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Gamma spectrometry of iodine-125 produced in IEA-R1 nuclear reactor, using HPGe detector and fixation into epoxy matrix disc

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ABSTRACT

Few places in the world produce iodine-125. In Brazil, the first production was achieved by using the IEA-R1 nuclear reactor located at Nuclear and Energy Research Institute – IPEN. To verify the quality of iodine-125 produced, and the amount of contaminants such as iodine-126, cesium-134 and caesium-137 among others, iodine-125 samples were immobilized into epoxy matrix disc, with the same geometry of a barium-133 reference radioactive source, used to calibrate an HPGe detector. The HPGe detector has a thin carbon composite window, which allows measure the iodine-125 photopeaks, between 27.1 and 35.4 keV. The method employed here was successful in producing and measurement of iodine-125.

1. Introduction

Iodine-125 is a versatile radionuclide which may applicable in diagnostic and therapy, such as radioimmunoassay, Auger electron therapy, and brachytherapy seeds implants (International Atomic Energy Agency, 2011; Isotope, 2020; Venkatesh and Banerjee, 2007). The IEA-R1 nuclear reactor in Nuclear and Energy Research Institute (IPEN) was used to produce iodine-125 by neutron irradiation of a xenon-124 target. The production follows $^{124}Xe(n,\gamma)^{125}Xe$, presenting a half-life of 16.9 h when xenon-125 decays to iodine-125 (Costa, 2015; Costa et al., 2015; Mirzadeh et al., 2011). The main impurity in this production route is the radionuclide iodine-126, $^{125}I(n,\gamma)^{126}I$, however, cesium-134 and cesium-137 among others, may be also produced during the irradiation process. The volatility of iodine is always a major concern to be considered. To verify the radionuclide quality of iodine-125 produced, it was developed a methodology to immobilize the radioactive iodine, avoiding contamination of the HPGe detector by the iodine volatile molecules (Costa, 2015; Mirzadeh et al., 2011).

2. Materials and methods

A planar radioactive source was designed to verify the radionuclide purity of I-125 produced (Fig. 1). This planar radioactive source consist of the radioactive material deposited on filter paper (Millipore, Ø 47 mm, SSWP004700), immobilized into an epoxy resin (Silaex Química, SQ2004 and SQ3131)(Silaex Química, 2020). The thickness of top and bottom parts may vary according to the specifications of geometry of measurement, as well as the outer diameter.

A silicone mold was manufactured to produce the planar radioactive sources (Fig. 2A). The silicone mold is used first to produce the bottom part of planar source (Fig. 2B). After that, the bottom part is turned upside up, the filter paper is placed on the bottom part (Fig. 2C), and iodine-125 NaOH solution is pipetted on the filter paper (1 - 10 kBq, 27 - 270 nCi, measured in an ionization chamber Capintec, CRC-15W) (Fig. 2D). The iodine-125 was produced by neutron irradiation in IEA-R1 nuclear reactor located at Nuclear and Energy Research Institute – IPEN. The filter paper with iodine-125 NaOH solution was dried in a period of 24 h, inside a fume hood. After the drying period, more epoxy resin was added on the filter paper and bottom part to form the top part of planar radioactive source, and to trap the iodine-125 inside the epoxy resin (Fig. 3A). The cure time was approximately 24 h. Five planar sources were manufactured.

After the epoxy resin cure (Fig. 3), leakage tests were performed according ISO 9978 standard:

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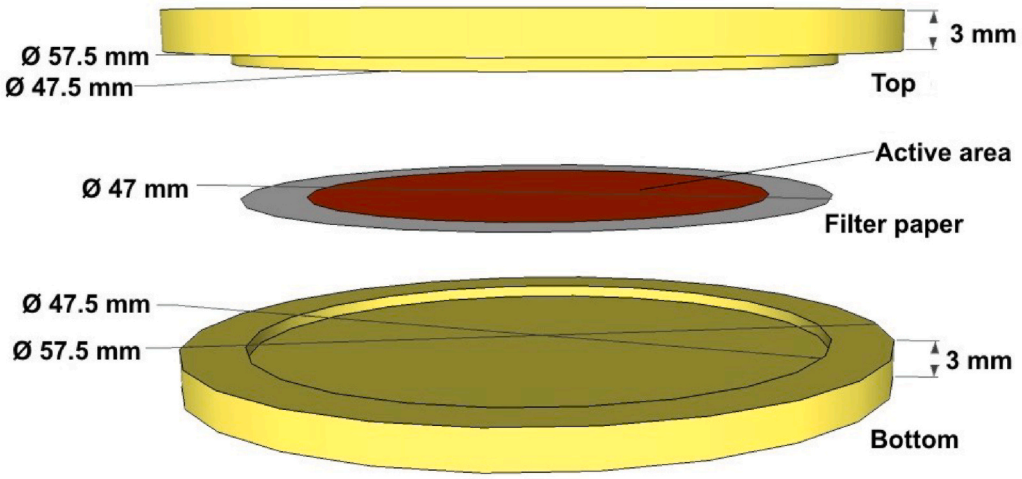

