Determination of the [121]Te gamma emission probabilities associated with the production process of radiopharmaceutical NaI[^{123}I]

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Abstract: The ^{123}I is widely used in radiodiagnostic procedures in nuclear medicine. According to Pharmacopoeia care should be taken during its production process, since radionuclidic impurities may be generated. The [121]Te is an impurity that arises during the ^{123}I production and determining their gamma emission probabilities (Pγ) is important in order to obtain more information about its decay. Activities were also obtained by absolute standardization using the sum-peak method and these values were compared to the efficiency curve method.

1. Introduction

The presence of impurities that emit gamma rays can be identified through the technique of gamma spectrometry. However, the determination of nuclear decay related parameters of this radioisotope will provide a greater insight into the method of production and the possible effects of the presence of this radionuclide when administered in the form of radiopharmaceutical to the patient. The determination of the gamma emission probability (Pγ) is fundamental for this control, as well as being an opportunity to confirm the efficiency of the spectroscopy system for determination of radionuclidic impurities enabling the measurement procedure both activity and Pγ. The sum-peak method was adopted here since it allows to obtain very low values for activity of radionuclides emitting gamma radiation, as is the case of [121]Te. This method had already been used by (Brinkman et al., 1977; Silva et al., 2006) for calibration of ^{123}I, providing good results with low uncertainty.

2. Methods

2.1. Sum-peak method

The spectrum of ^{121}Te with energy peaks can be observed in Figure 1. Gamma spectrometry system used here consisted of an HPGe coaxial detector, with 20 % relative efficiency, and a good resolution in the range from 3 to 300 keV.

The sum-peak coincidence method is an absolute method used in calibration processes of radionuclides that have emission lines x or gamma. Therefore, the combinations that will result in sum-peak can be of type: γ-γ, x-x or x-γ. Thus, the application of the method requires a well-calibrated spectrometry system, combined with associated electronic units, as well as a program for data acquisition, integrated with a multichannel analyzer. According to the decay scheme, ^{121}Te decays by electron capture to ^{121}Sb. The absolute value of the activity is calculated using the following equation:

$$N_0 = [N_T + N] + \frac{N_X N_\gamma}{N_{X\gamma}} \tag{1}$$

where N_0 is the activity; N_T is the total count; N is the counting corresponding to the extrapolation to zero in multichannel analyzer; Nx is the number of counts in the x photopeak; Nγ is the number of counts to the γ photopeak; Nxγ is the number of counts of coincidence due to x and γ peaks. The estimate of uncertainty is given taking into account uncertainty components of A- and B - type for an expanded uncertainty k = 1 (de Almeida et. Al., 2007).
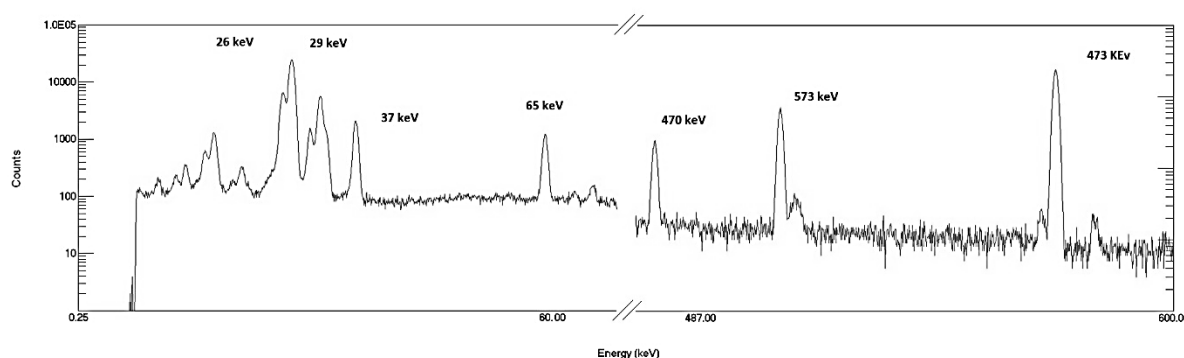

Figure 1   HPGe spectrum obtained by the spectrometry system with the respective energy peaks associated with the radionuclide ^{121}Te.

2.2. Spectral separation method

Here was developed a spectral separation method to remove the contribution of ^{125}I that appears in the sample due to the production mode of ^{123}I. This other radionuclidic impurity has a characteristic energy represented by the peak of 35.5 keV, which interferes with quantification of ^{121}Te x-ray due to its proximity and high intensity.

Point sources were prepared from the master solution of ^{123}I, the first step consisted in identifying all energy peaks in the region of 30-40 keV that appear in the spectrum, which is stored in the internal memory of the automated multichannel Analyzer. Then one obtains the gross count values for this region of the spectrum and subtract the corresponding background radiation. Hence, it is bounded on the region of interest in the spectrum that corresponds to the peak of 35.5 keV and, with appropriate command, gets the count for just this peak, generating a peak area correction factor of this region. This factor is obtained by division between the 35.5 keV peak area in the spectrum of the source to be calibrated (mixture) and the corresponding area in another spectrum of a source contained only ^{125}I

(pure) in the same conditions of measurements. This area factor obtained in the previous step is considered to subtract the contribution of the ^{125}I contaminant. Due to small discontinuity that appears at the beginning of the detector window is made the extrapolation to zero in multichannel to get the value of N, which will be added to total N so that, finally, is determined the value of N_0, according to equation 1.

