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Absolute measurement of ^{242g}Am sources activities in the ^{241}Am(n, γ) cross-section determination—Improvement by simulation

Nora L. Maidana[a,*], Mauro N. Takeda[b], Mauro S. Dias[b], Marina F. Koskinas[b], Vito R. Vanin[a]

[a]*Laboratório do Acelerador Linear, Instituto de Física, Universidade de São Paulo, Travessa R 187, Cidade Universitária, CEP: 05508-900 São Paulo, SP, Brazil*
[b]*Laboratório de Metrologia Nuclear, Instituto de Pesquisas Energéticas e Nucleares, IPEN-CNEN/SP Travessa R 400, Cidade Universitária, CEP: 05508-900 São Paulo, SP, Brazil*

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Abstract

A calibration system composed of a 4πβ-γ gas-flow proportional counter coupled to a pair of NaI(Tl) crystals operating in coincidence was used for absolute measurement of ^{242g}Am sources activities. The samples were produced by irradiating dried aliquots of ^{241}Am solution with epithermal neutrons in a research reactor. Efficiency tracing methodology using ^{60}Co as tracer was applied. Thick external absorbers were used to change the ^{242g}Am beta detection efficiency and to avoid counting alpha particles from the dominating activity of ^{241}Am. The resulting complex behavior of the beta detection efficiency curve was simulated by the Monte Carlo method, and the ^{242g}Am activity was obtained by least-squares method fitting. The activity was determined with an uncertainty of about 1%.

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1. Introduction

Successive neutron capture reactions can produce new elements in a nuclear reactor, in particular yielding a relatively high amount of americium and curium at high fuel burning rates. These isotopes must be considered in reactivity calculations, requiring the respective activation cross-section to evaluate their contribution to the overall radioactivity balance. This paper concerns the source standardization of one of ^{241}Am

*Corresponding author.
E-mail address: nmaidana@if.usp.br (N.L. Maidana).

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thermal neutron capture products, the radionuclide ^{242g}Am, whose accurate induced activity measurement is needed to determine experimentally the ^{241}Am neutron capture cross-section [1,2].

The measurements were taken with a system [3] composed of a 4π gas-flow proportional counter coupled to a pair of NaI(Tl) crystals operating in coincidence at the Laboratório de Metrologia Nuclear, IPEN (Nuclear Metrology Laboratory, Nuclear Energy Research Institute), in São Paulo, Brazil. Systems and procedures used here were also applied to other radionuclide standardization works [4,5] and activation cross-section measurements [6].

The special difficulty in standardizing ^{242g}Am, obtained from ^{241}Am irradiation in a reactor, by the tracer method in 4πβ-γ systems comes from the dominating ^{241}Am radioactivity, whose alpha particles are detected along with the electrons from ^{242g}Am β^- decay. The efficiency tracing methodology usually requires sources deposited on collodion films and thin electron absorbers to change the beta detection efficiency in a region near the extrapolation point. In this measurement, thick absorbers were required to block the alpha particles; hence, the experiment was performed with low β^- detection efficiencies, an undesired situation that can lead to ambiguities in the extrapolation to 100% electron detection efficiency, as can be seen in recent work [7]. To overcome this problem, an appropriate functional relationship between the coincidence and single efficiencies was determined by Monte Carlo simulation [8] that was successfully applied to this research.

2. Experimental method

The experiment consisted in the observation of the ^{242g}Am β^- decay radiation and ^{241}Am α radiation from specially prepared sources. The ^{242g}Am was obtained by the irradiation of ^{241}Am in a research reactor. As the ^{242g}Am gamma-ray emission probabilities are very low, it is not possible to apply the 4πβ-γ coincidence method; thus, the activity was determined using the tracer technique [9] employing ^{60}Co as tracer. Since the sample radioactivity was dominated by the alpha particles from ^{241}Am, then thick removable absorbers were used to allow the observation of electrons from ^{242g}Am β^- decay.

2.1. Decay schemes and observed radiations

The radionuclide ^{242g}Am decays by β^- emission with 665 keV end-point energy and by electron capture with probabilities 82.7(3)% and 17.3(3)%, respectively, as shown in Fig. 1 [10]. The electromagnetic transitions from the excited states of the daughter nuclides ^{242}Pu and ^{242}Cm, with energies 44.54(2) and 42.13(1) keV, respectively, have high internal conversion coefficients. For this reason, the gamma-ray emission probabilities per decay in both cases are negligible, making it a suitable radionuclide to be standardized by the tracer technique [9]. The radionuclide ^{60}Co was chosen as tracer because of its simple decay scheme and small internal conversion coefficients.

