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Disintegration rate and gamma ray emission probability per decay measurement of ^{123}I

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ABSTRACT

A series of ^{123}I measurements have been carried out in a $4\pi(e_A,X)-\gamma$ coincidence system. The experimental extrapolation curve was determined and compared to Monte Carlo simulation, performed by code ESQUEMA. From the slope of the experimental curve, the total conversion coefficient for the 159 keV total gamma transition, α_{159}, was determined. All radioactive sources were also measured in an HPGe spectrometry system, in order to determine the gamma-ray emission probability per decay for several gamma transitions. All uncertainties involved and their correlations were analyzed applying the covariance matrix methodology and the measured parameters were compared with those from the literature.

1. Introduction

One of the research areas under development by the Nuclear Metrology Laboratory (Laboratório de Metrologia Nuclear—LMN) at the Nuclear and Energy Research Institute (IPEN) in São Paulo, Brazil, is the primary standardization of radionuclides applied to Nuclear Medicine. In this context the radionuclide ^{123}I plays an important role because it is routinely supplied by IPEN to Nuclear Medicine Services for diagnosis of cardiac metabolism and functional and morphological thyroid studies. Moreover, it is being produced routinely at the IPEN Cyclone-30 cyclotron, by means of ^{124}Xe (p, 2n) ^{123}Cs reaction, followed by $^{123}Cs \rightarrow ^{123}I$ decay. For this reason, there was a need to develop its standardization by a primary method.

The radionuclide ^{123}I decays solely by electron capture process, with a half-life of 13.2234(37) h, mainly to the first excited state of ^{123}Te (97.18%) followed by a 158.99 keV gamma transition (Chisté and Bé, 2004a), as shown in Fig. 1. The intensity of this transition amounts to 99.22(30)% and it is partly converted with α_{159} equal to 0.1918(18). Several low yield gamma transitions are also present comprising the energy interval from 174 keV to 1068 keV and with probabilities per decay in the range from 0.0006% to 1.3%.

The LMN has two $4\pi\beta-\gamma$ coincidence systems composed of gas-flow or pressurized 4π proportional counters coupled to a single or a pair of NaI(Tl) scintillation counters. The latter may be replaced by an HPGe detector for high resolution measurements. A series of ^{123}I measurements have been carried out with one of these systems. The experimental extrapolation curve was determined and compared to Monte Carlo simulation, performed by code ESQUEMA (Takeda et al., 2005; Dias, et al., 2006). From the slope of the experimental curve, the total conversion coefficient for the 159 keV gamma transition was determined.

All radioactive sources were also measured in an HPGe spectrometry system. The ^{123}I radioactive solutions were checked for the presence of impurities by means of this system and a series of measurements were performed in order to obtain the emission probability per decay for several gamma-rays. All uncertainties involved and their correlations were analyzed applying the covariance matrix methodology (Smith, 1991) and the measured parameters were compared with those from the literature.

2. Methodology

2.1. Coincidence equations

Full description of the coincidence equations can be found elsewhere (E.G. Campion, 1959; Baerg, 1966, 1967, 1973). In the case of ^{123}I, the equations were already described in the literature (Reher et al., 1984). Neglecting low intensity gamma transitions, and selecting the gamma window to cover only the total energy absorption peak at 159 keV, the formulae can be given by:

$$N_{4\pi} = N_0\left\{\varepsilon_{EC} + (1-\varepsilon_{EC})\left[\frac{\alpha_{159}}{1+\alpha_{159}}\varepsilon_{ec} + \frac{1}{1+\alpha_{159}}(\varepsilon_{4\pi})_\gamma\right]\right\} \quad (1)$$

$$N_\gamma = N_0\varepsilon_\gamma \quad (2)$$

$$N_c = N_0\varepsilon_{EC}\varepsilon_\gamma \quad (3)$$

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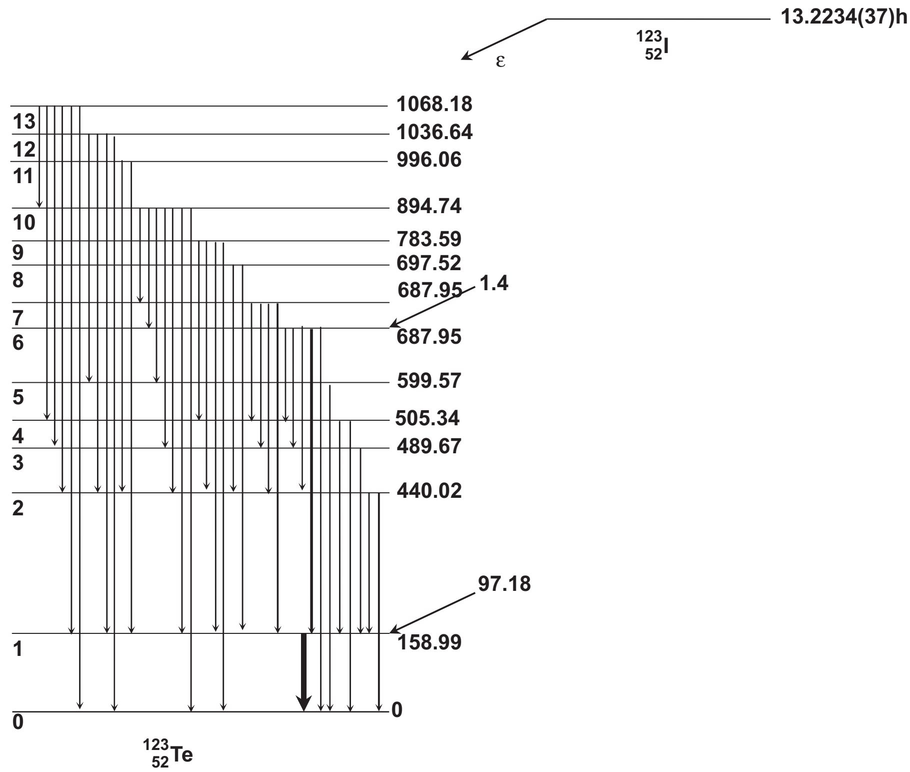