3 Experimental Procedure

3.1 Source preparation

The master solution used for these measurements was provided in ampoule by the producer (IEN/CNEN) and then was eluated in the form of iodide (^{123}I) with a sodium hydroxide solution for obtaining sodium iodide (NaI). Before starting the counts, was here set a timeout, around a few half-lives of ^{123}I, in order to reduce the intensity of the sources so that its secondary energy lines do not hide the ^{121}Te characteristic peaks observed in the spectrum. The masses were determined by gravimetric method with differential weighing.

Point sources were prepared with the help of a picnometer, depositing drops of solution of radionuclide in a polystyrene film, with a thickness of 0.05 mm, set in an acrylic ring. The ring has an external diameter of 25 mm, inner diameter of 4 mm and 1 mm thick. Once dried, the sources were covered with the same polystyrene film (Bernardes et al., 2002). HPGe spectrometry system has been calibrated in efficiency through point standards of ^{60}Co, ^{152}Eu, ^{166m}Ho and ^{226}Ra. The efficiency curve obtained for an energy range between 100 and 1000 keV can be seen in Figure 2, and the uncertainties for values of activity varied around 2% (k = 1).

3.2 Radionuclidic impurities assessment

For carrying out of measurements was used a system consisting of a planar HPGe detector with the standard electronic. This detector is known "d4" and the measurements were carried out in two different positions: the first at a distance of 10 cm, "p2", from the detector and the second on the detector "p_0". HPGe spectrometry system has been calibrated in efficiency through point standards of ^{60}Co, ^{152}Eu, ^{166m}Ho and ^{226}Ra.

The efficiency curve obtained for an energy range between 100 and 1000 keV can be seen in Figure 2, and the uncertainties for values of activity varied around 2% (k=1). Significant sum effects were not observed in for the preparation of the efficiency curve, at "p2", mainly for ^{152}Eu and ^{166m}Ho standards. This allowed the calibration point source, quantification of impurities and determining the gamma emission probability for ^{121}Te. The spectrum acquired in the position "p0" was used by the sum-peak absolute method to get the activity of ^{121}Te.

Radionuclidic impurities of ^{121}Te and ^{125}I, both with half-life more than ^{123}I, identified in the spectrum were quantified in order to assess possible damage to the patient during the period of incorporation. As the main radionuclide decay during the acquisition of multiple spectra it was possible to verify the presence of the peaks associated with impurities. Brazilian law follows the recommendation of the American Pharmacopoeia, which, for the production of ^{123}I, stipulates a limit of 15% for impurities in relation to the main radionuclide (^{123}I). And in this study the presence of ^{125}I was a negative contribution factor, which could be properly evaluated with the aid of the spectral separation technique, ensuring its content to be compared to the percentage indicated by the current recommendations.
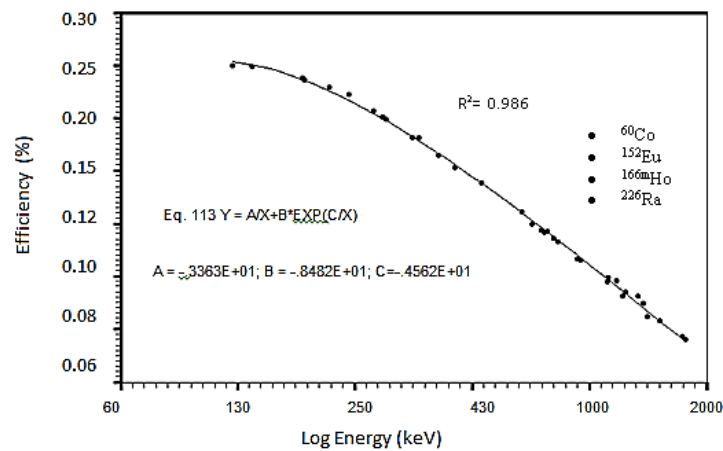
Figure 2. Efficiency curve obtained (linear-log scale) for the spectrometry system using the radionuclidic standards in point source geometry.

3.3 Gamma-ray emission probabilities measurement

In order to associate the main peaks of the spectrum to the radionuclidic is need to obtain the relation energy-channel, for calibrating in energy the spectrometer. After, the total absorption efficiency curve is determined, in function of energy, to calculate the radionuclidic activity from net areas under each interest peak. The expression of activity area is:

$$A = CPS_{(CORRECTED)}/(\varepsilon . P_\gamma) \tag{2}$$

where: CPS (CORRECTED) is the count rate of photopeak; εγ is the photopeak efficiency for specific energy; and Pγ is the emission probability for specific energy. However, as the source activity was obtained directly by sum-peak method, Pγ was calculated for two main energies of [121]Te, 507 and 573 keV, by means of the following expression, taking into account the corrections as decay, background, position:

$$P_\gamma = CPS_{(CORRECTED)}/(A . \varepsilon_\gamma) \tag{3}$$

where: CPS (CORRECTED) is the count rate of photopeak; A is the absolute activity measured by sum-peak method; εγ is the photopeak efficiency for specific energy. The precision to determine Pγ depends on the precision achieved in efficiency curve with the choice of interest energy of the standards: [60]Co, [152]Eu, [166m]Ho and [226]Ra.

