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DETERMINATION OF THE ^{60}Co SOURCE ACTIVITY BY USING THE SUM-PEAK METHOD

S. ILIE[1,2,4], C. A. UR[1,2], O. SIMA[1,3,4], G. SULIMAN[1], A. PAPPALARDO[1]
[1] Extreme Light Infrastructure – Nuclear Physics, ELI-NP, 30 Reactorului, P.O.Box MG-6, RO-077125 Bucharest-Magurele, Romania
E-mails: *simona.ilie@eli-np.ro*; *calin.ur@eli-np.ro*; *gabriel.suliman@eli-np.ro*; *alfio.pappalardo@eli-np.ro*
[2] University Politehnica of Bucharest, Doctoral School of Engineering and Applications of Lasers and Accelerators (S.D.I.A.L.A.), Bucharest, Romania
[3] University of Bucharest, Faculty of Physics, Bucharest–Magurele, Romania
E-mail: octavianalexandru.sima@g.unibuc.ro
[4] "Horia Hulubei" National Institute for Physics and Nuclear Engineering, P.O. Box MG-6, RO-077125 Bucharest-Magurele, Romania

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Abstract. The sum-peak method is one of the methods used for absolute activity measurement of radionuclides emitting gamma-ray photons in coincidence. This method is based on the use of acquired pulse-height distribution data only, without knowledge of the detector efficiencies. In this paper it is shown that by using Monte Carlo techniques for angular correlation corrections, pile-up peaks for corrections of accidental coincidences, low-level discriminator corrections, the activity of the ^{60}Co source can be measured with accuracy and reproducibility below 5% by using spectral data only.

Key words: sum-peak method, HPGe detector, coincidence-summing correction factors, Monte Carlo simulation.

1. INTRODUCTION

The sum-peak method is a primary activity measurement procedure. This method is based on the same principle as coincidence counting [1] but requires only one γ-ray detector with spectroscopy electronics, while the coincidence counting method requires two or more detectors with additional timing and coincidence modules. The method allows for absolute activity measurements for point sources containing radionuclides with simple decay scheme similar to ^{60}Co emitting two coincident gamma-ray photons. When two photons interact with a detector in a time interval shorter than the resolving time, each transferring the full energy to the detector, the detector response is proportional to the total deposited energy and a sum-peak appears in the spectrum. The count rate in the sum-peak, together with the count rates from the characteristic peaks of ^{60}Co source and from the integral

spectrum can be used for activity assessment. The method was introduced in a series of papers by Brinkman [2, 3] laying the foundation of the sum-peak coincidence counting. They presented the equations and their application to calculate the activity of ^{60}Co, ^{46}Sc, ^{22}Na nuclides with simple decay schemes.

The advantage of this method is that the activity of gamma cascade emitting sources can be determined based only on the use of measured spectra, without knowledge of the detector efficiencies. Most commonly, the method is applied to point-like sources measured in a close geometry, which improves the statistics of the counts in the sum-peak.

The application of the sum-peak method requires the knowledge of the total count rate together with the peak count rates and the sum-peak count rate. Since the total count rate should include the counts below the low-level discriminator (LLD), certain assumptions must be taken into account, such as the spectrum extrapolation to zero. Also, the background contribution to the total spectrum should be accurately known.

To overcome the problem regarding the difficult way of estimation of the total count rate, the modified sum-peak method [4] has been developed as a practical measurement technique, which estimates the activity solely from the peak and the sum-peak count rates without using the total count rate. The method has already been demonstrated experimentally by using ^{60}Co and ^{22}Na sources [4].

In this work different possibilities for the determination of the activity using the traditional sum-peak method, the coincidence-summing correction factors and the modified sum-peak method are presented.

The experimental values of activity obtained using these approximations were calculated using measurements of a point-like source of ^{60}Co with a high-purity germanium detector (HPGe) with 150% relative efficiency from the new facility Extreme Light Infrastructure – Nuclear Physics, ELI-NP.

The ELI-NP facility will host two major experimental set-up, 2 × 10 PW High Power Laser System and a high intensity Gamma Beam System with tunable energy in the range from 200 keV to 19.5 MeV [5, 6, 7].

