of solid epoxy was determined to be $\rho = 1.14 \text{ g cm}^{-3}$ in ethyl alcohol and pure water, respectively by using RADWAG density measuring kit. Shelf-life of epoxy matrix was observed during least two years and it was remained in transparent and rigid. There was observed little colourization in samples. In bend tests, its elasticity module is determined to be $1246 \pm 30 \text{ MPa}$, and the strength is about 173 N by Zwick Universal Material Testing Machine in Middle East Technical University (METU) in Ankara.

2.2. Efficiency transfer (ET) methods implemented in MEFFTRAN and ANGLE 4

In this section, the basics of ET function algorithms are described briefly and then their implementation in MEFFTRAN and ANGLE 4 softwares are outlined:

In order to calculate the efficiency for the specific sample geometry, one of the validated computing methods is EFFTRAN based on Monte Carlo (MC) simulation efficiency transfer, especially suitable for disk and cylindrical source to a HPGe detector arrangement. As mentioned in Introduction section, a new version of EFFTRAN-called MEFFTRAN which is suitable for many-dimensional systems such as Marinelli beakers was developed. MEFFTRAN contains the additional module which calculates coincidence correction factors based on the defined parameters of the detector and sample [7]. The MC efficiency transfer method needs to use directly the manufacturer supplied data in all MC calculations. In principle, the efficiency at a given energy, E can be calculated using the expression, $\epsilon_x = \epsilon_{\text{ref}} \times \left[\frac{\epsilon_x^{\text{MC}}}{\epsilon_{\text{ref}}^{\text{MC}}} \right]$ where ϵ_x is the *reference*

efficiency for a given geometry and energy, ϵ_{ref} is the experimentally determined efficiency for a reference geometry, $\epsilon_{\text{ref}}^{\text{MC}}$ is calculated efficiency for the reference geometry, and ϵ_x^{MC} is calculated efficiency for the sample geometry, respectively. Another efficiency computing method is ANGLE that uses a semi-empirical approach, by way of the efficiency transfer method, based on the calculated effective solid angle. In a physical model, the treatment of gamma-ray attenuation, geometry and detector response is defined by the expression: $\epsilon_x = \epsilon_{\text{ref}} \times \left[\frac{\bar{\Omega}_x}{\bar{\Omega}_{\text{ref}}} \right]$, where $\bar{\Omega}_{\text{ref}}$ is effective solid angle for the

reference geometry and $\bar{\Omega}_x$ is effective solid angle for the sample geometry of interest. Effective solid angle $\bar{\Omega} = \iint_{V_S, S_D} d\bar{\Omega}$ is semi-

empirically calculated where V_S is the source volume and S_D is the detector surface subtended at the source [4]. This approach can have an advantage over the traditional relative and stochastic methods by decreasing the chances for systematic errors and reducing sensitivity to uncertainties in detector parameters [10].

In both MEFFTRAN and ANGLE 4 softwares, the efficiency calculation starts with using a well-defined *reference efficiency* calibration curve which was already determined experimentally efficiency values obtained from a standard source containing either single radionuclide or multinuclide source. Additionally, the geometrical characteristics of the detector have to be defined in the detector module (pop-up menu in ANGLE 4) and the physical dimensions of the beaker and its material info have to be defined in the sample container module (previewed dimension details of Marinelli geometry in display). When we compared the MEFFTRAN, ANGLE 4 uses much more characteristics of the detector, and allows the user to define additional possible layered absorbing materials which might surrounding the end-cap.

The sample source (here called an in-house prepared source) for which the efficiency is calculated, as well as the radioactive “standard” source (here called a multinuclide standard source) is defined in the same module in Excel sheet named “Source” in MEFFTRAN but it was named “sample source” in ANGLE 4 menu. In both MEFFTRAN and ANGLE 4, the measurement geometry is defined as Marinelli beaker with adjustable radius and sample filling height by the user. Different matrices of the samples are defined in terms of the chemical composition (either elements or compounds, or mixtures in fraction or weight percent) and the density in the “Material” module of the software. In MEFFTRAN, the efficiency calculation is performed in the module “Transfer Efficiency” by ensuring the “reference efficiency calibration or standard source efficiency” data which was inserted in the Excel sheet. After pressing the transfer efficiency button, the efficiency calculation starts transferring from “Standard source efficiency” to “Sample efficiency”. The efficiency results are tabulated in the display, together with their estimated uncertainty values, and also visually displayed as the efficiency curves in the same Excel page. MEFFTRAN has also another capability to calculate true coincidence summing correction factors when the characteristics of detector and the source are appropriately defined in Excel based sheets.