Fig. 1. Planar radioactive source designed to immobilize iodine-125.

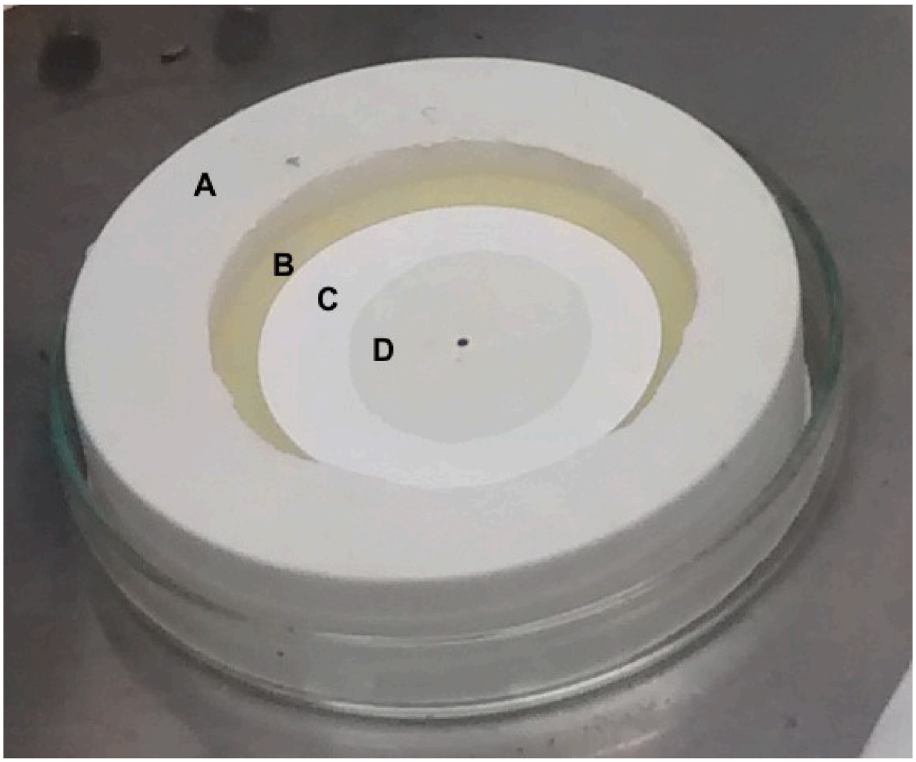

Fig. 2. (A) Silicone mold; (B) Bottom of the planar radioactive source; (C) Filter paper; (D) Iodine-125 deposited area.

- wet wipe test - all the surface of the sealed source was wiped with a paper moistened with detergent (Merck, Extran MA 02 neutral). The activity of the paper was measured in well counter (NaI(Tl) Scintillator Capintec, CRC-15W, t = 60 s);
- immersion test at room temperature - the sealed source was immersed in a detergent solution (water 95%, Merck Extran MA 02 neutral, 5%), and maintained at room temperature for 24 h. The sealed source was then removed and the activity of solution (1 mL sample) was measured in well counter (NaI(Tl) Scintillator Capintec, CRC-15W, t = 60 s).

The sealed source is considered to be leaktight if the activity detected in the paper and in the entire volume of solution does not exceed 200 Bq (5.4 nCi) (International Organization for Standardization, 1992). The paper moistened with detergent and the detergent solution samples (1 mL) were measured before and after the tests to calculate the final result without background influence.

The gamma spectrometry of planar sources was performed by using a high purity germanium detector (Canberra, Extended Range Coaxial Ge Detector – XtRa, GX2518), software Genie 2000 and module DAS 1000. This detector has a thin carbon composite window, which guarantees to give an intensity ratio greater than 20:1 in the 22–88 keV energy range (Canberra Industries, 2006), adequate to iodine-125, whose energy varies between 27.1 and 35.4 keV. The count periods range between 5000 and 65000 s. The system efficiency was calibrated with a reference barium-133 radioactive point source (Ø 1 mm), with 45.5 kBq activity on January 1, 1988 (12 h GMT) and stated accuracy 7%. The source is part of a set by Amersham, "(Gamma reference source set QCR.12)".

