PRIMARY STANDARDISATION OF ^{75}Se RADIOACTIVE SOLUTION

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ABSTRACT

The procedure followed by the Laboratório de Metrologia Nuclear (LMN) at the IPEN - CNEN/SP, in São Paulo, for the standardisation of the ^{75}Se is described. This radionuclide has been selected for an international comparison sponsored by the Bureau International des Poids et Mesures (BIPM) due its importance in medicine and other fields and also because of the complexity in decay scheme characteristics. The LMN have participaded in this comparison in collaboration with the Laboratório Nacional de Metrologia das Radiações Ionizantes (LNMRI), in Rio de Janeiro. The calibration system was composed of a 4π pressurized proportional counter coupled to a NaI(Tl) crystal operating in coincidence. The proportional counter efficiency was extrapolated to 100% by changing the electronic threshold above 2 keV. The results obtained by our laboratory agree with those from other laboratories, as shown in the BIPM report.

I INTRODUCTION

The ^{75}Se is an important radionuclide used in medical applications and also as a standard nuclide for energy and efficiency calibration of germanium spectrometers due to its numerous well defined gamma-ray energies in the range of 66 to 400 keV. Nevertheless due to its complex decay scheme, the determination of activity with a good accuracy is not straightforward. To solve the problem, the Bureau International des Poids et Mesures (BIPM) sponsored an international comparison of this radionuclide. The Laboratório de Metrologia Nuclear (LMN) at the IPEN-CNEN/SP took in part this comparison in collaboration with the Laboratório Nacional de Metrologia das Radiações Ionizantes (LNMRI), in Rio de Janeiro.

The decay scheme of ^{75}Se is shown in Figure 1 (1). This radionuclide decays by electron capture to several levels of ^{75}As, with a meta-stable state which has a half-life of 17 ms at 304 keV.

The LMN has undertaken the measurement using the 4π pressurized proportional counter 4π(PPC)β-γ coincidende system, described in the following section

II EXPERIMENTAL METHOD

Source preparation The ^{75}Se radioactive solution was supplied by the BIPM which distribuited flame-sealed ampoules filled with about 3.6 g of solution and having approximately 1.2 MBq/g, to the participant laboratories. The sources were prepared from the original solution by depositing appropriated amounts of radioactive solution in droplet form on metal-coated Collodion film 20μg/cm^2 thick. Barium chloride or hydrazine were used in order to precipitate the ^{75}Se as barium selenite or selenium metal. The sources had a mass range from 10 to 30 mg. A seeding agent (CYASTAT SM) was used to improve the deposit uniformity and the sources were dried in a dissecator. The accurate source mass determination was performed using the picnometer technique (2).

4π(PPC)β-γ MEASUREMENTS The measurements were carried out in a 4π(PPC) X-γ coincidence system consisting of a pressurized proportional counter, coupled to a 3"x3" NaI(Tl) crystal.

The proportional counter has been operated with a voltage of 3 kV in 4π geometry. A flow of argon/methane, at a mixing ratio 9:1 and a fixed pressure of 0.6 MPa has been mantained in the counter. A lower threshold has been set at 2 keV, to cut off electronic noise.

The measurements in the proportional counter has been made in integral mode, to avoid the lost of summing effects produced by electrons Auger and conversion electrons which have been detected simultaneously, yelding a higher pulse.

For gamma detection with the 3"x3" NaI(Tl) scintillator counter, two windows were selected from 150 to 450 keV and from 340 to 450 keV.

The activity of the solution has been obtained by means of the extrapolation method described by Baerg (3). The proportional counter efficiency was extrapolated to 100% by changing the electronic threshold above 2 keV.

The equations developed for discribing the count rates are described as follows.

