Available online at www.sciencedirect.com
ScienceDirect
Applied Radiation and Isotopes 64 (2006) 1238–1241
Applied Radiation and Isotopes
www.elsevier.com/locate/apradiso

Standardization of ^{241}Am solution

Marina F. Koskinas*, Eliezer A. Silva, Ione M. Yamazaki, Mauro S. Dias

Instituto de Pesquisas Energéticas e Nucleares (IPEN-CNEN/SP), Centro do Reator de Pesquisas-CRPq, C.P. 11049, Pinheiros, 05422-970-São Paulo-SP, Brazil

Abstract

The standardization of ^{241}Am solution has been undertaken using a $4\pi\beta-\gamma$ coincidence system. The 4π proportional counter used for alpha detection has a 0.1 mm thick Al window in the outside wall in order to minimize γ-ray attenuation. The extrapolation technique was applied to determine the activity of the solution. The variation of alpha efficiency has been made using external absorbers and by the electronic discrimination. The results from the two methods were compared and are in agreement within the experimental uncertainty.

Keywords: Activity; ^{241}Am; Coincidence system; Disintegration rate

1. Introduction

Am-241 decays with half-life of 432.6 y (Chechev and Kuzmenko, 2004), emitting alpha particles populating the excited levels of ^{237}Np which emit gamma-rays to achieve the stable level (Fig. 1). ^{241}Am is one of the most important radionuclides used as standard source for α and γ spectrometers. To γ spectrometers, its importance is due to the low gamma energy (59.54 keV) emitted with 35.78% intensity, which is in a region lacking in standards. For this reason ^{241}Am has been selected by the CCRI(II) (Comité Consultative pour les Etalons de Mesures des Rayonnements Ionisants II) for an international comparison.

The LMN, Nuclear Metrology Laboratory from IPEN has participated in this comparison in collaboration with the Laboratório Nacional de Metrologia das Radiações Ionizantes (LNMRI) from Rio de Janeiro.

The standardization of ^{241}Am presented in this paper was undertaken in a 4π (PC)-NaI(Tl) coincidence system. The 4π proportional counter used for alpha detection has a 0.1 mm thick Al window in the outside wall in order to minimize γ-ray attenuation. The extrapolation technique was applied in order to determine the activity of the solution (Baerg, 1967). The alpha efficiency was changed by two different methods: the first made use of external absorbers of Collodion films with 20 μg cm^{-2} gold coated, placed over and under the sources; the second was by electronic discrimination, changing the alpha spectrum threshold. The events were registered by a method developed by the LMN that makes use of a time to amplitude converter (TAC) associated with a Multichannel Analyser (Lavras et al., 2001).

2. Experimental method

2.1. Source preparation

The ^{241}Am solution was sent by the Bureau International des Poids et Mesures (BIPM) to the LNMRI, in Rio de Janeiro, and a fraction forwarded to LMN in São Paulo. The sources to be measured were prepared by dropping known aliquots of the solution on a 20 μg cm^{-2} thick Collodion film. This film had been previously coated with a 10 μg cm^{-2} gold layer in order to render the film conductive. A seeding agent (CYASTAT SM) was used to improve the deposit uniformity and the sources were dried in a desiccator. The mass determination has been performed using the pycnometer technique (Campion, 1975).

2.2. $4\pi\beta-\gamma$ Coincidence method

A 4π proportional counter filled with P-10 gas and operated at 0.1 MPa was used for alpha detection. Its

---

*Corresponding author. Tel.: +55 11 3816 9176; fax: +55 11 3816 9188.
E-mail address: koskinas@net.ipen.br (M.F. Koskinas).

0969-8043/$ - see front matter © 2006 Elsevier Ltd. All rights reserved.
doi:10.1016/j.apradiso.2006.02.028

