Applied Radiation and Isotopes 87 (2014) 192–194

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Standardization and determination of the total internal conversion coefficient of In-111

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HIGHLIGHTS

- Primary standardization of ^{111}In with a 4π(PC)-HPGe coincidence system.
- Software coincidence counting system applied.
- Extrapolation curves for 171 keV and 245 keV gamma-ray windows determined.
- Conversion coefficients for 171 keV and 245 keV transitions determined experimentally.

ARTICLE INFO

Available online 11 December 2013

Keywords:
Primary standardization of In-111
Software coincidence counting
Total internal conversion coefficient

ABSTRACT

The standardization of ^{111}In by means of a 4πβ-γ coincidence system, composed of a proportional counter in 4π geometry, coupled to a 20% relative efficiency HPGe crystal, for measuring gamma-rays is presented. The data acquisition was performed by means of the software coincidence system (SCS) and the activity was determined by the extrapolation technique. Two gamma-ray windows were selected: at 171 keV and 245 keV total absorption peaks, allowing the determination of the total internal conversion coefficient for these two gamma transitions. The results were compared with those available in the literature.

1. Introduction

In recent years, the Nuclear Metrology Laboratory (LMN) of IPEN, has been involved in developing procedures for standardization of radionuclides which decay by electron capture (EC) process, in a program for standardization of important radionuclides used in nuclear medicine (Brito et al., 2012; Koskinas et al., 2012). The standardization of ^{111}In is presented.

^{111}In decays with a half-life of 2.8049 days (Bé et al., 2006) by electron capture process, populating the excited levels of ^{111}Cd, and emitting two main gamma-rays with energies of 171 keV and 245 keV, as shown in Fig. 1.

The measurement was carried out using a 4πPC-HPGe coincidence system. The use of the HPGe instead of the usual NaI(Tl) makes possible the determination of the activity by means of the selection of the two gamma-rays separately and the determination of the total internal conversion coefficient of each gamma transition obtained by the slope of each extrapolation curve. The data acquisition was performed using the software coincidence system developed at LMN (Toledo et al., 2007).

The total internal conversion coefficients of 171 keV and 245 keV transitions were determined and compared with the values from the literature.

2. Experimental set up

The coincidence system is composed of a proportional counter in 4π geometry, filled with P-10 gas (90% argon + 10% methane) and operated at 0.1 MPa for detecting the Auger electrons and X-rays emitted from the electron capture process, coupled to a 20% relative efficiency HPGe detector, positioned close to the PC wall, for measuring gamma-rays.

The data acquisition was performed by means of the software coincidence system (SCS) based on a National Instruments PCI-6132 card (equipped with four independent analog inputs) and interfaced by means of LabView Version 8.5 acquisition program, details of this system can be found in Toledo et al. (2007) and in Dias et al. (this issue) from the present conference proceedings.

In this system, the signals from the two amplifiers (572 Ortec amplifier, with shaping time equal to 2 μs) are sent to an interface

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0969-8043/$ - see front matter
http://dx.doi.org/10.1016/j.apradiso.2013.11.136

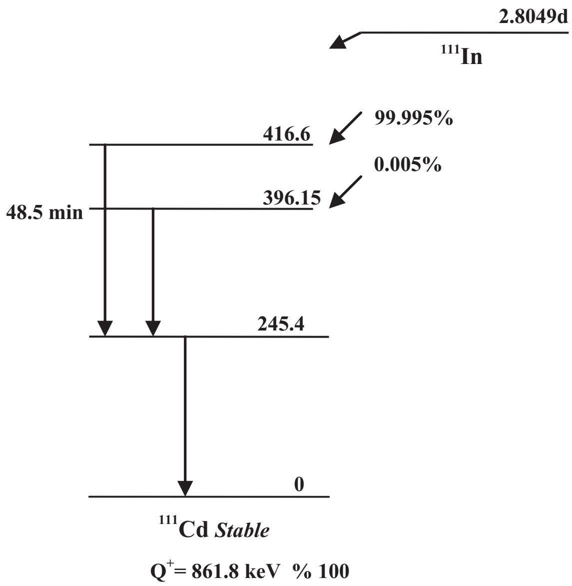

Fig. 1. Decay scheme of ^{111}In (Bé et al., 2006), all energies are given in keV.

connected by a special cable to the PCI 6132 card; the information on pulse height and time of occurrence are registered for both beta and gamma channels. After being acquired, this information is processed by SCTAC code version 6.0 (Dias, 2010), developed at LMN.

