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GAMMA SPECTROMETRY OF IODINE-125 PRODUCED IN IEA-R1 NUCLEAR REATOR, 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 happened in nuclear reactor IEA-R1 located at Nuclear and Energy Research Institute – IPEN. To verify the quality of iodine-125 produced, because contaminants as iodine-126, caesium-134 and caesium-137 among others, may be produced in irradiation process, 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.

1. INTRODUCTION

Iodine-125 is a versatile radionuclide which may applicable in diagnostic and therapy: radioimmunoassay, Auger electron therapy and brachytherapy seeds implants are current examples [1, 2, 3]. 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 [4, 5, 6]. 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 [4, 5].

2. MATERIALS AND METHODS

A planar radioactive source was designed to verify the radionuclide purity of I-125 produced (Figure 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)[7]. The thickness of top and bottom parts may vary according to the specifications of geometry of measurement, as well as the outer diameter.

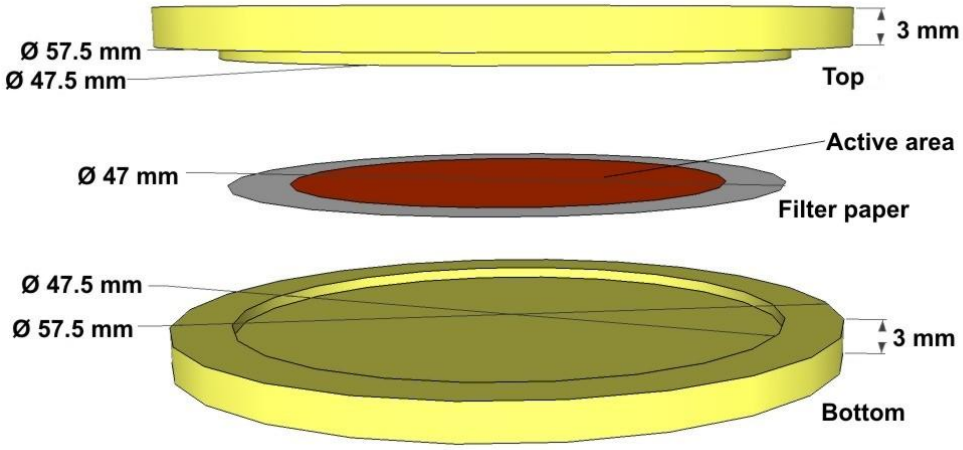

Figure 1: Planar radioactive source designed to immobilize iodine-125.

A silicone mold was manufactured to produce the planar radioactive sources (Figure 2A). The silicone mold is used first to produce the bottom part of planar source (Figure 2B). After that, the bottom part is turned upside up, the filter paper is placed on the bottom part (Figure 2C), and iodine-125 NaOH solution is pipetted on the filter paper (1 - 10 kBq, 27 - 270 nCi, measured in a well chamber Capintec, CRC-15W, t = 60 s) (Figure 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 (Figure 3A). The cure time was approximately 24 h. Five planar sources were manufactured.

