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Calibration of HPGe-HPGe coincidence spectrometer through performing standardisation of ^{125}I activity by X-ray-gamma coincidence spectrometry using two HPGe detectors

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Abstract

X-ray-gamma coincidence measurement method for efficiency calibration of a HPGe-HPGe system using the methodology for activity standardisation of ^{125}I has been developed. By taking one list-mode time-stamped measurement of ^{125}I source, six spectra are generated in post-processing: total spectrum, coincidence spectrum and energy gated coincidence spectrum for each of the two detectors. They provide enough observables so that source activity can be determined without a priori knowledge of the detector efficiencies. Once the source is calibrated in this way the same spectra can be used to perform efficiency calibration of the individual detectors in the low energy range. The source activity determination method is an alternative to the already established X-ray-(X-ray, gamma) coincidence counting method with two NaI(Tl) detectors and the sum-peak method using a single HPGe detector. The method was compared with coincidence counting method using two NaI(Tl) detectors. The advantage of this newly developed method is in combination of better energy resolution of HPGe detectors and measurement of only full peak areas, without the need for total efficiency determination, thus enabling activity determination even in presence of other gamma emitters in the sample. Standard coincidence counting with NaI(Tl) detectors provides lower uncertainties. The method has been used for calibration of a coincidence HPGe spectrometer in the low energy range of ^{125}I and fine adjustments of a Monte Carlo model of the coincidence system.

1. Introduction

A growing number of HPGe gamma coincidence systems [1–4], driven by the development of cheaper and easier to use digital acquisition systems, raises the question of calibration routines for such systems. Coincidence HPGe detectors are very sensitive to accurate and precise efficiency calibration. As a product of efficiencies is used for activity determination ($\varepsilon_{det1} \times \varepsilon_{det2}$), all errors and uncertainties contribute as squared values. Activity determination is either based on Monte Carlo (MC) calculations or reference sample. The first option is more versatile not needing a dedicated calibration for each isotope to be measured but generally introduces larger uncertainties. Full Energy Peak (FEP) efficiency calibration of a detector in low energy range is especially sensitive because of true coincidence summing (TCS) effects with X-rays and MC model sensitivity to dead layer

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thickness. Development of a MC model relies on proper estimation of the dead layer thickness of the germanium crystal, as this is the main parameter affecting low energy response. FEP efficiencies measured using a reference material need to be corrected for TCS effect in case of multi-gamma cascade emitters. There are only a few radioisotopes providing calibration points under ^{210}Pb 46.5 keV (26.3 keV ^{241}Am line needs a TCS correction as does 14 keV from ^{57}Co). Here we propose a method for FEP calibration of HPGe-HPGe coincidence spectrometer based on ^{125}I measurements. Basic principle is the same as used by Erikson et al. [5] for calibration of full energy peak and total efficiency by absolute measurement of ^{60}Co , except we calibrated our detector in the low energy range. No standardised source of ^{125}I is needed as the coincidence system is used for absolute measurement.

Different methods have been reported for ^{125}I absolute activity measurement and source standardisation. Pommé et al. [6] described and used seven techniques for ^{125}I solution standardisation. If we limit only to photon detection, two methods most widely used are photon-photon coincidence counting and the sum peak coincidence method [7–13]. In photon-photon coincidence counting method two NaI(Tl) detectors are used to identify coincidence events between X-rays originating from ^{125}I EC and gamma-rays or X-rays originating from the ^{125}Te de-excitation (isomeric transition). Knowing count rates on each of the detectors and the coincidence count rate give enough observables to determine the source activity [14,15]. This method provides good accuracy and precision, with the main uncertainty component coming from the uncertainties in nuclear data [6]. X-ray-(X-ray, gamma) photon coincidence counting with NaI detectors cannot be used with HPGe detectors because of Germanium X-ray escape events detected in both detectors introduce additional coincidences. The sum peak coincidence method relies on identification of coincidence sum count rate, when both photons coming from the EC and isomeric transition are absorbed in a single detector, and singles count rate when only one photon from a single decay is detected [8,16]. NaI(Tl) detectors as well as HPGe can be used for this method. Both methods work only for point sources [17], although there are various modifications correcting for voluminous source [18,19]. Abovementioned methods are sensitive to impurities presence as they rely on total count rate measurement (for total efficiency measurement).

