Index

PRIMARY STANDARDIZATION OF ^{153}Sm RADIOACTIVE SOLUTION

Marina F. Koskinas*, Wilson O. Lavras and Mauro S. Dias

Instituto de Pesquisas Energéticas e Nucleares - IPEN-CNEN/SP
Av. Lineu Prestes 2.242
05508-900 Butantã, São Paulo, SP, Brasil
* koskinas@net.ipen.br

ABSTRACT

The procedure followed by the Laboratório de Metrologia Nuclear (LMN) at the IPEN, in São Paulo, for the standardisation of ^{153}Sm is described. This radionuclide is part of the LMN programme for the standardisation of important radionuclides used in nuclear medicine. The activity measurement was carried out in a 4π (PC)β-γ coincidence system consisting of a proportional counter, coupled to a pair of 3” x 3” NaI(Tl) crystals. This radionuclide was obtained by means of the ^{152}Sm (n,γ) ^{153}Sm reaction in a thermal neutron flux at the IPEN 2 MW research reactor. The solution activity was determined by the extrapolation technique, using external absorbers over and under the ^{153}Sm sources. The events were registered using a Time to Amplitude Converter (TAC) associated with a Multichannel Analyser.

Keywords: samarium 153, coincidence method, standardization.

I. INTRODUCTION

The procedure followed by the Nuclear Metrology Laboratory (LMN) at the IPEN – CNEN/SP, in São Paulo, for the standardization of ^{153}Sm is described. This radionuclide was chosen as part of an ongoing program of standardization of radionuclides used in nuclear medicine, for improving the standards produced in Brazil. ^{153}Sm is a therapeutic agent indicated for pain relief in patients with confirmed osteoblastic metastatic bone lesions.

^{153}Sm decays with a half-life of 46.285 ± 0.004 h, by β^- transition, 79.3 % populating the excited states of ^{153}Eu and 20.7 % to the ground state. The main gamma-ray energy is 103 keV with 28.5 % intensity. The decay scheme of ^{153}Sm is shown in fig. 1[1].

The measurements were carried out in a 4πβ–γ coincidence system as described in section II. The disintegration rate was determined using the extrapolation efficiency technique using external absorbers over and under the sources. The events were registered by a method developed at the LMN which makes use of a Time to Amplitude Converter (TAC) associated with a Multichannel Analyser (MCA).

II. EXPERIMENTAL METHOD

Source Preparation. ^{153}Sm was produced in a thermal neutron flux of 2×10^{13} cm^{-2} s^{-1} obtained near the core of the IPEN 2 MW research reactor by means of the reaction ^{152}Sm(n,γ)^{153}Sm. The sample consisted of samarium oxide isotopically enriched with ^{152}Sm sealed in a quartz tube. After irradiation the oxide was dissolved into 0.1N HCl. From this master solution one solution with a diluting factor of 40 was prepared. From the two solutions, sources to be measured in the 4π β–γ system were prepared by dropping known aliquots of the solutions on a 20 μg/cm^2 thick Collodion film. This film had been previously coated with a 10 μg/cm^2 thick gold layer on each side, in order to render the film conductive. A seeding agent (CYASTAT SN) was used for improving the deposit uniformity and the sources were dried in a warm (45 °C) nitrogen jet [2]. The accurate source mass determination was performed using a Mettler 5SA balance by the pycnometre