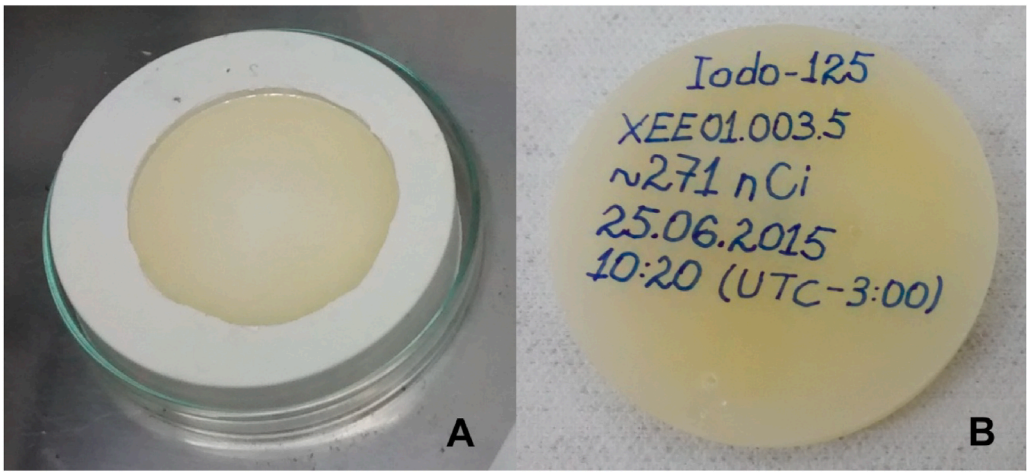

Fig. 3. (A) The planar radioactive source in the silicone mold after the cure process of the top part; (B) The planar radioactive source ready to use.

Table 1
Gamma lines of barium-133 used to determine the detection efficiency.

Energy (keV)	Energy uncertainty (keV)	Yield (%)	Yield uncertainty (abs. ±)
30.850	0.010	96.8000	0.1000
35.220	0.010	22.8000	0.1000
53.161	0.001	2.1400	0.0200
80.890	0.003	35.5000	0.3000
160.613	0.008	0.6380	0.0080
223.234	0.012	0.4530	0.0040
276.398	0.002	7.1640	0.0220
302.853	0.001	18.3400	0.0600
356.017	0.002	62.0500	0.1900
383.851	0.003	8.9400	0.0300

Source: Genie 2000 software library (Canberra Industries, 2006).

Table 1 shows the gamma lines of barium-133 reference source according to Genie (2000) software library (Canberra Industries, 2006).

Each planar I-125 sources was placed in an acrylic holder, with its center coaxial to the detector axis, at a distance $z = 12.7$ cm between the source and the detector surface. For the efficiency calibration, the Ba-133 reference point source was placed at the same position of the planar sources center, maintaining the distance z, in order to achieve the same effective solid angle as exactly as possible. In order to estimate the solid angle Ω, subtended by the source-detector geometry for both sources, a formulation by Ruby (1994) was employed. Values of Ω are given by Equation (1):

$$\Omega = 4\pi \frac{R_d}{R_s} \int_0^\infty \frac{e^{-kz}}{k} J_1(kR_s)\, J_1(kR_d)\, dk \quad (1)$$

where.

R_d is the radius of the detector surface, 28.25 mm;
R_s is the radius of each source, equal to 23.5 mm for the planar sources and 0.5 mm for the reference source;
J_1 is the Bessel function of the first kind, order 1.

By applying the expression, values of Ω = 0.0078393 for the reference point source and Ω = 0.0078375 for the planar source were obtained. The difference between both values is approximately 2.4%, less than the stated accuracy of the reference source; therefore no further corrections were applied.