The ^{241}Am decays by alpha particle emission followed by characteristic gamma rays of energies 26.345(1) and 59.537(1) keV and emission probabilities equal to 2.4(1)% and 35.9(4)%, respectively, as shown in Fig. 1 [10].

2.2. Applying the tracer method

In a 4πβ-γ coincidence experiment with a mixed source consisting in a β^--emitter nuclide combined with a β-γ tracer, the electron detector counting rate depends on the detection efficiencies for both the β^--emitter and the tracer. The tracer beta detection efficiency, related to the gamma, beta and coincidence counting rates, can be associated with the β^--emitter detection efficiency [9]. Specifically, the mixed source counting rates in the detector system are related by

$$\frac{N_\beta N_\gamma}{N_c} = N_{0Co} + N_{0Co}\frac{(1-\varepsilon_{\beta Co})}{\varepsilon_{\beta Co}}\left(\frac{\alpha_{Co}}{1+\alpha_{Co}}\right)\varepsilon_{CECo} + N_{0\beta Am}\frac{\varepsilon_{\beta Am}}{\varepsilon_{\beta Co}} + N_{0\beta Am}\frac{(1-\varepsilon_{\beta Am})}{\varepsilon_{\beta Co}} \times\left(\frac{\alpha_{Am}}{1+\alpha_{Am}}\right)\varepsilon_{CEAm} \quad (1)$$

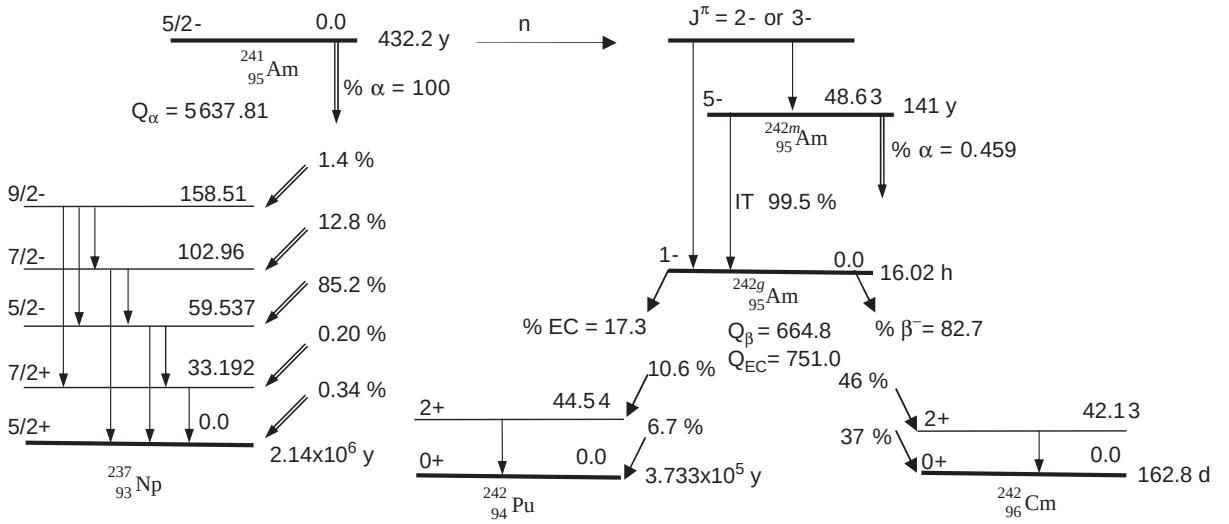

Fig. 1. ^{241}Am(n, γ)^{242g}Am reaction and decay schemes [10], with energies in keV. Only the main transitions are represented.

where N_β, N_γ, N_c are the beta, gamma and coincidence counting rates, respectively, corrected for dead time and chance coincidence; N_{0Co}, $N_{0\beta Am}$ the ^{60}Co and ^{242g}Am β^- branch disintegration rates, respectively; $\varepsilon_{\beta Co}$, $\varepsilon_{\beta Am}$ the beta detection efficiencies of the tracer and the ^{242g}Am β^--branch, respectively; α_{Co}, α_{Am} the ^{60}Co and ^{242g}Am electron conversion coefficients, respectively; ε_{CECo}, ε_{CEAm} the conversion electron detection efficiencies for ^{60}Co and ^{242g}Am β^- branch, respectively.