The Gamma Beam System diagnostics of ELI-NP requires a precise determination of the gamma beam parameters up to 19.5 MeV. Part of the diagnostics will involve the use of the high efficiency HPGe detector for in-beam measurements. Knowledge of the detector full energy peak efficiency over the 0.2 – 19.5 MeV range is very important [8, 9].

Usually, the efficiency of the detectors is assessed by making well controlled measurements of standard gamma-ray sources with known activity but may appear situations when the activity of the source is unknown. In such situations it is very important to firstly determine the activity of the source by using only the spectral data and secondly to determine the efficiency of the detector.

2. DIFFERENT POSSIBILITIES FOR ACTIVITY DETERMINATION INVOLVING THE SUM-PEAK COUNT RATE

To explain quantitatively the coincidence summing effects, a simple two photons cascade emitting source with a decay scheme as presented in Fig. 1 is considered.

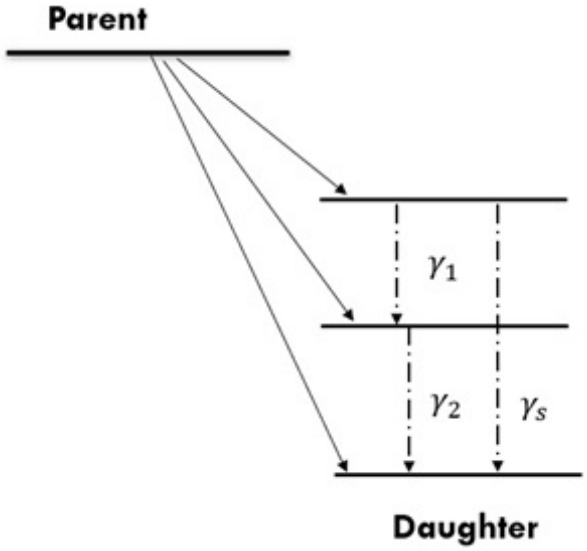

Fig. 1 – Simple gamma cascade decay scheme.

The coincidence summing effects produce two types of changes in the count rate from a given peak. In the case when a photon of energy E_1 (energy of γ_1) delivers its complete energy in the detector a pulse in the peak is always registered in the absence of coincidence-summing effects. However, if another photon, with energy E_2 (energy of γ_2), interacts simultaneously with the detector, then the pulse is lost from the peak of energy E_1, being moved to a higher energy channel, corresponding to the total energy deposited in the detector by both photons. The probability of such coincidence losses from the peak of energy E_1 depends on the probability of other photons being emitted simultaneously with photon of energy E_1 and on the total efficiency of the detector for the energy of the other photons. In the case when the transition resulting in the emission of the photon with energy E_S (energy of γ_S) can also take place in successive transitions in which photons with energy E_1 and E_2 are emitted, then simultaneous total energy deposition of the two photons in the detector would add a pulse in the peak of energy $E_S = E_1 + E_2$. The probability of such summing in (sum-peak) effects depends on the probability of emission of the corresponding group of photons and on the peak efficiency for the energies of these photons.

When the point source is measured by γ-ray spectrometry the count rates N_1, N_2 and N_S in the peaks corresponding to the energies E_1, E_2, E_S are given by the following relations:

$$N_1 = p_1\varepsilon_1\Lambda - p_{12}\varepsilon_1\eta_2 w_{\varepsilon\eta}\Lambda = p_1\varepsilon_1\Lambda F_1\,, \quad F_1 = \left(1 - \frac{p_{12}}{p_1}\eta_2 w_{\varepsilon\eta}\right) \tag{1}$$

$$N_2 = p_2\varepsilon_2\Lambda - p_{12}\varepsilon_2\eta_1 w_{\eta\varepsilon}\Lambda = p_2\varepsilon_2\Lambda F_2\,, F_2 = \left(1 - \frac{p_{12}}{p_2}\eta_1 w_{\eta\varepsilon}\right) \tag{2}$$