In this work, we present a photon-photon coincidence spectrometry method for standardisation of ^{125}I using two HPGe spectrometers with a digital list-mode acquisition system. After only one time-stamped measurement, six spectra are generated in post processing, single detector spectra, coincidence spectra and energy gated coincidence spectra, providing enough independent observables for the exact activity determination and thus for precise efficiency calibration of the individual detectors in the low-energy range. The method is not sensitive to presence of impurities.

2. Theory

^{125}I decays by electron capture (EC) to the 35.5 keV excited state of ^{125}Te which de-excites (half-life 1.48 ns) by either gamma emission (6.63%) to the ground state or by conversion electrons followed by X-ray emission, Fig.1. The decay thus results in coincident emission of characteristic X-rays from the EC process with the 35.5 keV gamma line as well as with

characteristic X-rays originating from the internal conversion process. Decay data were taken from [20] and the following values are used through the rest of this paper: $T_{1/2}=59.388(28)$ days, $X_2=P_K\omega_K\times P(XK\beta)/P(XK)=0.12935(65)$ for $K\beta$ X-ray emission probability in the EC branch and $X_\gamma=0.0663(6)$ for γ -ray emission probability.

Fig. 1. Decay scheme of ^{125}I (from [20]).

If using two HPGe detectors their single count rates of the 35 keV photopeak, N_1 and N_2 , for a point source with activity N_0 can be written:

$$N_1 = N_0(X_\gamma \varepsilon_1^{\gamma,p} - X_\gamma \varepsilon_1^{\gamma,p} \times X_1 \varepsilon_1^{X,tot}) \quad (1a)$$

$$N_2 = N_0(X_\gamma \varepsilon_2^{\gamma,p} - X_\gamma \varepsilon_2^{\gamma,p} \times X_1 \varepsilon_2^{X,tot}), \quad (1b)$$

with X_γ and X_1 for gamma the emission probability and the total KX-ray emission probability in the EC branch (the gamma line cannot be in coincidence with X-rays in the internal conversion process so that is why only X-ray-gamma coincidences are subtracted). $\varepsilon_k^{i,p}$ is the FEP efficiency of photon, KX or gamma-ray, detection by detector k. $\varepsilon_k^{i,tot}$ is the total efficiency of detecting KX or γ -ray in detector k, that is the probability of any deposition of energy higher than the low energy threshold (around 5 keV on our system).

For energy gated coincidence count rate on each detector we have:

$$N_3 = N_0 X_\gamma \varepsilon_2^{\gamma,p} \times X_2 \varepsilon_1^{X,p} \quad (2a)$$

$$N_4 = N_0 X_\gamma \varepsilon_1^{\gamma,p} \times X_2 \varepsilon_2^{X,p} \quad (2b)$$

N_3 is the count rate under the X-ray peaks on detector 1 that are in coincidence with the FEP gamma events on detector 2 and N_4 is the count rate under the X-ray peaks on detector 2 that are in coincidence with the FEP gamma events on detector 1, see Fig. 5. We used only the $K\beta$ X-rays (30.9-31.8 keV) to avoid the problem of a more complicated peak area calculation for the non-resolved $K\alpha$ X-rays lines from the escape peak, see Fig. 6, so X_2 stands for $K\beta$ X-ray emission probability in the EC branch.

Finally, for coincidence count rate under gamma peak we get:

$$N_5 = N_0 X_\gamma \varepsilon_1^{\gamma,p} \times X_1 \varepsilon_2^{X,tot} \quad (3a)$$

$$N_6 = N_0 X_\gamma \varepsilon_2^{\gamma,p} \times X_1 \varepsilon_1^{X,tot} \quad (3b)$$

To solve analytically the system of Eqs. (1) - (3) we need an additional assumption.
Connecting the FEP efficiencies for K β X-ray and γ -ray:

$$\varepsilon_1^{X,p} = z_1 \varepsilon_1^{\gamma,p} \quad (4a)$$

$$\varepsilon_2^{X,p} = z_2 \varepsilon_2^{\gamma,p} \quad (4b)$$

leads to two analytical solutions for decay rate N_0 :

$$N_{01} = \frac{z_1(N_1N_2 - N_5N_6)^2 X_2}{N_3(N_1 - N_5)(N_2 - N_6)X_\gamma} \quad (5a)$$

$$N_{02} = \frac{z_2(N_1N_2 - N_5N_6)^2 X_2}{N_4(N_1 - N_5)(N_2 - N_6)X_\gamma} \quad (5b)$$

If we assume that the FEP efficiencies $\varepsilon_1^{i,p}$ on X-ray energies and on gamma energy are proportional by factor z_1 on detector one, Eq. (4a), we get the solution Eq. (5a) and respective for the other detector.

