Validation of EFFTRAN for efficiency transfer to distant geometries and volume sources

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Abstract

The efficiency transfer procedure from a geometry where a volume source was placed directly on the endcap of a germanium detector to three different distant geometries was carried out using the EFFTRAN code. One of these distant geometries included absorbers consisting of poly(methyl methacrylate). The efficiency transfer to this geometry therefore had to be realized as a two-stage transfer, since a direct efficiency transfer is not possible using EFFTRAN in such a case. Efficiency transfer to all three distant geometries yielded results which can be considered as fit-for-purpose in e.g. most of the applications of gamma ray spectrometry.

Keywords Gamma spectrometry · Efficiency transfer · EFFTRAN · Volume source

Introduction

High resolution gamma spectrometry using high purity germanium (HPGe) detectors is an established measurement technique for measuring gamma emitting radionuclides in several applications like environmental monitoring, measurements after radiological incidents like reactor accidents, safeguards, and nuclear forensics. A prerequisite for activity measurements is that there is a valid calibration available for the specific geometry used for a measurement. Empirical calibration is usually done with a reference material that contains a few radionuclides with known activities with a stated metrological traceability and their related uncertainties, and emitting gamma photons over the energy interval of interest. Another approach is to do a semi-empirical calibration, where a detector model is first constructed and its parameters are optimized based on measurements. This detector model can then be used to calculate correction factors by applying an efficiency transfer procedure (ET) when the geometry of a sample deviates from the geometry for which the calibration curve was obtained, as well as correction factors for true coincidence summing. Common geometry deviations are differences between the calibration source and the sample such as the different chemical composition (matrix) and density, as well as the filling height of the containers. Deviation in geometry occurs also when the sample container is of different size compared to the container used in the calibration. There are a few specialised calculations codes that are in use for the calculation of such correction factors [1–3]. Corrections for geometry deviations within a specific sample container, as well as to different sample containers using different codes were earlier proven to work well [4].

Another geometry deviation is when samples are positioned at a different distance compared to the calibration source. For some applications it might be favourable to increase the sample-to-detector distance. These applications include specifically samples having large activities resulting in a high dead time of the measurement, and random summing if measured close to the detector. Adding (relatively thick) absorbing layers of low-Z materials will in addition reduce the generation of bremsstrahlung generated by high-energy beta emitters that contributes to the continuous background in the spectrum, in particular at relatively low energies.

Ultimately, only one calibration geometry might in principle be needed for an HPGe detector system. However, it is important to realise that validation measurements still have to be performed in different geometries in order to guarantee

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confidence in the efficiency transfer procedure [5]. Another important aspect regards traceability, since according to VIM (Vocabulary in Metrology) [6] it now constitutes an 'unbroken chain of calibrations' (earlier it was defined as 'an unbroken chain of comparisons), which opens the way for calculation codes to be used for the calibration in e.g. a deviating geometry [7].

In this work EFFTRAN was used and validated for the calculation of correction factors by ET from an endcap geometry to three distant geometries (76, 101 and 140 mm). One of the distant geometries (140 mm) included absorbers [poly(methyl methacrylate), PMMA] of which one was on the endcap of the detector and the other formed a support for the sample container. The efficiency transfer to this geometry required a two-stage transfer where an intermediate detector specification was established from which the second ET was done.

Experimental

All measurements were done using a p-type HPGe detector (Canberra, USA), having a relative efficiency of about 80% and a resolution of about 1.9 keV, both at 1332 keV. All electronics were analogue Nuclear Instrument Modules (Ortec, USA) and acquisitions and evaluations were done using Gammavision (Ortec, USA). A certified mixed radionuclide solution (Eckert & Ziegler, USA) emitting gamma photons in the energy interval 60–1836 keV was used to calibrate the detector for a geometry where the container ($D = 64.9$ mm, $H = 19.7$ mm) was placed directly on the endcap. The calibration method described in detail in [4] was used. This calibration method results in a detector model where the dead layer thickness (using a low gamma ray energy) and the detector-to-endcap distance (using a high gamma ray energy) are adjusted in order to fit calculated efficiencies to the empirical efficiencies. The two gamma lines were chosen as ones not affected by true coincidence summing (TCS). Using the optimized detector model correction factors for TCS could be calculated in order to obtain an efficiency calibration function free from this systematic effect. The resulting detector model from this semi-empirical calibration was used as the detector model in subsequent ET calculations using EFFTRAN. Thereafter the container was measured in three different distant geometries:

- 76.0 mm: In this geometry an extra bottom thickness of 2.14 mm of poly-propylene was added to the sample container to account for the support plate where the container was placed. The distance as measured using a calliper was (76.00 ± 0.10) mm. The uncertainty represents one standard deviation from five repeated measurements

of the distance as measured at five different positions along the perimeter of the sample holder.

- 101.08 mm: In this geometry an extra bottom thickness of 0.92 mm of poly-propylene was added to the sample container to account for the support plate where the container was placed. The distance as measured using a calliper was (101.08 ± 0.19) mm. The uncertainty is one standard deviation from five repeated measurements of the distance as measured at five different positions along the perimeter of the sample holder.

- 139.96 mm: This geometry consisted of a poly(methyl methacrylate) plate (9.50 mm) on the endcap and one as a support plate (7.16 mm) for the sample container. The distance from the top of the endcap plate to the bottom of the support plate as measured using a calliper was (139.96 ± 0.20) mm. The uncertainty represents one standard deviation from five repeated measurements of the distance as measured at five different positions along the perimeter of the sample holder.

EFFTRAN was used to correct for the deviation between the calibration and measurement geometries. In EFFTRAN this is done by describing the detector and the standard geometry (geometry of the calibration) and the sample geometry. Taking as input the efficiencies at different gamma ray energies from the measurement of the standard, the efficiencies for the sample geometry are calculated as output from EFFTRAN. For the first two distant geometries a direct ET was possible to perform. However, due to being a slightly more complex geometry, ET to the third geometry had to be done in a two-stage process. In the first stage, ET was done from the calibration geometry to a geometry consisting of a sample beaker with an extra 9.50 mm bottom thickness. The efficiencies for this 'intermediate geometry' was saved as a new detector description consisting of the original detector with an extra 9.50 mm poly(methyl methacrylate) plate as an absorber placed directly on the endcap of the detector. From this intermediate geometry a second ET was done to the measurement geometry. The detector model was the same as the one presented earlier [4]. In the distant geometries a small TCS effect (maximum of about 2% losses) exists for the gamma ray energies of ^{88}Y and ^{60}Co and was accounted for.

Due to a limitation in Gammavision, the peak at 514 keV from ^{85}Sr was fitted off-line in the measurement at the most distant geometry since this peak is not fully resolved from the annihilation peak at 511 keV, and due to the relative peak heights of these two peaks in such measurement conditions. The fitted parameters in the off-line peak fit included the two energies, the different widths of these peaks (the annihilation peak is Doppler broadened), a continuous background (no observable step for the background) and the amplitudes of the two peaks. Both peaks were fitted with Gaussian

functions having, as mentioned above, different FWHM. The fitting was implemented in a spreadsheet in Microsoft Excel and the error square sum was minimized by varying the above-mentioned parameters using the add-in function Solver. All physical data, photon emission probabilities and half-lives for decay corrections to a reference date were taken from the Decay Data Evaluation Project site [8].