The peak area evaluate method taken into account the channel integrations that define the region of each peak, after subtracts background. Spectra were analyzed by Maestro II code. The samples and standards were measured three times at least. All measurements were made with point sources in "p2" position using the same geometry.

4 Results and Discussion

The experimental results for the activity of [121]Te are presented in Table 1, accompanied by their respective uncertainties both for efficiency curve method, position p2 as to the sum-peak method, position p0.

Table 1: Results of activities for efficiency curve and sum-peak methods for ^{121}Te

SOURCE	Efficiency Curve (p2)			Sum-peak method (p0)		
	Activity (kBq)	u_A(%)	u_B(%)	Activity (kBq)	u_A(%)	u_B(%)
172S14	695.0	0.50	2.10	696.4	0.50	0.1
173S14	560.0	0.59	2.10	570.4	0.30	0.1
174S14	600.0	0.39	2.10	599.0	0.15	0.1
175S14	689.0	0.27	2.10	604.7	0.35	0.1
176S14	600.0	0.47	2.10	596.5	0.36	0.1

Positions: p2 = 10 cm (distance between source detector); p0 = 0 cm (top of detector)

 The result for the source 175S14 obtained by the efficiency curve method suffered some interference and unable to again measure the value of the activity of the source. The values obtained in both methods are compatible and this enabled the standardization of absolute ^{121}Te.

As can be seen in table 2, it should be noted also the low uncertainties associated with the sum-peak method, indicating values that are more reliable. The identification of impurity for the ^{125}I brought several problems in order to calibrate ^{121}Te absolutely.

Table 2: Uncertainties to ^{121}Te impurity determination by sum-peak method

Uncertainty Component	^{121}Te	
type	A	B
measurement statistic	0.15	
decay	-	< 0.1
live time	-	< 0.1
weighing		0.1
Relative standard uncertainty u (%)	0.18	
Relative expanded uncertainty	0.36	

U (%) (k=2)

Table 3 shows the results for the gamma emission probabilities compared to the values of the Physikalisch-Technische Bundesanstalt (PTB) and Laboratoire National Henry Becquerel (Nucleide-LARA/LNHB). In general, the values for the emission probabilities evaluated were consistent and uncertainty values obtained are slightly below the published values.

Table 3: Gamma emission probabilities and their relative standard uncertainties for 507 and 573 keV energies of ^{121}Te by efficiency curve method (k=1)

Energy (keV)	PTB	LARA/LNHB	This work
507	0.177±0.004	0.177±0.006	0.167±0.002
573	0.803±0.017	0.823±0.025	0.804±0.006

Finally, it was possible to develop a methodology for analysis of these impurities that took into account the interference contribution for ^{125}I during the data acquisition. The proper calibration of the photon spectroscopy system, as well as the successful implementation of sum-peak method, made it possible to obtain the values of the emission probabilities with associated uncertainty until 1.2 % for 507 keV and 0.5% for 573 keV. The Pγ value for 507 keV is 5.6 % lower than of reference publications. And for 573 keV the value Pγ is until 2% is compatible with the reference. However, the uncertainties here obtained are lower than of literature data.

Table 4: Percentage of radionuclidic impurities in the ^{123}I sample and associated uncertainties by efficiency curve method

Isotope Impurity	^{121}Te		^{125}I	
Percentage in ^{123}I sample (%)	0.326		0.394	
Uncertainties (μ) type	A	B	A	B
measurement statistic	0.47		0.88	
efficiency curve	-	0.60*	-	4.0
weight-geometry	-	0.20	-	0.20

half-life	-	0.40	-	0.02
live time	-	< 0.1	-	< 0.1
photon emission probability (Pγ)	-	2.1	-	2.0
relative standard uncertainty *u* (%)		2.3		4.6
Relative standard uncertainty associated with an activity of ^{123}**I ****		1.1		1.1
Relative expanded uncertainty *U* (%) (*k*=2)		5.1		9.5

* (Lépy et al., 2015) ** values obtained by ionization chamber method

6 Conclusions

The absolute standardization process of ^{121}Te here developed presented a new approach for the treatment of radionuclidic impurities. Activities obtained by efficiency curve were satisfactory and important enough for the comparison of two methods here adopted. The sum-peak method made it possible to obtain values of activity with uncertainties below 0.5 %. At the same time made it possible to obtain good values for both the activity and the determination of precise nuclear parameters as gamma emission probabilities for the two main radionuclide and its impurities.

7 References

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