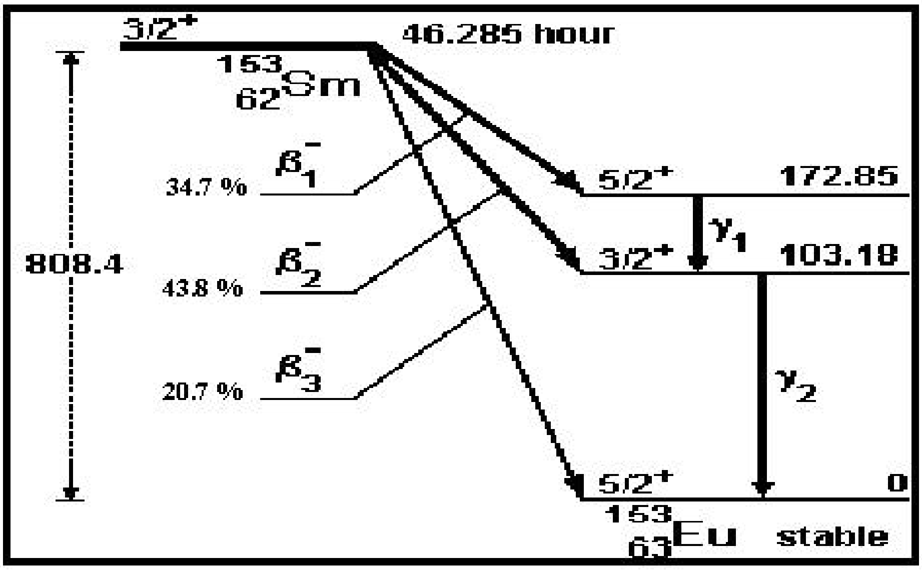

Figure 1. Simplified decay scheme of ^{153}Sm [1]; All energies are in keV.

technique [3]. Two flame-sealed ampoules with known mass were prepared for gamma ray spectroscopy and ionization chamber measurements.

Standardization Methods. A conventional $4\pi\beta-\gamma$ coincidence system was used [4], consisting of 4π proportional counter filled with P-10 gas and operated at 0.1 MPa, coupled to a pair of 3” x 3” NaI(Tl) crystals. The events were registered by a method developed at the LMN which makes use of a Time to Amplitude Converter (TAC) associated with a Multi-channel Analyser [5]. The electronic diagram is shown in figure 2. By this method, the pulses from the proportional counter, after passing by a single-channel analyser (SCA), which cuts off noise pulses, are sent to gate and delay generators 1 and 2. The gamma detector pulses after passing by the SCA, set for the selected gamma window, are sent as negative pulses to gates 1 and 2. Delayed pulses from the gate and delay generator are sent to a Time to Amplitude Converter (TAC) and are recorded by a Multi-Channel Analyser (MCA). Pulses from gate 1 send a signal start and pulses from gate 2 send a signal stop to TAC.

Figure 3 shows a typical spectrum obtained. In this figure the first peak corresponds to pulses from the proportional counter, the second one corresponds to coincidence pulses and the third one is due to gamma pulses. Both first and last peaks do not include coincidence events.

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

$$N_\beta = N_o \Sigma a_r [\varepsilon_\beta + (1-\varepsilon_\beta)[(\alpha_i \varepsilon_{ce} + \varepsilon_{\beta\gamma})/(1+\alpha_i)]] \quad (1)$$

For the γ channel a window was set at the most intense γ- energy (103 keV). The number of registered events N_γ is given by:

$$N_\gamma = N_o a_2 \varepsilon_{\gamma 2} \quad (2)$$

The number of coincident events is given by:

$$N_c = N_o a_2 \varepsilon_\beta \varepsilon_{\gamma 2} \quad (3)$$

In this equation N_o corresponds to the source activity, a_r is the branch ratio of r-beta, ε_β is the detection efficiency for beta particles, ε_{ce} is the detection efficiency for conversion electrons and α_i is the total conversion coefficient of branch i, a_2 is the branch ratio of β_2 and $\varepsilon_{\gamma 2}$ is the efficiency of γ_2.

Corrections for background, dead time and decay were applied. The corrections of accidental coincidences were made by spectrum subtraction.

Equations (1), (2) and (3) lead to the value of the activity by the well-known expression:

$$\frac{N_b N_g}{N_c} = N_o (1 + k_e) \quad (4)$$

where

$$k_\varepsilon = \frac{(1 - e_b)}{e_b} C$$

and

$$C = \left(\frac{1+\alpha_i}{\alpha_i}\right)\varepsilon_{ce}$$

The constant C was obtained by the efficiency extrapolation method described by Baerg [6].