The measurements with the reference source in the geometry described allowed to record a calibration curve (Equation (2), Fig. 4), to determine the iodine-125 activity from the gamma spectrum.

$$\ln(E_{ff}) = -13.00 + 5.097 \times \ln(E) - 0.8738 \times \ln(E)^2 + 0.04029 \times \ln(E)^3 \quad (2)$$

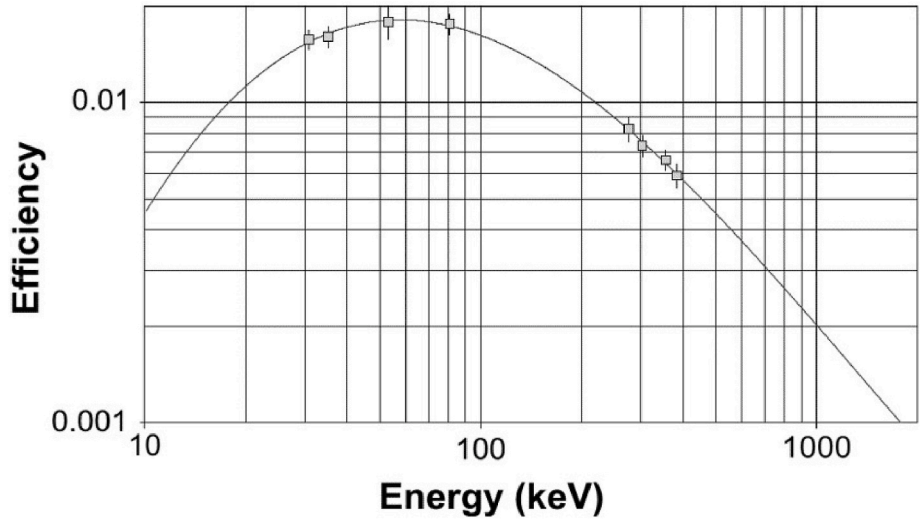

Fig. 4. Calibration curve of spectrometry recording efficiency with HPGe detector, using barium-133 reference source, to iodine-125 gamma spectrometry.

Table 2
Gamma lines used to determination of iodine-125 and iodine-126.

Radionuclide	Half-life (d)	Energy (keV)	Intensity (%)	Efficiency	Net peak area[a]
Iodine-125	59.407	27.38	112.50	0.0144	3.27×10^6
		31.18	25.44	0.0155	7.24×10^5
		35.49	6.63	0.0164	1.85×10^5
Iodine-126	12.93	388.63	34.00	0.0059	3.06×10^2
		666.33	33.10	0.0033	1.96×10^2

Source: Genie 2000 software library (Canberra Industries, 2006).
[a] Net peak area relative to planar source 2 in Table 3.

Table 3
Leakage tests results for iodine-125 radioactive sealed source.

Planar source	I^{125} NaOH solution activity (Bq)[(a)]	Wet wipe test activity(Bq)[(b)]	Immersion test at room temperature (Bq)[(b)]
1	$14.66(44) \times 10^3$	0.33(740)	3.51(999)
2	$3.11(10) \times 10^3$	59.31(166)	0.18(92)
3	$12.59(38) \times 10^3$	30.63(144)	1.55(118)
4	$23.93(72) \times 10^3$	0.93(136)	0.03(99)
5	$14.52(43) \times 10^3$	5.44(122)	0.03(96)

Detector: Capintec CRC-15W.
[a] Ionization Chamber.
[b] Well Counter.

Where E_{ff} is the detector efficiency and E is the energy.

The gamma energies and the respective probabilities of gamma emission of the radionuclides of interest are compiled in a file (Table 1), with software and nuclear data supplied by the Genie 2000 software library (Canberra Industries, 2006). Only two gamma lines (160 and 223 keV) stayed out of calibration curve due to low emission intensity (0.6 and 0.4% respectively). The gamma lines used to determine the activity of iodine-125 and iodine-126 are in Table 2.

In order to validate the application of the efficiency calibration, at energies higher than the point of maximum gamma energy used in the curve fit, measurements with a Cs-137 point source, from the same set of the Ba-133 source, were performed, as Cs-137 emits gamma photons with energy 661.65 keV, very close to the 666 keV emission from I-126. This source has the following specifications: 35.3 kBq activity on January 1, 1988 (12 h GMT) and stated accuracy 6%.

The measurement of the Cs-137, with application of the efficiency curve previously determined, resulted in an activity of 37.2(13) kBq corrected for the reference date. This figure agrees with the reference value within the uncertainties, thus enabling the applicability range of the curve up to this energy.

With the efficiency curve and the spectrum, the sample activity correspondent at each gamma energy line $E\gamma$ is obtained by Equation (3).

$$A = \frac{C}{\varepsilon(E\gamma)\cdot I\gamma\cdot t_L} \tag{3}$$

where A is the source activity to a specific radionuclide; C, net area counting from gamma peak of energy $E\gamma$; $\varepsilon(E\gamma)$, detection efficiency for gamma ray energy $E\gamma$ obtained by calibration curve; $I\gamma$, emission probability per disintegration for each gamma photon; t_L, counting live time.

