Disintegration Rate and Gamma Ray Probability per Decay Measurement of ^{166m}Ho

D. S. Moreira, M. F. Koskinas, M. S. Dias and M. N. Takeda

Abstract–**The procedure followed by the Nuclear Metrology Laboratory (Laboratório de Metrologia Nuclear - LMN) at the IPEN-CNEN/SP, in São Paulo, for the standardization in a 4πβ-γ coincidence system and the determination of gamma ray emission probabilities per decay of ^{166m}Ho are described. The standardization by means of a 4πβ-γ system was performed selecting two γ-ray windows: one set at the 184 keV total absorption peak and another one covering 700-830 keV energy range, both following β^- decay processes. The ^{166m}Ho sources were also measured in a ReGe spectrometer previously calibrated with standard sources of ^{241}Am, ^{133}Ba, ^{57}Co, ^{60}Co, ^{152}Eu and ^{54}Mn standardized in the 4πβ–γ coincidence system. The experimental results were compared with data from literature.**

I. Introduction

Gamma-emitting radionuclides are used in several applications such as nuclear medicine, agriculture and industry.

The gamma radiation detection is usually performed by means of spectrometers using HPGe semiconductor detectors or NaI(Tl) scintillation crystals.

For good accuracy, the efficiency calibration must be performed by means of primary standard sources within a gamma energy range specified by the application. These standards sources correspond to radionuclides which have well known decay parameters and suitable half-life. However, in some energy regions there is a need of new standards and in this case secondary standards may be necessary.

The secondary standard ^{166m}Ho has half life of 1200 years and decays by β^- emission, populating the excited levels of ^{166}Er, as shown in Fig. 1. This radionuclide is suitable for calibrating gamma ray spectrometers because it has a long half-life and several gamma-rays in the range of 80 to 1000 keV. Therefore it becomes important to determine experimentally the gamma-ray probability per decay with good accuracy. To contribute with more data the Nuclear Metrology Laboratory (LMN) at the IPEN, São Paulo has carried out measurements of the gamma-ray emission probabilities per decay of ^{166m}Ho which are presented in this paper.

The ^{166m}Ho radioactive solution was first standardized in a 4πβ–γ coincidence system in order to determine the specific activity. Subsequently, radioactive sources prepared from the calibrated solution were measured in a HPGe spectrometer, previously calibrated with standard sources. Monte Carlo methods were used to predict the behavior of the coincidence system and to predict the gamma spectrometer efficiency curve.

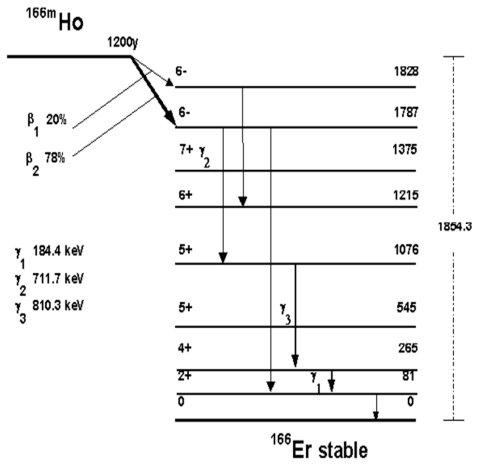

Fig. 1 Simplified decay scheme of ^{166m}Ho [1]. All energies are in keV.

II. Methodology

The ^{166m}Ho solution used for these measurements was taken from an ampoule sent by the Laboratório Nacional de Metrologia das Radiações Ionizantes (LNMRI), from Rio de Janeiro, which in turn have received it from the National Metrology Institute of Japan (NMIJ formerly ETL). This ampoule came from a batch belonging to a regional comparison sponsored by the Bureau des Poids et Mesures (BIPM) in France.

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A. Experimental setup

The system for absolute standardization consisted of a gas-flow proportional counter with 4π geometry and using 90% Ar + 10% CH_4 gas at 0.1 MPa, as the β detector, coupled to a single NaI(Tl) crystal for gamma-ray detection. The events were registered by means of a Time to Amplitude Converter (TAC) associated with a Multi-channel Analyzer [2]. The disintegration rate was obtained by application of the extrapolation technique [3].

The calibration on the 4πβ-γ system was performed selecting two γ-ray windows, one set at the 184 keV total absorption peak and another covering 700 to 830 keV energy range, both following β^- decay process.

The efficiency parameter was varied in the range from 67% to 88 % for the 184 keV gamma window and in the range from 71 % to 88 % for the 700 keV to 830 keV window, by using external absorbers. The extrapolated value gives the disintegration rate N_0.

The observed counting rates were corrected for background, dead time and decay in the usual way. The coincidence rates were corrected for accidentals by using Cox-Isham formalism adapted by Smith [4].