The symmetric combination of Eqs. (5a) and (5b) is the final solution for the decay rate:

$$N_0 = \sqrt{N_{01}N_{02}} \quad (6)$$

Eq. (6) is derived under the assumption that total detection efficiencies are the same for all X-ray photons (27-31 keV) in each of the detectors and the same is valid for X-ray FEP efficiencies. This means that the different detectors can be used. Ratios of X-ray to γ ray FEP efficiencies, Eqs. (4a) and (4b), will be determined by Monte Carlo simulations (Chapter 4), but as it will be mentioned in Section 5.3 they can be roughly determined by simple attenuation calculations. X-ray emission from EC and γ -ray emission are independent processes so there is no angular correlation between the photons coming from the two. Another parameter of interest is the solution for the FEP efficiency:

$$\varepsilon_1^{\gamma,p1} = \frac{N_3(N_1 - N_5)}{z_1(N_1N_2 - N_5N_6)^2 X_2} \quad (7a)$$

$$\varepsilon_1^{\gamma,p2} = \frac{N_4(N_1 - N_5)}{z_2(N_1N_2 - N_5N_6)^2 X_2} \quad (7b)$$

Depending on the assumption (4a) or (4b) we get Eq. (7a) or (7b). For the efficiency on detector two just exchange N_1 with N_2 and N_5 with N_6 . Similar as for the count rate, the final solution for the efficiency is the symmetric combination of Eqs. (7a) and (7b):

$$\varepsilon_k^{\gamma,p} = \sqrt{\varepsilon_k^{\gamma,p1} \varepsilon_k^{\gamma,p2}} \quad (8)$$

Eq. (8) gives the coincidence summing corrected value for gamma photon FEP on detector k.

3. Materials and methods

A dual HPGe detector system, Nutech Coincidence Low Energy Germanium Sandwich (NUCLEGeS) [21], recently installed at DTU Center for nuclear technologies (Nutech) has been used for the measurements. The system consists of two low-energy HPGe (Canberra LEGe GL3825R) detectors with carbon windows, Fig. 2a. The distance between the detectors is adjustable and can be varied from 0 to 6 cm, Fig. 2b. A CAEN N6781A digital multichannel analyser is used for data acquisition, enabling time-stamped list-mode data collection with 10 ns time resolution and 15 bit ADC resolution. All spectra are generated using MATLAB based coincidence analysis software and produced in Genie 2000 compatible format using Genie batch commands. Non-extending dead time and random coincidence corrections [22] and acquisition time decay correction have been implemented in the calculation software.

Fig. 2. Schematic representation of EGSnrc model for LEGe 2 detector, upward looking one in the system (a), and two detectors with plastic source holder in between (b).

A NaI(Tl) detector system, consisting of two Bicron 3' NaI(Tl) detectors placed in a long lead shield of 10 cm thickness enabling detectors to be positioned from 0 cm to 15 cm window-to-window distance, was used for X-ray-(X-ray, gamma) coincidence measurements. A CAEN DT5780P digital multichannel was used for the acquisition, similar to the one used with the HPGe system in all parameters except that it is not a NIM unit but a standalone desktop also providing high voltage for the photomultiplier tubes.

