PRIMARY STANDARDIZATION OF ^{242}Am RADIOACTIVE SOURCES

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ABSTRACT

The procedure followed by the Laboratório de Metrologia Nuclear in São Paulo, Brazil, for the standardization of ^{242g}Am is described. The calibration system was composed of a 4π gas-flow proportional counter coupled to a pair of NaI(Tl) crystals operating in coincidence. The samples were produced by irradiating dried aliquots of ^{241}Am with thermal and epithermal neutrons at the IEA-R1 research reactor. The efficiency tracer technique has been applied using ^{60}Co as tracer. The beta detection efficiency was changed by external absorbers and extrapolated to unity by linear least square fitting applying covariance methodology.

Key Words: Proportional counters, Coincidence, efficiency tracer technique, Americium.

I. INTRODUCTION

Successive neutron capture reactions can produce new elements in a nuclear reactor. At high fuel burning rates, a relatively high amount of Americium and Curium can be formed. These isotopes must be taken into account in reactivity calculations. Moreover, the presence of these radioisotopes makes fuel element handling more difficult due to high dose rates. The exact estimate of this effects depend on the value of activity of the reaction products.

One example is ^{242g}Am (ground state reaction product), produced by thermal neutron capture reaction of ^{241}Am. In this case an accurate measurement of ^{242g}Am induced activity is needed in order to estimate experimentally the ^{241}Am neutron capture process [1].

The Laboratório de Metrologia Nuclear (LMN – Nuclear Metrology Laboratory) at the IPEN, in São Paulo, Brazil, has achieved large experience in developing radionuclide standardization procedures.

The present work describes the technique adopted by LMN for the standardization of ^{242g}Am.

This radionuclide decays 82.7% by β^- emission and 17.3% by electron capture, as shown in Figure 1 [2]. The transitions from excited states of ^{242}Pu and ^{242}Cm, 44.52 and 42.13 keV respectively, have high internal conversion coefficients. For this reason, the gamma ray probabilities per decay in both cases are very small. This feature makes it in a suitable radionuclide to be standardized by the tracing technique [3]. The radionuclide ^{60}Co was chosen as tracer because of its simple decay scheme and low internal conversion coefficients.

The calibration system was composed of a 4π gas-flow proportional counter coupled to a pair of NaI(Tl) crystals operating in coincidence [4,5].

II. METHODOLOGY

Irradiation

The irradiations were performed at the IPEN 2 MW pool-type research reactor. The target consisted of a ~40 MBq ^{241}Am source sealed inside a double 0.2 mm thick polyethylene envelope. The sample was placed inside a polyethylene rabbit, transported to the reactor core by means of a pneumatic tube and irradiated in a thermal neutron flux of 2.8 x 10^{12} $cm^{-2}.s^{-1}$ for 30 min. The time interval between the end of irradiation and counting was approximately 60 min.

