Measurement of the gamma-ray probability per decay of ^{186}Re

Kátia Aparecida Fonseca, Marina Fallone Koskinas, and Mauro da Silva Dias

Abstract–**In this paper the procedure followed by the Nuclear Metrology Laboratory (Laboratório de Metrologia Nuclear - LMN) at the IPEN, in São Paulo, for the standardization of the ^{186}Re and determination of the gamma emission probabilities per decay of the main gamma-ray energies of ^{186}Re, 137.15 keV from β^- decay and 122.6 keV from EC are presented. The activity measurement was carried out in a 4π (PC) β-γ coincidence system. The contribution from electron capture events in the proportional counter has been estimated by means of additional 4π(PC)β–γ HPGe coincidence measurements. The activity of the solution was determined by the extrapolation technique. The events were registered using a Time to Amplitude Converter (TAC) associated with a Multichannel Analyzer. The gamma emission probabilities per decay were determined by means of an HPGe spectrometer calibrated in the 81 keV to 1408 keV energy range, by measuring sealed ampoules of ^{60}Co, ^{133}Ba, ^{137}Cs and ^{152}Eu, standardized at the LMN.**

I. Introduction

THE accurate knowledge of gamma-ray emission probabilities per decay of radionuclides is important in several applications. Usually, the procedure consists of standardization of the radionuclide by a primary system, in order to obtain the activity, and measurements of the gamma-rays in an accurately calibrated HPGe spectrometer.

In this paper the procedure developed at the Nuclear Metrology Laboratory (Laboratório de Metrologia Nuclear - LMN) at the IPEN, in São Paulo, for the standardization of the ^{186}Re by means of a 4π (PC)β-γ coincidence system is presented. In addition, the gamma–ray emission probabilities per decay of the most intense ^{186}Re transitions were determined in a HPGe spectrometer.

This radionuclide was selected due its importance in nuclear medicine. The decay scheme of ^{186}Re is shown in Fig. 1 [1]. It decays with a half-life of (90.64 ± 0.09) h, 93.1% by β^- transition, populating the excited states of ^{186}Os, and 6.9 % by EC populating the excited states of ^{186}W. Both branches are followed by gamma-ray emission. The main gamma-ray energies are 137.15 keV from β^- decay and 122.6 keV from EC.

The measurement was carried out in a 4π (PC) – (NaI) coincidence system as described in section II. The disintegration rate was determined using the extrapolation efficiency technique using external absorbers on both sides of the radioactive 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 Analyzer. The contribution from electron capture events in the proportional counter has been estimated by means of a 4π (PC) – (HPGe) coincidence system [2].

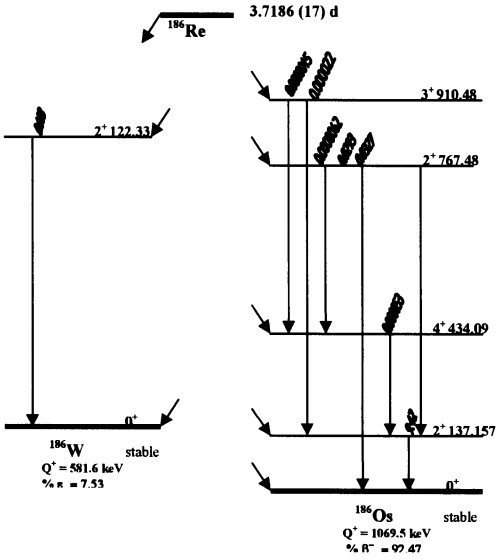

Fig. 1. Decay scheme of ^{186}Re [1]. All energies are in keV.

Manuscript received November 12, 2008. This work was supported in part by the Brazilian National Council for Science and Technological Development under Grant No. 301146/2007-01.

