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Characterization of an itinerant NaI(Tl) spectrometric system for calibrating activimeters

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ABSTRACT
In radiopharmaceutical production centres and nuclear medicine services, the use of short-lived radionuclides as radiotracers is essential, with applications seeing significant growth and attracting the interest of national metrology laboratories. These laboratories are responsible for ensuring the traceability of radioactive standards, which is crucial for the precise determination of the activity of administered radiopharmaceuticals. However, the short half-life of radionuclides complicates direct traceability for nuclear medicine services (NMS). To overcome this limitation, the National Laboratory for Ionizing Radiation Metrology (LNMRI) is developing a mobile system based on an NaI(Tl) scintillation detector. This study aims to present the schematic arrangement of the proposed system for calibrating dose calibrators, as well as to conduct characterization tests of its main parameters and determine calibration factors for specific radionuclides. This development seeks to ensure traceability for key radiopharmaceuticals used in nuclear medicine services, providing greater accuracy and reliability in diagnosis and treatment.

Section: RESEARCH PAPER

Keywords: radionuclide metrology; activimeters; NaI(Tl) arrangement; characterization

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1. INTRODUCTION

There is a pressing need to trace the dose calibrators used in nuclear medicine practices. Compliance with equipment calibration standards is a fundamental regulatory requirement for the authorization of radiopharmacies in production and nuclear medicine facilities [1]. Difficulties and limitations in precise measurements arise due to the properties of radiopharmaceuticals, especially their short half-lives, which prevent the use of in situ radioactive standards [2]. To overcome these challenges, the National Laboratory of Ionizing Radiation Metrology (LNMRI) is developing a mobile system to provide traceability for gamma-emitting radiopharmaceuticals, based on a NaI(Tl) scintillation detector [3].

NaI(Tl) detection systems, widely used as radiation monitors in various applications and facilities, can provide relevant information to operators and users about the activity of radioactive sources, thanks to their portability, robustness, and precision in the obtained data. In this context, they are extensively used as relative (or indirect) calibration systems in metrology laboratories for measuring monoenergetic gamma-emitting sources.

Experimental tests and comparisons were conducted on a mobile system to evaluate the performance of the NaI(Tl) detector, to ensure traceability for different radionuclides with simple decay schemes that emit gamma radiation between 100 and 660 keV [4], [5]. This energy range is predominant in most radiopharmaceuticals supplied by production centres and used in hospitals and clinics for diagnosis or treatment. Standards of ^{241}Am and ^{137}Cs were tested to verify the main performance indicators, such as environmental conditions, dead time, energy calibration, background radiation, energy resolution, sample positioning test, detection efficiency, and minimum detectable activity (MDA). Calibration factors for specific radionuclides were also determined. The characterization results, along with the obtained calibration factors, will demonstrate the potential use of the proposed system to calibrate dose calibrators that monitor the main radiopharmaceuticals.

2. MATERIALS AND METHODS

Some of the parameters used to characterize NaI(Tl) scintillation detectors will be addressed, with a primary focus on their ability to measure gamma radiation emitters accurately and

Figure 1. Setup of the gamma spectrometry system with NaI(Tl), assembled for characterization.

precisely, particularly considering the main radiopharmaceuticals used in the country's production centres, or in clinics and hospitals where they are administered to patients. This characterization must be performed and documented to demonstrate that the gamma spectrometry system with sodium iodide meets the specifications and is suitable for its intended purpose.

2.1. Calibration system setup

The system setup consisted of a planar, portable 6.2 × 10.8 cm² NaI(Tl) inorganic scintillator detector, model OSPREY – DTB (Canberra), as shown in Figure 1. The Osprey features an integrated multichannel analyser (MCA) tube that supports scintillation spectrometry. Designed for both laboratory and field use, this compact system includes a high-voltage power supply (HVPS), a preamplifier, and a complete digital MCA. Spectrum acquisition is performed using the Genie 2000 data acquisition and analysis software, which can automatically adjust to a low-energy threshold, defining the start and end of the spectrum. The crystal is coupled to a photomultiplier along with associated electronics, and a tripod supports the detector with an aluminium ring adapted for position adjustment. The detector is encased in a thin shielding layer of lead, tin, and copper, which is necessary to attenuate low-energy photon radiation in the spectrum. The adopted setup can be controlled via USB or Ethernet, requiring only a single connection cable for the control and data acquisition system [6].