$$N_S = p_S\varepsilon_S\Lambda + p_{12}\varepsilon_1\varepsilon_2 w_{\varepsilon\varepsilon}\Lambda = p_S\varepsilon_S\Lambda F_S\,,\ F_S = 1 + \frac{p_{12}}{p_S}\frac{(\varepsilon_1\varepsilon_2)}{\varepsilon_S}w_{\varepsilon\varepsilon} \tag{3}$$

where, p_i are the emission probabilities of the photons γ_i with energies E_i, $\varepsilon_i(E)$ are the full energy peak efficiencies for the energies E_i, Λ is the activity of the source, η_i are the total efficiencies for the energies E_i, $w_{\varepsilon\eta}$ is the correction term for angular correlation factor when γ_1 photon is fully absorbed (first subscript ε), while the γ_2 photon is partially absorbed (second subscript η), $w_{\eta\varepsilon}$ is the correction term for the angular correlation factor when γ_1 is partially absorbed while γ_2 is fully absorbed and $w_{\varepsilon\varepsilon}$ is the correction term for the angular correlation when both photons are fully absorbed.

The full energy peak efficiency $\varepsilon(E)$ is defined as the probability of a photon of energy E emitted from the source to deposit its full energy in the detector, whereas $\eta(E)$ is the probability of that photon to deposit its energy in the detector in absence of the coincidence summing effects.

The coincidence summing corection factors (F) describe the magnitude of coincidence summing effects, representing the distorsion of the counts in the full energy peak due to these effects. Indeed, in the presence of coincidence summing effects, in the count rate equation, the peak efficiency $\varepsilon(E)$ should be replaced by the apparent efficiency $\varepsilon^{app}(E) = \varepsilon(E) \cdot F$, with F the coincidence summing correction factor, depending on the nuclide and the γ transition.

In the case when there is no transition with energy E_S, *i.e.* $p_S = 0$, the peak of energy E_S is called a pure sum-peak, being completely the result of coincidence summing effects; in this case the last term in eq. (3) can still be applied, as well as the relation for the apparent efficiency if the convention that $p_S = 1$ and $F_S = P_{12}\frac{(\varepsilon_1\varepsilon_2)}{\varepsilon_S}w_{\varepsilon\varepsilon}$ is adopted. It is important to mention that the F factors can be computed with good accuracy, for example by Monte Carlo simulation.

In the following we will consider the case of pure sum-peak.

For the same source, the count rate in the entire spectra, N_T is given by:

$$N_T = \left(p_1\eta_1 A - p_{12}\eta_1\eta_2 w_{\eta\eta} A\right) + \left(p_2\eta_2 A - p_{12}\eta_1\eta_2 w_{\eta\eta} A\right) + \\ + \left(p_{12}\eta_1\eta_2 w_{\eta\eta} A\right) \tag{4}$$

The first paranthesis in eq. (4) represents the count rate when only the first photon is detected, the second paranthesis the count rate when only the second photon is detected, while the last paranthesis corresponds to the case when the signal is produced by the simultaneous detection of the two photons.

The sum-peak method can be applied by solving the system of equations (1) to (4) for the nuclides with a simple decay scheme such as ^{60}Co with no direct transition of energy E_S. It is clear that in the case of more complex decay schemes coincidence effects due to other photons may invalidate equations (1) to (4) [10]. These equations are also not valid if the other nuclides are presented in the source, because N_T is affected by the counting rate of other nuclides. To address this, Ogata *et al.*, [11] proposed a new approximation which was applied for ^{134}Cs determination when the source contains both ^{134}Cs and ^{137}Cs, as is the case of many samples collected after Fukushima accident. In this method, the contribution of ^{137}Cs to the total spectrum is subtracted by using the count rate in the 661.6 keV peak of ^{137}Cs and the peak to total ratio of ^{137}Cs previously measured with pure ^{137}Cs sources.

2.1. ACTIVITY DETERMINATION BY USING THE TRADITIONAL SUM-PEAK METHOD

The experimental parameters which are required for activity determination using equations (1) to (4) are the full energy peak efficiencies, (ε_1, ε_2) and total efficiencies (η_1, η_2). These parameters cannot be computed very accurately by Monte Carlo simulation because of their dependence on the detector construction details which are not precisely known.