Kátia Aparecida Fonseca is with the Health Physics Departament SPR IPEN – Instituto de Pesquisas Energéticas e Nucleares, São Paulo, Brazil, (e-mail: kfonseca@ipen.br)

Marina Fallone Koskinas is with the Nuclear Metrology Laboratory - CRPq/Nuclear Physics Division IPEN – Instituto de Pesquisas Energéticas e Nucleares, São Paulo, Brazil (e-mail: koskinas@ipen.br).

Mauro da Siva Dias is with the Nuclear Metrology Laboratory - CRPq/Nuclear Physics Division IPEN – Instituto de Pesquisas Energéticas e Nucleares, São Paulo, Brazil (e-mail: msdias@ipen.br)

II. METHODOLOGY

A. Source Preparation

The ^{186}Re sample was produced by ^{185}Re(n,γ) ^{186}Re reaction in a thermal neutron flux of $2x10^{13}$ cm^{-2} s^{-1} obtained near of the core of the IPEN 2 MW research reactor. The material consisted of 100 μg of natural rhenium oxide sealed in a quartz tube. It has been irradiated for 30 h and was left to decay for about 14 days in order to reduce the activity of ^{188}Re (17 h half-life) to minimum level. After irradiation the oxide was dissolved into 20 mg of 1M HNO_3, and the tube washed with distilled water. The final volume was 2 ml with 0.1 M. Radioactive sources to be measured in the 4π β–γ system were prepared immediately after dissolution by dropping known aliquots on a 20 $\mu g.cm^{-2}$ thick Collodion film. This film had been previously coated with a 10 $\mu g.cm^{-2}$ 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 desiccator. The accurate source mass determination was performed using a Mettler M5SA balance by the pycnometer technique [3]. A flame sealed ampoule was prepared for impurity checks and for measuring the gamma ray emission probability per decay.

B. 4πβ–γ coincidence measurement

A conventional 4πβ–γ coincidence system was used. The proportional counter of the LMN, presented in Fig. 2 has a pill-box shape. It is composed by two symmetrical brass parts with 3 mm thickness each, interfaced by a sliding plate with two holes for positioning the sources inside of the detector. Its internal volume is delimited by a brass block in a truncated cylinder shape, having 30 mm of diameter and 75 mm of length. In each half of the detector there is a thin anode which consists of a stainless steel wire 50 μm thick, fixed to one of the extremities by Teflon plugs and connected to BNC connectors at the other. The gas mixture composed of 10% methane and 90% argon is flowing through the pill-box at a pressure of 0.1 MPa. The proportional counter is coupled to a 76 x 76 mm^2 NaI(Tl) scintillators on the upper and lower sides.

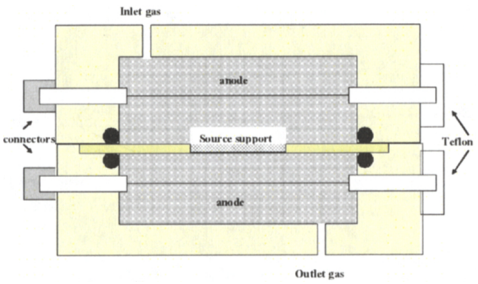

Fig. 2. Proportional counter diagram [4]

The electronic diagram used in this measurement is presented in Fig. 3. In this diagram power supplies modules are not shown.