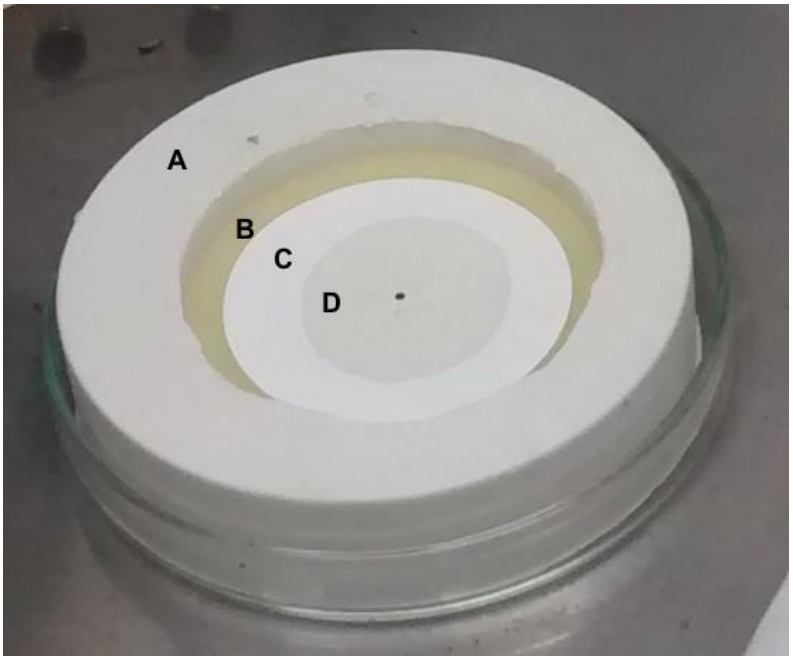

Figure 2: (A) Silicone mold; (B) Bottom of the planar radioactive source; (C) Filter paper; (D) Iodine-125 deposited area.

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

- 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 chamber (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 chamber (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) [8]. 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.

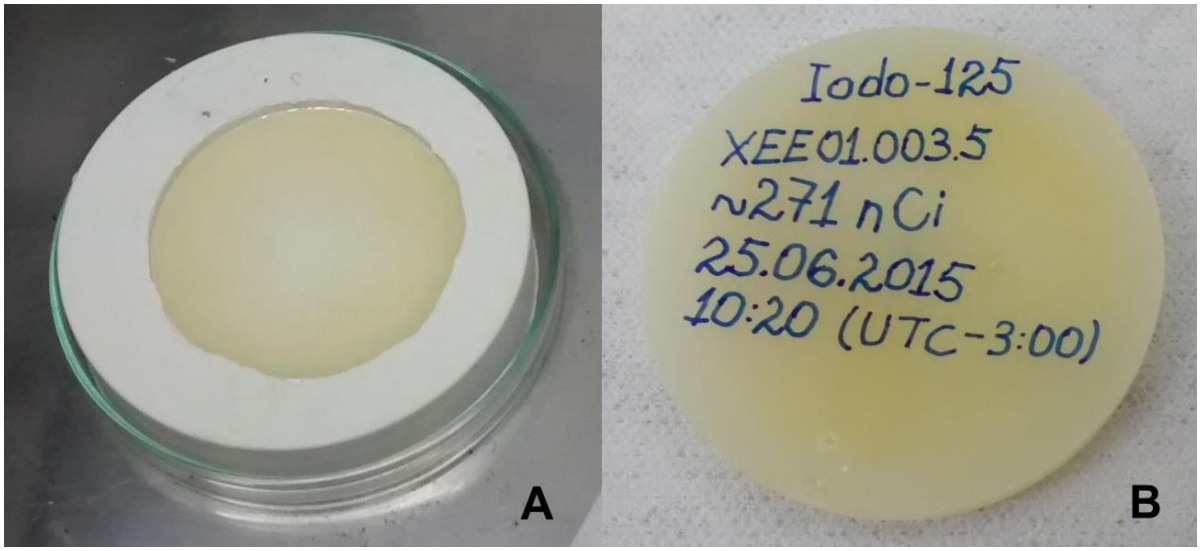

Figure 3: (A) The planar radioactive source in the silicone mold after the cure process of the top part; (B) The planar radioactive source ready to use.

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 a 22 to 88 keV intensity ratio of greater than 20:1 [9], 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 an Amersham reference barium-133 radioactive source, 1.230 µCi ± 7 %, January 1, 1988 (12 h GMT). Table 1 shows the gamma lines of barium-133 reference source according to Genie 2000 software library [9].

Table 1: Gamma lines used to determine barium-133.

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 [9].

