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Research Article

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Development of a 2D photon-photon coincidence digital system for absolute activity measurement

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HIGHLIGHTS

- A 2D photon-photon coincidence digital system is designed using for the primary standardization of radionuclides.
- 2D energy spectrum of Co-60 was measured.
- The application of list mode data acquisition for absolute calibration was described.
- Formulas for the activity measurement of cascade gamma-ray emitter radioisotopes were presented.

ABSTRACT

Primary standardization of radioactivity is related to the direct measurement of activity in radioactive decay. A large variety of primary standardization techniques have been developed in the past years. The photon-photon coincidence counting is one of the methods for activity determination. This method is particularly applied for the standardization of I-125 using the detection of X-ray and gamma-ray coincident counting. In this paper, a 2D photon-photon coincidence digital system with two similar $2'' \times 2''$ NaI(Tl) detectors for absolute activity measurement is developed. The system is established based on a 100 MHz CAEN waveform digitizer (DT5724) which directly records the pre-amplifier output signals of the two NaI(Tl) detectors. The sampled signals was transformed to trapezoidal signals using pulse height analyzer firmware and coincidence events were recorded in a list file. The list file was analyzed offline using a Matlab code to realize correlated gama lines of Co-60 source. The Volkovitsky formulas were used for the activity calculation and the details of the experimental setup were also discussed. Standardization of the two Co-60 standard sources was performed using this system. Results are in good agreement with the reference activity of Co-60 sources. The presented formula can be modified for absolute calibration of the other medical radioisotopes. The technique can be generalized for absolute activity measurement of I-125 which uses for ophthalmic plaque radiation therapy.

KEYWORDS

Absolute activity
Coincidence counting
Co-60 radioisotope
Metrology

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1 Introduction

Primary standardization of radioactivity is related to the direct measurements of activity in radioactive decay (Quinn, 1997). Different types of primary standardization techniques have been developed in the past years (Colle, 2009). The choice of the appropriate method is based on the decay scheme of the radionuclide and it may be different from one radionuclide to another radionuclide. The primary standardization techniques lie at the center of the activities that are used by national nuclear metrology laboratories to ensure the quality of radioactivity measurements. (Pommé, 2007) reviews different Methods for primary standardization of activity. The techniques are divided into high-geometry (4π or 2π) methods, defined solid-angle counting methods, and coincidence counting methods.

The coincidence technique is one of the method to assay a large variety of radionuclides that decay through two or more types of radiations. These techniques involve sum peak coincidence counting, photon-photon coincidence counting, and $4\pi\beta$-gamma coincidence counting. Photon-photon coincidence counting method for activity measurement has been applied in standard laboratories

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for many years (Barnothy and Forro, 1951). The limitation of the technique is the angular correlation between the emitted gamma rays in the cascade. To overcome this problem, Brinkman et al. have been introduced the sum-peak method (Brinkman et al., 1963). In this technique, by moving the source farther away from the detector, accurate activity measurement is possible (Bikit et al., 2009). However, this technique suffers a low count rate at the summing region and is not suitable for sources containing low-level and short half-life radioisotopes.

In particular photon-photon coincidence technique is suited for the standardization of I-125 (Taylor, 1967; Schrader and Walz, 1987). The technique has also been used for the characterization of Hg-197 and I-125 radioisotopes with I-125 as a tracer. Other possibilities, such as In-111, Cd-109, and I-124 with I-125 as an impurity (Schrader, 2006) and Co-60 have been studied (Volkovitsky and Naudus, 2009).