The iodine-125 activity in each source was determinated by means of weighted average by variance from activities defined for each gamma peak detected.

Table 4
Correlation between iodine-125 and iodine-126 produced in IEA-R1 nuclear reactor (calibration 10 days after irradiation).

Planar source	I^{125} NaOH solution activity (Bq)[(a)]	I^{125} planar source activity (Bq)[(b)]	I^{126} planar source activity (Bq)[(b)]	I^{126}/I^{125} (%)
1	$14.66(44) \times 10^3$	$5.5(2) \times 10^3$	32(8)	0.6
2	$3.11(10) \times 10^3$	$5.03(18) \times 10^3$	38(6)	0.7
3	$12.59(38) \times 10^3$	$11.4(4) \times 10^3$	81(12)	0.7
4	$23.93(72) \times 10^3$	$22.5(8) \times 10^3$	127(34)	0.5
5	$14.52(43) \times 10^3$	$13.3(4) \times 10^3$	88(28)	0.6

[a] Detector: Ionization Chamber Capintec CRC-15W.
[b] Detector: Canberra HPGe.

3. Results and discussion

The five iodine-125 sources were approved in the wet wipe test and in immersion test at room temperatures, as is shown in Table 3. All the results remained below 200 Bq which is the limit value according to ISO 9978 (International Organization for Standardization, 1992).

Table 4 presents the data about the radionuclide analysis, which detected only iodine-126 as contaminant in iodine-125 samples produced. Although authors such as Kar (2007) propose different methods to purify the iodine-125 solution after the production, to separate aluminum alloy metallic contaminants, carried by NaOH solution, it was not possible to detect traces of radionuclides from aluminum alloy in the samples.

The correlation between the quantity of iodine-125 and iodine-126 produced presented values between 0.5 and 0.7%, which ensure radionuclide purity greater than 99%. These values make the iodine-125 produced suitable to radioimmunoassay laboratory applications. To reach medical grade radionuclide purity (I-126 < 0.005%), it is necessary to decrease the irradiation time or to increase the decay time. However, in both cases the final activity of iodine-125 produced will decrease. Chemical methods of purification may also be applied, however, demand a detail study and infrastructure.

In Table 4, a comparison between the activity in I^{125} NaOH solution, measured in ionization chamber of Capintec CRC-15W detector, and the activity in I^{125} planar source, measured in HPGe detector, shows a learning curve along the manufacture of planar sources. The first two sources manufacture presented differences greater than 60%. However, the three last sources presented differences lower than 10%. The activity value of planar source tends to be lower than the value of I^{125} NaOH solution, because there are inevitable losses during the transport from solution vial to filter paper (pipette tip and vial surface), and in the desiccation process (sublimation of iodine).

During the manufacture of planar source 2 an incident happened: along the cure process, the silicone mold was uneven, part of iodine-125 was dragged from the filter paper to the edge of planar source by epoxy resin. The planar source 2 was decontaminated and a new layer of epoxy resin was overlapped to level the source. The activity of I^{125} NaOH solution was recalculated. However, it was not enough to avoid the differences of measurements.

It should be noticed that, apart from the statistical uncertainty derived from the peak area measurements, the Ba-133 reference source has a 7% stated accuracy, which should be considered as an additional irreducible systematic uncertainty in the present work, since all efficiency points were obtained from the same source.

Fig. 5 shows the gamma spectrum of an iodine-125 planar source measured with HPGe detector. One should note, that the number of scattered gamma photons, as a function of energy, quickly increases, from the cutoff point at 21 keV, up to approximately 25–26 keV and then slowly decreases, leading to an apparent peak at that energy range. The two photopeaks of 27.47 keV [73.4(10) %] and 27.2 keV [39.4(5) %] are represented as one photopeak of 27.38 keV, the photopeak of 31.18 keV is a sum of photopeaks near 31 keV, the gamma photopeak of 35.49 keV