B. Source Preparation

The radioactive sources to be measured in the 4πβ-γ system were prepared by dropping known aliquots of the radioactive solution onto a 20 μg.cm^{-2} thick Collodion film. This film had been previously coated on both sides with a 10 μg.cm^{-2} thick gold layer in order to render the film conducting. 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 Sartorius MC21S balance by the pycnometre technique [5].

C. Gamma spectrometer measurements

The spectrometer system is an ReGe spectrometer with a 500 μm thick Be window, with 1.79 keV FWHM at 1332.5 keV. The gamma-ray full efficiency peak curve was measured at 17.6 cm of source-detector distance, in the range of 59 to 1400 keV, with Collodion sources of ^{241}Am, ^{133}Ba, ^{57}Co, ^{60}Co, ^{152}Eu and ^{54}Mn previously standardized in the 4πβ–γ coincidence system. The ^{166m}Ho sources measured in the ReGe spectrometer were the same used for calibrating the radioactive solution.

The area under the peak was evaluated by code ALPINO [6]. The gamma peak area is obtained by means the simple integration of the counts under the total absorption peak. Dead time and pile-up corrections were applied by measuring a reference pulser peak near the upper edge of the gamma-ray spectrum simultaneously with the sources. The cascade-summing effects were determined by a Monte Carlo calculation [7].

III. Results and Discussions

A. Standardization

Figure 2 and 3 shows the extrapolation curve obtained for the first and second selected γ-windows, respectively.

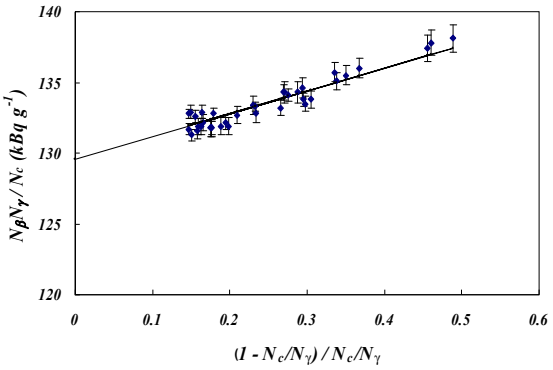

Fig. 2. Extrapolation curve for 184 keV gamma window.

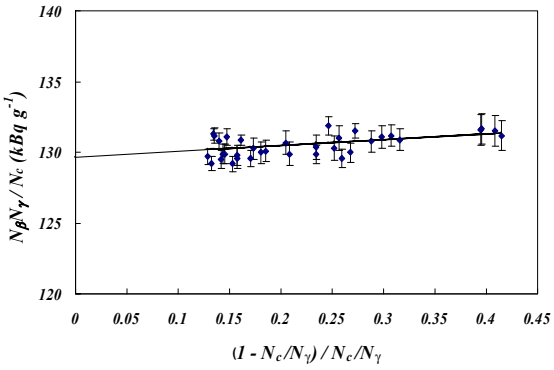

Fig. 3: Extrapolation curve for 700 to 830 keV gamma window.

The final activities were obtained using the program LINFIT [8]. In the fitting the uncertainties were considered as partial errors in: weight, beta efficiency, background and dead time. Table I present typical uncertainties in percentage. The final uncertainty is given by means the covariance methodology [9].

Table I. Typical uncertainties in percentage of the two gamma windows selected.

COMPONENT	UNCERTAINTY (%)		
	TYPE A		TYPE B
GAMMA WINDOW	1	2	BOTH
COUNTING STATISTICS (INCLUDED IN THE FITTING)	*	*	
WEIGHT			0.10
DEAD TIME			0.05
BACKGROUND	0.30	0.16	
EXTRAPOLATION CURVE	0.22	0.26	
TOTAL	0.37	0.31	0.11

Table II presents the fitting parameters obtained for each extrapolation curve, the activity for the two gamma windows and the average activity value.

Table II. Fitting parameters obtained with two gamma windows.

GAMMA WINDOW	ACTIVITY ($kBqg^{-1}$)	SLOPE
1	129.54 (28)	0.124(9)
2	129.66 (34)	0.031(12)
AVERAGE	129.60 (35)	

The reliability of the activity measurement was checked by comparing this result with those from "The 2000 Regional Comparison of ^{166m}Ho" [10] sponsored by BIPM . The degree of equivalence of each laboratory with respect the SIR reference value is given by the expression Di = [Aei - KCRV]/ MBq , where KCRV is the Key Comparison Reference Value which is derived from the unweighted mean of all the results submitted to the SIR (International Reference System) and Aei is the equivalence activity of each laboratory. The degree of equivalence is presented in the Fig. 4, adding the result of the present experiment.