As well as MEFFTRAN, ANGLE 4 makes use of the detector and source characteristics and produces an efficiency transfer function from the “reference efficiency or standard source efficiency” data to “sample efficiency” data by solving complex analytical formulae. This is possible if the user is already defined the “reference efficiency curve” in the menu. In the logarithmic scale, the *reference efficiency curve* can be defined in either a single one energy region, or in two (or more) energy regions if there exists sufficient number of the measured efficient points. This tool can provide the user to

establish more accurate the reference efficiency curve data input before the final efficiency calculation for the sample. In ANGLE 4, the efficiency results are tabulated without any uncertainty components. But it provides the effective solid angles corresponding to the energies in the display. All calculated the results can be saved, stored in files and transferred by copy-clipboard to another programs, such as Excel for further use them. It is to say that ANGLE 4 has no yet a capability to calculate true coincidence summing correction factors.

Both MEFFTRAN and ANGLE 4 softwares have user-friendly features for the gamma-spectrometrists. For instance, both can convert the calculated efficiency functions to the data formats read by Ortec GammaVision and Canberra Genie 2000™ data acquisition programs. This feature provides a practical benefit to the gamma-analysts in data evaluation.

2.3. Gamma-ray spectrometry systems

An n-type and closed-end coaxial high purity Ge (Ortec GMX70P4S Model) detector (No.1) has a relative efficiency of 78.5% and a P/C ratio of 74:1 at 1332.5 keV (⁶⁰Co). Its energy resolution is 2.08 keV at 1332.5 keV (⁶⁰Co) and 0.8 keV at 122.1 keV (⁵⁷Co). A p-type, well geometry Ge (Canberra GCW4023 Model) detector (No.2) was used only for homogeneity tests to measure the samples in small ampoules/vials and tubes. It has a 44.8% relative efficiency and its energy resolution is 2 keV at 1332.5 keV (⁶⁰Co). Its P/C ratio is 60.8:1 at 1332.5 keV (⁶⁰Co). This detector has a well with a diameter of 23 mm and a depth of 35 mm. As given in Table 1, the main physical characteristics of n-type HPGe detector (No.1) was used in the detector modelling implemented in ANGLE 4 and MEFFTRAN versions. These data were taken from the manufacturer technical sheets and they are not optimised for the characterization.

Each detector has separately installed in its cylindrical lead shield of 10 cm thickness, graded with Sn and Cu liners, jacketed by about 1 cm low-carbon steel outer casing. The base of lead shield has an annular lead plug in which accommodate only a dipstick cryostat and the detector cables.

Data acquisition from Ortec n-type Ge detector was carried out in the 4096 MCA channels by means of a 16 K ADC/MCA digital signal processing (DSPEC jr 2.0) spectrometer through a Gamma Vision acquisition software. A lower level discriminator (LLD) was set at 26 channels to offset low energy electronic noise of below 5 keV and soft low γ - and x-rays of below 15 keV in the fine

Table 1
Main physical characteristics of detectors.

Detector	Detector No.1 n-type Ge; ORTEC	Detector No.2, p-type, Ge well; CANBERRA
Crystal diameter [mm]	69.9	66.5
Crystal length [mm]	82.6	67
Inactive side thickness [mm]	0.003	0.003
Contact side thickness [mm]	0.7	0.76
Crystal hole length [mm]	73.8	35 (well depth)
Crystal hole diameter [mm]	9.2	23 (well radius)
Crystal material	Ge	Ge
End cap diameter [mm]	82.6	88.9
End cap thickness [mm]	0.508	0.762
End cap material	Al	Al
End cap lower thickness and inside thickness [mm]	N/A	1
Window thickness [mm]	0.5	1
Vacuum gap from window to upper top of crystal [mm]	4	7.874
Vacuum gap from lower top to crystal [mm],	4	4.064
Vacuum gap from side to crystal [mm],	4	2.738
Window material	Be	Al
Mount cup thickness [mm]	0.76	0.005
Mount cup material	Aluminized Mylar	Al

N/A:Not Applicable.