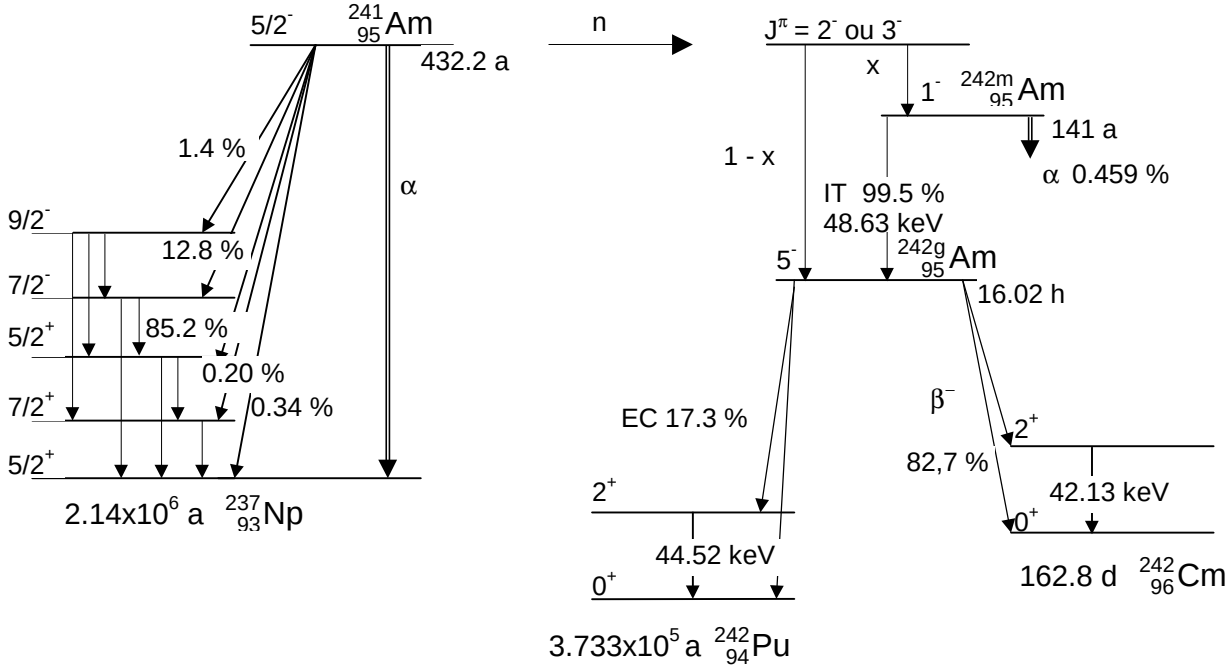

Figure 1: Scheme of the $^{241}Am(n,\gamma)^{242g}Am$ reaction [2].

Source Preparation

After irradiation, the ^{241}Am + ^{242g}Am sample was diluted in 1 M HNO_3 solution. The sources to be measured in the 4πβ-γ system were prepared by dropping known aliquots of each radioactive solution (~6000 Bq of ^{241}Am, ^{242g}Am and ^{60}Co) on a 20 μg.cm^{-2} thick Collodion film.

This film was previously coated with a 10 μg.cm^{-2} thick gold layer in order to render the film conducting. Three mixed sources ($^{241,242g}Am$ + ^{60}Co) prepared from each irradiated target using a 2:1 ratio (β-pure and β–γ, respectively), in order to obtain similar counting rates for both radionuclides. A seeding agent (CYASTAT SN) was used for improving the deposit uniformity and the sources were dried in a warm (45ºC) nitrogen jet [6]. Several sources of ^{60}Co and ^{241}Am were standardized by the 4πβ–γ coincidence and 4πα systems respectively, for calibrate the HPGe spectrometer system.

4πβ-γ Coincidence Measurements

The absolute standardization system [3,4] consisted of a gas-flow proportional counter with 4π geometry using 90% Ar + 10% CH_4 gas filling at 0.1 MPa, as the β detector and coupled to a pair of 76 mm x 76 mm NaI(Tl) scintillation counters, as γ detectors. The selected γ-channel window covered both 1173 and 1332 keV peaks of ^{60}Co. The proportional counter bias was 1400 V for alpha sources, 1520 V for mixed sources and 2050 V for beta sources. Figures 2 and 3 show β spectra obtained with the absorbers and spectra of mixed and purely α sources, respectively.

The number of detected events in the proportional counter is given by:

$$\mathbf{N_{\beta(Am+Co)} = N_{0Co}\,\varepsilon_{\beta Co} + N_{0\beta Am}\,\varepsilon_{\beta Am}} \qquad \textbf{(1)}$$

where:

- $\varepsilon_{\beta Co}$ is the tracer efficiency in the mixed source, obtained from the ratio N_c/N_γ;
- N_γ, N_c are the γ and coincidence counting rates;
- $N_{\beta(Am+Co)}$ is the mixed source counting rate in the proportional counter;
- N_{0Co} is the activity of ^{60}Co tracer deposited on the mixed source;
- $N_{0\beta Am}$ is the ^{242g}Am beta-branch disintegration rate
- $\varepsilon_{\beta Am}$ is the ^{242g}Am beta efficiency.