The first batch of ^{125}I point sources was prepared by pipetting 5-20 μL of ^{125}I solution (PerkinElmer, NaI, pH9) onto sticky paper with plastic back support (0.1 mm thickness, 1.5 $\times$ 3.0 cm), the droplet of ^{125}I solution was evaporated to dryness at room temperature for 1-4 hours. The dried sources were sealed by folding the sticky foil, which was then put into a plastic bag (0.05 mm thickness) for measurement. A large variation of the measured activity in these point sources was observed (up to 10%), which might be attributed to the loss of ^{125}I during evaporation even at room temperature. That is why the second batch of point sources were prepared by immediately sealing the source by folding the sticky paper after pipetting ^{125}I solution (2-5 μL) on a small filter paper (0.3 $\times$ 0.3 cm^2) attached to the sticky paper without evaporation. The folded sources were sealed in plastic bag for measurement. Additional ^{125}I point source was prepared using ^{125}I standard solution of certified activity provided by Eckert & Ziegler (California, USA)) and diluted to 5.0 ml using water.

1. Monte Carlo simulations

MC model was derived based on manufacturers specifications, Fig.2. MC simulations were done using the EGSnrc package [23] with additional decay generator [24] reproducing decays from ENSDF data. Rough model optimization was done by adjusting the MC model to reproduce experimental ^{125}I spectrum (fine tuning is explained in section 2.2). Dead layer thickness and window to crystal distance were roughly estimated by adjusting the areas of X-ray escape peaks and sum peaks while keeping the gamma peak normalized to experimental area of γ peak. It was needed to adjust the curvature of C window (Fig. 2) as the attenuation in air is higher than attenuation in vacuum. The upper and side germanium dead layers are not visible in the figure as their thickness is in submicron range. Fig. 3 shows good overlap of

experimentally measured spectrum and MC simulation of ^{125}I for a 0.9 μm Ge dead layer and a 5.5 mm window to crystal distance for each of the detectors.

The model with monoenergetic photon source of 31.1 keV and 35.5 keV energies was used for calculation of X-ray to γ ray FEP efficiency ratio (z in Eqs. (4a) and (4b)). As FEP efficiencies of X-ray and γ energies are two strongly correlated quantities, relative uncertainty of their ratio is much lower than the relative uncertainty of each quantity¹. By this it was possible to determine the coefficient z to 0.5% relative uncertainty (conservative estimate) with efficiencies varying by 10%. Table 1 gives the values of total efficiencies and their ratios for different detector parameters. Values for 0.9 μm Ge dead layer and 5.5 mm window to crystal distance are used as reference (z_1^* and $\varepsilon_1^{\gamma,p*}$) in the last two columns based on MC model optimization in Fig. 3. For standard carbon window detectors the ratio of FEP efficiencies on $\text{K}\beta$ X-ray energies and on gamma-energy is around 0.97² but it depends on detector dead layer and window material/thickness.

Fig. 3. MC spectrum (black) added on exponential background over experimentally measured spectrum (red). γ peak area of simulated spectrum is normalized to the experimental one ($15 \cdot 10^6$ decays simulated).

Table 1 FEP efficiencies at 31.1 keV (X-ray) and 35.5 keV (γ -ray) and their ratios for different detector geometry parameters ($6 \cdot 10^6$ simulated gamma events for each geometry).

Dead layer thickness (μm)	Crystal to window distance (mm)	$\varepsilon_1^{\gamma,p}$	$\varepsilon_1^{X,p}$	z_1	$\frac{z_1^* - z_1}{z_1^*}(\%)$	$\frac{\varepsilon_1^{\gamma,p*} - \varepsilon_1^{\gamma,p}}{\varepsilon_1^{\gamma,p*}}(\%)$
0.1	5	0.3489	0.3443	0.9868	0.0278	-3.8008
0.1	7	0.3258	0.3217	0.9874	-0.0409	3.0885
0.1	9	0.3036	0.3006	0.9903	-0.3293	9.6978
0.5*	5.5*	0.3362	0.3318	0.9870	0.0000	0.0000

¹ For $u = x/y$ the uncertainty is $\frac{\sigma_u}{u^2} \approx \frac{\sigma_x}{x^2} + \frac{\sigma_y}{y^2} - 2 \frac{\text{cov}(x,y)}{xy}$ [27]

² The values for efficiencies we calculate using LABSOCS on a factory calibrated Canberra BE5030 detector, for point source geometry, are 0.3241 and 0.3348 on 31.0 keV and 35.5 keV energies respectively. And that is probably the trend on all similar detectors.