Table 2
Homogeneity test results for epoxy based ^{152}Eu source.

	Sample Number	Specific count rate (cps/g)	Homogeneity, %
From 121.8 keV	1	0.042±0.3	99.7
	2	0.057±0.7	99.3
	3	0.048±0.6	99.4
	4	0.055±2.1	97.9
	5	0.064±0.4	99.0
	6	0.066±1.6	98.4
	7	0.069±1.3	98.7
	8	0.065±0.6	99.4
	9	0.081±1.5	98.5
	10	0.082±0.7	99.3
		Average: 0.06±1.4	98.9
From 344.2 keV	1	0.041±0.8	99.2
	2	0.058±0.7	99.3
	3	0.049±0.6	99.4
	4	0.054±1.0	99.0
	5	0.062±0.4	99.2
	6	0.065±0.3	99.7
	7	0.068±0.4	99.6
	8	0.065±0.5	99.5
	9	0.077±0.6	99.4
	10	0.080±1.1	98.9
		Average: 0.06±0.7	99.3
From 1408.0 keV	1	0.004±5.3	94.7
	2	0.005±0.9	99.1
	3	0.004±4.4	95.6
	4	0.005±1.8	98.2
	5	0.006±1.7	98.3
	6	0.006±5.7	94.3
	7	0.006±4.9	95.1
	8	0.005±3.7	96.3
	9	0.007±3.4	96.6
	10	0.007±0.1	99.9
		Average: 0.006±3.2	96.8

adjustments. Data acquisition from Canberra GCW GCW4023 well detector was performed in 4096 MCA channels with the associated standard nuclear instrumentation modules (2025 Advanced Amplifier and 16 Multiport II ADC/MCA; Canberra Inc.) through Genie 2000™ acquisition software. In the spectrometer calibration, appropriate adjustments were also performed to cover up the interested energy range, and P/Z cancellation for pulse shaping and offsetting low-energy electronic noise and soft low γ - and x-rays of below 15 keV by setting at LLD channels.

The used Marinelli beaker has a nominal 500 ml sample volume as a re-entrant container. The physical dimensions and made-material (polystyrene) of Marinelli beakers used in this work are identical those of a silicon resin based multinuclide standard source (code: CBSS2, purchased from Czech Metrology Institute (CMI), based on silicon matrix and its density, $\rho = 0.985 \text{ g cm}^{-3}$). The filling heights of the Marinelli beakers are always kept the same.

3. Results and discussion

3.1. Homogeneity tests of in-house prepared epoxy based source

Homogeneity tests were carried out on the samples with small amounts taken from the prepared epoxy resin based ^{152}Eu mixtures by measuring a well type-Ge detector (No.2). A few ml solution of ^{152}Eu stock solution was thoroughly mixed epoxy resin with its hardener, and then filled in a 5 mL glass ampoule and 6 mL cylindrical vials, at the same heights.

The filling heights were kept the same in the vials as well as in the ampoules. At least 10 aliquots were prepared from different parts of the whole bulk mixture and they were waited for

solidification in the vials and glass ampoules. All these test samples were then measured in the well of detector No.2. The count rates per sample mass (in cps/g) were obtained for the most intense peaks of ^{152}Eu : 121.78 keV (28.41%), 344.28 keV (26.59%) and 1408.01 keV (20.85%). Since the samples were measured in the well of the detector, i.e., approx. 4π solid angle geometry, thus providing to a high gamma efficiency, and therefore the counting rates can be determined more accurately, as given in Table 2 in which their uncertainties are about 0.7–4.9%. For Marinelli beaker, the homogeneity of the epoxy based source was estimated to be 96.3–99.3% from three different peaks of ^{152}Eu . On the average, the homogeneity is better than of 98% for in-house produced ^{152}Eu source. It is assumed to be satisfactorily for the homogeneity tests.