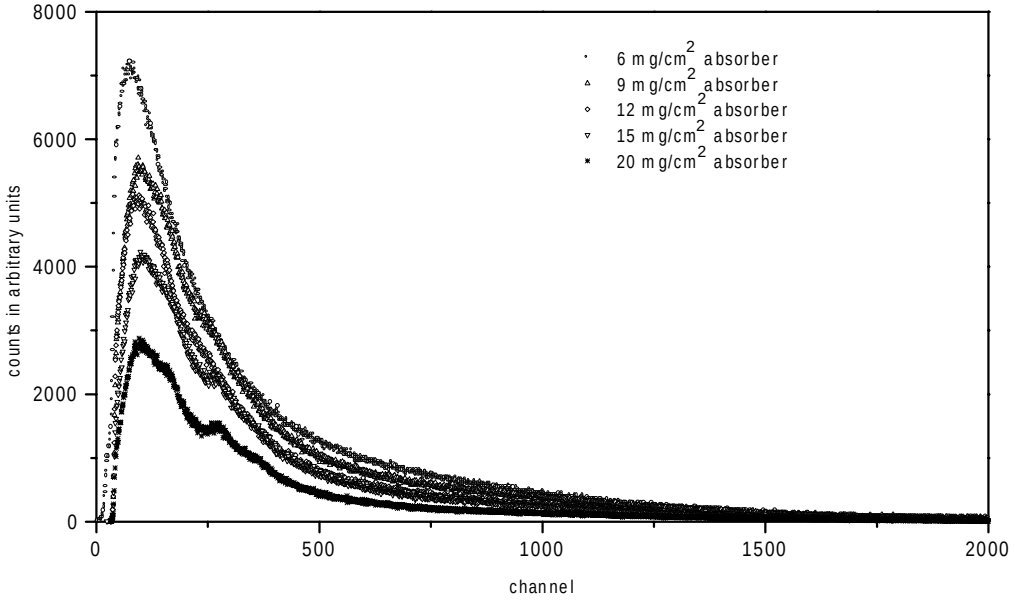

Figure 2: Variation of the β detection with the 6 to 20 mg/cm^2 absorbers.

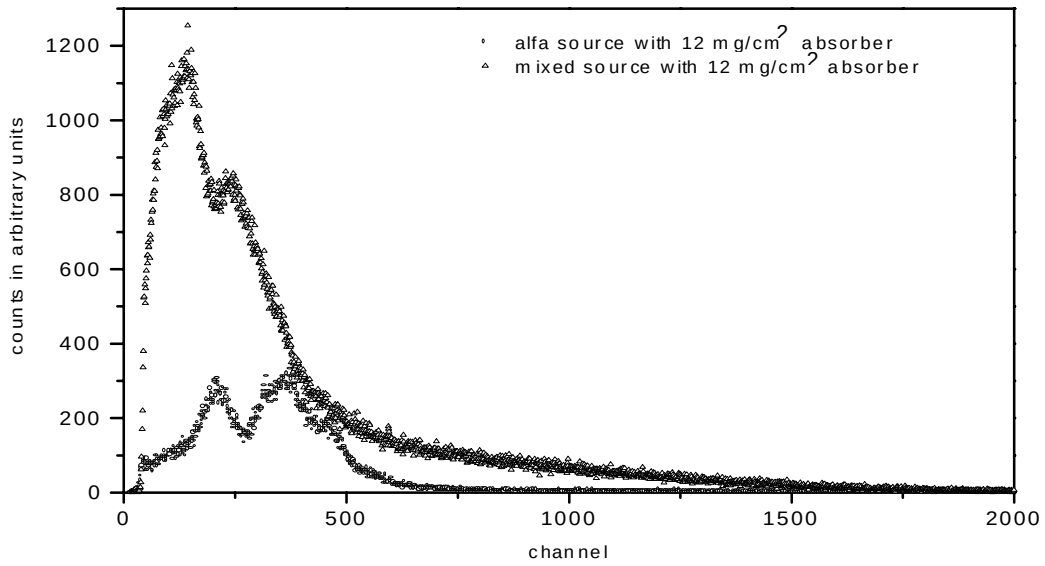

Figure 3: Particles detection from mixed and pure α sources with 12 mg/cm^2 absorbers.

Applying the linear extrapolation technique [3] the final expression is given by:

$$\frac{N_{\beta(Am+Co)}\,N_{\gamma}}{N_c} - N_{0Co} = N_{0\beta Am}\left[1 + C\left(\frac{1-\varepsilon_{\beta Co}}{\varepsilon_{\beta Co}}\right)\right] \qquad (2)$$

where: C is a constant.