1.0	5	0.3463	0.3404	0.9830	0.4093	-3.0029
1.0	7	0.3236	0.3184	0.9842	0.2833	3.7512
1.0	9	0.3017	0.2971	0.9849	0.2168	10.2566
Square root of quadratic sum of minimum and maximum value:					0.5	11

2. Results and discussion

2.1 Certified source measurement

The method was tested by measurement of a ^{125}I point source (prepared from E&Z solution) with activity of (936 ± 22) Bq ($k=2$) on the reference date of 1 February 2017. After the source had been measured all spectra were generated in post processing analysis of the time-stamped data file. First parameter needed for coincidence identification is the coincidence window width. Fig. 4 shows the coincidence time spectrum generated by plotting the time difference between the coincident signals in the two detectors. There is a sharp coincidence peak symmetric around 0, as the detectors are the same (very similar) there is no difference in charge collection and signal formation, so there was no need for a delay. A coincidence window width of $1.4 \mu\text{s}$ was selected and used in the further analysis. With this count rate the random coincidence effect was negligible.

Fig. 4. Coincidence delay spectrum showing the time difference in signal generation between the detectors. Coincidence width is taken to cover the area of coincidence peak.

By taking all the events detected within the selected time window a two-dimensional coincidence spectrum can be generated showing the coincidence fingerprint of ^{125}I , Fig. 5. An energy gated coincidence spectrum is produced by projecting the two dimensional spectrum along one detector axis around selected energy gate, i.e. by selecting all coincident events where the energy loss in the other detector is within the energy gate. The energy range used as gate to produce coincidence gamma gated spectra was 34.5 keV - 36.5 keV.

Fig. 5. Two-dimensional coincidence spectrum of a ^{125}I source. $\text{K}\alpha$ – $\text{K}\alpha$ X-ray coincidences around 27 keV on both detectors are the most abundant coincidence events. Energy gated (34.5 keV - 36.5 keV) coincidence regions used for production of spectra from which the count rates N_3 and N_4 are calculated are shown in red rectangles.

A single spectrum, coincidence spectrum and an energy gated coincidence spectrum for one of the detectors are shown in Fig. 6. In single spectrum all the X-ray peaks, γ peak, true coincidence sum peaks, germanium X-ray escapes and various combinations can be seen. In the coincidence spectrum only a few events above the 35.5 keV γ -ray energy are present and those can be explained by the recapture of the escaped Ge X-ray from the other detector as can be seen from Fig. 5 (full energy of the two coincident photons – 9.9 keV deposited in one detector and 9.9 keV in the other). The energy gated coincidence spectrum has two wide lines belonging to X-ray $\text{K}\alpha$ and $\text{K}\beta$ and their germanium X-ray escapes shifted 9.9 keV to lower energies.

Fig. 6. ^{125}I point source spectra. Single detector spectrum (black), coincidence spectrum (red) and energy gated coincidence spectrum (blue).

Count rates defined in Eqs. (1) - (3) have been determined following peak area calculation by dividing it with the live measurement time. For the peak location and area calculation Canberra Genie 2000 software [25] was used via the interactive peak fit package [26]. Finally, when all the count rates are calculated, Eq. (6) is used and after decay correction to reference date and correction for decay during the measurement time for total ^{125}I we got the value of $(938 \pm 13) \text{ Bq}$ ($k=1$) which is in agreement with the one provided by Eckert&Ziegler. Here the source was measured for 86400 s to get the counting statistic contribution to uncertainty as low as possible. If a shorter counting time of 14400 s would be selected we calculate an activity value of $(931 \pm 18) \text{ Bq}$ ($k=1$). Uncertainty was calculated with the use of propagation formula for dependent function³ under the assumption that the observables are independent. Uncertainty budget, consisting of 10 parameters, and presented in a way similar as in [15], is given in Table 2 where sources of uncertainty are divided in five groups. Relative uncertainty of MCs model used for the determination of parameters z_1 and z_2 is comparable to relative uncertainties of decay constants used so there is no point in further improving that part. The main uncertainty component is the energy gated coincidence count rate because of low probability for detection of γ -ray and $\text{K}\beta$ X-ray in two detectors.

Table 2 Uncertainty budget ($k=1$) for 86400 s measurement. Total uncertainty has been estimated as the square root of the sum of quadratic components (correlations neglected – conservative approach).