3.2. A reference efficiency calibration curve for a HPGe detector-Marinelli beaker geometry

The full energy peak efficiency for each photopeak at a given energy, E was calculated by the following the well known expression:

$$\epsilon(E) = \frac{N_p/t_c}{P_\gamma(E) \cdot A \cdot D} \cdot F_c \cdot F_m \quad (1)$$

Where N_p is the net counts after subtracted background counts, t_c is the live-time for the measurement $P_\gamma(E)$ is the gamma-emission probability at a given energy, F_c is the true coincidence summing correction factor, F_m is the measurement factor for correcting the count losses during the measurement, especially for short half-life radionuclides, A (Bq) is the activity at the reference date, D is decay

Table 3

The measured efficiency values for CBSS2 standard multinuclide source contained in a Marinelli Beaker.

Nuclide	Activity ^a (Bq)	Energy (keV)	Gamma-ray emission probability, %	True coincidence summing correction factors, F _c		Experimental efficiency for Marinelli geometry, ε _{ref} (%)
				From GESPECOR	From MEFTRAN	
²¹⁰ Pb	20250 ± 284	46.54	4.252	1	1	15.02 ± 0.21
²⁴¹ Am	3559 ± 39	59.54	35.92	1	1	14.67 ± 0.18
¹⁰⁹ Cd	16440 ± 230	88.03	3.626	1	1	15.05 ± 0.21
⁵⁷ Co	890 ± 10	122.06	85.51	0.9996	1	13.98 ± 0.15
⁵⁷ Co	890 ± 10	136.47	10.71	1	1	12.64 ± 0.14
¹³⁹ Ce	1104 ± 12	165.86	79.9	0.9342	0.9569	12.71 ± 0.14
²⁰³ Hg	2014 ± 48	279.2	81.48	1	1	8.58 ± 0.21
¹¹³ Sn	3186 ± 70	391.7	64.97	1	1	6.53 ± 0.14
⁸⁵ Sr	5610 ± 73	514	98.5	0.9992	1	5.36 ± 0.07
¹³⁷ Cs	2034 ± 26	661.66	84.99	1	1	4.44 ± 0.06
⁸⁸ Y	4158 ± 62	898.04	93.9	0.8894	0.8881	3.54 ± 0.05
⁶⁰ Co	2046 ± 23	1173.23	99.85	0.8833	0.8795	2.88 ± 0.03
⁶⁰ Co	2046 ± 23	1332.49	99.9826	0.8795	0.8756	2.59 ± 0.03
⁸⁸ Y	4158 ± 62	1836.05	99.32	0.8763	0.8742	2.04 ± 0.03

^a As of the reference date:30.07.2014, and Marinelli beaker filled with a 470 ml silicon resin mixed with above 11 different radionuclides.

factor. F_c factors calculated from both MEFTRAN and GESPECOR softwares based on Monte Carlo method, and there is a consistency between F_c values, as seen in Table 3. The measurements were repeated at least five times with use of CBSS2 standard source containing ²⁴¹Am, ⁵⁷Co, ¹⁰⁹Cd, ¹³⁹Ce, ¹³⁷Cs, ⁶⁰Co, ¹¹³Sn, ⁸⁵Sr, ⁸⁸Y, ²⁰³Hg and ²¹⁰Pb radionuclides. The energy range of the gamma-rays covers from 46.5 to 1836.05 keV.

As a reference efficiency curve (or standard efficiency), the experimental efficiency values given in Table 3 were then provided in both MEFTRAN and ANGLE 4 softwares as input to establish for ET algorithms, thus resulting in an accurate sample source efficiency (here either an in-house epoxy based ¹⁵²Eu source or a sand matrix spiked with ²¹⁰Pb, ¹³⁷Cs and ⁶⁰Co) in the efficiency calibration of the Ge detector No. 1.

3.3. Characterization of an in-house epoxy based ¹⁵²Eu source for Marinelli geometry by ET methods

An in-house epoxy based ¹⁵²Eu source in a Marinelli plastic beaker with a filling volume of a 470 mL was put on the end-cap of the detector No.1. Since one of our aims is to demonstrate that an HPGe detector can be calibrated by using new epoxy based in-house prepared source. The efficiency measurements were repeated at least 10 times and then for the interested photopeaks of ¹⁵²Eu calculated by MEFTRAN and ANGLE 4 softwares, respectively. The results are given in Table 4.

The efficiency curves for measured and calculated efficiencies for ¹⁵²Eu source based on epoxy matrix in Marinelli beaker are also shown in Fig. 1. In the energy range of 121.7–1408.0 keV, a good

agreement is observed between experimental and calculated efficiencies for epoxy based ¹⁵²Eu source prepared in a Marinelli beaker.