The reference source was put in an acrylic holder at the same counting geometry of planar sources, with the same solid angle from the source to detector. The radioactive sources (reference and samples) were positioning in a centralized manner, coaxially to the detector axe, with 12.7 cm between the source and the detector surface. This geometry allowed to record a calibration curve (Equation 1, Figure 4), to determine the iodine-125 activity from the gamma spectrum.

$$ln(E_{ff}) = -13.00 + 5.097 \times ln(E) - 0.8738 \times ln(E)^2 + 0.04029 \times ln(E)^3 \quad (1)$$

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

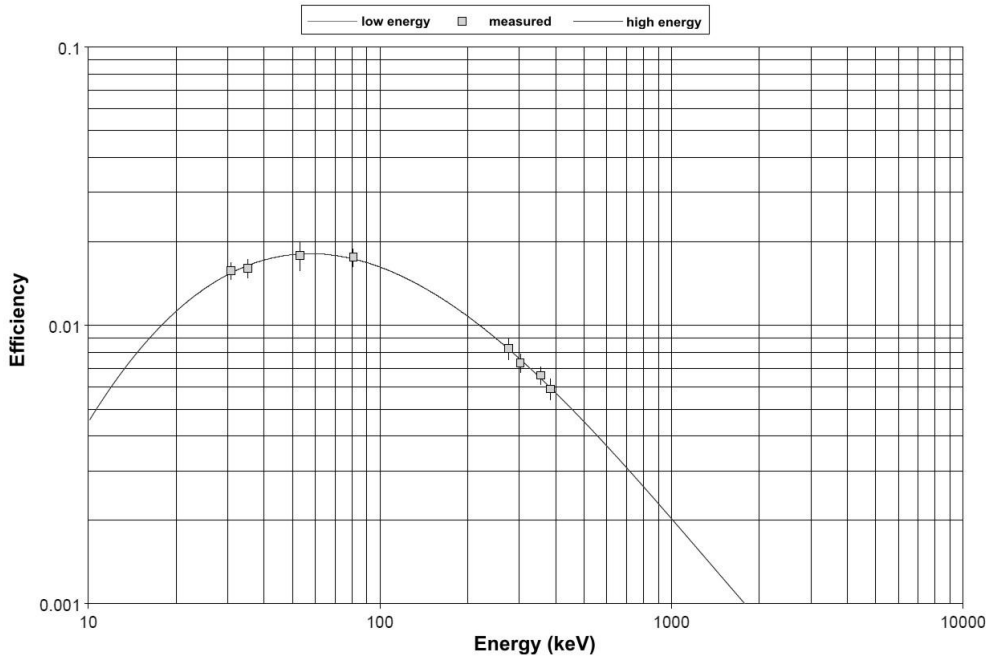

Figure 4: Calibration curve of spectrometry recording efficiency with HPGe detector, using barium-133 reference source, to iodine-125 gamma spectrometry.

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 [9]. Only two gamma lines (160 and 223 keV) stayed out of calibration curve due to low emission intensity (0.6 and 0.4 % respectively). With the efficiency curve and the spectrum, the sample activity correspondent at each gamma energy line $E\gamma$ is obtained by Equation 2.

$$A = \frac{C}{\varepsilon(E\gamma)\cdot I\gamma\cdot t_L} \quad (2)$$

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 in gamma energy $E\gamma$ obtained by calibration curve; $I\gamma$, emission probability per disintegration for each gamma photon; t_L, counting live time. The gamma lines used to determine the activity of iodine-125 and iodine 126 are in Table 2.

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

Iodine-125		Iodine-126	
Gamma energy (keV)	**Intensity (%)**	**Gamma energy (keV)**	**Intensity (%)**
27.38	112.5	388.63	34.0
31.18	25.44	666.33	33.1
35.49	6.63		

Source: Genie 2000 software library [9].

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

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 [8].

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

Planar source	I^{125} NaOH solution activity (Bq)	Wet wipe test activity(Bq)	Immersion test at room temperature (Bq)
1	$14.66(146) \times 10^3$	0.33(740)	3.51(999)
2	$3.11(31) \times 10^3$	59.31(166)	0.18(92)
3	$12.59(125) \times 10^3$	30.63(144)	1.55(118)
4	$23.93(239) \times 10^3$	0.93(136)	0.03(99)
5	$14.52(145) \times 10^3$	5.44(122)	0.03(96)

Detector: Capintec CRC-15R.

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 [10] 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 Capintec CRC-15R 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.