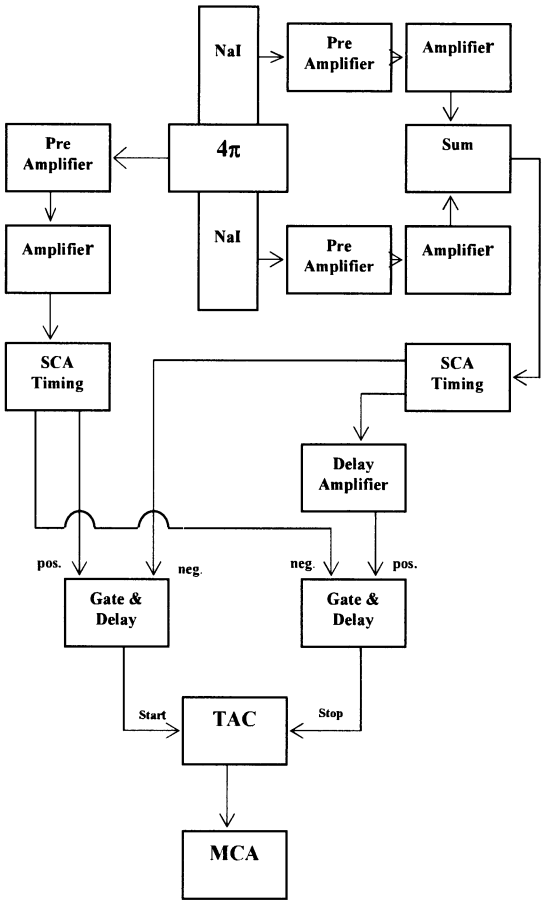

Fig. 3. Electronic diagram of coincidence system 4πβ–γ.

The gamma window was set by gating the gamma-rays from both β^- and EC branches (137 keV + 122 keV), due to the poor resolution of NaI(Tl) and low intensity of 122 keV gamma-ray. The events from the proportional counter and from the scintillators were registered by a method developed at the LMN which makes use of a Time to Amplitude Converter (TAC) associated with a Multichannel Analyzer [5].

The formulae applied to the coincidence measurement are the following:

$$N_\beta = N_0 a\left[\varepsilon_\beta + (1-\varepsilon_\beta)\, I_{Os(2)}\left(\frac{\alpha\,\varepsilon_{ec}+\varepsilon_{\beta\gamma}}{1+\alpha}\right)_{Os(2)}\right] + N_0 b\left[\varepsilon_{(X,A)} + (1-\varepsilon_{(X,A)})\, I_{W(1)}\left(\frac{\alpha\,\varepsilon_{ec}+\varepsilon_{\beta\gamma}}{1+\alpha}\right)_{W(1)}\right] \quad (1)$$

$$N_\gamma = N_0\left[a\, I_{\gamma 2}\frac{\varepsilon_{\gamma 2}}{1+\alpha_2} + b I_{\gamma 1}\frac{\varepsilon_{\gamma 1}}{1+\alpha_1}\right] \quad (2)$$

where:

N_o is the disintegration rate,
a β^- branching ratio,
b electron capture (EC) branching ratio,
N_β proportional counter counting rate,
N_γ γ-channel total counting rate (including β^- and electron capture processes),
N_c coincidence rate between β^- and γ events,
ε_β proportional counter efficiency,
ε_γ NaI(Tl) crystal efficiency for γ-rays,
$\varepsilon_{X,A}$ proportional counter efficiency for X-rays or Auger electrons,
ε_{ce} proportional counter efficiency for conversion electrons,
$\varepsilon_{\beta\gamma}$ proportional counter efficiency for γ-rays,
α total conversion coefficient,

$I_{Os(2)}$ and $I_{W(1)}$ are the probabilities of γ-ray transitions of the γ-window selected, ^{186}Os and ^{186}W branches, respectively.

$$N_\gamma = N_0\varepsilon_\gamma\left[a\,\frac{I_{\gamma 2}}{1+\alpha_2} + b\,\frac{I_{\gamma 1}}{1+\alpha_1}\right] \quad (3)$$

$$N_C = N_0\varepsilon_\gamma\left[a\,\varepsilon_\beta\,\frac{I_{\gamma 2}}{1+\alpha_2} + b\,\varepsilon_{(X,A)}\,\frac{I_{\gamma 1}}{1+\alpha_1}\right] \quad (4)$$

In these equations corrections for background, dead time and decay were applied. The corrections for accidental coincidences were performed by spectrum subtraction.