In this calibration procedure of Detector No.1 with a Marinelli beaker, true coincidence summing correction factors (F_c) were estimated for the interested peaks of ¹⁵²Eu from GESPECOR and MEFTRAN, respectively. The slightly different F_c factors are estimated when we used MEFTRAN and GESPECOR programs due their algorithms. Hence, the average values of F_c factors from these programs are used in activity calculations in this study. As expected, the larger F_c factors for the gamma-rays of ¹⁵²Eu radioisotope are estimated to be F_c = 0.76 for 121.7 keV, 0.68 for 244.7 keV, 0.89 for 344.2 keV, 0.71 for 443.9 keV, 0.85 for 778.9 keV, 0.81 for 964.1 keV and 0.79 for 1408.0 keV in Marinelli beaker geometry. This is mainly due to complex decay scheme of ¹⁵²Eu, thus leading to cascading transitions for the case of a close counting geometry and a relatively large coaxial crystal used in Detector No.1.

In this work, a few of in-house sources based on epoxy matrix were prepared in Marinelli and to show their reproducibility. As an example, a pre-determined activity of 940 ± 18 Bq of ¹⁵²Eu radioisotope containing in the epoxy matrix was measured at least 10 times repetition, then the mean activity results were obtained from the most intense photopeaks of ¹⁵²Eu and given in Table 5. These measured activity results were corrected for self-absorption and true coincidence summing correction factors.

It is worth noting that the efficiencies calculated by Monte Carlo based MEFTRAN software give more discrepancies from the experimental ones by 9.62% for 1408.0 keV peak as seen in Table 4. This might due to the non-optimised physical dimensions of the

Table 4The experimental and calculated efficiencies for epoxy based ¹⁵²Eu source contained in Marinelli beaker.

Energy (keV)	Experimental efficiency based on CBSS2 ^a	Calculated efficiency by MEFTRAN	Calculated efficiency by ANGLE ^b	Difference from Experimental Efficiency, %	
				MEFFTRAN	ANGLE
121.7817	0.1359±(2.88%)	0.1357±(3.16%)	0.1345	0.14	1.01
244.6974	0.0968±(2.89%)	0.0968±(3.17%)	0.0937	-0.06	3.30
344.2785	0.0741±(2.88%)	0.0753±(3.14%)	0.0720	-1.67	2.85
443.96	0.0604±(2.97%)	0.0618±(3.30%)	0.0591	2.40	2.18
778.9045	0.0391±(2.88%)	0.0393±(3.17%)	0.0387	-0.44	0.87
964.079	0.0333±(2.88%)	0.0327±(3.17%)	0.0331	2.02	0.61
1408.013	0.0250±(2.87%)	0.0228±(3.16%)	0.0248	9.62	0.61

^a Filled source matrix volume: 470 ml, source height: 92 mm, Beaker inner diameter:118 mm.

^b No uncertainty calculated by ANGLE 4 software.

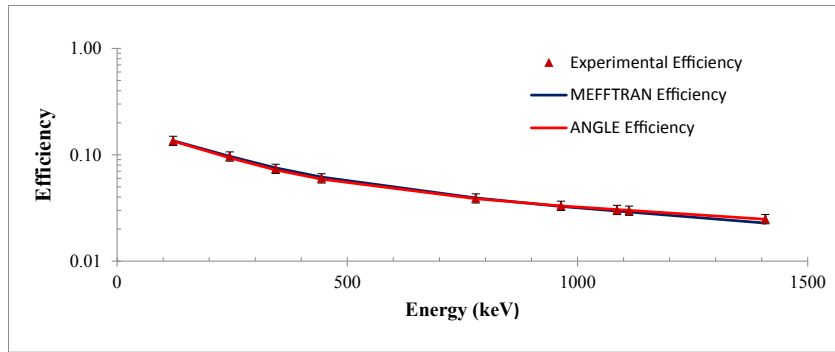


Fig. 1. Comparison of efficiency results obtained from transfer functions of ANGLE and MEFFTRAN for ¹⁵²Eu-epoxy source.

Table 5

Validation of the ¹⁵²Eu activity contained in a Marinelli beaker filled with an epoxy resin mixed with ¹⁵²Eu source by MEFFTRAN and ANGLE softwares.