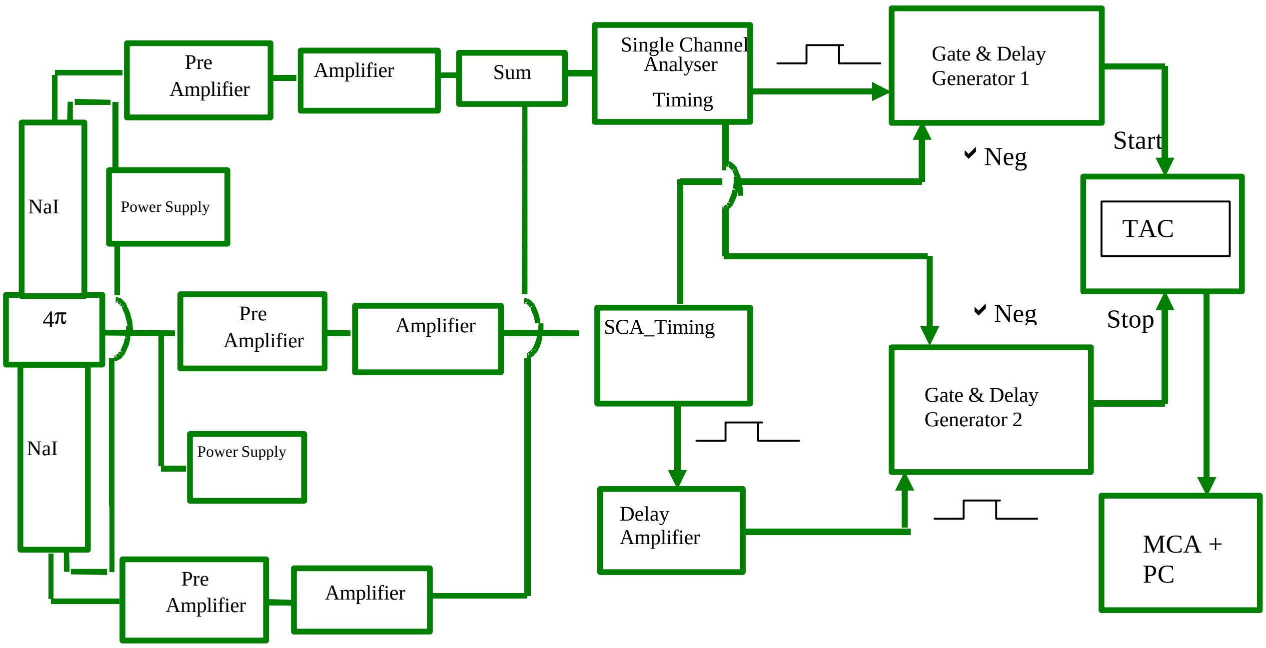

Figure 2. Electronic Diagram using TAC

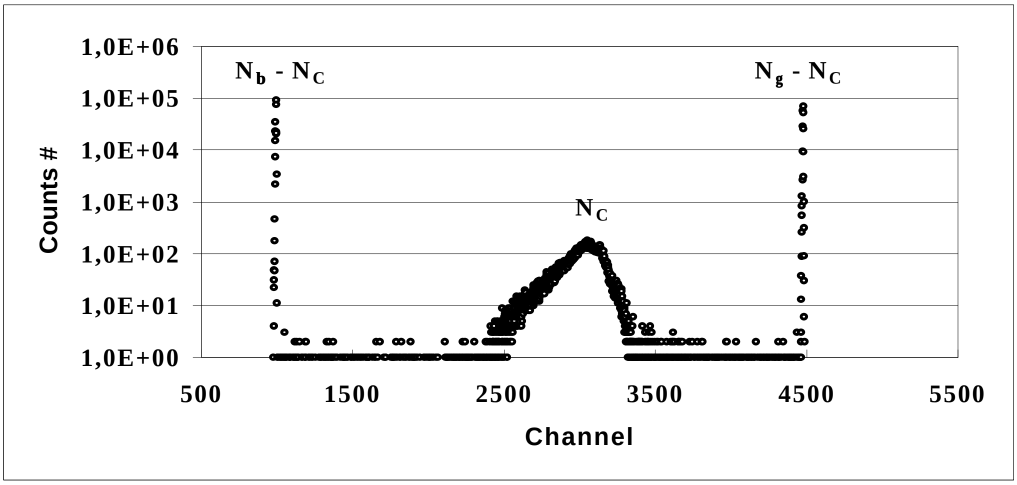

Figure 3. Typical spectrum obtained with TAC.

Index

III. RESULTS

The measurements in the coincidence system of sources prepared from two solutions (master and diluted) were normalized by measurements of ampoules in the gamma spectrometer. After normalization a plot of $N_\beta N_\gamma/N_c$ versus inefficiency parameter $(1 - N_c/N_\gamma)/N_c/N_\gamma$ was obtained in the range from 63 % to 92 %, using external absorbers, as shown in Fig.4. The extrapolation curve was obtained by linear least square fitting using code LINFIT[7] which incorporates covariance matrix methodology. To obtain this curve were used 24 measurements from 8 sources.

The parameters obtained with the covariance matrix are presented in table 1.