Equations (1), (3) and (4) lead to the activity value by the following expression:

$$\frac{N_\beta N_\gamma}{N_C} = N_0\left\{a\left[1+\frac{(1-\varepsilon_\beta)}{\varepsilon_\beta}K_{Os}\right] + b\left[\frac{\varepsilon_{X,A}}{\varepsilon_\beta} + \frac{(1-\varepsilon_{(X,A)})}{\varepsilon_\beta}K_W\right]\right\} * \left[\frac{a\left(I_{\gamma 2}/1+\alpha_2\right) + b\left(I_{\gamma 1}/1+\alpha_1\right)}{a\left(I_{\gamma 2}/1+\alpha_2\right) + b\left(\varepsilon_{X,A}/\varepsilon_\beta\right)\left(I_{\gamma 1}/1+\alpha_1\right)}\right] \quad (5)$$

where

$K_{Os} = I_{Os}\left(\frac{\alpha\,\varepsilon_{ec}+\varepsilon_{\beta\gamma}}{1+\alpha}\right)_{Os}$, $K_W = I_W\left(\frac{\alpha\,\varepsilon_{ec}+\varepsilon_{\beta\gamma}}{1+\alpha}\right)_W$ and

$B = a\,K_{Os} + b\,f_X$ and $\varepsilon_{(X,A)} + (1-\varepsilon_{(X,A)})\,K_W = f_X$

In the limit where $\varepsilon_\beta \to 1$:

$$\left[\frac{\left(I_{\gamma 2}/1+\alpha_2\right) + \frac{b}{a}\left(I_{\gamma 1}/1+\alpha_1\right)}{\left(I_{\gamma 2}/1+\alpha_2\right) + \frac{b}{a}\left(I_{\gamma 1}/1+\alpha_1\right)\varepsilon_{(X,A)}}\right] \cong 1$$

yields

$$\frac{N_\beta N_\gamma}{N_C} = A + \left(\frac{1-\varepsilon_\beta}{\varepsilon_\beta}\right)B \quad (6)$$

where: $A = N_0\,(a + bf_x)$.

The factor f_X is an additional correction, obtained experimentally, to account for the sensitivity of the proportional counter to X-rays.
The values of a and b were taken from the literature [6] and [7] and the factor f_x has been measured by the spectroscopic method, as described below.

C. Spectroscopic method

Factor f_X has been determined by an 4π (PC) – (HPGe) system applying the spectroscopic method [2]. This system consisted of a 4π(PC) proportional counter coupled to a HPGe detector. In this system the pulses proceeding from proportional detector, after being amplified, discriminated and gated, were sent to a Multichannel Analyzer input gate. The pulses originated from the HPGe detector, after being amplified passed through a delay module in order to be synchronized with the signals from the proportional counter. After this, they were sent directly to the ADC input of the Multichannel Analyzer. Fig. 4 shows the electronic diagram used for this measurement.

The selected gamma-ray energy was 122 keV. The single and coincidence spectrum peak areas were called A_S and A_C, respectively. The ratio between them corresponds to the efficiency of the proportional counter, given by:

$$A_C/A_D = \varepsilon_{(X,A)} + (1-\varepsilon_{(X,A)})\,I_W\left(\frac{\alpha\,\varepsilon_{ec}+\varepsilon_{\beta\gamma}}{1+\alpha}\right)_W \quad (7)$$

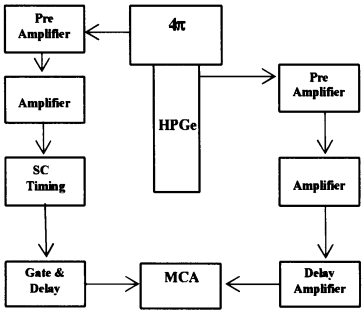

Fig. 4. Electronic spectroscopy system diagram.