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)(1)	I^{125} planar source activity (Bq)(2)	I^{126} planar source activity (Bq)(2)	I^{126}/I^{125} (%)
1	$14.66(146) \times 10^3$	$5.5(2) \times 10^3$	32(8)	0.6
2	$3.11(31) \times 10^3$	$5.03(18) \times 10^3$	38(6)	0.7
3	$12.59(125) \times 10^3$	$11.4(4) \times 10^3$	81(12)	0.7
4	$23.93(239) \times 10^3$	$22.5(8) \times 10^3$	127(34)	0.5
5	$14.52(145) \times 10^3$	$13.3(4) \times 10^3$	88(28)	0.6

(1) Detector Capintec CRC-15R.
(2) Detector Canberra HPGe.

Figure 5 shows the gamma spectrum of an iodine-125 planar source measured with HPGe detector. 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 either is a sum of photopeaks near 31 keV, the gamma photopeak of 35.49 keV [6.68(13) %] is a single photopeak [11].

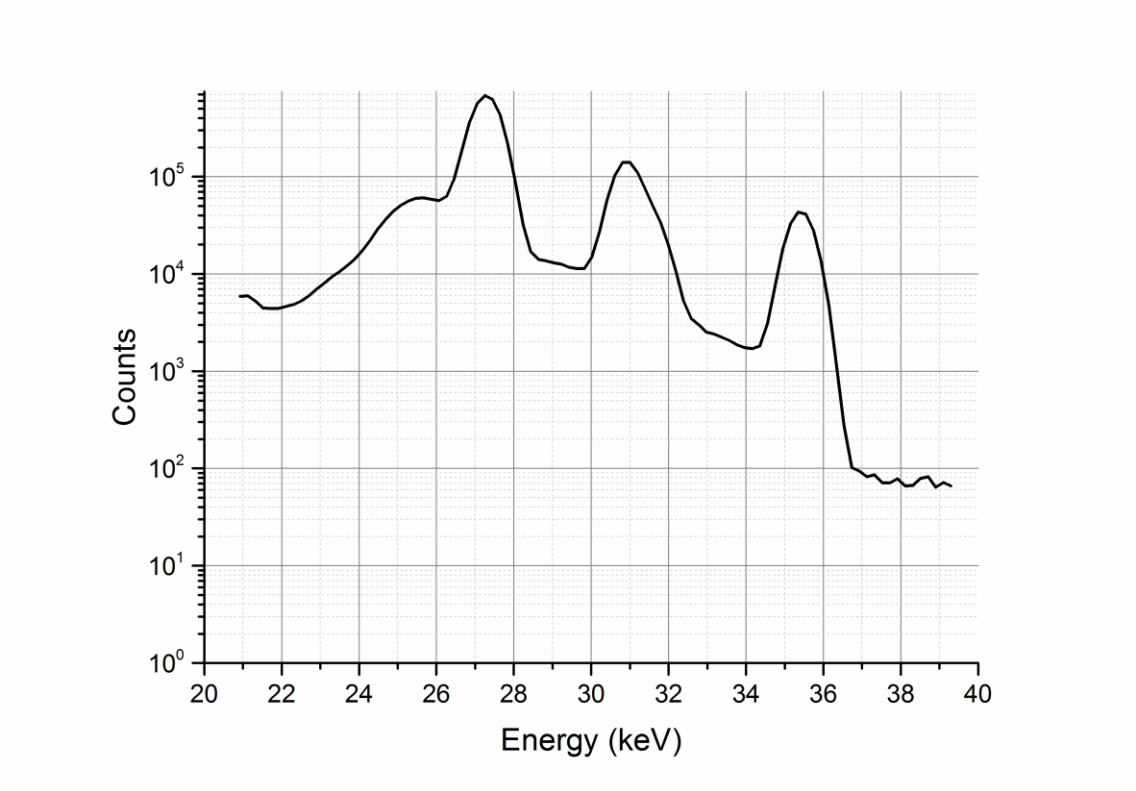

Figure 5: Iodine-125 planar source gamma spectrum with 27, 31 and 35 keV photopeaks measured with HPGe detector.

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 [8].

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.

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