Fig. 1. Decay scheme of ^{123}I (Chisté and Bé, 2004a). The numbers on the right are the energy levels, in keV. The numbers close to the arrows correspond to electron capture decay probability for the main transitions, in percent.

where N_0 is the source disintegration rate; $N_{4\pi}$ is the 4π(PC) counting rate; N_γ and N_c are the gamma and coincidence counting rates, respectively; ε_{EC} is the PC total efficiency including X-rays and Auger electrons; ε_γ is the 159 keV gamma-ray peak efficiency; α_{159} is the total conversion coefficient, ε_{ec} is the conversion electron efficiency and $(\varepsilon_{4\pi})_\gamma$ is the PC gamma-ray efficiency for 159-keV transition. Combining Eqs. (1)–(3), the final expression is given by:

$$\frac{N_{4\pi}N_\gamma}{N_c} = N_0\left\{1+\frac{(1-\varepsilon_{EC})}{\varepsilon_{EC}}\left[\frac{\alpha_{159}}{1+\alpha_{159}}\varepsilon_{ec}+\frac{1}{1+\alpha_{159}}(\varepsilon_{4\pi})_\gamma\right]\right\} \quad (4)$$

In the extrapolation limit where $(1-\varepsilon_{EC})\rightarrow 0$, the value of N_0 can be determined. From the slope of the experimental extrapolation curve, it is possible to obtain α_{159}. For this purpose, the value of $(\varepsilon_{4\pi})_\gamma$ has been calculated by Monte Carlo code MCNPX (ORNL, 2006), as explained in Section 2.4. The calculated value of ε_{ec} by MCNPX resulted close to unity, as expected. Possible contribution from Compton scattered photons with higher energies might affect the slope of the extrapolation curve. However, this effect was estimated to be negligible by Monte Carlo simulation, as described in Section 2.4. Corrections for dead time and accidental coincidences were applied according to formulae taken from the literature (Smith, 1978).

2.2. Gamma-ray emission probability per decay

In the case of ^{123}I, the total gamma transition from the first excited state of ^{123}Te to ground state (159 keV) has probability per decay close to 99%. Therefore, if the probabilities per decay of the other transitions that reach the ground state are known, the value for 159 keV can be obtained by subtraction from 100%. The gamma-ray emission probability per decay for this transition will depend on the total gamma transition probability and on the value of the internal conversion coefficient α_{159}. On the other hand, this latter parameter can be obtained from the slope of the extrapolation curve, as explained in Section 2.1. Therefore, in the present paper, the gamma emission probability per decay for the 159 keV transition was obtained from the gamma transition probabilities per decay of other low yield transitions, combined with the experimental value of α_{159}, as follows:

$$I_\gamma(159\,\text{keV}) = \frac{1}{1+\alpha_{159}}\left(1-\sum_{i=1}^{n}P_{\gamma i}\right) \quad (6)$$

where $P_{\gamma i}$ is the gamma transition probability per decay of the *i*-th gamma transition which reaches the ^{123}Te ground state; *n* is the total number of these gamma transitions. From Fig. 1, it can be observed that the corresponding gamma-ray energies are 440.02, 505.34, 599.69, 687.95, 783.62, 894.8, 1036.64 and 1068.18 keV. All of these transition probabilities were determined in the present paper, considering the gamma-ray emission probabilities per decay determined experimentally and the conversion coefficients taken from the literature (Chisté and Bé, 2004a). The theoretical internal conversion coefficients were calculated by ICC Computer code (program Icc99v3a-GETICC) (Chisté and Bé, 2004b). The validity of Eq. (6) depends on the assumption that there is no direct feeding of the Te ground state during the ^{123}I electron capture decay. This assumption was considered on the basis of decay scheme information from the literature (Chisté and Bé, 2004a). The gamma-ray emission probability per decay for the 159 keV transition obtained directly from the activity value and HPGe measurements has not been included in the present results because its uncertainty turned out to be 0.89% which is much higher than if Eq. (6) is used (0.27%).

In order to obtain the gamma-ray emission probability per decay for the low yield transitions, a series of measurements were carried out in a gamma-ray spectrometry system consisting of a thin Be window HPGe detector with 20% relative efficiency. The radioactive sources were positioned in a well-defined geometry, approximately 18 cm away from the detector front face. At this distance, the corrections for cascade summing are expected to be