The correction factors F_1, F_2, F_S can be computed with a higher accuracy, because they are less sensitive to uncertainties in detector data, having a weak dependence on the detector model [12].

The first approach for the activity determination consists in solving the system of equations (1) to (4) by removing the dependence on all four experimental parameters.

The value of the activity calculated from equations (1) to (4) can be deduced as follow:

$$A = \left(\frac{(N_1 N_2)}{N_S} + N_T\right) K_T \text{ , with } K_T = \frac{(p_{12} w_{\varepsilon\varepsilon})}{(p_1 p_2)} \tag{5}$$

The approach described in Eq. (5) to determine the value of the activity by taking also into account the count rate in the total spectrum represents the traditional

sum-peak method. This method is very advantageous because only the uncertainties of the direct experimental quantities (count rates) contribute significantly to the activity's uncertainty. The coefficient K_T depends only weakly on the detector parameters, through the angular correlation factor and can be computed accurately by Monte Carlo simulations.

The sources of uncertainty in the activity determination are discussed in the following.

The count rate corresponding to the sum-peak energy has several sources of uncertainty including counting statistics, evaluation of the net peak area and random coincidences. In general, it is better to measure the source far from the detector to reduce the uncertainty of the angular correlation factor and the contribution of random coincidences. This leads in exchange to longer measurement times to assure a convenient level of statistical uncertainty.

The count rates N_1, N_2, also have statistical uncertainties, but they are less significant than in the case of N_S because N_1 and $N_2 >> N_S$.

The most important source of uncertainty is represented by the count rate in the total spectrum, N_T, due to spectrum extrapolation to zero energy and background subtraction. The possible fluctuations of the background should be taken into account, whereas if the background is dominated by the radon decay products, then a control of the main peaks of these nuclides in the spectrum of the source relative to the values from the background spectrum may help partly to remove the fluctuations.

2.2. ACTIVITY DETERMINATION BY USING COINCIDENCE-SUMMING CORRECTION FACTORS

To avoid the high uncertainty of the count rate in the total spectrum, the activity can be estimated by using the computed values of the coincidence summing correction factors F_1, F_2 and F_S. These factors can be evaluated with higher precision than the efficiencies ε_1, ε_2. So, the activity can be computed by using the system of equations (1) to (3) as folllows:

$$A = \left(\frac{(N_1 N_2)}{N_S} \right) K_F, \text{ with } K_F = \frac{(p_{12} w_{\varepsilon\varepsilon})}{(p_1 p_2)} \frac{1}{(F_1 F_2)} \tag{6}$$

In order to remove the uncertainty resulting from the difference between the value of the distance set in the computations and the actual distance from the source to detector, a particular correlation between the apparent efficiency and the peak count rates can be used [13, 14].

As compared to K_T from Eq. (5), the coefficient K_F depends stronger on the detector model, on the source to detector distance through the total efficiencies

which enter in the F factors and through the angular correlation correction factor. However, the sensitivity of the F factor to the uncertainty of the detector parameters is weaker than the uncertainty of the peak efficiencies, so that the uncertainty of the coefficient K_F is generally convenient.

2.3. ACTIVITY DETERMINATION BY USING THE MODIFIED SUM-PEAK METHOD

To solve the problem regarding the difficult estimation of the total count rate in the entire spectrum, the modified sum-peak method [4] has been developed as a practical measurement technique in which the activity is estimated without the need of the total count rate in the spectrum. This approach, together with the expected decrease of the uncertainties (except for the statistics of the count rates) as source to detector distance increseas (at the expense of increasing the measurement times) suggests obtaining the activity by extrapolation of the variable Λ_0 computed as:

$$\Lambda_0 = \frac{\left(p_{12} w_{\varepsilon\varepsilon}\right)}{\left(p_1 p_2\right)} \frac{\left(N_1 N_2\right)}{N_S} \tag{7}$$

for large distances. Obviously, N_T decreases when the source to detector distance increases. Asymptotically, for the distance increasing towards infinity, N_T should be zero. Therefore, asymptotically for the activity determination only the first term in the first parenthesis in Eq. (5) contributes. The method proposed by Ogata *et al.* [4] is based on the extrapolation of the quantity Λ_0, as a function of N_1 by taking the final value of the activity as the limit of the above expression for N_1 going to zero. Because the values of the F factors tend to 1 at large distances, the asymptotic values provided by both measuring procedures (Sect. 2.2 and 2.3) are equal.