For the proportional counter the number of collected events is given by

$$N_X = N_0\left[\varepsilon_c + (1 - \varepsilon_{co})p_p + p_{Do}\varepsilon_{Do} + p_{Di}\right] \qquad (1)$$

For each window set in γ channels, the number of registered events $N_{\gamma i}$ is:

$$N_\gamma = N_0 \sum_{i \neq \gamma 11} \varepsilon_{\gamma i} p_i \qquad (2)$$

The number of coincidence events is given by:

$$N_c = N_0 \varepsilon_c \left[\sum_{i \neq \gamma 11} \varepsilon_{\gamma i} p_i + \varepsilon_{\gamma 11} p_{\gamma 11}\right] \Big/ \sum_{i \neq \gamma 11} \varepsilon_{\gamma i} p_i \qquad (3)$$

In these three equations, N_0 refers to the source activity; ε_c is the detection efficiency for X-ray, electrons Auger and prompt conversion electrons, ε_{co} is the detection efficiency of events at the beginning of electron spectrum, p_P is the emission probability of prompt conversion electrons (ce), p_{Do} is the emission probability of delayed conversion electrons, ε_{Do} is the detection efficiency of the delayed events of probability p_{Do}, $\varepsilon_{\gamma i}$ is the gamma efficiency of the i-th gamma-ray and p_i its corresponding emission probability. The terms have been corrected for the background, dead time , decay, as usual. For accidental coincidences the Cox-Isham (4) formalism has been applied.

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

$$N_0 = \frac{N_x N_\gamma}{N_c}(1 - k_\varepsilon)^{-1} f_p \qquad (4)$$

where $k_\varepsilon = \dfrac{(1 - \varepsilon_c)}{\varepsilon_c} C$,

$$C = (1 - \varepsilon_{co})p_p + p_{Do}\varepsilon_{Do} + p_{Di}$$

and $f_p = 1 + (1 - \varepsilon_{co})p_p + p_{Do}\varepsilon_{Do} + p_{Di}$

when $\varepsilon_c = 1$.

Figure 2 shows a plot of the $N_x N_\gamma / N_c$ versus inefficiency parameter $(1 - \varepsilon_c)/\varepsilon_c$ for the two gamma windows. The curve A represents the measurement of the window set from 150 to 450 keV and the curve B the window set from 340 to 450 keV.

As shown in the equation 4, the extrapolation to 100% efficiency do not lead to the true activity, because delayed events are present in the X-channel but are not present in the coincidence channel. To reach the true value of the activity, corrections for this delayed events have been applied, making use of values of the intensity taken from the literature (1). These coretions led to a higher uncertainty because the feeding of the delayed level is not well defined.

For the window set between 150 to 450 keV another correction has been applied due to the detection of delayed gamma-rays.

An additional correction has been applied to correct for the efficiency of the proportional counter to gamma rays emitted by ^{75}Se.

III RESULTS AND DISCUSSION

Table 1 shows the results obtained for the two gamma windows, as well as values of the applied corrections.

Table 1 Results of activity of ^{75}Se

gamma window (keV)	Correction for delayed events (%)	corr. for $\varepsilon_{\beta\gamma}$ (%)	Activity* (Bq/mg)
150-450	7.35±1.24 [a] 1.32±1.24 [b]	0.26	1233 ± 17
340-450	7.35±1.24 [a]	0.26	1223 ± 15
		Mean	1228 ± 16

* ref. Date 06/01/92 0 h UT

a - electrons

b - gamma

Table 2 shows the partial uncertainties envolved in the measurement . As it can be seen, the main source of uncertainty are from the correction of delayed events, that suggests that these values should be obtained with better accuracy.

Table 3 shows the results obtained by our laboratory and the result of an unweighted mean value of results from all laboratories which took part this comparison, without and with corrections for delayed events. As it can be seen, our results are in agreement with these values. The uncertainty of BIPM mean value is greater than our uncertainty because of the large spread among the results from different laboratories as described in the BIPM report (5).