All measurement results were evaluated using two measures: The common zeta-score (ζ) (representing the difference between the certified activity in the sample and the evaluated one normalized by the combined uncertainty of the certified and the reported activities) and the evaluation used by the IAEA in proficiency tests (PT). For the zeta-score the results are usually accepted when $|\zeta| \leq 2$. One drawback of using this measure is that the results might get accepted due to an overestimated reported uncertainty [9]. In PT arranged by the IAEA a maximum allowed relative bias (%MAB) for the different analytes are set by the PT provider. If then the combined relative uncertainty of the certified and reported activities are larger than %MAB, a warning is assigned to the reported result. Of course, %MAB should not be smaller than the reported bias [10]. This evaluation method encourages the particpants not to overestimate uncertainties. For too high reported uncertainties a warning is assigned to the specific analyte. However, it also gives a warning if the uncertainty is too small. In this work a %MAB of 10% was set as a criterion for accuracy, which is lower than usually set by IAEA in a PT for gamma spectrometry where it usually is $\geq 15\%$ depending on the activity of the sample and the complexity of the task. To account for uncertainties due to imperfections in the detector and sample models, an additional uncertainty of 5%, $k = 1$, was set in the spectrum evaluations for the distant geometries. This extra uncertainty was not added arbitrarily but based on our previous experiences of ET when the measurement geometry has a large deviation compared to the reference geometry, i.e. the calibration geometry. Using this extra uncertainty will still make activity measurements with this added uncertainty fit-for-purpose for most applications of gamma ray spectrometry.

Results and discussion

The results of the measurements at the three different distances are shown in Table 1 and Figs. 1, 2 and 3. For the 76 mm distance, the maximum (absolute) bias was −7.1% (^{241}Am), and the average bias was −0.8%. The measurement done at a distance of 101 mm had a maximum bias of +7.1% (^{203}Hg) and the average bias of +1.8%.

For the most distant geometry (140 mm) the maximum bias was +13%, in this case for ^{113}Sn (^{203}Hg was not possible to evaluate in this measurement since the hatch of the Pb-shield could not be closed, resulting in an increased

Table 1 Certified and evaluated activities for the different distant geometries

Radionuclide	A_{cert} (Bq)	u_{cert} (Bq)	$A_{Measured76\ mm}$ (Bq)	$u_{Measured76\ mm}$ (Bq)	$A_{Measured101\ mm}$ (Bq)	$u_{Measured101\ mm}$ (Bq)	$A_{Measured140\ mm}$ (Bq)	$u_{Measured140\ mm}$ (Bq)	$A_{Measured140\ mm}$ (Bq) Off-line peak fits	$u_{Measured140\ mm}$ (Bq)
^{241}Am	217.1	3.4	202	10	208	11	218	14	211	14
^{109}Cd	3061	37	2881	145	3032	152	3164	201	3045	201
^{57}Co	115.6	1.3	112.6	5.7	114.5	5.8	121.7	7.6	116.7	7.6
^{139}Ce	151.6	1.7	146.1	7.5	149.7	7.6	166	15	152	15
^{203}Hg	434.3	5.0	460	29	465	29	NM	NM	NM	NM
^{113}Sn	570.8	6.6	569	29	584	30	642	43	599	43
^{85}Sr	711.4	8.2	715	36	747	38	754^a	60	754	60
^{137}Cs	512.9	5.9	514	26	523	26	529	27	527	27
^{88}Y	1125	13	1145	58	1169	58	1218	67	1160	67
^{60}Co	601.8	6.9	622	31	629	31	645	33	639	33

NM Not measured

a ^{85}Sr had to be fitted off-line here due to the large Annihilation peak relative the peak at 514 keV in the most distant geometry
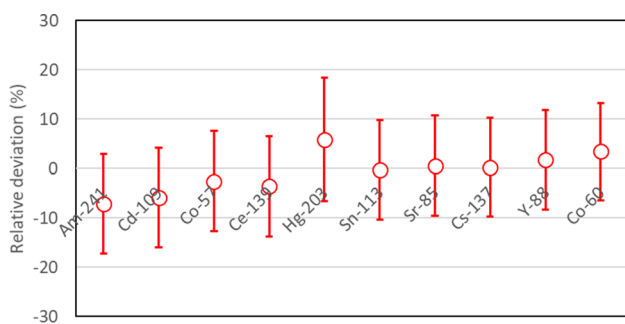
Fig. 1 Results for the 76 mm distant geometry. Uncertainties are presented for a coverage factor $k = 2$
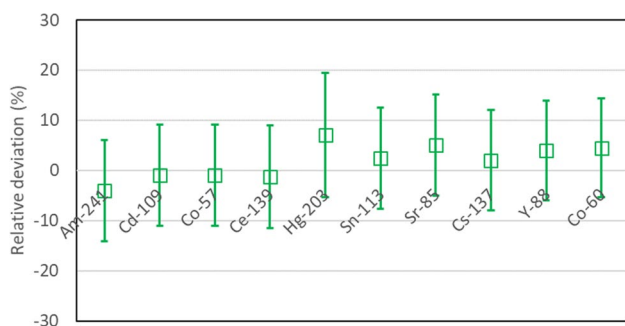