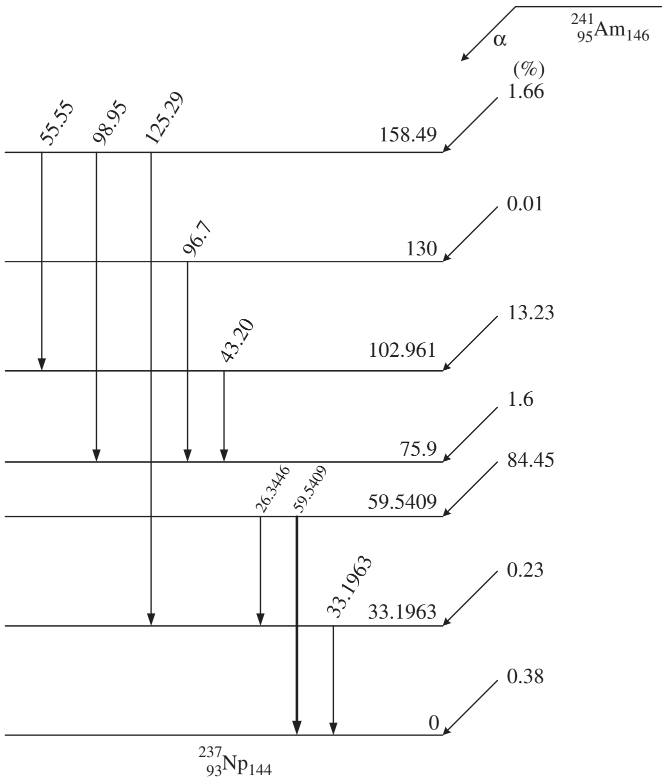

Fig. 1. Simplified decay scheme of ^{241}Am. All energies are in keV.

outside wall (with a 0.1 mm thick Al window) was coupled to the crystal scintillator, in order to minimize γ-ray attenuation. The gamma detector was a 58 mm × 58 mm NaI(Tl), instead of the usual 76 mm × 76 mm, in order to reduce the background in the 59 keV region selected for the gamma channel window.

The formulae applied to the coincidence measurement is given by the well-known relationship (Baerg, 1967):

$$\frac{N_\alpha N_\gamma}{N_c} = N_0\left\{1 + \left(\frac{1-\varepsilon_\alpha}{\varepsilon_\alpha}\right)\left(\frac{\alpha\varepsilon_{ec} + \varepsilon_{\alpha\gamma}}{1+\alpha}\right)\right\},$$

where N_0 is the disintegration rate, N_α the proportional counter counting rate, N_γ the γ-channel counting rate, N_c the coincidence rate, ε_α the proportional counter efficiency for alpha particles, $\varepsilon_{\alpha\gamma}$ the proportional counter efficiency for γ-rays, ε_{ec} the proportional counter efficiency for conversion electron, α the total conversion coefficient.

The observed counting rates N_α and N_γ were corrected for background, dead time and decay in the usual way. The coincidence rate N_c was corrected for dead time and accidental coincidences using the Cox-Isham formalism (Cox and Isham, 1977) adapted by Smith (1978).

The electronic diagram system used is presented in Fig. 2. In the TAC method, the signals after selection in the SCA Timing are sent to two gates and delay generators that yield the start and stop information to the TAC. A typical spectrum obtained in the multichannel analyzer is shown in Fig. 3. In this figure the first peak is due to the alpha counts without coincidences, the middle peak corresponds to the coincidences counts and the last peak is due to the gamma counts without coincidences. The total alpha counts are given by the integration of two first peaks, the total gamma counts are given by the integration of the second and third peaks and the coincidence counts are given by the integration of the middle peak. For the dead time correction, the ratio live time/real time obtained from the multi channel analyzer was used. The correction for accidentals was performed by subtracting the expected accidental counting rate given by $N_{ACC} = \tau_r N_\alpha N_\gamma$, where

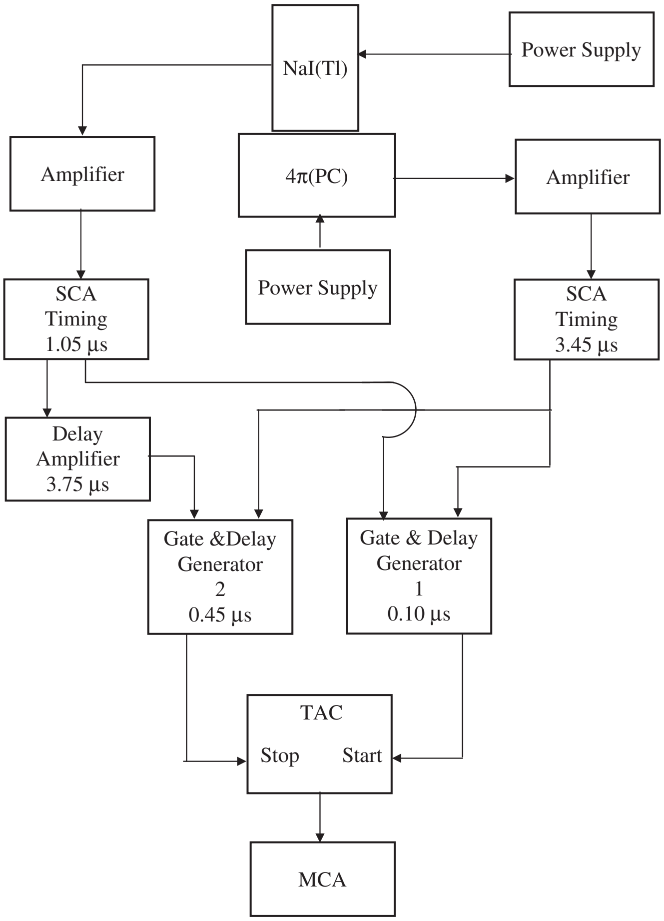

Fig. 2. Schematic diagram of the electronic system.