Expression (1) assumes that there is no noticeable contribution to the counting rates from the isomer ^{242m}Am decay at the time of measurement, due to its long half-life.

The ratio $N_\beta N_\gamma / N_c$ is related to an inefficiency parameter that gives how much the used absorber reduces the efficiency. Since N_c/N_γ is proportional to the beta detection efficiency, the quantity $(1 - N_c/N_\gamma)$ goes to zero when the beta detection efficiency increases, and the ratio

$$\frac{(1 - N_c/N_\gamma)}{N_c/N_\gamma} \quad (2)$$

gives a normalized, slowly varying, measure of what is called beta detection inefficiency, a parameter that goes to zero when the efficiency goes to one.

2.3. Target preparation and irradiation

The target was made from aliquots of carrier-free ^{241}Am (40 MBq) solution dropped on a 0.2 mm thick polyethylene envelope. When the solution was completely dried, the envelope was sealed, and placed inside another 0.2 mm thick polyethylene envelope, which was also sealed.

The irradiations were carried out at the IPEN-CNEN (Nuclear Energy Research Institute, National Energy Nuclear Agency) pool-type research reactor in São Paulo, SP, Brazil, operating at 2 MW. The sample was covered with Cd foils, 0.5 mm thick, and placed inside a polyethylene rabbit, transported to the reactor core by means of a pneumatic tube and irradiated during 60 min in an epithermal neutron flux of 1.1×10^{12} cm^{-2} s^{-1}. The neutron fluence was monitored using the ^{197}Au(n, γ)^{198}Au and ^{59}Co(n, γ)^{60}Co reactions with alloy Al–Au foils and Al–Co wires containing 0.1% of gold and 0.475% of cobalt, respectively. The time interval between the end of irradiation and beginning of counting was approximately 60 min. This paper deals with data obtained using samples made from three out of several irradiated targets used to measure the ^{241}Am neutron absorption cross-section [1].

2.4. Sources preparation

Three kinds of sources were made: (a) mixed sources with aliquots of $^{241}Am + ^{242g}Am$ and of ^{60}Co; (b) pure ^{241}Am; and (c) pure ^{60}Co.

Two mixed sources were prepared from each irradiation, with the procedure described as follows. First, the $^{241}Am + ^{242g}Am$ in the target were diluted with 1 M HNO_3 solution and collected in a pycnometer. One or two drops of this dilution, containing about 6 kBq of ^{241}Am, were deposited in 20 $\mu g\,cm^{-2}$ thick collodion films, previously coated on both sides with a 10 $\mu g\,cm^{-2}$ thick gold layer to render the film conducting. Just after deposition, an amount of ^{60}Co solution was dropped, containing about half of the ^{242g}Am activity. A seeding agent (CYASTAT SN) was added to improve the deposit uniformity, and the sources were dried in a jet of warm (45 °C) nitrogen [11].

One source of pure ^{241}Am and one of pure ^{60}Co were prepared by dropping aliquots of the respective radionuclides in collodion films, followed by addition of the seeding agent, and then dried with nitrogen.

2.5. Detection systems

The 4πβ-γ absolute standardization system [3] consisted of a gas-flow proportional counter in a 4π geometry using P10 gas (90% Ar + 10% CH_4) filling at 0.1 MPa, as the electron and α-particle detector, coupled to a pair of 76 mm × 76 mm NaI(Tl) scintillation counters used as gamma-ray detectors, as shown in Fig. 2. The selected γ-channel window covered both 1173 and 1332 keV gamma-ray transitions from ^{60}Co decay. In coincidence mode, the proportional counter bias was +1520 V for $^{241}Am + ^{242g}Am + ^{60}Co$ mixed sources and +2050 V for the pure ^{60}Co source.

The same proportional counter described above was used to obtain the ^{241}Am alpha-particle activity in the mixed and pure ^{241}Am sources, with +1400 V bias.

