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# Coincidence system for standardization of radionuclides using a 4π plastic scintillator detector

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## Abstract

A coincidence system using a plastic scintillator detector in 4π geometry has been developed and applied for the standardization of radionuclides. The scintillator shape and dimensions have been optimized for maximum charge particle detection efficiency, while keeping background low and a nearly constant gamma-ray efficiency for different points from the radioactive source. The gamma-ray events were measured with a NaI(Tl) scintillation counter. The electronic system for processing pulses consisted of logic gates and delay modules feeding a time-to-amplitude converter with output to a multichannel analyzer. The alpha detection efficiency measured with $^{241}$Am was around 95% and the beta detection efficiency for $^{60}$Co was around 67%. Activity measurements of $^{241}$Am and $^{60}$Co were performed and the results showed good agreement when compared with a conventional coincidence system employing a 4π proportional counter.

*Keywords:* Plastic scintillator; Americium-241; Cobalt-60; Coincidence; Standardization

## 1. Introduction

For many years, $4\pi(\alpha,\beta,X)$–$\gamma$ coincidence systems have been considered primary standards due to high accuracy and because the results depend only on observable quantities. The validity of the coincidence method enforces strict conditions for the experimental setup, as high efficiency for one type of radiation (e.g. alpha or beta) and insensitivity to the other type of radiation (e.g. gamma). The basic method is used for simple decay schemes, and can be extended for complex decay schemes by applying the linear extrapolation technique (Baerg, 1973).

The Laboratório de Metrologia Nuclear of IPEN, founded in 1964, has developed several radionuclide standardization systems (Dias and Koskinas, 1997; Fonseca, 1997; Fonseca et al., 2001; Hilario, 2002; Koskinas, 1988; Lavras et al., 2001; Moura, 1969; Simões, 2001) and participated in several international comparisons sponsored by the *Bureau International des Poids et Mesures, France*. The present detector system (Baccarelli et al. 2001) is intended to perform primary calibration without need of a coating metal layer on the radioactive source film in order to render it conductive.

Two radionuclides, namely $^{241}$Am and $^{60}$Co, were chosen for testing the system. These radionuclides can be easily standardized by the conventional $4\pi\beta$(PC)-$\gamma$ coincidence system employing a gas flow proportional counter. $^{241}$Am decays by alpha emission with 432.2 yr half-life, promptly followed by a 59.537 keV gamma-ray. $^{60}$Co decays by $\beta^-$ emission with 5.271 yr half-life, followed by two gamma-rays with 1173.239 and 1332.503 keV, respectively. The latter radionuclide is particularly interesting because it combines a low-energy

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beta-ray with a high-energy gamma-ray. Therefore, the contribution of these gamma- rays in the beta detector counting rate may be significant.

## 2. Experimental procedure

### 2.1. Detector description

Different plastic scintillator shapes and designs were tried in order to approximate 4π geometry, to ease the radioactive source insertion and removal and to allow the use of simple electronics. The best results were obtained with a double plastic cylindrical scintillator. Its lower part is 6 mm thick and has a coaxial depression of 3 mm where the radioactive source is placed (Fig. 1). The upper part is 3 mm thick and is used as a lid to improve light collection efficiency and to assure 4π geometry.

The scintillator was wrapped with Teflon tape for producing diffused light reflection (Fig. 1). The lower end was coupled to a RCA 8850 photomultiplier tube by means of silicone grease. The whole set was coupled to a $3^{in} \times 3^{in}$NaI(Tl) scintillator counter for gamma- detection from $^{60}$Co, and with a $2^{in} \times 2^{in}$NaI(Tl) scintillation counter for the case of $^{241}$Am. The smaller crystal was used in the latter case in order to reduce room background.

### 2.2. Source preparation

The sources were prepared by depositing quantitative aliquots of radioactive solutions onto a 20 μg/cm$^2$ thick Collodion film, previously coated with 10 μg/cm$^2$ gold layer. This gold layer is not necessary for the detection system proposed in this paper and was applied only to allow comparisons with a conventional 4π proportional counter coincidence system where this procedure is necessary. The accurate source mass determination was performed by the pycnometer technique using a Mettler 5SA balance (Campion, 1975). A seeding agent (Cyastat SN) was used to improve the deposit uniformity and the sources were dried in a desiccator. This procedure avoids crystal growing that may lead to self-absorption of the source. The Collodion film was held by a stainless-steel ring, 20 mm in external diameter and 10 mm in internal diameter, which proved to be the best holder material for this application. Transparent plastic rings were tested and showed better light collection efficiency. However, the geometric efficiency turned out to be lower because of the greater ring thickness.

