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Parameters for Validating a Spectrometric System with Itinerant NaI(Tl) in the Activimeter Calibration

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Abstract: In radiopharmaceutical production centers and nuclear medicine services, short half-life radionuclides are essential. They have been used as radiotracers since their use increased significantly and aroused the interest of national metrology laboratories, which are responsible for providing traceability to radioactive standards. Therefore, knowledgement of the activity of administered radiopharmaceuticals must be obtained accurately. However, their short half-lives prevent the provision of traceability. To overcome these obstacles, an itinerant system based on a NaI(Tl)-type scintillator detector is being developed at the National Laboratory for Ionizing Radiation Metrology (LNMRI). This work aims to show the schematic arrangement of the system developed to calibrate activimeters, as well as the tests and experimental comparisons carried out in this study using performance indicator parameters to characterize and validate the itinerant NaI(Tl) detection system in the calibration of activimeters.

Keywords: NaI(Tl), Method validations, Performance indicator parameters, Activimeter calibrations.

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Parâmetros para Validação de um Sistema Espectrométrico com NaI(Tl) Itinerante na Calibração de Ativímetros

Resumo: Nos centros de produção de radiofármacos e serviços de medicina nuclear, radionuclídeos de meia-vida curta são essenciais. Eles têm sido utilizados como radiotraçadores desde que seu uso aumentou significativamente e despertou o interesse dos laboratórios nacionais de metrologia, responsáveis por fornecer a rastreabilidade para padrões radioativos. Portanto, o conhecimento da atividade dos radiofármacos administrados deve ser obtido com precisão. No entanto, suas meias-vidas curtas impedem a garantia de rastreabilidade. Para superar esses obstáculos, um sistema itinerante baseado em um detector cintilador do tipo NaI(Tl) está sendo desenvolvido no Laboratório Nacional de Metrologia das Radiações Ionizantes (LNMRI). Este trabalho tem como objetivo apresentar o arranjo esquemático do sistema desenvolvido para calibrar activímetros, assim como os testes e comparações experimentais realizados neste estudo, utilizando parâmetros de indicadores de desempenho para caracterizar e validar o sistema itinerante de detecção NaI(Tl) na calibração de activímetros.

Palavras-chave: NaI(Tl), Validação de métodos, Parâmetros indicadores de desempenho, Calibrações de activímetros.

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1. INTRODUCTION

There is a pressing need to calibrate the activimeters used in nuclear medicine practices. This calibration of the equipment is one of the main regulatory requirements to authorize the operation of radiopharmacies in production centers as well as in nuclear medicine services [1]. Difficulties and limitations arise due to the properties of radiopharmaceuticals, especially because of their short half-lives that prevent the use of radioactive standards *in situ* [2]. To overcome these obstacles, the National Laboratory of Ionizing Radiation Metrology (LNMRI/IRD) is developing an itinerant system to provide traceability to radiopharmaceuticals based on a NaI(Tl) type scintillator detector [3,4]. Thus, to ensure traceability, the validation of the laboratory method used represents a step that will be in harmony with analytical reliability. In other words, it is to ensure, through evidence and measurement, those specific requirements for the intended use. NaI(Tl) detection systems, which are widely used as radiation monitors in different applications and installations, can provide operators and users with relevant information about the activity of radioactive sources, mainly due to their portability, robustness and accuracy of the data obtained.

Tests and experimental comparisons were carried out in this study to evaluate the performance and validate the NaI(Tl) detection system in order to ensure traceability for different radionuclides that have a simple decay scheme and emit gamma radiation between 100 and 700 keV [5,6]. The choice of ^{241}Am and ^{137}Cs standards meets one of the validation criteria, namely performing energy calibration for the detection system, covering both the lower and upper regions of the spectrum of interest. Additionally, these standards are readily available due to their well-defined energies and long half-lives. This energy range is predominant in the majority of radiopharmaceuticals supplied by production centers and used in hospitals and clinics for diagnosis or treatment.

Standards of ^{241}Am and ^{137}Cs were tested to verify the main parameters indicating their performance with the aim of validating laboratory methods to be in harmony with the analytical reliability of the method developed. In other words, it is to ensure, by means of analysis, measurements and evidence, those specific requirements for the intended use [7]. The characterization results will demonstrate the potential use of the proposed system to provide calibration of the activimeters that monitor the main radiopharmaceuticals.