The LNMRI standard radioactive sources used in the tests were point-type and vial-type sources for ^{241}Am (59.5 keV) and ^{137}Cs (661.7 keV). The activities ranged from 8 kBq to 45 kBq for point sources and from 9 kBq g^{-1} to 47 kBq g^{-1} for vial geometry, respectively. These sources were positioned directly above the top of the detector using PVC supports specifically designed for this system. For the measurements, a counting time of 2,000 seconds was used for background radiation and 100 seconds for the radioactive sources, which was sufficient to achieve uncertainties below 1 %.

2.2 System performance characterization

The main characteristic parameters and performance indicators that must be tested include the following:

- The effects of variations in environmental conditions, such as temperature, humidity, and atmospheric pressure, on the results, which can be assessed by monitoring the results of quality control checks over an extended period.
- Dead time, related to the accuracy of the correction technique, defined by the difference between real time and live time. According to ISO Standard 20042, a dead time of less than 10 % is usually sufficient for energy calibrations and less than 5 % for other measurements [7].
- Energy calibration, which identifies different radionuclides based on their emission energy. Calibration for photopeaks of unknown energies is performed via microcomputer, once the channel number corresponding to the photopeak is known.
- Background radiation refers not only to the events in the spectrum that form a smooth curve, over which the photopeaks are superimposed, but also to the contribution of external environmental radiation to the detection system. The number of counts in the photopeak is calculated by summing the total counts in a region of interest around the photopeak and subtracting the counts from the continuous background radiation [7].
- Energy resolution (FWHM) allows the detector to distinguish between two neighbouring photopeaks. This test must be performed within the energy range of interest for the measurements. Resolution (Ri) indicates the system's ability to discriminate radiations with close energies in the spectrum [7].
- The sample positioning test consists of the degree of agreement of the results obtained under different measurement conditions related to sample positioning. The reproducibility of the counts or, if applicable, the efficiencies of the total energy photopeak must be periodically verified using a long half-life reference source emitting gamma ray that cover the energy range of interest [7], [8].
- Detection efficiency specifies the number of photons detected in the photopeak relative to the number of photons emitted as a function of emission energy. The experimental efficiency must be calculated as a function of energy, according to the following relation:

$$\varepsilon(E_i) = \frac{cps(E_i)}{A_{(p)} \cdot I(E_i)}, \tag{1}$$

where $cps(E_i)$ is the counting rate at each energy; $A_{(p)}$ is the activity of the standard corrected to the reference date, in Bq; and $I(E_i)$ is the gamma emission intensity for each energy of interest [9].

For this characterization, two radioactive standards of the same nature (^{137}Cs in vial geometry) can be used, considering one as the reference source and the other as the sample, in order to compare and then verify the consistency of the results obtained in terms of activity at the reference date.

- Characteristic limits such as minimum detectable activity (MDA); for low-activity measurements, these indicate the limitations of the measurement. They are used to decide whether activity is present in the sample and to estimate the sensitivity of the measurement. Therefore, they depend on the measurement system, background radiation, and detection efficiency for a given energy [10]. According to the specialized literature, the MDA for a given radionuclide can be expressed as

$$MDA \cong 5 \cdot \frac{\sqrt{B_k}}{\varepsilon \cdot I \cdot m \cdot \Delta t}, \tag{2}$$

where MDA is the minimum detectable activity concentration for the radionuclide, given in Bq kg^{-1}; B_k is the area of the counts corresponding to background radiation in the region of interest for the gamma energy k; ε is the detection efficiency interpolated on the curve for the considered gamma energy; I is the probability of photon emission by gamma decay (intensity), obtained from updated nuclear data tables; m is the mass of the sample, in kg; and Δt is the measurement time interval (live time), in seconds [11]–[13].

It is important to emphasize that a quality control and maintenance program should be defined, along with the operational procedure. However, other tests can be performed as recommended by the manufacturer, such as the peak-to- Compton ratio [6].

For the analysis of the performance of these parameters, appropriate statistical tests (such as control charts, ANOVA, and the Chauvenet rejection test) were used to demonstrate compliance with the expected specifications for the spectrometry system [14]–[16].

- Determination of calibration factors (CF): Calibration factors represent the response of the NaI(Tl) detector to photons incident on its surface. Thus, calibration factors will be used to convert the net counting rates for each photopeak of interest present in the sample spectrum into activity per unit mass of the radiopharmaceutical. The CF value is determined by positioning the standardized source of the radionuclide of interest in the predefined geometry. The spectrum is obtained by accumulating counts from the standard over a set time. The region of interest must be carefully selected around the photopeak to obtain counting rates for both the standard and the sample. Therefore, CF is determined by the ratio of the net count under the standard's photopeak (in cps kg^{-1}) to the activity per unit mass of the standard ($kBq\ kg^{-1}$). With the CF value, the sample activity can be determined by dividing the sample's counting rate in the same region of interest by the CF:

$$F\left(\frac{cps}{\mathrm{kBq}}\right) = \frac{C - B}{A}\,, \tag{3}$$

where F is the calibration factor; cps is the count rate of the standard source in the region of interest (ROI) for the target gamma energy; C is the count rates; B is the background count rate; A is the certified activity of the standard solution at measurement time (decay-corrected).