Fig. 2 Results for the 101 mm distant geometry. Uncertainties are presented for a coverage factor $k = 2$
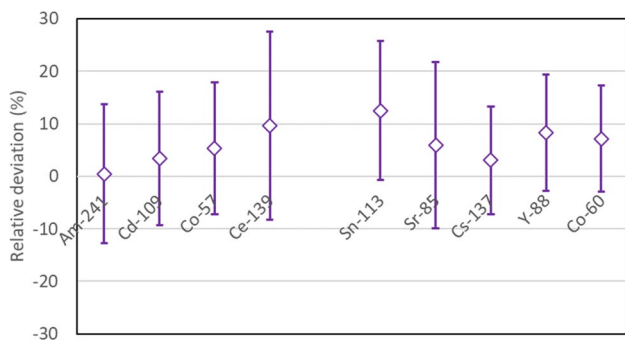

Fig. 3 Results for the 140 mm distant geometry. This was the geometry for which a two-stage ET was needed. Uncertainties are presented for a coverage factor $k = 2$

background of the measurement and in combination with the low activity of ^{203}Hg), and the average bias of $+6.2\%$. For all these three geometries the tests showed $|\zeta| \leq 2$, and relative biases $\leq 13\%$, see Table 2. In addition, all measurement results passed the IAEA criterium, which usually is $\geq 15\%$. However, ^{113}Sn did not fully met the criterium of 10% set in this work since it was 13%), for precision showing that evaluated measurement uncertainties could neither not be considered too large nor too low. These biases are in about the same range as

reported by Lépy et al. [11], which might be an indication of the limitation of the efficiency transfer method.

There seems to be an increase in both the maximum and the average bias with distance from the detector, although no statistically significant deviation from certified values could be observed. If this is a trend resulting in possible significant deviation for even larger distances is not known. This might have to be explored in further research. There are a few possible explanations for why this trend eventually could yield measurement results in error. First, the detector model might become less valid when the distance is increased, generating an error in the ET that becomes eventually too large, and in the end statistically significant. Second, there might be limitations in how the detector and geometries can be described in EFFTRAN resulting in deviations. Of course, there might also be a combination of these two causes. However, there is also a third possibility. Although the everyday user of gamma ray spectrometry most often relies on the detector manufacturers software for evaluating spectra from measurements, these software packages also have limitations of their own in e.g. peak fitting. In an attempt to evaluate this as a possible explanation the measurement at the 140 mm distance was evaluated with peak fitting done off-line (like the peak fitting of ^{85}Sr presented above). Except for ^{85}Sr, no significant interfering peaks should be expected for the other radionuclides measured in this work. The result for the 140 mm geometry and with off-line fits of all peaks is shown in Fig. 4. Performing the peak fitting off-line resulted in a maximum relative bias of about $+6.3\%$ (^{60}Co) and an average relative bias of $+1.7\%$. The latter being more than a factor of three less than when all evaluations were done using GammaVision. It can therefore not be excluded that the evaluation software may be part of the reason for the weak trend in both maximum and average biases when the distance becomes larger. It is possible that the software performs less well in the fitting of smaller peaks, in this case due to larger distances. Moreover, this triggered an attempt to perform an off-line fit of the weakest peak (279.2 keV from ^{203}Hg) in the spectra from the the 76 and 101 mm distant geometries. As a result, the deviation decreased from 5.9 to 1.0% in the 76 mm geometry and from 7.1 to 2.3% in the 101 mm geometry. However, at least up to the most distant geometry explored in this work, the software for evaluation of the measurement in combination with EFFTRAN for calculation of correction factors in the ET process can be considered as fit-for-purpose in most application of gamma ray spectrometry, and in particular for routine measurements.