The gamma-ray measurements to obtain the ^{60}Co activity in the mixed source and ^{198}Au and ^{60}Co activities in the neutron monitors were made with a HPGe detector (20% Intertechnique),

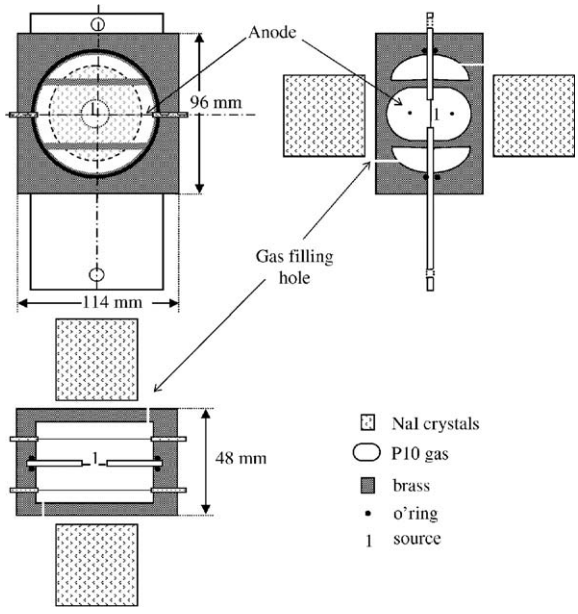

Fig. 2. 4πβ-γ detector sketches. The top and bottom left-hand drawings are the upper and side views, respectively; the right-hand drawing is the upper view after a 90° rotation around a horizontal axis through the source holder and perpendicular to the anodes. The source holder, although made of brass, is not shown shaded for clarity of the sketch.

shielded by a 10 cm-thick lead wall, using live-time counting methodology. The sources were placed in holders on the detector axis in well-defined distance, 88 mm, between source and detector capsule.

3. Measurements

In the experiment, six mixed sources and four absorbers were used in a relatively complex measurement procedure. We will identify the values obtained with a specific source and a particular absorber by subscripts i and v, respectively, like in $N_{\beta(iv)}$.

3.1. Coincidence mode in the 4πβ-γ and half-life check

The mixed sources were measured in the 4πβ-γ system using removable aluminum absorbers

9, 12, 15 and 20 mg cm^{-2} thick, placed above and below the radioactive sources to vary the beta detection efficiency. These relatively thick absorbers reduced significantly the counting of ^{241}Am alpha particles, and avoided counting conversion and Auger electrons following ^{242g}Am electron capture, making the last term of Eq. (1) negligible.

Fig. 3 shows β spectra of a mixed source and different absorbers, acquired to confirm the assumptions on the behavior of the charged particle detector; these spectra had no other use in the data analysis.

Special measurements were performed with the sole purpose of evaluating the contribution of the absorber non-uniformity to the electron detection efficiency uncertainty in the 4πβ-γ system. Many successive coincidence data were recorded using all the absorbers and one of the mixed sources after waiting more than 10 ^{242g}Am half-lives, obtaining a counting data set for each absorber. The system was opened and the absorbers manually exchanged during the measurement breaks to position the absorber relative to the source at random. The ratio of the standard deviation to the mean of each data set was used as the relative standard deviation of the electron detection efficiency for the respective absorber.

The ^{60}Co pure source was standardized in the 4πβ-γ system operating in coincidence mode, with +2050 V bias. In the following sections, this source will be called "reference ^{60}Co source", and its activity, named $N_{0\mathrm{Co}(s)}$, will be used to determine the amount of tracer.

The counting rate in the proportional counter from the irradiation product was followed during seven days in order to determine ^{242g}Am half-life, providing an additional nuclide identification. A mixed source was specially made for this experiment. The measurement, with 7.2 mg cm^{-2} external absorbers, gave 16.02(2) h [2], in close agreement with the adopted value [10].

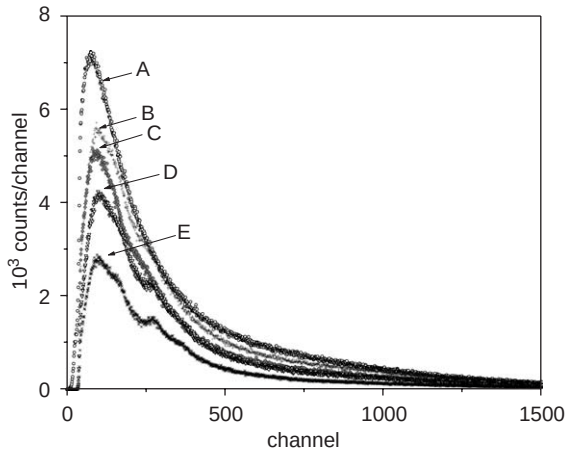

Fig. 3. Proportional counter spectra of mixed sources with 6, 9, 12, 15 and 20 mg cm^{-2} aluminum absorbers, labelled A–E, respectively.