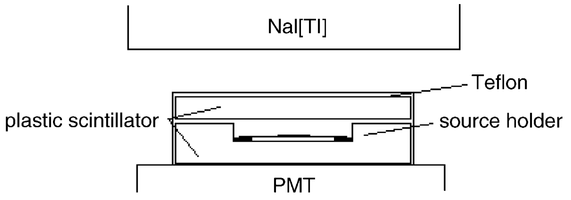

Fig. 1. Coincidence system using 4π plastic scintillator coupled to a NaI(Tl) crystal.

### 2.3. Typical spectra

Typical alpha and beta spectra are shown in Figs. 2 and 3, respectively. In Fig. 2, the alpha spectrum from $^{241}$Am, shows two peaks. The one to the right corresponds to pulses produced by the lower scintillator while the one to the left corresponds to pulses produced by the upper scintillator. In Fig. 3, the beta spectrum from $^{60}$Co shows a high tail in the region of low-amplitude pulses. This shape indicates that many low-energy beta-rays produce pulses in the noise region. As a result the maximum beta-ray efficiency for $^{60}$Co is lower as compared with a 4π proportional counter.

The electronic system diagram is shown in Fig. 4. The pulses, after being amplified and discriminated, were fed into gate and delay modules. The logic pulses were sent

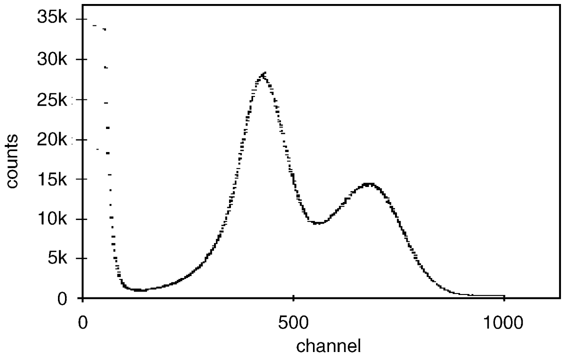

Fig. 2. Typical $^{241}$Am spectrum from 4π plastic scintillator.

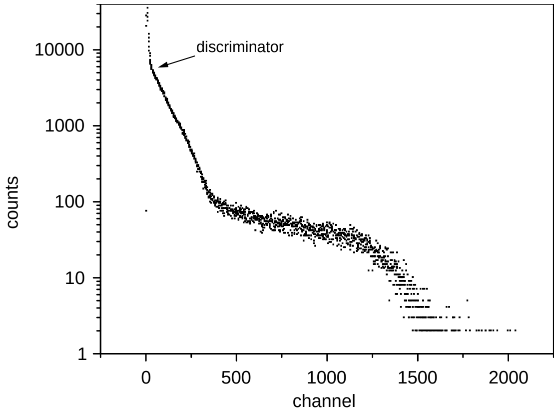

Fig. 3. Typical $^{60}$Co spectrum from 4π plastic scintillator.

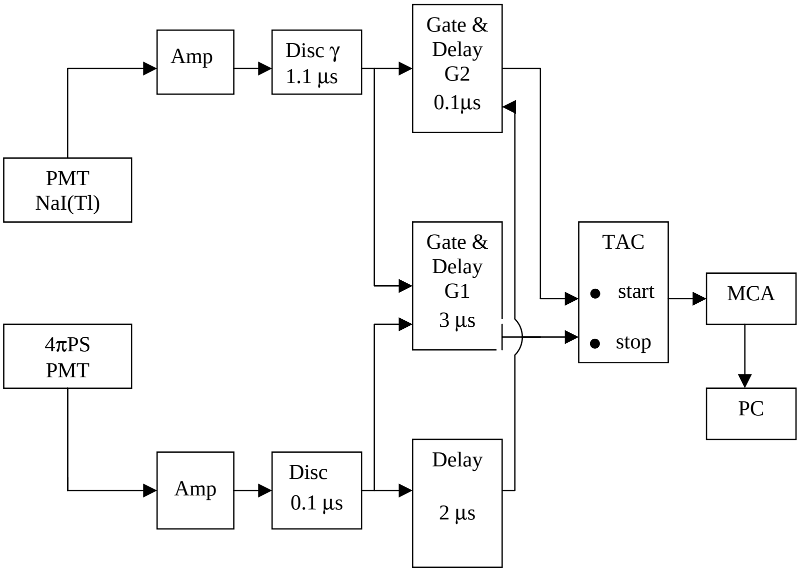

Fig. 4. Schematic diagram of the electronic system.