Table 1 Parameters of fitting

Parameters	Values x10^7	Covariance Matrix x10^{10}	
A	(2.906 ± 0.013)	1.78089	
B	(0.719 ± 0.026)	-2.33195	6.50816

The main uncertainties involved in the measurements are: counting statistics, weighing, dead time, half-life and extrapolation curve efficiency. All these uncertainties are included in the fitting, uncertainties type A (statistic) were included as not correlated and uncertainties type B (systematic) as correlated. For this measurement the final activity was obtained with an overall uncertainty of 0.46% which is satisfactory for calibrating secondary standard systems.

Ampoules with the standard solution were measured in an ionization chamber $4\pi\gamma$ in order to determine the calibration factor. Additional gamma spectrometry measurement is underway for determining the emission probability per decay of 103 keV gamma ray of ^{153}Sm. This result will be compared with the literature.

ACKNOWLEDGEMENT

The authors would like to thank to IAEA for partially financial support in BRA02/014 project, and to IPEN Radioisotope Production Center for supplying the ^{153}Sm master solution.

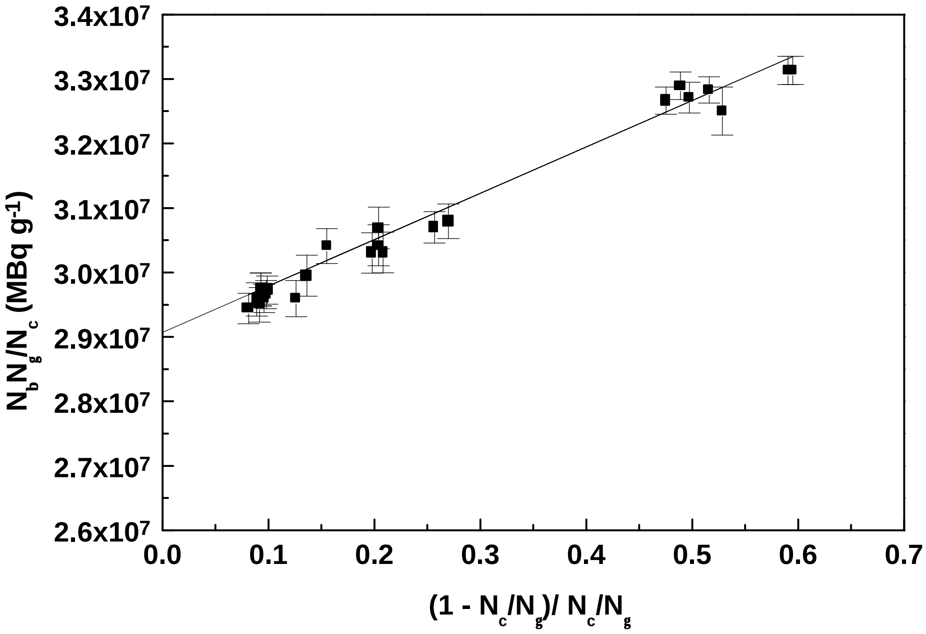

Figure 4. Extrapolation Curve

Index

REFERENCE

[1] Bowles, N. E.; Woods, S. A.; Woods, D. H.; Jerome, S. M.; Woods, M. J.; De Lavison, P.; Lineham, S.; Keightley, J.; Poupaki, I. **Standardization of ^{153}Sm, National Physical Laboratory, U.K.** Appl. Radiat. Isot. Vol49, no 9-11, pp. 1345-1347, 1998.

[2] Wyllie, H.A.; Johnson, E.P. and Lowenthal, G.C. (1970) **A procedure for stirring aliquots of radioactive solutions when making thin 4p counting sources.** Int. J. Appl. Radiat. Isot., **21**, 497.

[3] Campion, P.J. (1975*)* **Procedures for accurately diluting and dispensing radioactive solutions.** Bureau International des Poids et Mesures, Monographie BIPM 1.

[4] Moura, L.P. **Generalized coincidence method for absolute activity measurements of radionuclides. Application for determining the internal conversion coefficient of Tl-203 279-keV transition.** Ph.D. Thesis, University of Campinas, 1969.

[5] Lavras, W. O.; Koskinas, M.F.; Dias, M.S. and Fonseca, K.A. **"Primary Standardization of Cr-51 Radioactive Solution"**, V Regional Congress On Radiation Protection And Safety -Regional Irpa Congress, Recife - PE, BR. April 29 to May 04, 2001.

[6] Baerg, A. P. **Absolute measurement of radioactivity.** Metrologia, 3 (4), p. 105 - 108, 1967.

[7] Dias, M. S. **Programa LINFIT.** Internal Report, IPEN, 2000.

.