3. EXPERIMENTAL DETERMINATION OF ACTIVITY OF THE ^{60}CO SOURCE

The experimental approach presented in this paper applies to a ^{60}Co source which emits two coincident gamma-ray photons. A simplified decay scheme is presented in Fig. 2. ^{60}Co decays into an excited state of ^{60}Ni which in turn decays to the ground state through the emission of two γ rays E_1 (1173 keV) and E_2 (1332 keV). The intermediate state has a very short half-life (10^{-13} s) with respect to the time response of a Ge detector that is of the order of several tens of ns (10^{-7} s) and therefore the interactions of the two gamma rays are seen as a single event by the detector.

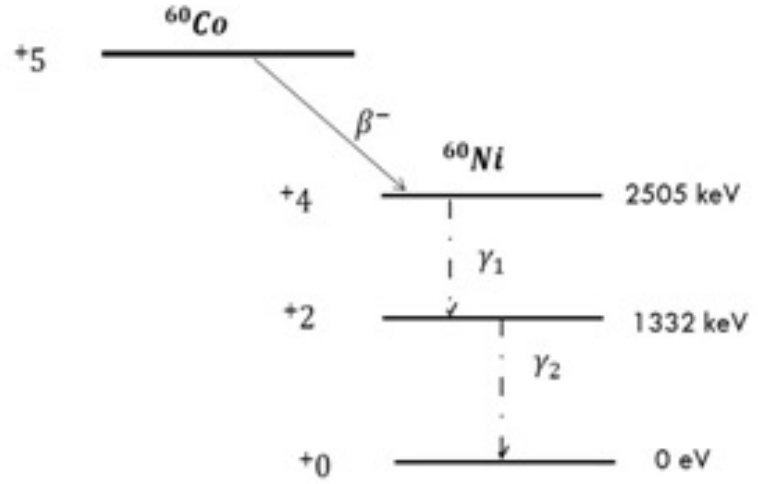

Fig. 2 – Simplified decay scheme of ^{60}Co.

In a single detector experiment the effective time in which the detection system cannot resolve two events due to chance of coincidence or pulse pile-up is represented by the time resolution τ. Due to the finite time resolution of the detection system some counts will be lost from the full energy and true coincidence peaks and collected in other parts of the spectrum [15].

In the ^{60}Co spectrum this will lead to count losses from the full energy peaks that will appear in the spectral continuum. The gain in the sum-peak due to random coincidence events from totally absorbed gamma rays of energies 1173 keV and 1332 keV emitted by different nuclei and losses from peaks due to chance coincidence events can be described by making use of τ as the time resolution of the spectrometer system. Also, counts are lost from the total spectrum because one count will be recorded instead of two, if the time interval between the two interactions is less than τ. If the two gamma rays, emitted from different nuclei, are totally absorbed in the detector within the time interval τ, in the spectrum will also appear peaks due to chance coincidence only. So, there is the posibility that in addition to the sum-peak, N_S, in the ^{60}Co spectrum peaks corresponding to the energies of 2 × 1173 kev and 2 × 1332 keV (random sum-peaks) to be present [16].

The value of τ can be estimated from the count rate of the random sum-peaks:

$$N_{(2x1173)} = 2\tau N_1^2, \qquad N_{(2x1332)} = 2\tau N_2^2 \tag{8}$$

Then the corrections corresponding to the count rates N_1, N_2, N_S and N_T due to pile-up were calculated in the first order as follows:

$$N_{1cor} = \frac{N_1}{1 - 2\tau N_T} \tag{9}$$

$$N_{2\,cor} = \frac{N_2}{1 - 2\,\tau N_T} \tag{10}$$

$$N_{Scor} = \frac{\left(N_S - \left(N_1 N_2 2\tau\right)\right)}{\left(1 - 2\tau N_T\right)} \tag{11}$$

$$N_{Tcor} = \frac{N_T}{1 - 2\tau N_T} \tag{12}$$

A more rigurous description of the pile-up effects in the application of the sum-peak method is presented in [17].