The source of uncertainty	Relative uncertainty of input quantity, $u_i(x_i)/x_i$ (%)	Relative uncertainty of output quantity, $u_i(y)/y$ (%)
EGSnrc model	0.5	0.4
Counting statistics in singles count rate	0.2	0.2
Counting statistics in energy gated coincidence count rate	0.8	0.7
Counting statistics in coincidence count rate	0.3	0.1
Difference between the two solutions (Eqs. (5a) and (5b))	0.4	0.4
Uncertainty of decay constants	0.45	1.0
Total uncertainty (%)		1.4

Using both $\text{K}\alpha$ and $\text{K}\beta$ X-rays in energy gated coincidence mode significantly improves counting statistics but introduces additional uncertainty in the final result that is harder to account for. The reason is not only complicated peak area determination for $\text{K}\alpha$ X-rays but also harder relying on the assumption of equivalence between efficiencies on different energies, Eqs. (4a) or (4b), extending over the almost double energy range than if only $\text{K}\beta$ X-rays are used. If the source activity is high enough we thus recommend using only $\text{K}\beta$ X-rays since this introduces less error. If both $\text{K}\alpha$ and $\text{K}\beta$ X-rays are going to be used X_2 needs to be changed to X_1 in Eqs. (5).

³ For $u = f(x, y)$ combined uncertainty is $\sigma_u^2 = \left(\frac{\partial f}{\partial x}\right)^2 \sigma_x^2 + \left(\frac{\partial f}{\partial y}\right)^2 \sigma_y^2 + 2\text{cov}(x, y) \frac{\partial f}{\partial x} \frac{\partial f}{\partial y}$, Eq. (4.64) from [27], extended to 6 variables used in this work.

2.2 Full energy peak efficiency

If we calculate the FEP efficiency on detector 1 using Eq. (8) and $z=0.9870$ from MC simulation, a value of (0.3276 ± 0.0033) counts/photon is obtained. A value obtained by dividing the count rate under gamma peak with the calculated photon flux is (0.2448 ± 0.0033) counts/photon. With the help of EFFTRAN code [28,29] the later value is corrected for X- γ coincidence summing with correction factor of a 1.331 using all sample and detector parameters (distance to window, absorbers and with dead layer set to zero) as in the measurement. The resulting value after the correction becomes 0.33 counts/photon, proving the EFFTRAN can be used for TCS with ^{125}I .

Now we use calculated FEP efficiencies (TCS corrected and uncorrected value) for fine adjustment of the geometry parameters in MC simulation, by minimizing the value of difference between the experimental and simulated quantities: $\sigma =$

$$\sqrt{(\epsilon_{\text{noTCS}}(\text{sim}) - \epsilon_{\text{noTCS}}(\text{exp}))^2 + (\epsilon_{\text{TCS}}(\text{sim}) - \epsilon_{\text{TCS}}(\text{exp}))^2}$$

Dead layer thickness was changed for different crystal to window distances. The final value obtained by this procedure is $\sigma = 0.0035$, that is within the uncertainty bounds of calculated efficiencies, for dead layer thickness 0.9μ and window to crystal distance 5.5 mm.

Comparison of measured and simulated spectra shows very good agreement not only for the main peaks but also sum and X-ray escape peaks, Fig. 3. Dead layer thickness is an important parameter of MC simulations of low-energy detectors as it plays a significant role in TCS correction of X-ray- γ summing. The model is only a representation of the detector with best agreement to the experimental measurements, not necessary the reality, as the dead layer thickness is probably non-uniform [30] and here we used uniform thickness representation.

2.3 Effects of geometry and absorbers

Comparison of the results with point sources (from the second batch) is shown in Fig. 7. All the measurements were done for 18000 s counting time. Point sources had different ^{125}I activities, with solution mass proportional to the source activity. When looking at the point source geometry it is clear that the higher activity of the source gives better counting statistics and thus lower uncertainties (at least in our activity ranges). The highest activity was around 250 Bq at the time of measurement giving a detector dead time of less than 0.1 %.