Suitable corrections for accidental coincidences, dead time and decay were taken into account. The proportional counter efficiency has been changed by applying external absorbers over and under the sources with thickness in the range of 6-20 mg/cm^2.

The value of C in equation (2) is expected to be large because of the difference between ^{242g}Am and ^{60}Co endpoint beta energies. This effect was reduced dividing this equation by the following factor:

$$r = \frac{\varepsilon_{\beta Am}}{\varepsilon_{\beta Co}} \cong \exp\left\{ -\ln\left(\varepsilon_{\beta Co}\right)\left[1 - \left(\frac{E_{Co}}{E_{Am}}\right)^{1.14}\right]\right\} \quad (3)$$

The final expression was fitted by weighed least squares using code LINFIT [7]. This code makes use of covariance matrix methodology and takes into account all partial errors involved. The extrapolation to $(1- \varepsilon_{\beta Co})/ \varepsilon_{\beta Co})=0$ yielded the expected $N_{0\beta Am}$ value (as we see in Fig. 4).

Since the isomer ^{242m}Am half-life is long, its decay contribution to the ^{242g}Am activity at the time of measurement has been estimated to be negligible. The beta branching ratio correction was applied in order to obtain the final ^{242g}Am activity:

$$N_{0Am} = \frac{N_{0\beta Am}}{I_{\beta Am}} \quad (4)$$

III. RESULTS AND DISCUSSION

The extrapolated ^{242g}Am activity was obtained with 2.7% standard deviation. The main source of this uncertainty is the alpha background due to ^{241}Am. The attempts to reduce this background by applying external absorbers and reducing the proportional counter bias were moderately successful in reducing the alpha background.

Inclusion of factor r described in equation (3) reduced considerably the slope in equation (2) and contributed to reduce the final uncertainty. This factor appears to be an interesting alternative to improve the extrapolation technique.

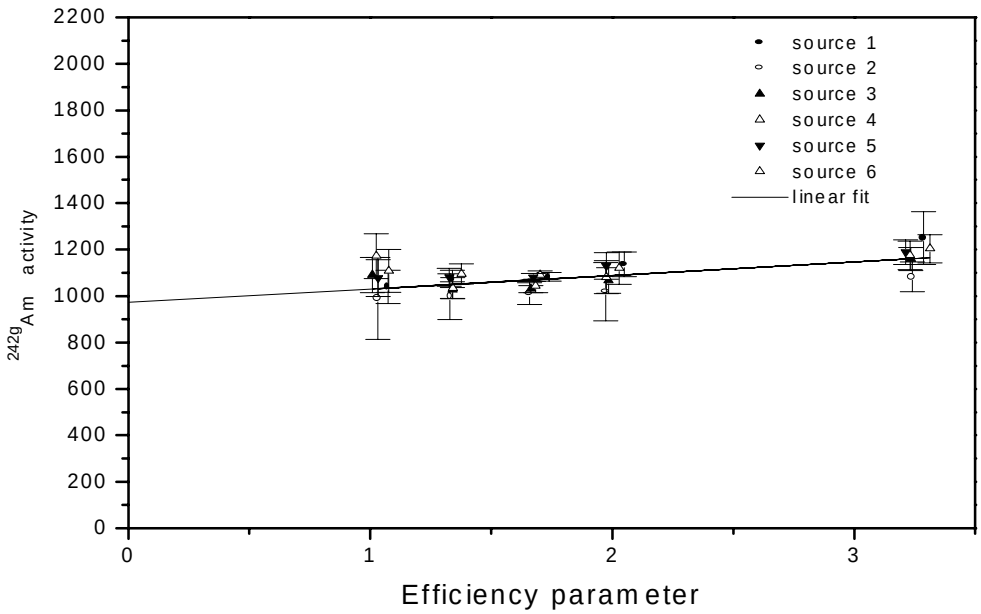

Figure 4: Observed activity as a function of the efficiency parameter: $(1- \varepsilon_{\beta Co})/ \varepsilon_{\beta Co}$.

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