By means of the SCTAC code it was possible to reconstruct the beta and the gamma spectra, select the gamma-ray windows, as well as to vary the beta discrimination, if necessary. The corrections for dead time (non-extendable), decay and resolution time were also calculated by this code according an off-line processing for source activity calculation.

The 4π(PC) detection efficiency was changed by using external absorbers on both sides of the radioactive sources. These absorbers were made of collodion films previously coated with a 20 μg cm^{-2} gold layer, or thin aluminum foils. The detection efficiency in the β-channel given by $(N_c/N_\gamma)_i$ was varied between 13% and 40% for the 171 keV total absorption energy peak, and between 15% and 42% for the 245 keV total absorption energy peak.

For the ^{111}In, there are two gamma transitions in cascade and, in addition of the coincidence between the gamma ray selected with the electron capture process, we have to consider the coincidences occurring between both gamma transitions due to total internal conversion events. Therefore the formulae to be applied are those suggested by Kawada and Hino (1985)

$$\left(\frac{N_{4\pi}N_\gamma}{N_c}\right)_i = N_0\left\{1+\left(\frac{1-\varepsilon_{4\pi}}{\varepsilon_{4\pi}}\right)\left(\frac{\alpha\varepsilon_{ce}+(\varepsilon_{4\pi})_\gamma}{1+\alpha}\right)\right\}_i \quad (1)$$

where:

$$(\varepsilon_{4\pi})_i = \left(\frac{N_c}{N_\gamma}\right)_i = \varepsilon_{ec} + (1-\varepsilon_{ec})\sum_i \times\left[\prod_j\left(1-\frac{\alpha_T\varepsilon_{ce}+(\varepsilon_{4\pi})_\gamma}{1+\alpha_T}\right)_{j\neq i}\left(\frac{\alpha_T\varepsilon_{ce}+(\varepsilon_{4\pi})_\gamma}{1+\alpha_T}\right)_i\right] \quad (2)$$

The suffixes i and j indicate as suggested by Kawada and Hino (1985) that each notation is to be made for the ith and jth transition, respectively.

N_o is the disintegration rate;
$N_{4\pi}$ proportional counter counting rate;
N_γ γ-channel counting rate of the ith gamma selected;
N_c coincidence counting rate;
ε_{ec} proportional counter efficiency for electron capture events (Auger electrons and X-ray emission);
$(\varepsilon_{4\pi})_{\gamma i}$ proportional counter efficiency for γ-rays;
α_{Ti} total internal conversion coefficient of the ith transition;
ε_{cei} proportional counter efficiency for conversion electron from the ith transition.

The plot of $(N_{4\pi}\,N_\gamma/N_c)_i$ versus $((1-N_c/N_\gamma)/N_c/N_{\gamma i}$ results in a straight line. From the slope B to intercept A ratio ($C=B/A$) the α_{Ti} for the ith transition can be obtained.

$$C = \left(\frac{\alpha_T\varepsilon_{ce}+(\varepsilon_{4\pi})_\gamma}{1+\alpha_T}\right)_i \quad (3)$$

For this purpose, was considered the value of $\boldsymbol{\varepsilon_{ce\ i}}=1$ and the value of $(\varepsilon_{4\pi})_{\gamma t}$ has been calculated by Monte Carlo code MCNPX (ORNL, 2006).

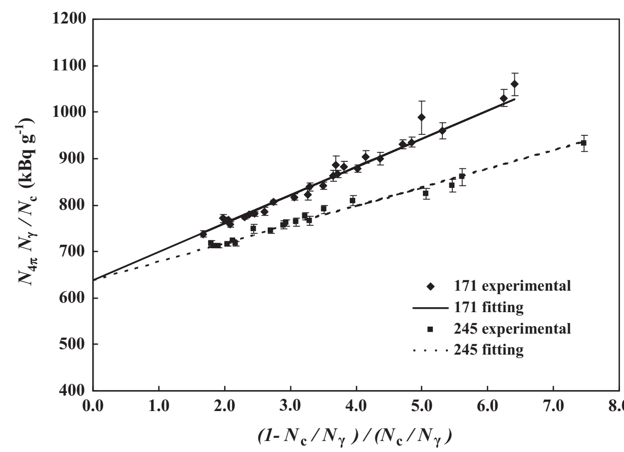

Fig. 2. Extrapolation curves of $N_{4\pi}\,N_\gamma/N_c$ as a function of $(1-N_c/N_\gamma)/N_c/N_\gamma$ from the two windows.

Table 1
Fitting parameters obtained for the two selected γ-windows, by means of least squares fitting, with the corresponding correlation matrix.