To obtain experimental results by taking into account all the considerations presented in the above sections, the equations (5), (6) and (7) with the correction factors calculated by using the Monte Carlo technique were applied to measurements of a calibrated point source of ^{60}Co with a coaxial HPGe detector with a relative efficiency of 150%.

To collect and analyse the data, an analog acquisition system consisting of a spectroscopic amplifier (Canberra model 2026) coupled to a MCA (ORTEC) model and ORTEC Gamma Vision data acquistion software was used.

Acquisition times from 3 000 to 80 000 s were chosen to obtain comparable statistics in the main peaks. For the measurements a shaping time of 6 μs and pole zero adjustments were used. The resolving time of the electronic system was in the order of 1.2 μs. The maximum total count rate on the detector in our measurements did not exceed $8 \cdot 10^3$ s^{-1}.

The number of counts in the peaks were obtained by integration of the peaks with the GASPware data analysis package [18] and background substraction was applied.

Anoher consideration in our calculations was that the spectrum had a low energy threshold; the value of N_T inserted in Eq. (5) included spectrum extrapolation to zero energy.

In order to determine the correction factors F, Monte Carlo simulations were run with the GESPECOR package [19]. For the simulation the parameters of the detector model were taken from manufacturer's data sheet.

Table 1 presents some spectral data which include the source to detector distances, the acquisition times, the count rates in the peaks at energy E_1, E_2, E_s, and the correction factors F_S.

Table 1

The spectral data of measurements of the ^{60}Co source

d [cm]	time [s]	N_{1cor} [s^{-1}]	N_{2cor} [s^{-1}]	N_{Scor} [s^{-1}]	F_S
5	3000	$(1144 \pm 5) \cdot 10^{-4}$	$(1059 \pm 6) \cdot 10^{-4}$	$(31 \pm 0.003) \cdot 10^{-3}$	1.22
10	5000	$(519 \pm 6) \cdot 10^{-4}$	$(486 \pm 6) \cdot 10^{-4}$	$(5.3 \pm 0.006) \cdot 10^{-3}$	1.15
15	7000	$(300 \pm 7) \cdot 10^{-4}$	$(280 \pm 7) \cdot 10^{-4}$	$(1.7 \pm 0.009) \cdot 10^{-3}$	1.13
20	9000	$(191 \pm 7) \cdot 10^{-4}$	$(170 \pm 7) \cdot 10^{-4}$	$(0.7 \pm 0.01) \cdot 10^{-2}$	1.12
25	15000	$(132 \pm 7) \cdot 10^{-4}$	$(124 \pm 8) \cdot 10^{-4}$	$(0.3 \pm 0.02) \cdot 10^{-2}$	1.12
30	20000	$(95 \pm 8) \cdot 10^{-4}$	$(89 \pm 8) \cdot 10^{-4}$	$(0.2 \pm 0.02) \cdot 10^{-2}$	1.11
35	30000	$(71 \pm 8) \cdot 10^{-4}$	$(68 \pm 8) \cdot 10^{-4}$	$(0.1 \pm 0.02) \cdot 10^{-2}$	1.11
40	40000	$(59 \pm 8) \cdot 10^{-4}$	$(55 \pm 8) \cdot 10^{-4}$	$(0.06 \pm 0.02) \cdot 10^{-2}$	1.11
50	60000	$(41 \pm 8) \cdot 10^{-4}$	$(36 \pm 8) \cdot 10^{-4}$	$(0.03 \pm 0.02) \cdot 10^{-2}$	1.11
70	80000	$(20 \pm 8) \cdot 10^{-4}$	$(19 \pm 8) \cdot 10^{-4}$	$(0.007 \pm 0.04) \cdot 10^{-2}$	1.11

The summary of the results taking into consideration all the factors discussed above is presented in Table 2. The uncertainties from Table 2 include only the counting statistics.