I-125 is a radioisotope of iodine with a half-life of 59.43 days. It emits 27 (113%), 31 (21%), 32 (4%) keV X-rays, and the 35 (7%) keV gamma-rays following after 100% electron capture, and it also emits conversion and Auger electrons (Lagoutine et al., 1984). The I-125 is a major diagnostic tool that is widely applied in clinical tests and in particular to the diagnosis of thyroid disorders. Auger electrons of I-125 radionuclide represent an appropriate alternative to beta emitters for cancer therapy, in particular, if they can be placed close to nuclear DNA (Bodei et al., 2003). With their ability to deposit energy in extremely small volumes, they also serve as attractive probes of radiobiological phenomena (Adelstein et al., 2003). The I-125 seed implantation has been successfully applied in radiation therapy as brachytherapy to treat several types of tumors, including uveal melanomas, prostate cancer, brain tumors, rectal carcinoma (Wang et al., 2011b), advanced pancreatic cancer (Zhongmin et al., 2010), and non-small cell lung cancer (Martínez-Monge et al., 2008; Wang et al., 2011a). Moreover, I-125 is also used for photon detector calibration at the low energy scale. Hence, there is a need for accurate radioactivity measurements of reference sources. In general, photon-photon coincidence counting represents a cheap and efficient technique for the direct measurement of nuclear transitions occurring per unit of time if an appropriate electronic coincidence measuring system is available in a laboratory. In this paper, a 2D photon-photon coincidence system was developed based on a CAEN digitizer and two NaI(Tl) detectors for absolute activity measurement.

2 Standardization method

The theory and standardization method of photon-photon coincidence counting for activity measurement of radionuclides are described by many authors. This method is particularly applied for the standardization of I-125 using X-ray detection and gamma-ray coincident counting (Taylor, 1967; Eldridge and Crowther, 1964; Horrocks and Klein, 1975; Martin and Taylor, 1992).

In the case of radionuclides such as I-125 and Hg-197 unlike gamma-gamma coincidence counting at higher energies, X-X and X-gamma coincidence counting does not require corrections for angular correlations or for Compton scattering (due to the low energies of both X-rays and gamma-rays in I-125 and Hg-197, only photopeak is visible) (Taylor, 1967). The details of the theory of the used formulas for the activity calculation have been discussed by (Taylor, 1967). Following the reference (Taylor, 1967), the nuclear transitions occurring per unit time of I-125 (N_0) can be written as:

$$N_0 = \frac{4K}{(1+K)^2}\left[N_1 + \frac{N_c(1-\frac{N_c}{2}N_2)}{2(1-\frac{N_c}{2}N_1)}\right] \times \left[N_2 + \frac{N_c(1-\frac{N_c}{2}N_1)}{2(1-\frac{N_c}{2}N_2)}\right]\frac{1}{2N_c} \tag{1}$$

where N_0, N_i, and N_c are the disintegration rate of radionuclides, the counting rate in channel i ($i = 1, 2$), and the coincidence rate, respectively. In the case of radionuclides that emit coincident gamma-rays of higher energies, in particular for Co-60, the equations are slightly different. The first difference between the description of Co-60 and I-125 decays appears due to the Compton scattering contribution. Another difference relates to the careful account of the coincident events. Following the approach that has been presented in (Volkovitsky and Naudus, 2009), formulas for the coincident count rate in the two detectors for calculation of the activity of the Co-60 source will be as:

$$N_0 = \sqrt{\bar{N}_0 \widetilde{N}_0} \tag{2}$$

where

$$\bar{N}_0 = \frac{\left(N_1^{(2,p)}N_2^{(2,p)} - N_{1c}^{(2,p)}N_{2c}^{(2,p)}\right)\left(N_1^{(1,p)}N_2^{(1,p)} - N_{1c}^{(1,p)}N_{2c}^{(1,p)}\right)}{N_c^{(1,2)}\left(N_1^{(2,p)} - N_{1c}^{(2,p)}\right)\left(N_2^{(1,p)} - N_{2c}^{(1,p)}\right)} \tag{3}$$

and

$$\widetilde{N}_0 = \frac{\left(N_1^{(2,p)}N_2^{(2,p)} - N_{1c}^{(2,p)}N_{2c}^{(2,p)}\right)\left(N_1^{(1,p)}N_2^{(1,p)} - N_{1c}^{(1,p)}N_{2c}^{(1,p)}\right)}{N_c^{(2,1)}\left(N_1^{(1,p)} - N_{1c}^{(1,p)}\right)\left(N_2^{(2,p)} - N_{2c}^{(2,p)}\right)} \tag{4}$$

where N_0 is the disintegration rate of Co-60 into the channel with two coincident gamma-ray and $N_i(j,p)$ is the count rate in the detector i ($i = 1, 2$) of events in the photopeak of the gamma-ray j ($j = 1, 2$).