Sample	Energy (keV)	P _γ %	Predetermined Activity, A _{pre} (Bq)	Self-absorption factors, F _s	True coincidence summing factor, F _c	Experimental activity based on CBSS2 standard source ^a , A _{exp} (Bq)	% Difference, A _{pre} /A _{exp} %	Calculated activity based on MEFFTRAN ^a , A _{mef} (Bq)	% Difference, A _{pre} /A _{mef}	Calculated activity based on ANGLE ^a , A _{ang} (Bq)	% Difference, A _{pre} /A _{ang}
Epoxy resin mixed with ¹⁵² Eu	121.78	28.41 (13)	940 ± 18	1.30	0.756	913 ± 26	3	885 ± 28	6.2	903 ± 26	4.1
	344.28	26.59 (12)		1.21	0.894	913 ± 26	3	885 ± 28	6.2	921 ± 26	2.1
	778.9	12.97 (6)		1.15	0.845	941 ± 27	-0.1	915 ± 29	2.7	944 ± 27	-0.4
	1408.01	20.85 (8)		1.11	0.805	964 ± 28	-2.5	986 ± 31	-4.7	980 ± 28	-4.1

^a Filling volume: 470 ml, Efficiency calibrations were based on CBSS2 standard multinuclide source, MEFFTRAN and ANGLE 4 softwares for Marinelli Beaker geometry measured with an n-type detector (No.1). Gamma-ray self-absorption factors, F_s and true coincidence summing factors F_c for the interested gamma-rays, were applied to all measured and calculated activities of ¹⁵²Eu.

Table 6

The uncertainty budget together with standard uncertainty values or ranges.

Uncertainty components	Standard uncertainty typical value or range (%)
Gamma emission probability, P _γ	0.03–9.41
Masses of samples in preparation, m (g)	0.01
True coincidence summing correction, F _c	0.75–1
Gamma-ray self-absorption factors, F _s	0.8–1.1
Pipetting from stock solution	0.9–1.2
Full energy peak area (including repeated measurements, fitting and background correction) determinations, N _p	0.03–0.52
Full energy peak efficiency (including repeated measurements, fitting and background correction) determinations, e _p	1.10–3.30
Variations in sample heights (Type B)	0.01–0.2
Total uncertainty range	1.8–10.2

The total uncertainties are expressed at 68.4% confidence level(i.e., ±1σ confidence interval).

detector and the beaker. This is because MEFFTRAN has yet no detailed modelling. As a cost free-software, it uses only the main dimensions in modelling option. However, commercial ANGLE 4 uses the more detailed description of the whole detector,

surrounding materials and the used beaker. Therefore, the efficiencies calculated by ANGLE 4 agree well with the experimental ones given in Table 4 by 0.61–3.3% in whole energy range. In ANGLE 4, there is no calculation option to calculate the uncertainties

Table 7

Results for sample source based sand matrix contained in a Marinelli beaker by using MEFFTRAN and ANGLE softwares.

Sample	Nuclide	Energy (keV)	Pre-determined activity ^b , A _{pre} (kBq)	Self-absorption factor, F _s	True coincidence summing factor, F _c	Measured and calculated activity (kBq)					
						In-house calibration source activity, A _{exp} (kBq)	% Difference, A _{pre} /A _{exp}	In-house calibration sources-based on MEFFTRAN A _{mef} (kBq)	% Difference, A _{pre} /A _{mef}	In-house calibration source-based on ANGLE A _{ang} (kBq)	% Difference, A _{pre} /A _{ang}
Sand matrix ^a	²¹⁰ Pb	46.54	50.35±%1.6	2.42	1	41.5 ± 9.9%	21.4	48.3 ± 5.2%	4.2	50.44 ± 3.0%	-0.17
	¹³⁷ Cs	661.66	7.85±%5	1.28	1	7.77 ± 0.3%	1	8.15 ± 3.0%	-3.7	8.13 ± 3.0%	-3.41
	⁶⁰ Co	1173.23	9.3±%1.3	1.21	0.88	9.13 ± 3.0%	2.1	9.53 ± 3.0%	-2.1	9.42 ± 3.0%	-1.46
		1332.49	9.3±%1.4	1.2	0.876	9.13 ± 3.0%	2.1	9.53 ± 3.0%	-2.1	9.42 ± 3.0%	-1.46

^a This source was purchased from Eckert and Ziegler Inc.