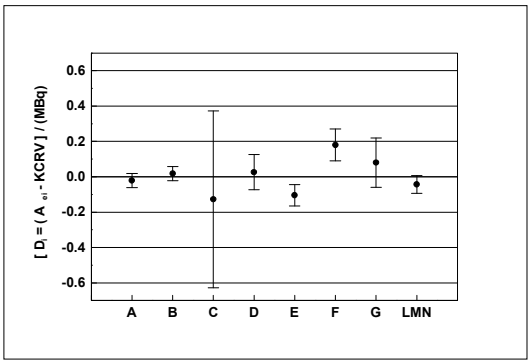

Fig.4: Results of the 2000 Regional Comparison of ^{166m}Ho. The Laboratories are identified in [2]

The average activity of the radioactive solution presented in Table I is in excellent agreement within the experimental uncertainty with the reference value of the SIR (KCRV) 130.16 (25) kBq g^{-1} [10].

B. Gamma probability per decay

The peak efficiency curve as a function of the gamma-ray energy was determined by a least square method, fitting a 5th degree polynomial and applying the covariance methodology by means of the LOGFIT code [11]. The reduced chi-square was 1.16, the interpolated efficiency uncertainty ranged from 0.92% to 0.38%. The peak efficiency curve is shown in Fig. 5, compared with a theoretical efficiency curve calculation carried out by means of the Monte Carlo simulation using MCNP-4C code [12].

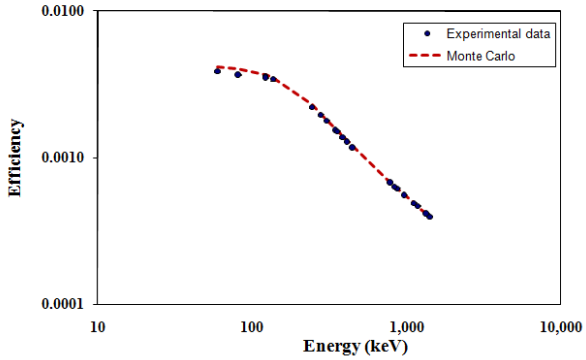

Fig 2. Peak efficiency curve as a function of the gamma-ray energy. The dots are the experimental data and the Monte Carlo curve is presented as a dot line.

The residues of the experimental efficiency curve are presented in Fig. 6. As can be seen the experimental curve does not present any bias when compared with the fitted value.

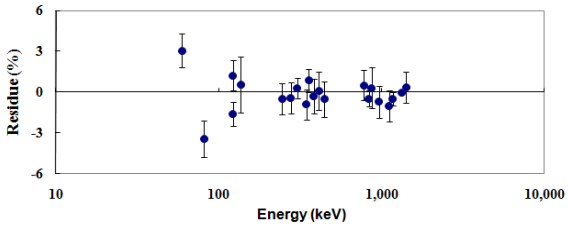

Fig.6: Residues in percentage in the HPGE peak efficiency curve as function of the gamma-ray energy, the errors bars in the figure correspond to the experimental uncertainty (u=1).

TABLE III ^{166m}Ho gamma emission probabilities per decay in comparison with the literature.

Gamma Energy (keV)	This work	Morel et al. 1996[13]	Hino et al. 2000[14]	Bernardes 2002[15]	BIPM-5 2004[16]
80.6	12.31(14)	12.06(8)	11.84(16)	11.68(10)	12.66(23)
184.4	71.64(96)	70.21(35)	72.4(7)	72.60(47)	72.5(3)
280.46	29.01(22)	28.55(14)	29.7(3)	29.30(15)	29.54(25)
410.96	11.27(9)	11.10(6)	11.39(13)	11.17(6)	11.35(17)
451.54	3.01(5)	2.852(26)	-	-	2.915(14)
529.83	9.65(8)	9.36(5)	9.63(11)	9.35(5)	9.4(4)
571.00	5.49(6)	5.41(3)	5.54(8)	5.42(3)	5.43(20)
670.53	5.35(6)	5.31(3)	5.65(9)	5.32(3)	5.34(21)
711.70	54.43(32)	53.6(3)	56.0(5)	53.8(2)	54.9(9)
752.28	12.16(11)	11.92(6)	12.27(15)	11.98(6)	12.2(3)
778.83	2.96(5)	2.978(18)	-	3.019(18)	3.01(8)
810.29	57.31(40)	56.4(3)	58.2(5)	56.6(3)	57.3(11)
830.57	9.75(9)	9.58(5)	9.77(12)	9.56(5)	9.72(18)
950.99	2.71(5)	2.663(16)	-	2.693(19)	2.744(19)

The number inside the parenthesis correspond to the uncertainty in the last digits (u=1).

In Table III, the results obtained for the emission probabilities of the most intense gamma lines are presented with literature data for comparison.

IV. CONCLUSIONS

The ^{166m}Ho standardization method adopted by the LMN can be considered reliable because it showed good consistency when compared with results from other laboratories, as shown in Fig. 4.

In the gamma emission probabilities per decay of the ^{166m}Ho, the values are not consistent within the experimental uncertainties, suggesting the need of more measurements.

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