Table 2

Experimental results

d [cm]	Λ [kBq] Eq. 5	Λ [kBq] Eq. 6	Λ_0 [kBq] Eq. 7
5	63.7 ± 1.6	63.8 ± 1.5	63.1 ± 1.3
10	64.1 ± 2.3	64.5 ± 2.1	63.8 ± 2.0
15	64.2 ± 2.6	64.6 ± 2.7	63.9 ± 2.1
20	64.3 ± 2.7	64.7 ± 2.7	64.3 ± 2.1
25	64.6 ± 2.7	64.7 ± 2.7	64.5 ± 2.2
30	64.7 ± 2.8	64.7 ± 2.7	64.6 ± 2.2
35	64.7 ± 2.8	64.7 ± 2.7	64.6 ± 2.3
40	64.7 ± 2.8	65.3 ± 2.9	64.7 ± 2.4
50	64.8 ± 2.9	65.8 ± 3.1	64.7 ± 2.6
70	64.8 ± 3.1	65.9 ± 3.2	64.7 ± 2.9

The reference activity of the ^{60}Co source at the measurements date was (64.9 ± 3) kBq.

The results summarized in Table 2 confirm that the sum-peak method can provide source activities with accuracy by using only the spectral data.

The dependence of the calculated values of the activity on the source to detector distance is plotted in Fig. 3.

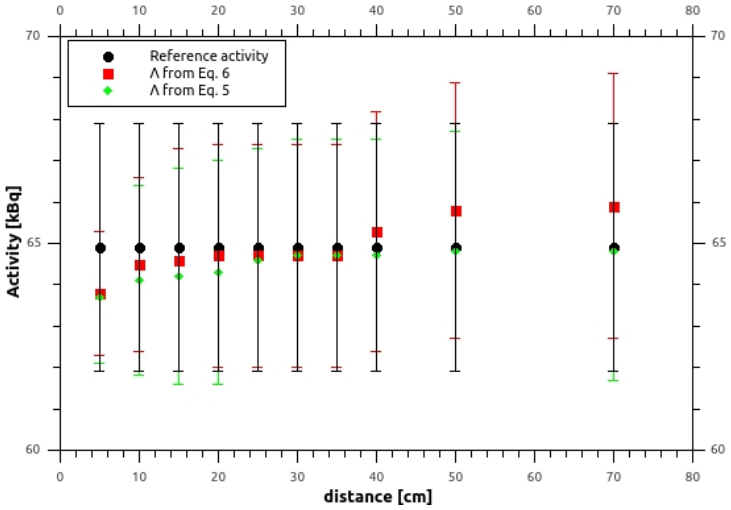

Fig. 3 – The dependence of the calculated activity as a function of the distance.

As can be seen in Fig. 3 the values of the calculated activity approach very well the known source activity.

The reason of slight decrease of calculated activity at shorter distances is considered to be the extrapolation to zero in the total spectrum, the imperfect correction of pile-up effects, the correction of the dead time.

The dependence of the ratio of the measured and reference source activity on source to detector distance is plotted in Fig. 4.

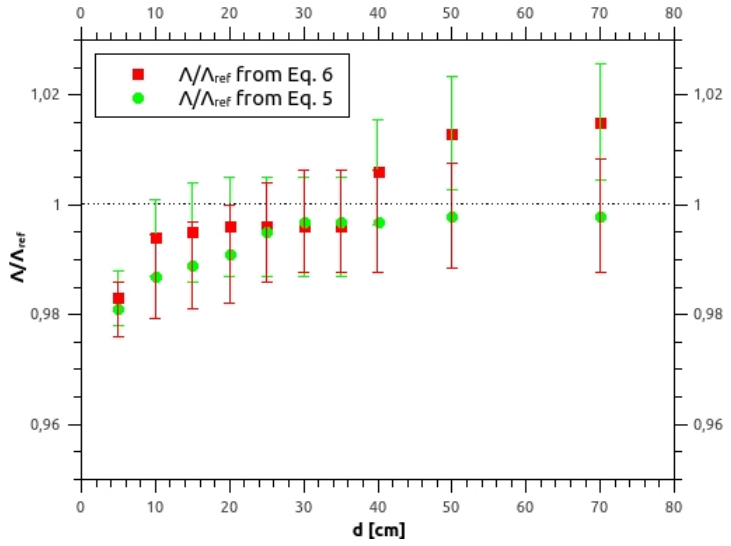

Fig. 4 – The measured and reference source activity ratio.