Window	Fitting parameters		Correlation matrix	
171 keV	A	$6.391(54)\times10^5$	1	
	B	$6.07(21)\times10^4$	−0.963	1
	C	0.0951(34)		
245 keV	A	$6.380(37)\times10^5$	1	
	B	$4.00(16)\times10^4$	−0.957	1
	C	0.0627(25)		

Table 2
Typical partial uncertainties in the activity, in percent ($k=1$).

Component	Uncertainty (%)	Comments
Counting statistics	-	Statistics in $N_{4\pi}$ and N_c/N_γ included in the extrapolation fitting
Weighing	0.15	Balance certificate
Dead time	0.10	Software
Background	0.20	Counting statistics
Decay	0.01	Half-life
Resolving time	0.10	Statistics in accidental coincidence corrections
Extrapolation of efficiency curve	0.80	Least square fit error
Combined uncertainty	0.85	

Table 3
Results of total internal coefficient of 171 keV and 245 keV transitions, in comparison with the literature.

Energy (keV)	This work	Steffen 1956	Sparrman et al. (1966)	Shevelev et al. (1975)	Kawada and Hino (1985)	Raman et al. (2006) (theory)	Chechev (2006) (evaluation)
171	0.1038 (34)	0.099 (3)	0.100 (3)	0.124 (7)	0.1018 (13)	0.1068 (15)	0.1036 (24)
245	0.0657 (26)	0.0621 (15)	0.0618 (15)	0.0634 (30)	0.0620 (9)	0.0637 (9)	0.0625 (7)

The observed counting rates $N_{4\pi}$ and N_{γ} were corrected for background, dead time and decay. The coincidence rate N_c was corrected for dead time (non-extendable, defined by software) and accidental coincidences applying Cox and Isham (1977) formalism and adapted by Smith (1978).

2.1. Source preparation

The $^{111}InCl_3$ solution was supplied by the IPEN Radiopharmaceutical Center and diluted in distilled H_2O. The radioactive sources were prepared by dropping known aliquots on collodion substrate, previously coated with 10 μg cm^{-2} gold layer on both sides to make the film conductive. The source masses were determined by the pycnometer technique. A seeding agent (Cyastat SM) was used to improve the deposit uniformity and the sources were dried under a nitrogen jet at 45 °C (Wyllie et al., 1970). Two batches of different solutions were prepared with four sources prepared for each batch with masses ranging from 15 mg to 60 mg. The measurements were normalized by means of each extrapolation curve intercept.

3. Results and discussion

The extrapolation curves obtained for two gamma ray selected, are presented in Fig. 2. The fitting parameters for each curve were obtained by linear least-squares by means of code LINFIT (Dias, 1999) which incorporates covariance matrix methodology. Table 1 shows the extrapolation parameters and slope values obtained for the two selected gamma windows, together with the corresponding correlation matrix. The specific activity (noted A in Table 1) obtained with the two gamma windows are in agreement within the experimental uncertainties.

The slope of each extrapolation curve, after subtraction of $(\varepsilon_{4\pi})_{\gamma i}$ (calculated by MCNPX to be 0.0014(4) for the 171 keV and 0.0012 (3) for the 245 keV, respectively), yielded the total internal conversion coefficient α_T for the 171 keV and 245 keV transitions. SCS method has the advantage, due to off-line analysis, of reducing the contribution from Compton events from the 245 keV gamma-ray by subtracting the background below the 171 keV gamma-ray peak.

The main uncertainties involved in the measurements were: counting statistics, dead time, half-life and extrapolation curve efficiency. All these uncertainties were included in the uncertainty budget shown in Table 2. The main component to the overall uncertainty of the total internal conversion coefficients is caused by the large extrapolation interval due to the low efficiency of the proportional counter.

The comparison between the present results for the total internal conversion coefficient of the 171 keV and 245 keV and the values from the literature are shown in Table 3. As it can be seen our result for the total internal conversion coefficient of the 171 keV transition is in agreement with the values from the literature except with the Shevelev et al. (1975) value. The present result for the total internal conversion coefficient of the 245 keV is in agreement with the other literature values within the experimental uncertainties.

4. Conclusions

The ^{111}In solution was standardized successfully with good accuracy by means of the software coincidence system. The total internal conversion coefficient of 171 keV and 245 keV transitions were measured by an absolute method and the results are in agreement with the literature.

Acknowledgements

The authors are grateful to the IPEN Radiopharmacy Center for supplying the ^{111}In solution and to the National Council for Scientific and Technological Development (CNPq) for partial support to the present research project.

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