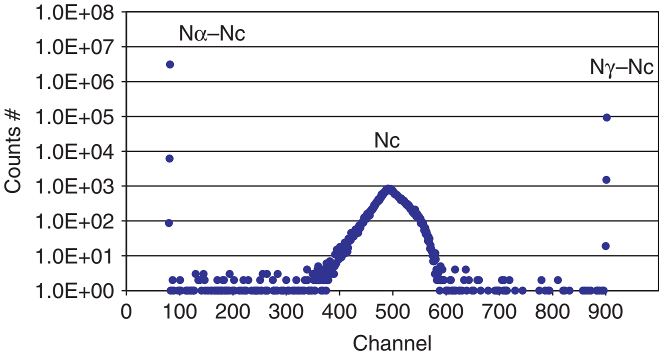

Fig. 3. Typical TAC spectrum.

τ_r is the resolution time corresponding to the selected time window from TAC.

3. Results and discussion

The extrapolation efficiency curve obtained with external absorbers is presented in Fig. 4. The extrapolation efficiency curve obtained by changing the threshold level of alpha spectrum is shown in Fig. 5. A linear least-square fitting using code LINFIT (Dias, 1999), which incorporates covariance matrix methodology was used. The curves obtained by the two methods have the same behavior and the best fit was a constant as expected, because the pulses from conversion electrons and gamma detection in the alpha counter have fallen in the noise region of the spectrum. The relative deviation as a function of $(1 - N_c/N_\gamma)/(N_c/N_\gamma)$ for both curves are presented in Figs. 6 and 7, confirming the fitting result. The extrapolation values from the two methods are in agreement within the experimental uncertainty, as shown in Table 1. These results indicate that both methods are valid to standardize alpha-gamma emitters. The activity of ^{241}Am solution is reported as the weighted average between the two methods: $295.88 \pm 0.80\,\mathrm{kBq\,g^{-1}}$. In Table 1 the values obtained by the LNMRI (Iwahara et al., 2005) are also presented and are in agreement with our result. Table 2 gives the partial uncertainties involved in the absolute standardization of ^{241}Am.

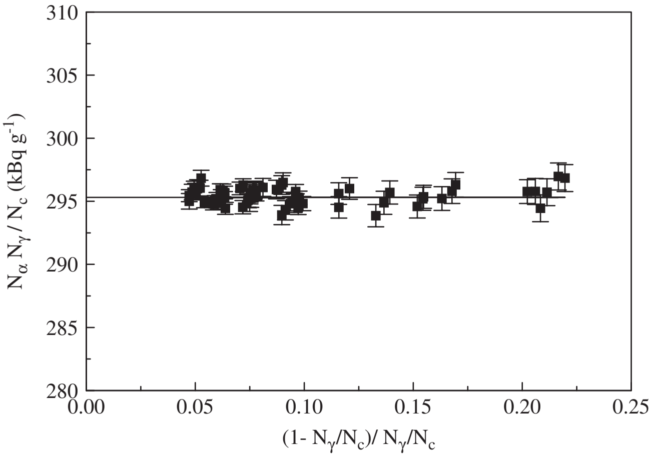

Fig. 4. Extrapolation curves of $N_\beta N_\gamma/N_c$ as a function of $(1 - N_c/N_\gamma)/(N_c/N_\gamma)$ from external absorbers.

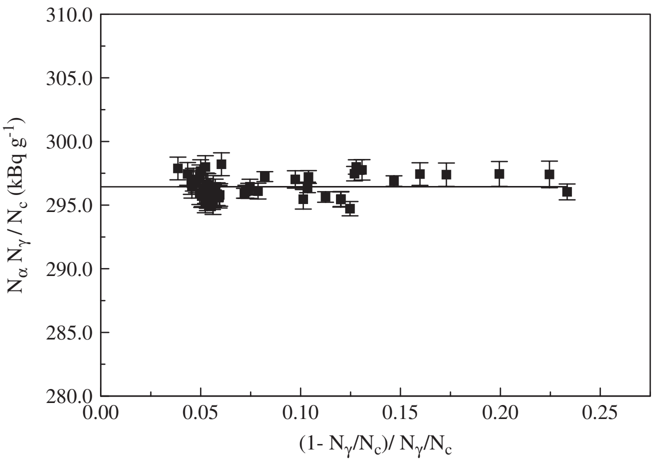

Fig. 5. Extrapolation curves of $N_\beta N_\gamma/N_c$ as a function of $(1 - N_c/N_\gamma)/(N_c/N_\gamma)$ from electronic discrimination.