D. Gamma-ray spectrometry measurements

The gamma-ray measurements were performed in a REGe spectrometer, previously calibrated in the energy range between 81 keV to 1408 keV by measuring sealed ampoules of ^{60}Co, ^{133}Ba, ^{137}Cs and ^{15}Eu, standardized at the LMN. The source-detector distance was around 17.9 cm. Dead time and

pile-up corrections were applied by measuring a reference pulser peak near the upper edge of gamma spectrum together with the radioactive sources. The total absorption peak area was determined by simple spectra integration with linear background subtraction from both sides of the peak applying code ALPINO [8]. The calibration curve was obtained by a forth order polynomial function in log-log scale, applying least square fitting and considering covariance methodology in order to determine all uncertainties.

III. Results and Discussion

Fig. 5 shows the extrapolation curve for the activity determination obtained by linear least square fitting using code LINFIT [9] which incorporates covariance matrix methodology. This curve was obtained by changing the beta efficiency from 64.9% to 88.7% with the aid of external absorbers, placed above and below the radioactive sources.

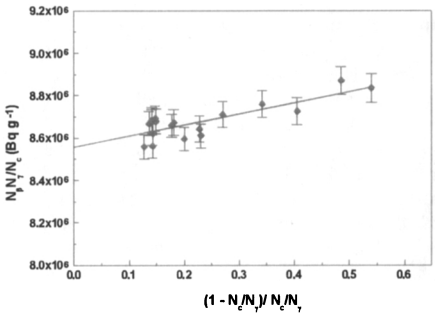

Fig. 5. Extrapolation curves of $N_\beta N_\gamma/N_C$ as a function of $(1- N_C/ N_\gamma)/ (N_C/ N_\gamma)$.

The efficiency parameters and experimental values with uncertainties are presented in Table I.

For the determination of the final activity value it was necessary to use *a* and *b* branching ratio values taken from the literature [6], [7]. However, there are two values which are not in agreement within the claimed uncertainty; therefore we decided to present our results using each one separately. These values, together with the resulting activity and the f_x factor, are presented in Table II. The main uncertainties involved in the measurement are: counting statistics, weighing, dead time, half-life and extrapolation curve efficiency. All these uncertainties are included in the fitting. For this measurement the final activity was obtained with an overall uncertainty of 0.55% which can be considered satisfactory for a secondary calibration system.

TABLE I

VALUES OBTAINED WITH GATE GAMMA (γ = 122 + 137KEV).

Measurement	N_c/N_γ	$(1-N_c/N_\gamma)/N_c/N_\gamma$	$N_\beta N_\gamma/N_C$ (Bq.g^{-1})	σ(%)
1	8 8019E-01	1 3612E-01	8 6668E+06	0.66
2	8 1256E-01	2 3068E-01	8.6105E+06	0 66
3	8 3360E-01	1.9962E-01	8 5942E+06	0 66
4	8 4928E-01	1 7746E-01	8 6577E+06	0 65
5	8 7508E-01	1 4275E-01	8 6201E+06	0 65
6	8 8690E-01	1 2753E-01	8 5596E+06	0 66
7	8 7615E-01	1 4136E-01	8.6831E+06	0.67
8	8.7564E-01	1 4202E-01	8.6736E+06	0.67
9	8 7159E-01	1 4733E-01	8 6914E+06	0 67
10	8 7040E-01	1 4890E-01	8 6777E+06	0 67
11	8 7508E-01	1 4276E-01	8 5607E+06	0 67
12	8.4668E-01	1 8108E-01	8 6711E+06	0 70
13	7 8731E-01	2 7015E-01	8 7087E+06	0 71
14	8 1466E-01	2 2750E-01	8 6423E+06	0 71
15	7 4544E-01	3 4149E-01	8 7583E+06	0 73
16	7 1174E-01	4 0500E-01	8.7252E+06	0 73
17	6.7329E-01	4.8524E-01	8 8702E+06	0 75
18	6 4923E-01	5 4029E-01	8 8347E+06	0 76

TABLE II

VALUES OF *A* AND *B* BRANCHING RATIOS TAKEN FROM THE LITERATURE [6] AND [7]. THE RESULTING ACTIVITY OF THE SOLUTION IS GIVEN IN THE LAST COLUMN AND FACTOR F_X IS SHOWN IN THE LOWEST ROW.