Conclusions

Validation of ET using EFFTRAN from the calibration geometry to three distant geometries of an HPGe detector system showed agreement between certified and measured

Table 2 Zeta-scores and relative biases for the three distant geometries

Radionuclide	E_γ (keV) (evaluation)	Geometry: 76 mm		Geometry: 101 mm		Geometry: 140 mm		Geometry: 140 mm (off-line peak fits)	
		zeta-score	Relative bias (%)	zeta-score	Relative bias (%)	zeta-score	Relative bias (%)	zeta-score	Relative bias (%)
^{241}Am	59.54	− 1.4	− 7.1	− 0.80	− 4.1	0.07	0.46	− 0.42	− 2.9
^{109}Cd	88.03	− 1.2	− 5.9	− 0.18	− 0.95	0.50	3.4	− 0.077	− 0.52
^{57}Co	122.06	− 0.51	− 2.6	− 0.18	− 0.94	0.79	5.3	0.14	0.93
^{139}Ce	165.86	− 0.72	− 3.6	− 0.24	− 1.2	0.98	9.7	0.023	0.23
^{203}Hg	279.20	0.87	5.9	1.0	7.1				
^{113}Sn	391.70	− 0.06	− 0.30	0.45	2.4	1.66	13	0.66	5.0
^{85}Sr	514.00	0.11	0.56	0.93	5.0	0.70	5.9	0.70	5.9
^{137}Cs	661.66	0.05	0.26	0.39	2.0	0.57	3.1	0.50	2.7
^{88}Y	1836.07	0.33	1.7	0.75	3.9	1.4	8.3	0.51	3.1
^{60}Co	1173.23	0.65	3.4	0.85	4.5	1.3	7.2	1.1	6.3

Results when all peaks were fitted off-line are also presented for the most distant geometry
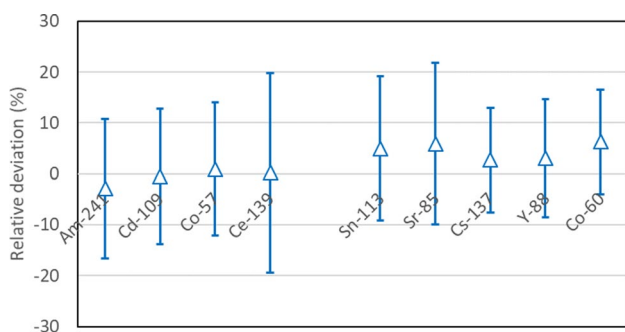

Fig. 4 Results for the 140 mm distant geometry. This was the geometry for which a two-stage ET was needed. The results in this figure are for the case when all peaks were fitted off-line. Uncertainties are presented for a coverage factor $k = 2$

activities. In one of the geometries a two-stage ET was needed, since that geometry was not possible to describe directly in EFFTRAN. The results of this work show that EFFTRAN is fit-for-purpose also for ET to somewhat more complex geometries including absorbing materials in different positions relative to the detector. In an earlier work [4] EFFTRAN showed to be fit-for-purpose for ET within a sample geometry as well as between different sample containers. Combined with the results in the work presented herein, only one calibration of a detector system might in the end be needed from which correction factors for deviation in geometry can be calculated, including different distances. Still, any application of the ET procedure with its models of the detector and the measurement geometries in a given laboratory ultimately needs to be validated against measurements of certified reference materials.

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