$N_c^{(1,2)}$ is the count rate of coincident events for which the first gamma-rays is detected by the first detector and the second gamma-ray is detected by the second detector. Similarly, $N_c^{(2,1)}$ is the count rate of coincident events for which the second gamma-ray is detected by the first detector and the first gamma-ray is detected by the second detector. $N_{1c}^{(1,p)}$ is the count rate of coincident events in the first detector when the first gamma-ray is detected in the area of the photopeak and the second gamma-ray with any energy is detected by the second detector. $N_{1c}^{(2,p)}$ is

Figure 1: Block diagram of a photon-photon coincidence counting system.

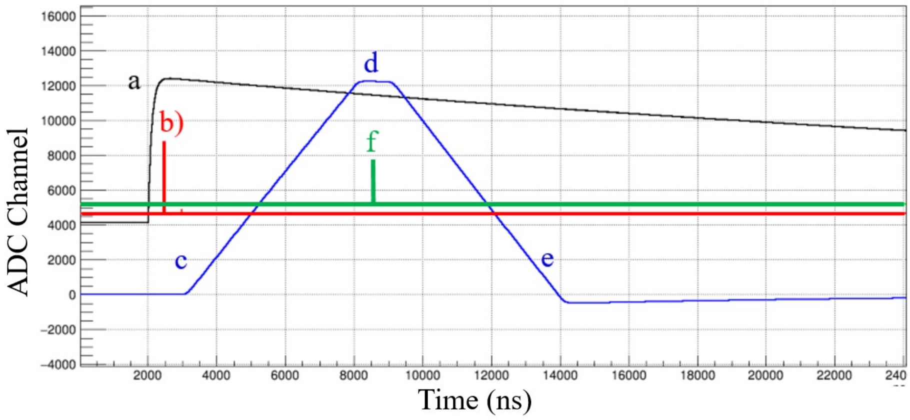

Figure 2: The digital oscilloscope: a) the dector signal, b) time tigger c) trapezoid rise-time, d) flat-top time, e) decay time, f) peaking time.

the count rate of coincident events in the first detector when the second gamma-ray is detected in the area of the photopeak and the first gamma-ray with any energy is detected by the second detector. $N_{2c}^{(1,p)}$ is the count rate of coincident events in the second detector when the first gamma-ray is detected in the area of the photopeak and the second gamma-ray with any energy is detected by the first detector. Similarly, $N_{2c}^{(2,p)}$ is the count rate of coincident events in the second detector when the second gamma-ray is detected in the area of the photopeak and the first gamma-ray with any energy is detected by the first detector.

3 Experimental setup

The block diagram of a photon-photon coincidence counting system is presented in Fig. 1. The data acquisition system for photon-photon coincidence counting was set up by two similar $2'' \times 2''$ NaI(Tl) detectors. The detectors were installed in front of each other and the source was placed on their common axis at a distance of 1 cm from the detectors. The acquisition system was established based on a 14-bit waveform digitizer (CAEN DT5724) that samples directly from the preamplifier output of the two NaI(Tl) detectors. The digital acquisition system was used to record the data in time-stamped list mode. In this configuration, all the nuclear electronic modules are omitted and the system is loaded with a Pulse Height Analyzer (PHA) firmware. The setting of the time trigger and trapezoidal energy filter were done by software (Caen, 2022). Figure 2 shows the digital oscilloscope of the digitizer. The sampled signal of the detector (Fig. 2-a) transforms into a trapezoidal signal (Fig. 2-c) based on the Jordanov algorithm (Jordanov and Knoll, 1994). The height of the flat-top region (Fig. 2-d) at the peaking time window (Fig. 2-f) reveals the energy of gamma rays. The advantages of digital signal processing for gamma ray spectroscopy compared to analog spectroscopy have been discussed in our previous paper (Biganeh and Kakuee, 2021). The calibration of the energy spectrum was carried out using gamma lines of Ba-133 (276 keV), Cs-137 (662 keV), and Co-60 (1332 keV). To obtain the optimum value for the coincidence time window, the list mode data of detectors were collected in pseudo coincidence mode (2 μs timing window). Then, the FWHM of the timing spectrum was measured and the value of 80 ns (twice the FWHM) was selected as an optimum coincidence time window. Pulses from both detectors were recorded in coincidence and normal modes. After the correct configuration of the setting for each channel of the digitizer, the MC2 software records the energy and the time stamp of all detected events in two detectors and stores the energy and time of each event in a text file (Caen, 2022). The problem that arises during data acquisition is the record of double coincidence events on the list data due to the large coincidence time window. To overcome this problem, the trigger hold-off parameter was set