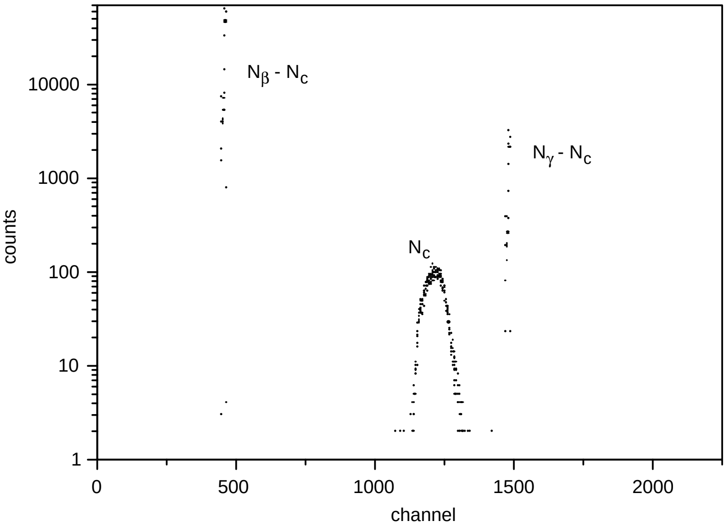

Fig. 5. Typical TAC spectrum.

to a time-to-amplitude converter (TAC) and then to a multichannel analyzer (MCA). This system eliminates the need of individual counters for each detection line.

The plastic scintillator pulses were discriminated to cut off noise. The gamma-ray window was set up to accept only total energy absorption pulses from $^{241}$Am or $^{60}$Co gamma-rays, in order to eliminate gamma–gamma coincidence.

A typical TAC spectrum registered in the MCA is shown in Fig. 5. The central peak corresponds to coincidences. The left and right peaks correspond to alpha or beta and gamma counting rates, respectively, without the coincidence rate contribution. The activity and efficiency parameters were obtained using code CONTAC (Dias, 2001) which makes corrections for decay, background, resolving time and dead time.

### 2.4. Coincidence method

The value of source activity $N_0$ was obtained by means of the generalized coincidence formula

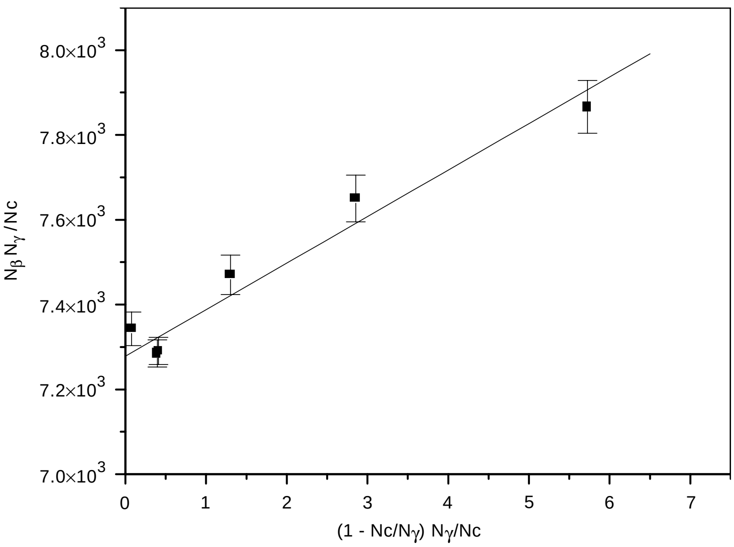

Fig. 6. Extrapolation curve of $N_\beta N_\gamma/N_c$ as a function of $(\mathbf{1}-N_c/N_\gamma)/(N_c/N_\gamma)$ for $^{241}$Am.

(Baerg, 1073):

$$\frac{N_\beta N_\gamma}{N_c} = N_0 G\left[\frac{1-N_c/N_\gamma}{N_C/N_\gamma}\right], \tag{1}$$

where $N_\beta, N_\gamma$, $N_c$ are the beta, gamma and coincidence counting rates respectively. $\boldsymbol{G}$ is a function of the efficiency parameter

$$\left[\frac{1-N_c/N_\gamma}{N_c/N_\gamma}\right]$$

which goes to zero when $N_c/N_\gamma$ approaches unity. In this extrapolation limit, the left side of Eq. (1) gives the radioactive source activity. Several measurements have been performed using external absorbers. The alpha and beta efficiency was changed by using 0.080 mm thick Makrofol external foils over and under the source.