3.2. Alpha activity and its role in counting

Fig. 4 shows the proportional counter energy spectrum of a mixed source—electrons from ^{60}Co and ^{242g}Am and alpha particles from ^{241}Am—compared to the pure ^{241}Am source, measured with an absorber of 12 mg cm^{-2}. These spectra, which were not used in the final analysis, show that not all alpha particles from ^{241}Am were blocked by the absorbers; hence, appropriate corrections had to be applied.

The number of counts in the proportional counter due to alphas from ^{241}Am decay in the mixed sources was determined from a measure-

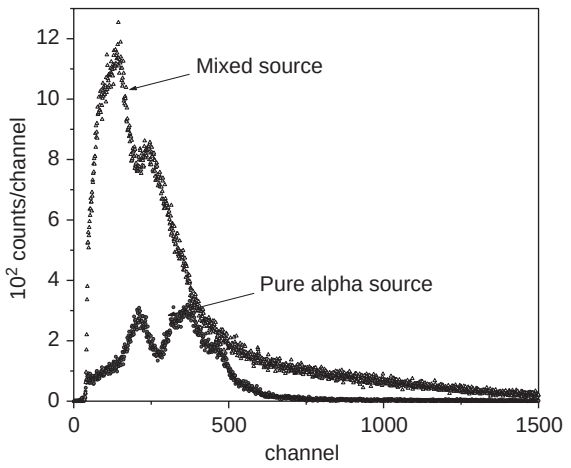

Fig. 4. Mixed and pure ^{241}Am sources spectra observed with 12 mg cm^{-2} aluminum absorber. The shape of the mixed source spectrum is somewhat different from that observed with the same absorber and source in Fig. 3, because most of the ^{242g}Am had decayed at the time of this measurement.

ment of the pure ^{241}Am source in the 4πβ-γ system using the same absorbers and voltage conditions (+1520 V bias). Since the amounts of ^{241}Am and ^{242g}Am in different mixed sources were not the same, proper normalization was required using the pure ^{241}Am source as reference. First, the alpha activity ratio of the mixed to the pure ^{241}Am sources was obtained from measurements in the proportional counter (+1400 V bias) without external absorbers and during a fixed time interval. This ratio was used as the ^{241}Am activity normalization factor,

$$D_{(i)} = \frac{\alpha_D}{\alpha_{(i)}} \tag{3}$$

where α_D and $\alpha_{(i)}$ are the number of alpha particle counts due to the reference and the mixed sources, respectively. Then, the expected contribution of ^{241}Am alpha activity to the electron detector counting of the mixed source i is given by

$$B_{(iv)} = \frac{N_{D(v)}}{D_{(i)}} \tag{4}$$

where $N_{D(v)}$ is the counting rate in the proportional counter measured with the pure ^{241}Am source and the absorber v.

3.3. Tracer activity and neutron flux normalization

The ^{60}Co activity in the mixed source i, $N_{0\mathrm{Co}(i)}$ used in Eq. (1), is given by

$$N_{0\mathrm{Co}(i)} = \frac{\mathrm{Area}_{\mathrm{Co}(i)}}{\mathrm{Area}_{\mathrm{Co}(s)}} N_{0\mathrm{Co}(s)} \tag{5}$$

where $\mathrm{Area}_{\mathrm{Co}(i)}$ and $\mathrm{Area}_{\mathrm{Co}(s)}$ are the 1173 and 1332 keV peak areas of the mixed source i and the reference ^{60}Co source in the gamma-ray spectra measured with the HPGe detector, respectively, and $N_{0\mathrm{Co}(s)}$ is the activity of the reference ^{60}Co source, previously measured with the 4πβ-γ coincidence system as mentioned above.

The neutron fluence was normalized to a reference irradiation, based on 411.8 keV total energy absorption peak areas per unit mass from Al–Au monitor, $\mathrm{Area}_{\mathrm{Au}(h)}$, where the subscript h corresponds to one of the irradiations. Calling r the reference irradiation, the neutron fluence normalization factor is

$$F_{(i)} = \frac{\mathrm{Area}_{\mathrm{Au}(h)}}{\mathrm{Area}_{\mathrm{Au}(r)}} E_{(h)} \tag{6}$$

where $E_{(h)}$ is the correcting factor for decay and counting time. An analogous factor was obtained with the Al–Co fluence monitors, using the 1173 and 1332 keV total energy absorption peak areas of ^{60}Co, in order to check the normalization factor obtained with the Al–Au, taking into account the different neutron cross-sections. The neutron fluences obtained from both monitors were in good agreement; therefore, the most precise values from Eq. (6) were used.