3. RESULTS

The analysis of the results describes the behaviour and performance of the proposed calibration system for each selected parameter, using statistical methods.

3.1. The effects of variation in environmental conditions: temperature, humidity, and atmospheric pressure

These parameters were monitored over a specified period, and, in accordance with the technical specifications outlined in the equipment's operating manual, they consistently remained well within the specified range of -10 °C to 50 °C. This demonstrates the robustness of the measurement system in maintaining accuracy despite variations in environmental conditions.

3.2. Dead time

The dead time for all measurements conducted with this system did not exceed 3.9 %, indicating that, according to Standard 20042, for energy calibration, it should be less than

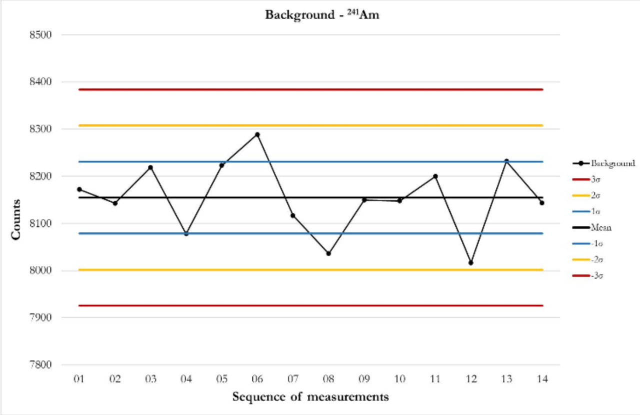

Figure 3. Behaviour of the background around 59.5 keV for the region corresponding to the ^{241}Am energy line, using a control chart.

10 %, and for other measurements, this dead time should be less than 5 %. Therefore, for all the experimental measurements conducted here, the dead time remained sufficiently adequate.

3.3. Energy calibration

Energy calibration was performed with energies of 59.5 and 661.7 keV, making it possible to identify the radionuclides ^{241}Am and ^{137}Cs by their emission energies, according to the channel number where the photopeak appears. This energy range was chosen because it covers the region where the emission of the main radiopharmaceuticals predominates.

3.4. Background radiation

For background radiation, Figure 3 presents the results of the control chart for individual values and moving range.

The data obtained for the background radiation show that the points remained within the upper and lower control limits.

Therefore, there was no significant contribution from external environmental radiation to the detection system, indicating that the background radiation is under control and should be considered in the corrections of the measurements to be performed.

3.5. Energy resolution

Measurements were taken for the energies of 661.7 keV for the resolution (FWHM) control of the proposed system. Figure 2 presents the control chart, which highlights the system's good ability to discriminate energies in the spectra obtained within the considered range. The range considered is within the specifications set for NaI(Tl).

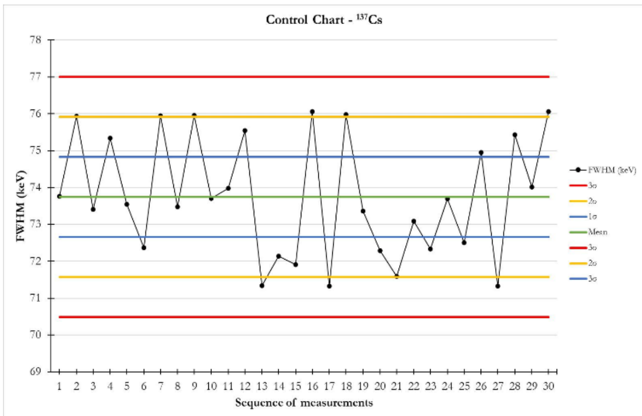

Figure 2. FWHM behaviour for a point source of ^{137}Cs.

Table 1. Measurements obtained for the study of the positioning of the ^{137}Cs source.

Group	Measurements	Counting	Mean	Variance
Position 1	10	2322597	232259.7	1251298.90
Position 2	10	2318030	231803.0	550089.78
Position 3	10	2321625	232162.5	1130854.28
Position 4	10	2319312	231931.2	1424076.62

Table 2. Results of the one-way ANOVA showing no statistically significant differences between the study groups.