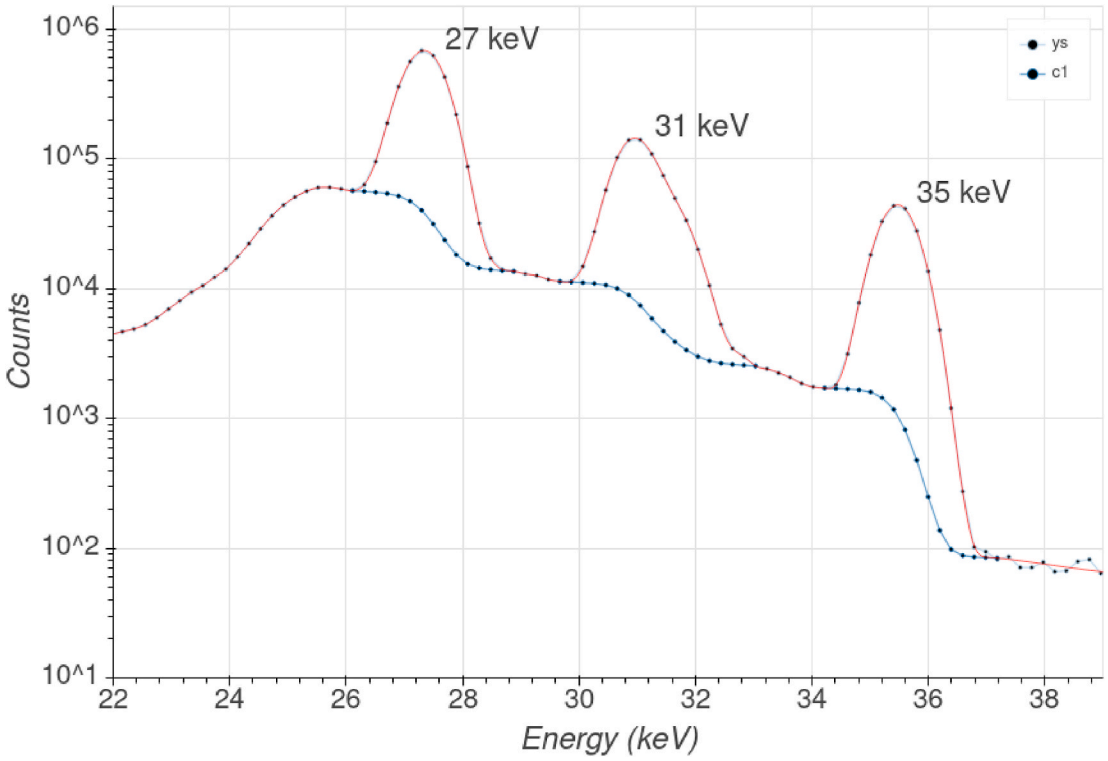

Fig. 5. Iodine-125 planar source gamma spectrum with 27, 31 and 35 keV photopeaks measured with HPGe detector. The blue line shows the baseline fit and the red line, the baseline plus the gaussian fit. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

[6.68(13) %] is a single photopeak (International Atomic Energy Agency Nuclear Data Section, 2020).

4. Conclusions

The epoxy resin discs were efficient to immobilize the iodine-125. The five iodine-125 planar sources passed in wet wipe test and immersion test at room temperature with values lower than 200 Bq according to ISO 9978 (International Organization for Standardization, 1992).

The iodine-125 planar sources were assembled with the same geometry of barium-133 reference source, enabling the calculus of source activity.

The calibration of HPGe detector allowed to verify the percentage of iodine-126 (between 0.5 and 0.7%, 10 days after irradiation), ensuring the categorization of the iodine-125 produced in IPEN IEA-R1 nuclear reactor.

The method presented in this work was considered efficient and safe to measure the iodine-125 produced by neutron irradiation.

The next step is to implement iodine-125 production in a semi-industrial route (for radioimmunoassay use) and continue to develop purification and concentration methodologies to use the iodine-125 for brachytherapy sources production.

CRediT authorship contribution statement

Osvaldo L. da Costa: Conceptualization, Design, Experimental activities, Data curation, Formal analysis, Investigation, Methodology, Project administration, Validation, Visualization, Writing - original draft, Writing - review & editing. **Daiane C.B. de Souza:** Experimental activities, Writing - review & editing. **Fabio G. Castanho:** Writing - review & editing. **Anselmo Feher:** Experimental activities, Data curation, Formal analysis, Writing - review & editing. **João A. Moura:** Experimental activities, Writing - review & editing. **Carla D. Souza:** Writing - review & editing. **Henrique B. Oliveira:** Writing - review & editing. **Marcelo F. Máduar:** Formal analysis, Gamma spectrum analysis, Writing - review & editing. **Carlos A. Zeituni:** Funding acquisition, Resources, Supervision, Visualization. **Maria Elisa C.M. Rostelato:** Funding acquisition, Resources, Supervision, Validation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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