4. Experiment simulation and data analysis

Usually, the absolute activity determination relies on the extrapolation to 100% beta detection efficiency of Expression (1). Due to the ensuing low electron detector efficiency, the extrapolation method would require guidance from detector efficiency simulation to find the appropriate extrapolation function. In this case, however, we preferred to simulate the counting rates with the different absorbers—and consequent inefficiencies—effectively used and then fitted the available experimental counting rates, leaving the activity as the free fitting parameter. The fit was performed by the least-squares method, considering the correlations arising from the experimental procedure.

4.1. Monte Carlo simulation

The behavior of Eq. (1) was simulated by the Monte Carlo technique. The response of the 4πβ-γ coincidence system was calculated with code MCNP-4C [12] considering all detector materials, geometry and dimensions. The absorbed energy spectra were obtained from 30 eV up to 3 MeV for electrons in the 4πβ proportional counter and from 3 keV to 4 MeV for gamma rays in the NaI(Tl) scintillator. From these spectra, a reference library for monochromatic electrons and gamma quanta was created.

The code ESQUEMA [8,13] was developed to simulate the $^{60}Co + {}^{242g}Am$ mixed source coincidence experiment. The Fermi theory was used to evaluate the ^{60}Co and ^{242g}Am beta spectra. The kinetic energy of electrons coming out the source holder was determined assuming isotropic emission and using range-energy tables from the literature; this was done to simplify the procedure and avoid building a multitude of different electron response function tables, which is a time-consuming task to be performed by MCNP code for each pair of absorbers. All transitions from the parent to the daughter radionuclide were followed tagging each one and checking whenever a coincidence event occurs. As a result, code ESQUEMA provides the behavior of the observed activity in function of the beta efficiency, as given by Eq. (1).

4.2. Collecting the experimental data to apply the method

The experimental data required corrections to apply Eq. (1). Moreover, the use of the same absorber with different sources and the use of the same source with different absorbers generated statistical correlations between the corrected experimental values. In the formula below, accidental coincidences and dead time in the quantities $N_{\beta(iv)}$ and $(N_\gamma/N_c)_{(iv)}$ were already taken into account, with almost negligible changes.

Eq. (1) does not take into account the ^{241}Am alpha activity. Therefore, this alpha background must be subtracted from the electron detector counting rate, giving the net beta counting rate from $^{60}Co + {}^{242g}Am$, $N_{\beta(iv)}$:

$$N_{\beta(iv)} = C_{\beta(iv)} - B_{(iv)} \tag{7}$$

where $C_{\beta(iv)}$ is the total beta counting rate, while $B_{(iv)}$ is defined in Eq. (4).

The value resulting from Eq. (7) corrected for decay is proportional to the ^{242g}Am activity. Further normalization for the amount of ^{241}Am in the target and the neutron fluence is required and gives the quantity to be fitted by the model:

$$y_{(iv)} = \left[N_{\beta(iv)} \left(\frac{N_\gamma}{N_c} \right)_{(iv)} - N_{0\mathrm{Co}(i)} \right] D_{(i)} F_{(i)} E_{(iv)} \tag{8}$$

where $D_{(i)}$ and $F_{(i)}$ are given by formulas (3) and (6), respectively, and $E_{(iv)}$ is the correcting factor for decay and counting time.

4.3. Activity determination and uncertainty calculation

The experimental data are shown in Table 1, arranged in order of increasing absorber thickness, with the source identification numbers in the first column. The inefficiency parameter based on the N_c/N_γ value measured with ^{60}Co appears in column 2. Column 3 shows the ratio between the experimental activity given by Eq. (8) and the corresponding Monte Carlo value per unit activity as described in Section 4.1. Each of these ratios corresponds to an estimate of $N_{0\beta\mathrm{Am}}$, the ^{242g}Am β^- disintegration rate, because the simulation procedure takes into account the effect of the absorbers; the average, $\overline{N}_{0\beta\mathrm{Am}}$, is the value that minimizes the function

$$\chi^2 = (\vec{y} - N_{0\beta\mathrm{Am}} \vec{y}_{\mathrm{MC}})^{\mathrm{T}} \mathbf{V}^{-1} (\vec{y} - N_{0\beta\mathrm{Am}} \vec{y}_{\mathrm{MC}}) \tag{9}$$