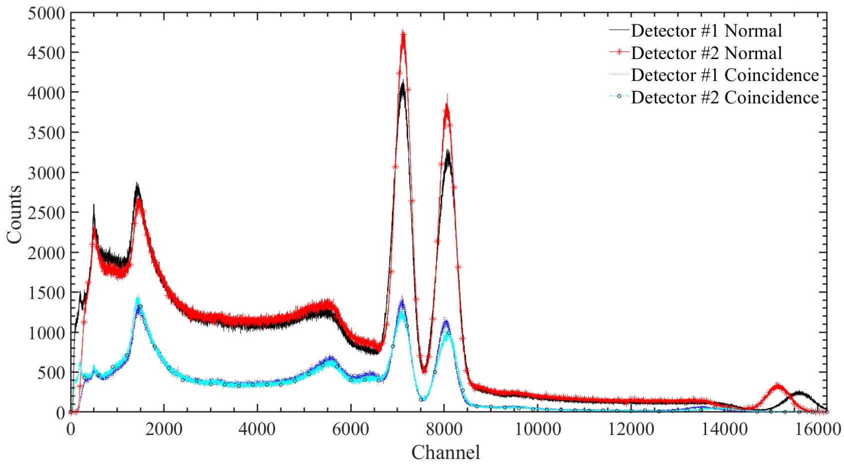

Figure 3: One-dimensional energy spectra of Co-60 source measured by the two 2″ × 2″ NaI(Tl) detectors in coincidence and normal modes.

Table 1: The results of the calculated activities for two Co-60 sources by photon-photon coincidence counting method.

A_0: **Reference activity (kBq)**	5.87 ± 0.17	13.6 ± 0.40
$N_1^{(1,p)}$	2196285 ± 1481	2182069 ± 1477
$N_1^{(2,p)}$	1857770 ± 1363	1877626 ± 1370
$N_2^{(1,p)}$	2064117 ± 1436	2060040 ± 1435
$N_2^{(2,p)}$	1761371 ± 1327	1730309 ± 1315
$N_{1c}^{(1,p)}$	637822 ± 798	522498 ± 722
$N_{1c}^{(2,p)}$	541643 ± 735	447664 ± 669
$N_{2c}^{(1,p)}$	635791 ± 797	520614 ± 721
$N_{2c}^{(2,p)}$	521578 ± 722	436976 ± 661
$N_c^{(1,2)}$	186260 ± 431	146410 ± 382
$N_c^{(2,1)}$	188514 ± 434	147779 ± 384
Measurement time (s)	5103	2709
A: **Calculated activity (kBq)**	6.76 ± 0.20	14.80 ± 0.31
A/A_0	1.14	1.08

greater than twice the coincidence time window (Biganeh and Shakeri Jooybari, 2022).