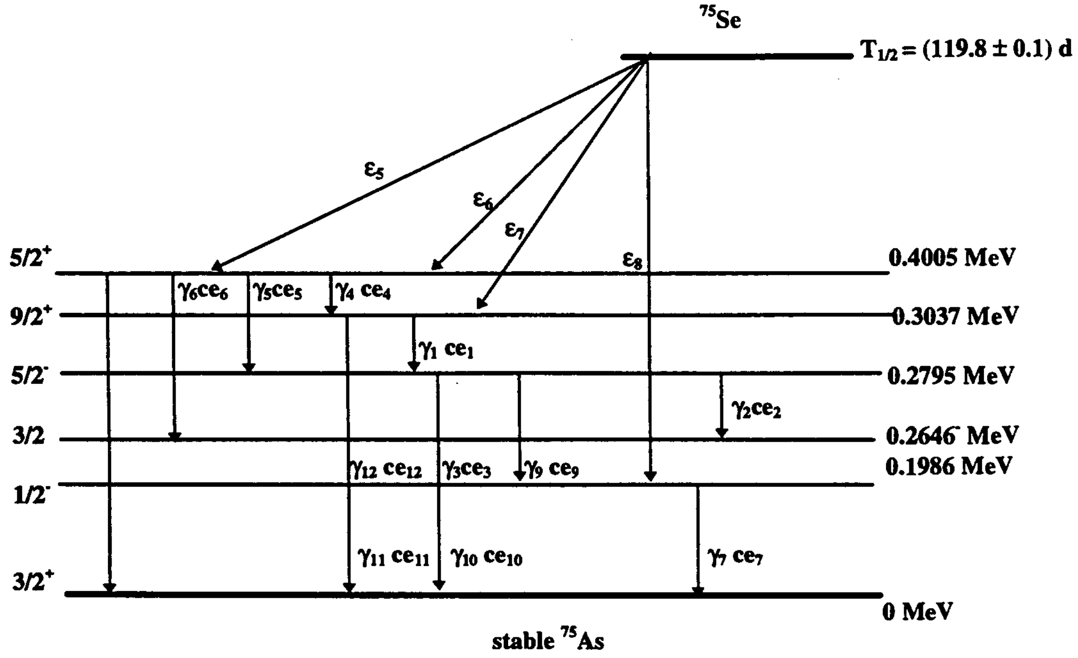

Figure 1 Decay scheme of ^{75}Se

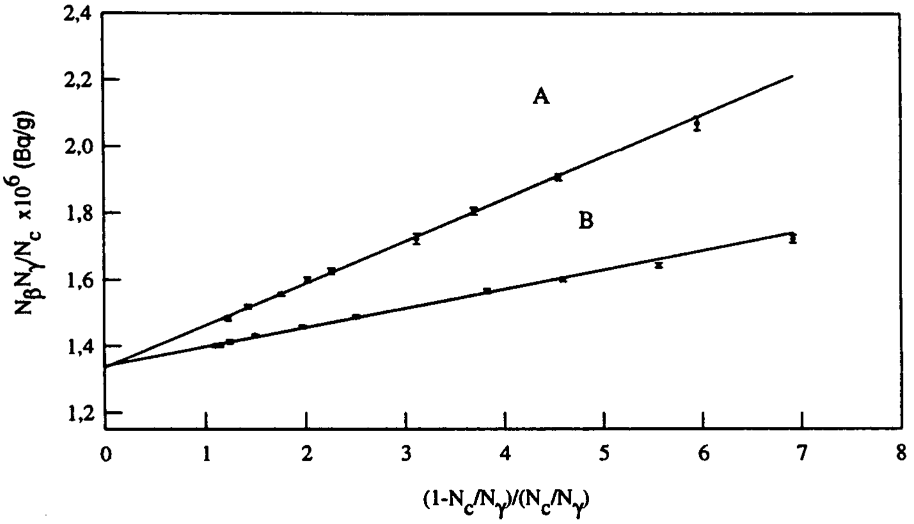

Figure 2 Extrapolated curves obtained for two gamma windows

Table 2 Partial uncertainties envolved in the activity of ^{75}Se

Source of uncertainty	γ window A (%)	γ window B (%)
statistics	0.20	0.05
mass	0.15	0.15
dead time	0.0001	0.0001
background	0.034	0.030
measurering. time	0.02	0.02
resolution time	0.002	0.002
extrapolation	0.48	0.24
half-life	0.043	0.043
scheme parameters*	1.2	1.2
self absorption	0.58	0.58
delayed γ-rays	0.1	0.1
$\varepsilon_{\beta\gamma}$	0.02	0.02
Total (c)	0.80	0.65
Total (d)	1.44	1.36

(c) without uncertainties of corrections for delayed events
(d) with uncertainties of corrections for delayed events
* delayed events.

Table 3 Comparison with results from the BIPM international comparison

	Activity* (Bq/mg)	
	Without	With
This work	1330 ± 11	1.228 ± 16
BIPM mean	1338 ± 26	1.253 ± 21

* ref. Date 06/01/92 0 h UT

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