where the symbol T means matrix transposition, $\vec{y}$ is the experimental data vector arranged as in column 3 of Table 1, $\vec{y}_{\mathrm{MC}}$ is the simulated counting rate calculated for ^{242g}Am unitary activity, and $\mathbf{V}$ is the covariance matrix

$$\mathbf{V} = \mathbf{V}_{\mathrm{exp}} + \overline{N}_{0\beta\mathrm{Am}}{}^2 \mathbf{V}_{\mathrm{MC}} \tag{10}$$

where $\mathbf{V}_{\mathrm{exp}}$ and $\mathbf{V}_{\mathrm{MC}}$ stand for the experimental and simulated values variance matrices, respectively. Note that $\vec{y}$ and $N_{0\beta\mathrm{Am}}$ have the physical dimension of activity, while the $\vec{y}_{\mathrm{MC}}$ is non-dimensional, being the expected counting rate per unit of ^{242g}Am activity.

$\mathbf{V}_{\mathrm{exp}}$ is calculated from the experimental variances arranged in a matrix, $\mathbf{V}_p$, whose diagonal elements are the variances in: $N_{\beta(iv)}$, $(N_\gamma/N_c)_{(iv)}$, $N_{D(v)}$, $D_{(i)}$, $F_{(i)}$, $E_{(iv)}$, and in the beta efficiency—arising from absorber non-uniformity—calculated as described in Section 3.1. Most of these

Table 1
Inefficiency and the experimental coincidence ratio $y_{(iv)}$ divided by the corresponding Monte Carlo value per unity activity is given in the second and third columns, respectively, for each source whose identification number is in the first column. The fourth and last columns give the main relative experimental uncertainties in percent; see text for explanation and uncertainties in the other experimental quantities

Source id	Inefficiency	$\frac{y_{(iv)}}{y_{MC}}$	Tracer subtraction (%)	Replication (%)
3	2.777	888	1.0	
1	2.788	830	5.0	
2	2.824	845	1.1	3.7
4	2.826	853	1.2	
6	2.910	904	1.8	
5	2.956	974	0.5	
1	4.022	794	4.0	
2	4.127	825	0.9	
3	4.171	867	0.8	0.9
4	4.211	840	0.9	
5	4.298	875	0.5	
6	4.475	865	1.4	
1	5.664	812	3.2	
3	5.669	901	0.6	
4	5.675	857	0.8	5.5
2	5.718	847	0.7	
5	5.962	880	0.4	
6	6.079	891	1.1	
3	15.026	861	0.3	
4	15.192	848	0.4	
2	15.218	834	0.4	3.5
6	15.749	918	0.6	
5	16.011	886	0.5	

experimental quantities are statistically independent, and $\mathbf{V}_p$ was assumed to be diagonal. The covariances in $\mathbf{V}_p$ generated by: $D_{(i)}$, correlated through the common reference source activity, $<0.1\%$ relative uncertainty; $F_{(i)}$, correlated through the common reference flux monitor activity, $<0.2\%$ relative uncertainty; $E_{(iv)}$, correlated through the common half-lives with 0.1% relative uncertainty, are much smaller than the experimental variances and were neglected. The variance matrix in $\overrightarrow{y}$ is given by

$$\mathbf{V}_{\text{exp}} = \mathbf{G}\mathbf{V}_p\mathbf{G}^{\text{T}}$$

where the transformation matrix G is given by

$$\mathbf{G}_{(iv)J} = \frac{\partial y_{(iv)}}{\partial p_J}$$

with J running over all the raw experimental data. Columns 4 and 5 in Table 1 give the relative standard deviation in the experimental value, $y_{(iv)}$, due to the uncertainty in the tracer activity subtraction as expressed by Eq. (8), and in the counting rates resulting from the absorber non-uniformity, respectively.

Relative standard deviation in counting statistics is 1% in the worst case and, in the inefficiencies, always smaller than 0.6%. All the remaining causes of uncertainty result in relative uncertainties of about 0.3%: background and detection of alpha particles by the proportional counter, Formula (7); fluence, Expression (6), and activity, Expression (3), normalization factors; and decay corrections.