Literature	Parameters	Values	Activity (kBq g^{-1})
Firestone.1996	a (β⁻ branch)	0.9310 ± 0.0020	9049 ± 50
	b (EC branch)	0.0690 ± 0.0020	
Baglin.1997	a (β⁻) branch	0.9253 ± 0.0010	9093 ± 47
	b (EC branch)	0.0747 ± 0.0010	
	Factor f_x	0.2072 ± 0.0059	

Table III compares the results obtained in the present work with the literature. These results are in agreement with those from Woods [10] and Schönfeld [11] within the experimental uncertainties, using branching ratio values from Firestone and Baglin, but are not in agreement with those by Miyahara [12] values.

TABLE III

:RE-186 GAMMA RAY EMISSION PROBABILITY PER DECAY USING BRANCHING RATIO VALUES FROM FIRESTONE AND BAGLIN IN COMPARISON WITH THE LITERATURE.

Gamma ray energy (keV)	This work	Woods [9]	Schöönfeld [10]	Miyahara [11]
122.61	0.00595 (8)^F 0.00592 (8)^B	0.00597(8)	0.00603(6)	0.00604(3)
137.16	0.0938 (13)^F 0.0934 (13)^B	0.0935(10)	0.0939(9)	0.0949(3)

F - Firestone [6] branching ratios
B – Baglin [7] branching ratios

REFERENCES

[1] M. M. Bé , V. Chisté V. et al. *Table of radionucléides*, Monographie BIPM-5, 2005.

[2] S. Baba, et. al., "A method for determination of the ^{152}Eu activity," *Nuclear Instrum. and Methods* A203, pp. 273 -280, 1982.

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

[4] D. S. Moreira *Padronização dos radionuclídeos muiti-emissores gama ^{166m}Ho e ^{72}Ga e determinação de suas intensidades gama por decaimento"*. PhD Thesis (in portuguese) – University of São Paulo, 2005.

[5] A. M. Baccarelli, M. S. Dias, M. F. Koskinas, F. Brancaccio, "Standardization of F-18 by means of 4π(PS)-gamma plastic scintillator coincidence system," *IEEE Transaction on Nuclear Physics* Vol 55, no. 3, pp 1767- 1772, 2008.

[6] R. B. Firestone, and V. S. Shirley, *Table of Isotopes*, 8th ed. John Wiley & Sons, New York, 1996.

[7] C. M. Baglin, *Nuclear Data Sheets for A=186*, 82 1. 1. Article nº DS970019, 1997.

[8] M. S. Dias, *Internal Report - Alpino* - LMN (IPEN), 2001.

[9] M. S. Dias, *Internal Report - Linfit-LMN* (IPEN), 1999.

[10] D. H. Woods, M. Ciocanel, L. J. Husband, J. D. Keightley, P. de Lavison, S. Lineham, M. J. Woods, S. A. Woods, "Standardization and decay data of ^{186}Re," *Applied Radiation and Isotopes*, 52 no 3, pp. 581-584, 2000.

[11] E. Schönfeld et. al. "Standardization and decay data of ^{186}Re," *Nuclear Instruments and Methods*, 339, no 2-3, pp 174-179, 1994.

[12] H. Miyahara, G. Wurdiyanto, H. Nagata, A. Yoshida, K. Yanagida, C. Mori, "Precise measurements of gamma-ray emission probabilities per decay of ^{186}Re and ^{188}Re," *Applied Radiation and Isotopes*, 52 no 3, pp. 573-579, 2000