Fig. 7. Relative ^{125}I massic activities for different amounts of radioactive ^{125}I solution. Weighted mean value of the massic activities calculated for the four samples was used in the denominator to determine the relative massic activity. Vertical bars represent standard uncertainties ($k=1$).

A set of measurements with different detector-detector distances and three absorbers (0.75 mm Al absorber on one side, 1.45 mm Al absorber on one side and on both sides of the point source) have been undertaken, Fig. 8 also shows normalized activities relative to the weighted mean of all measurements at different window to window distances (distance between the two HPGe detectors). When detectors are closer count rates are higher giving lower uncertainties. Table 3 shows attenuation factors for $\text{K}\beta$ X-ray and γ -ray of ^{125}I (31.2 keV and 35.5 keV photon energies are compared) for two different Al thicknesses calculated using XCOM [31] and EGSnrc simulation. Data obtained by simple XCOM calculation underestimate the differences in attenuation as a ratio of $I(\text{K}\beta)$ over $I(\gamma)$ was used to estimate z , considering that on average, path lengths are greater than absorber thickness. Additional factor is difference in photon attenuation in detectors dead layer and window (visible for zero absorber thickness).

Fig. 8. Relative ^{125}I activities of a point source measured at different window to window distances (black) and with different Al absorbers (red). Vertical bars represent standard uncertainties ($k=1$).

Table 3 Attenuation factor for ^{125}I photons in aluminium calculated by XCOM and using EGSnrc model of the detector.

Al thickness (mm)	XCOM			EGSnrc
	$I/I_0(\text{K}\beta)$	$I/I_0(\gamma)$	z	z
0	1	1	1	0.9870
0.75	0.8304	0.8748	0.95	0.9051
1.45	0.6982	0.7721	0.90	0.8403

2.4 Comparison to photon-photon coincidence with two NaI(Tl) detectors

When compared with photon-photon coincidence counting measurements using two NaI(Tl) detectors the new method gives much higher uncertainties (Table 4). This is due to the fact

that in the spectra obtained using NaI detectors all the lines are summed so the relative uncertainty of each input parameter is lower, and there are fewer input parameters for the same reason. Also when taking the energy gated coincidence spectrum, the additional energy condition significantly reduces the probability for coincidence events thus reducing the statistics. When applied to a pure ^{125}I source standardisation, photon – photon coincidence counting significantly outperforms the method proposed in this work, but it fails in the presence of impurities or mixture of radionuclides due to the poor energy resolution of NaI(Tl) detector (Fig. 9) and the need for total efficiency measurement.

Table 4 Comparison of two different photon – photon coincidence counting methods for standardisation of ^{125}I activity. Measurement time is 18 000 s for both. Standard uncertainties ($k=1$).

Method	Activity (Bq)
Coincidence with HPGe detectors [Eq. (6)]	298.4 ± 6.5
Coincidence with NaI detectors [6]	300.0 ± 1.3

Fig. 9. ^{125}I gamma spectra measured by HPGe detector (black) and NaI(Tl) detector (red). Coincidence spectrum on NaI(Tl) detector is shown in blue. Low-level discriminator on both detectors is above XL-rays so those are not visible.

3. Conclusion

When developing this method the motivation was to develop the counterpart to the well-established NaI-NaI source standardisation method that would work with our HPGe coincidence system. The method shows some promising characteristics, such as good energy resolution enabling source standardisation measurement even in the presence of other radionuclides or impurities. The aim was not to produce the most accurate, precise and time effective/fastest source standardisation method. X-ray-(X-ray, gamma) coincidence counting method with two NaI detectors provides sufficient performance with much simpler and cheaper detector system. Total uncertainty of the newly developed coincidence method for HPGe-HPGe system is around 1.5% compared to 0.3% for NaI-NaI method.

Low photon energies such as from ^{125}I usually are not included in standard multi-gamma calibration solutions and sometimes it might be useful to perform an activity standardisation measurement and extend the efficiency curve of the detector system towards lower energies. The efficiency calibration of HPGe coincidence systems is very sensitive so this method (or equivalents for other nuclides) could be used to perform the calibrations. Also it should be possible to extend the method to other cascade emitting nuclides and by that to expand the applicability of HPGe-HPGe coincidence systems not only for low level measurements as it has already been used, but also into the field of activity standardisation or absolute activity measurements.

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