### 2.5. *Beta–gamma efficiency determination*

The gamma efficiency of the beta detector ($\varepsilon_{\beta\gamma}$—beta–gamma efficiency) was measured for the gamma energies involved. For 59 keV determination, a point 7.272 kBq $^{241}$Am source was used, covered with a plastic layer 0.4 mm thick to absorb alpha particles. For 1.25 MeV (averaged) gamma-ray, a 3 mm long wire having 445 kBq of $^{60}$Co was used, wrapped inside a 0.4 mm aluminum foil in order to absorb beta rays. The beta-gamma efficiency was calculated by

$$\varepsilon\beta\gamma = \frac{S}{tI\gamma A} \tag{2}$$

where: $S$ are the counts above discriminator threshold; $I_\gamma$ is the gamma ray probability per decay; $t$ is the MCA live time and $A$ is the source activity measured by a calibrated HPGe spectrometry system.

## 3. Results and discussion

The source activity was obtained by linear least-square fitting between $N_\beta N_\gamma/N_c$ as a function of

$$\left[\frac{1-N_c/N_\gamma}{N_c/N_\gamma}\right]$$

using code LINFIT (Dias, 1999), which incorporates covariance matrix methodology and takes into account all partial errors involved. Figs. 6 and 7 shows a plot of the $N_\beta N_\gamma/N_c$ versus

$$\left[\frac{1-N_c/N_\gamma}{N_c/N_\gamma}\right]$$

for $^{241}$Am and $^{60}$Co, respectively. The curve slopes resulted (1.11 ± 0.59)% for $^{241}$Am and (4.32±0.13)% for $^{60}$Co.

The alpha detection efficiency measured with $^{241}$Am resulted around 95% and was considered satisfactory. The observed beta efficiency was rather low due to light absorption in the gold layer applied to the Collodion film, in order to render it conductive, for comparison with the conventional 4πβ(PC)–γ counter. Additional measurements performed without any gold layer showed maximum beta efficiencies around 67% for $^{60}$Co. New measurements are planned using two photomultiplier tubes coupled to the 4π plastic scintillator counter, in order to improve the light collection efficiency.

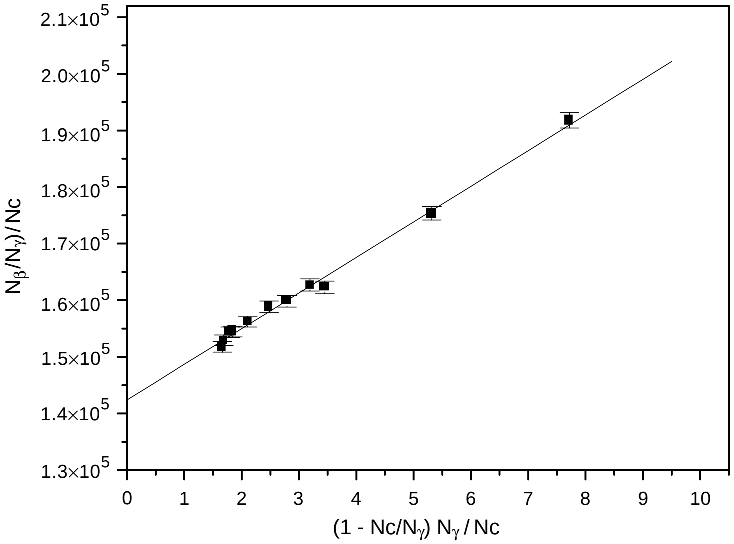

Fig. 7. Extrapolation curve of $N_\beta N_\gamma/N_c$ as a function of $(\mathbf{1}-N_c/N_\gamma)/(N_c/N_\gamma)$ for $^{60}$Co.

Table 1
Activity values obtained with conventional and present systems

| Radionuclide | System | Activity (kBq/g) |
|---|---|---|
| $^{241}$Am | 4πβ(PC)–γ | (7.272±0.009) |
| $^{241}$Am | 4πβ(PS)–γ | (7.276±0.012) |
| $^{60}$Co | 4πβ(PC)–γ | (142.72±0.29) |
| $^{60}$Co | 4πβ(PS)-γ | (142.42±0.54) |

The resulting 59 keV beta-gamma efficiency was (0.152±0.017)% and for 1.25 MeV ($^{60}$Co) the result was (4.90±0.64)%. The value for $^{60}$Co are in good agreement with the slope of the extrapolating curve of Fig. 7. In the case of $^{241}$Am, the beta–gamma efficiency is much lower than the slope in Fig. 6. This difference may be due to detection of internal conversion electrons from $^{241}$Am daughter decay.

The results of activities obtained in the present system are compared to a conventional system 4πβ(PC)–γ, in Table 1. It can be seen that they are in excellent agreement within the experimental uncertainty.

Measurements of radionuclides which decay by electron capture and positron emission, will be performed in near future for validating the proposed system.

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