The data obtained using the modified sum-peak method, Eq. 7, are plotted in Fig. 5 as a function of the distance.

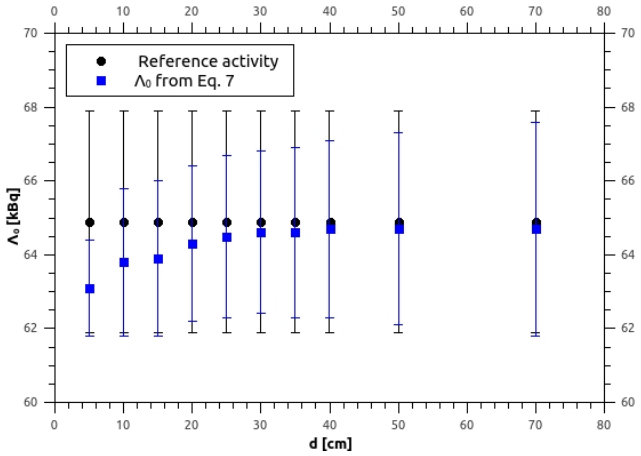

Fig. 5 – The dependence of the calculated activity Λ_0 as a function of the distance.

To obtain a better extrapolation, the dependence of the calculated activity Λ_0 as a function of the count rate in the 1173 keV peak is plotted in Fig. 6. We emphasize that in the modified sum-peak method the uncertainty associated to the total count rate is eliminated, at the expense of making measurements at several distances and extrapolating the results.

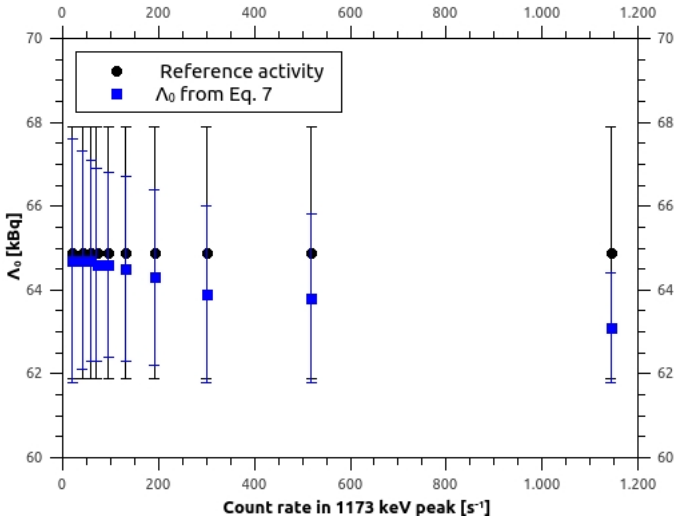

Fig. 6 – The dependence of the calculated activity Λ_0 as a function of the count rate in the 1173 keV peak. The correct value of the activity Λ is given by the asymptotic value of the Λ_0 for N_1 approaching zero.

It can be seen that the values of the activity using all the methods presented to determine the activity of ^{60}Co source agreed well with each other.

4. CONCLUSION

In this paper it was shown that the sum-peak method can be successfully applied to estimate the activity of nuclides with simple decay schemes similar to that of ^{60}Co using different approaches involving the use of spectra measured at various source to detector distances.

All these approximations demonstrate that by using only the spectral data, several different methods can be applied to obtain the ^{60}Co source activity with accuracy.

In conclusion this work showed that the activity of ^{60}Co could be calibrated with excellent accuracy not exceeding 5% by using a single HPGe detector and the sum-peak method by taking into considerations the corrections due to angular correlations and the pulse pile-up.

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