The uncertainties in $\overrightarrow{y}_{\text{MC}}$ came from: (1) Monte Carlo statistics, limited by the number of histories followed; (2) uncertainty in the electrons response functions calculated by MCNP for the proportional counter, and (3) uncertainty in the electron range-energy tables. The resulting relative standard deviations in $\overrightarrow{y}_{\text{MC}}$ are $0.3\%, 0.1\%$, and 0.2%, respectively; the last and the second to last quantities were calculated by changing the values in the input range-energy tables in $\pm 20\%$. The matrix $\mathbf{V}_{\text{MC}}$ was assumed diagonal, because the covariances arising from factors (2) and (3) above are much smaller than the variances.

The fitting procedure is interactive because the total variance matrix $\mathbf{V}$ depends on the fitting parameter, $N_{0\beta\text{Am}}$. However, it converges in a single iteration because the variances due to Monte Carlo calculation are much smaller than the experimental variances.

With the ^{242g}Am β^--branch disintegration rate obtained in the fit, the activity of ^{242g}Am, $\overline{N}_{0\text{Am}}$, is given by

$$\overline{N}_{0\text{Am}} = \frac{\overline{N}_{0\beta\text{Am}}}{I_\beta} \tag{11}$$

where I_β is the β^- branch decay fraction.

5. Results and discussion

The observed and simulated 4πβ-γ singles to coincidence ratio of mixed sources are shown in Fig. 5 as function of the inefficiency parameter; the overall trend was well described by the Monte Carlo simulation. The value resulting from the fit is 864(9) Bq, which substituted in Expression (11) gives the ^{242g}Am activity, equal to 1.045(11) kBq. Besides being an absolute measurement, it is interesting to notice that this 1% relative precision was obtained for the ^{242g}Am source mixed with the ^{241}Am matrix.

It was not possible to standardize ^{242g}Am sources neither by gamma-ray spectroscopy nor by the coincidence method because the intensity of gamma rays following its decay is very low. For this reason, we applied the tracer method using ^{60}Co, which proved to be a good choice.

The measurement of the ^{241}Am neutron capture cross-section requires the determination of the ^{242g}Am activity mixed with its precursor isotope, because they cannot be separated; as a consequence it was unavoidable to deal with the strong ^{241}Am background. Besides using external Al absorbers in the range 9–20 mg cm^{-2}, it turned out necessary to reduce the proportional counter bias to allow proper detection of electrons from ^{242g}Am beta decay.

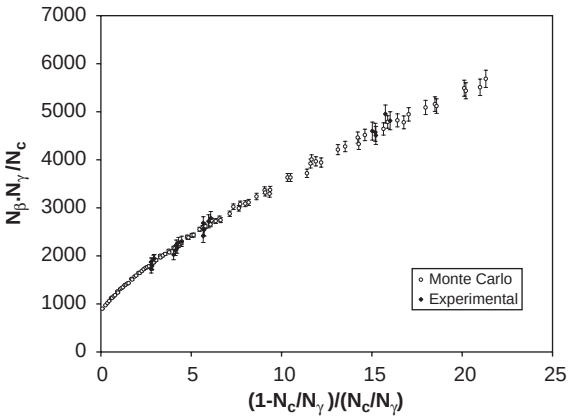

Fig. 5. Singles to coincidence counting rate as function of the ^{60}Co inefficiency parameter. The filled dots are the experimental data in Bq, normalized and corrected for the tracer activity as given in Formula (8). The open dots are the Monte Carlo values, already multiplied by the fitted $\overline{N}_{0\beta Am}$ value; the values used in the fitting procedure were calculated with better precision than these. Note that the data in this figure are not in Table 1, where the ratios of experimental to simulated value for unity activity were given.

In absolute standardization by the 4πβ-γ methodology, the activity is usually obtained by extrapolating, to the 100% electron detection efficiency limit, the singles to coincidence counting rate of a series of measurements obtained by changing slightly the electron detection efficiency, so that a polynomial model gives accurate results. In this experiment, however, the data were taken with low electron detection efficiency, making unreliable any polynomial extrapolation. Therefore, it was necessary to model the detection system, which was done by Monte Carlo simulation. The simulated counting rate in function of the inefficiency parameter is shown in Fig. 5, where it can be seen that it deviates from a straight line. Note that the method presented here relies in the comparison between simulated and experimental values for the different absorbers used and not in an extrapolation of experimental values.

In conclusion, the absolute standardization tracer method can be extended to circumstances where thick absorbers are required, achieving a precision of about 1% in the source activity when the performance of the detection system is simulated by the Monte Carlo method.

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