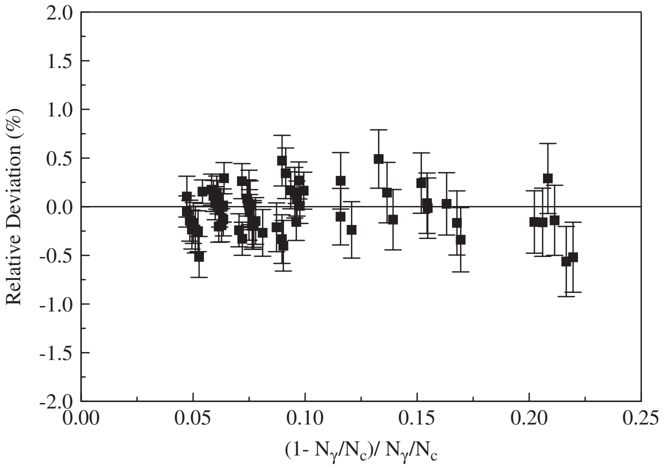

Fig. 6. Relative deviation in % as a function of $(1 - N_c/N_\gamma)/(N_c/N_\gamma)$ from external absorbers.

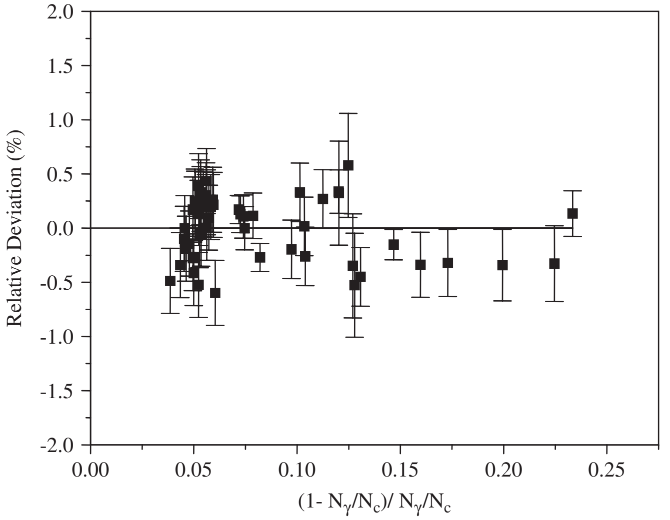

Fig. 7. Relative deviation in % as a function of $(1 - N_c/N_\gamma)/(N_c/N_\gamma)$ from electronic discrimination.

Table 1
Values obtained by LMN from methods of changing the alpha efficiency compared with the value obtained by LNMRI (Iwahara et al., 2005)

Efficiency method	Activity extrapolated (kBq g^{-1})
External absorbers	295.31 ± 1.06
Electronic discrimination	296.44 ± 1.07
Weighted average	295.88 ± 0.80
LNMRI	295.57 ± 0.96

Table 2
Typical uncertainties components in % of the activity concentration

Components	Uncertainty (%)
Counting statistics	0.10
Weighing	0.10
Dead time	0.05
Background	0.10
Resolving time	0.10
Extrapolation of efficiency curve	0.30
Combined uncertainty	0.36

Acknowledgment

The authors are grateful to the LNMRI (Rio de Janeiro) for supplying the ^{241}Am solution. We are also grateful to FAPESP (Fundação de Amparo à Pesquisa do Estado de São Paulo, Brazil) for partial support.

References

Baerg, A.P., 1967. Absolute measurement of radioactivity. Metrologia 3 (4), 105–108.

Campion, P.J., 1975. Procedures for accurately diluting and dispensing radioactive solutions. Bureau International des Poids et Mesures, Monographie BIPM-1.

Chechev, V.P., Kuzmenko, N.K., 2004. Table of Radionucléides. Laboratoire National Henry Becquerel, CEA/Saclay, France.

Cox, D.R., Isham, V., 1977. A bivariate point process connected with electronic counters. Proc. R. Soc. A 356, 149.

Dias, M.S., 1999. LINFIT: a code for linear least square fit with covariance analysis. Internal Report, IPEN-CNEN/SP.

Iwahara, A., Silva, M.A.L., Carvalho Filho, A.E., Bernardes, E.M.O., Delgado, J.U., 2005. Determination of disintegration rates and γ-ray emission probabilities of ^{65}Zn and ^{241}Am. Appl. Radiat. Isot. 63, 107–113.

Lavras, W.O., Koskinas, M.F., Dias, M.S., Fonseca, K.A., 2001. Primary standardization of ^{51}Cr radioactive solution. In: Fifth Regional Congress On Radiation Protection And Safety Regional IRPA Congress, Recife. V 1, 1-3 (CD ROM).

Smith, D., 1978. Improved correction formulae for coincidence counting. Nucl. Instrum. Methods 152, 505–519.