4 Results and discussion

The performance of the system for absolute activity measurement was tested by two Co-60 standard sources. Pulses from both detectors are recorded in coincidence and normal modes. The activity of the reference sources at the date of measurements was 13.6 kBq and 5.87 kBq. For these measurements, the time of measurement was set to be 2709 s. The spectrum of the weaker Co-60 source was recorded for 5103 s. The energy spectra of the two detectors in coincidence and normal modes are presented in Fig. 3. Assuming that E1 and E2 are the recorded energy by detectors #1 and #2, respectively. A 15000 × 15000 matrix array of E_1 and E_2 events was built-up. Figure 4 shows the 2D coincidence spectrum of the gamma rays. In this spectrum, the width of each channel is 165 eV. The circles at the minor diagonal array of the matrix are photopeak-photopeak events that are recorded free from uninterested events such as Compton scattering, incomplete charge collection, and pile-up events. The ridges parallel to the 1173 and 1332 keV axes originate from the coincidence events between a photopeak in one detector and a Compton scattered gamma-ray in the other detector. The chance summing coincidence of the two 1173 keV gamma-rays and the Compton edge events are also marked in Fig. 4. The summing coincidence at higher energies (2×1332 keV and $1173 + 1332$ keV) is ignored due to the limitations in the Random-Access Memory (RAM) of our computer. The recorded events in the 2D and 1D spectra were analyzed offline by a homemade Matlab code. The determination of the total counts of the peaks for substitution in Eqs. (3) and (4) were also performed by the Gamma Vision software via the total summation method. This peak area calculation maintains precision as the peak gets smaller, is less sensitive to the random fluctuation in the data and is less sensitive to the differences between the spectrum peak shape and the calibrated peak shape. The details of the calculations have been presented in (ORTEC, 2000).

Table 1 shows the activities of the sources that are calculated using Eq. (2) for the two standard sources. As listed in Table 1, the measured and reference source activity ratio (A/A_0) values are close to unity. The uncertainty in the results origin from the statistical error, the uncertainty of the reported reference activity values (about 2%), and the muon cosmic radiations (Biganeh and Kakuee, 2021). Further to this work, we are going to design a muon shielding veto system using an 8-channel digitizer (DT5730B) to decrease the error caused by cosmic radiations.

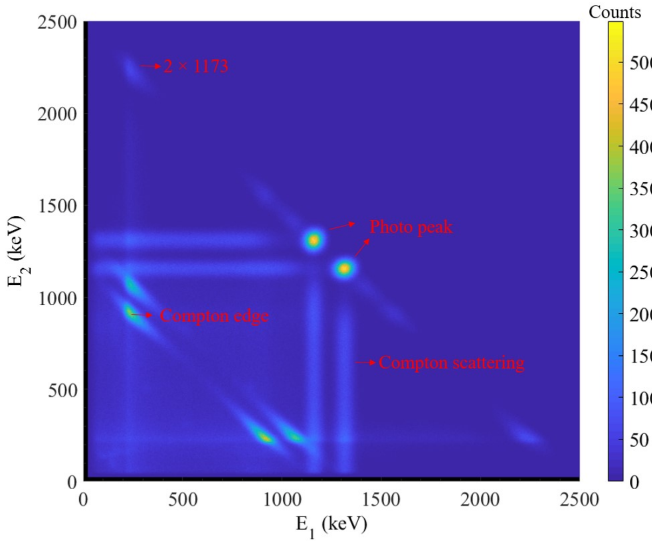

Figure 4: The two-dimensional energy spectrum of the coincident events of the Co-60 source.

5 Conclusions

The activity of a radionuclide can be calculated by applying different techniques. The choice of the method depends on the decay scheme of the radionuclide. In the present paper, we developed a 2D photon-photon coincidence digital system for absolute activity measurement by two NaI(Tl) detectors. The system was established based on digital signal processing. Digital data acquisition system allowed event-by-event time-stamped data collection. Data analysis was performed offline using MATLAB software and the activity of the two point sources of Co-60 was measured. The measured activities found to be in good agreement with the reference values. Under well-chosen measuring conditions, photon-photon coincidence counting shows compatible results for Co-60 in comparison with other methods. In general, this technique represents a cheap and impressive activity standardization method for all cascade gamma ray emitter radioisotope sources and can fill the gap between the existence of technology and its application.

Conflict of Interest

The authors declare no potential conflict of interest regarding the publication of this work.

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