Variation	*SQ*	*gl*	*MQ*	*F*	Value-*P*	*F* critic
Between groups	1312775	3	437591.80	0.402	0.753	2.866
Within Groups	39206876	36	1089079.89			
Total	40519652	39				

3.6. Sample positioning test

Four different positions were taken for the vial set and source supports with the radionuclide of half-life in vial geometry. To evaluate this parameter, each source was rotated clockwise in 90° increments, totalling four distinct positions. For each configuration, ten measurements were performed, and mean values were calculated based on the average of these readings.

In each position, 10 measurements were performed with a duration of 200 s. The statistical test applied (ANOVA, F-test) verified that the averages obtained in each position are considered significantly equal, according to Table 1 and Table 2.

Therefore, there was no variation in the positioning of the samples for the adopted liquid standard of ^{137}Cs, indicating a degree of consistency in the results obtained under different measurement conditions.

According to the F-test (ANOVA) used for a standard solution of ^{137}Cs, where the F value is much lower than the critical F value, it can be observed that the averages are significantly equal for the 4 variations taken regarding the positioning test. However, a statistical analysis of variance was conducted to test the null hypothesis (H_0) that sample position has no effect on the results. The test failed to reject H_0 at the $\alpha = 0.05$ significance level (value $p = 0.753$). This value p indicates that any observed differences are attributable to random chance rather than a systematic effect of sample position. Consequently, these findings justify, and will be followed by, more detailed investigations into the potential subtle effects of sample positioning under varied experimental conditions.

3.7. Detection efficiency

For this characterization test, two-point radioactive standards of ^{137}Cs and ^{241}Am, certified by the LNMRI, were used: one as the sample and the other as the reference. The response to these results can be seen in Table 3, where the adequacy of the activity values was observed, indicating consistency for the efficiencies as a function of energy. In the table, the total uncertainties are given as percentages, and D indicates the percent deviation from the certified value.

Table 3. Values of activities in relation to a radioactive standard.

Radionuclide	Measurements value (kBq g^{-1})	Certified value (kBq g^{-1})	Δ (%)
^{241}Am	7.79 ± 1.1	7.73 ± 0.32	-0.7
^{137}Cs	24.26 ± 1.0	24.53 ± 0.49	1.1

It can be observed from Table 3 that for ^{241}Am, the experimental uncertainty (±1.1 kBq g^{-1}) significantly exceeds the certified uncertainty (±0.32 kBq g^{-1}). Similarly, for ^{137}Cs, the values are ±1.0 kBq g^{-1} versus ±0.49 kBq g^{-1}. These disparities, while suggesting lower metrological precision at first glance, reflect fundamental distinctions: specific experimental conditions, distinct instrumentation, differentiated analytical procedures, mainly, the calibration of reference standards in primary systems—notably characterized by lower uncertainties.

3.8. Characteristic limits – MDA

The MDA was determined here for ^{241}Am and ^{137}Cs, with the values obtained being 4 and 9 Bq on the reference date for vial geometry, respectively.

These values demonstrate the minimum capability of this system to detect and determine the activity of radiopharmaceuticals. Clearly, the maximum detection capability is limited by the dead time, which is less than 5 %.

4. CONCLUSION

The control parameters adopted here proved to be suitable for evaluating performance regarding the characterization of the proposed detection system. According to the monitoring performed on the performance parameters, via statistical tests, control charts, a comparison of detection efficiencies, and the monitoring of environmental conditions, the parameters were found to be compatible, not only in terms of portability, but also robustness and accuracy of the data obtained for the presented system. Furthermore, the determination of calibration factors for ^{241}Am and ^{137}Cs vials indicates that other factors can be similarly determined for radiopharmaceuticals of interest, properly calibrated at the LNMRI by primary measurement systems or by the ionization chamber.

In summary, this phase of characterizing the spectrometric setup demonstrated the potential use and suitability of the proposed system to provide in situ calibration of the activimeters that monitor the main radiopharmaceuticals used in nuclear medicine services in the future.

AUTHORS' CONTRIBUTION

All authors contributed to the development of this study. Dayana A. Conceição (first author) was responsible for the study's conception and planning, as well as the collection and analysis of experimental data. José U. Delgado (second author) collaborated in drafting the methodology section, analysing the results, and contributing to the discussion. Octávio L. T. Filho (third author) contributed to the statistical analysis and the organization of tables and graphs. In the final revision of the manuscript, which included the review of technical and editorial aspects, all authors worked together to finalize and present this article.

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