^b As of the reference date: 12.4.2016.

assigned for the efficiency results.

The main uncertainty components are mainly due to the counting statistics, the detection efficiency, weight determination, solution pipetting, self-absorption correction and true coincidence summing factors, and the uncertainties in the certified activities, given in Table 6. Other type B uncertainties are also considered such as filling height of the beaker. The uncertainty components are combined according to normal error propagation law. Total uncertainty ranged from 1.8% to 10.2%.

Additionally, the activity concentrations of the ^{210}Pb , ^{137}Cs and ^{60}Co in the spiked sand (SiO_2) matrix ($\rho = 1.7 \text{ g cm}^{-3}$) were determined using the efficiency values obtained by the epoxy based in-house ^{152}Eu source and compared with the target values given in its certificate. The measured activity results for ^{210}Pb , ^{137}Cs and ^{60}Co contained in sand matrix in 470 ml Marinelli beaker are given Table 7.

The activity results in Table 7 confirmed the validity of efficiency calculations obtained by MEFFTRAN and ANGLE softwares. Only ^{210}Pb activity determined by in-house epoxy source was deviated than its pre-determined reference value. This is because there lacks of low energy experimental efficiency points below 121.7 keV. Below to 40–50 keV in the low energy region there are only x-rays and strong coincidence gamma-rays which are not suitable efficiency calibration using ^{152}Eu source. The corrections required for self-absorption as well as true coincidence summing effects for the gamma-rays given in Table 7 were also applied to the results obtained for a such close counting geometry condition.

4. Conclusion

For the preparation of a gamma-ray calibration source with low density matrix, an epoxy resin with $\rho \approx 1.14 \text{ g cm}^{-3}$ was chosen because of its high stability, durability after it has been solidified. It has low Z elemental composition resulting in a low effective atomic ($Z \approx 6.1$) number, thus leading to less self absorption effects especially for low energy gamma-rays, below 200 keV. Additionally it provides a well homogeneity for mixing radionuclide of interest. In this work, 98.5–99% homogeneous standard sources was prepared from a ^{152}Eu stock solution. It is worth noting that other physical properties of epoxy and its very low cost makes it a good candidate matrix to produce radioactive standard source. The present results also suggest that in-house standard sources from different radionuclides can be prepared by using suitable laboratory techniques and then they can subject to standardize to National Metrology Institute. In this way, relatively very low cost-in-house standard sources can be used for the measurement of environmental samples and foodstuff samples. In study, the suitability of the prepared ^{152}Eu source based on epoxy matrix was tested for a Marinelli beaker geometry, which is a very close counting geometry condition. Because, in gamma-ray spectrometry this counting geometry is commonly used in environmental samples. Efficiency calibration of a coaxial HPGe detector-Marinelli beaker geometry was performed by using efficiency transfer (ET) function methods implemented in MEFFTRAN and ANGLE 4 softwares. A good agreement is obtained between the experimental efficiencies for ^{152}Eu and the corresponding ones calculated by using either MEFFTRAN or ANGLE softwares. It was shown that they are appropriate for this purpose although MEFFTRAN and ANGLE use two different efficiency transfer algorithms to calculate the sample efficiencies.

For further validity of the employed method, the activity

concentrations of ^{210}Pb , ^{137}Cs and ^{60}Co in the another radioactive standard source based on sand matrix ($\rho = 1.7 \text{ g cm}^{-3}$) were also determined using the efficiency values obtained by our epoxy based in-house ^{152}Eu source and then compared with their activity values given in its certificate. The results confirmed the validity of efficiency calculations obtained by MEFFTRAN and ANGLE softwares. It is worth noting that especially for the case of a coaxial HPGe detector with a Marinelli beaker geometry, both MEFFTRAN and ANGLE 4 provide accurate the efficiency results to quantify the radionuclides in every type of the samples. In the efficiency calibration procedure, it is essential that the necessary corrections for self-absorption and true coincidence summing effects be applied to the measured results obtained for especially a such close counting geometry condition.

Conflicts of interest

All authors have no conflicts of interest to declare.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.net.2018.09.024>.

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