2. MATERIALS AND METHODS

The system setup consisted of a 2.44 x 4.25 inch OSPREY - DTB (Canberra) planar, portable, inorganic Nal(TI) scintillator detector, as shown in figure 1. The Osprey has an integrated multichannel analyzer (MCA) tube that supports scintillation spectrometry. The spectra are obtained using a data acquisition and analysis program such as Genie 2000, which can be automatically adjusted to a low energy threshold, defining the start and end of the spectrum. The crystal is coupled to a photomultiplier plus associated electronics, with the detector supported by a tripod using an aluminum ring adapted for position adjustment. The detector is surrounded by a thin layer of lead, tin and copper shielding necessary to attenuate the radiation from low energy photons in the spectrum. The arrangement adopted can be controlled via USB or Ethernet, with only one connection cable for the control and data acquisition system [8].

LNMRI standard radioactive sources includes the effects of variations in sample: preparation, geometry and composition [9]. The standard sources used in the tests were of the punctiform and ampoule type for ^{241}Am (59.5 keV) and ^{137}Cs (661.6 keV). The activities on the reference date were 8 kBq - 45 kBq for point source and 9 kBq g^{-1} and 47 kBq g^{-1} for ampoule geometry, respectively. These sources were positioned directly above the top of the detector using PVC supports specially made for the system. The standard sources used in

the various tests were measured between May 23 and July 25, 2023, with counting times of 100 seconds and 10 repetitions for each. For the background measurements, conducted during the same period, a counting time of 2000 seconds was adopted. The indicator parameters used to characterize the NaI(Tl)-type scintillator detector will be discussed, with the main focus on its ability to accurately and precisely measure gamma emitters. To ensure the analytical reliability of the NaI(Tl) itinerant gamma spectrometry measurement system developed here, certified standards of ^{241}Am and ^{137}Cs were tested to verify the main parameters indicating its performance in the validation stage, such as: energy resolution, detection efficiency, reproducibility of sample positioning, effects of varying environmental conditions, characteristic limits - minimum detectable activity (AMD), linearity, dead-time, background radiation and robustness [4].

Figure 1: Arrangement of the gamma spectrometry system with NaI(Tl), containing the detector crystal, associated electronics, source holder, and lead shielding.

3. RESULTS AND DISCUSSIONS

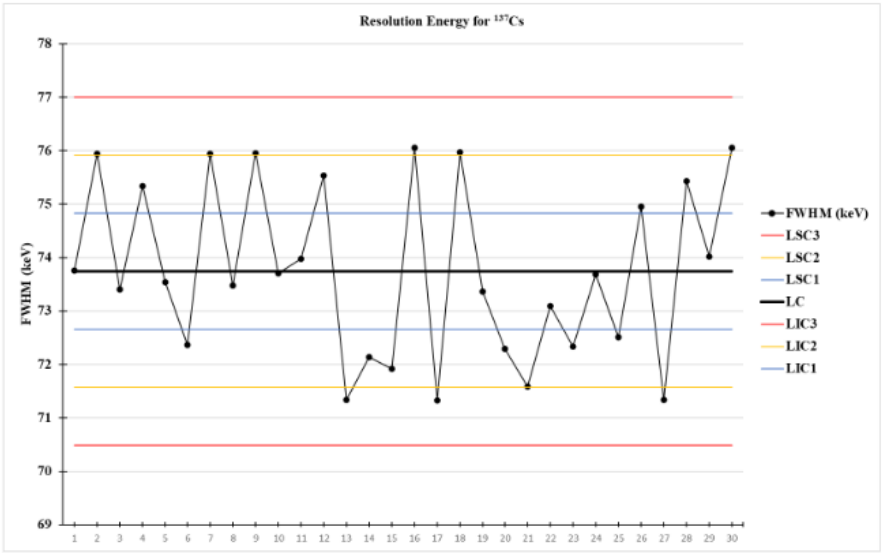
The analysis of the results describes the behavior and performance of the proposed measurement system for each selected indicator parameter using statistical methods. Previously, the system was energy-calibrated using the 59.6 keV and 661.6 keV peaks from the standard sources counted simultaneously. These peaks, selected in their respective channels of the spectrum, provided the Genie 2000 software with the necessary information to establish

the linear relationship between the channel and energy. The calibration result was stored in the system's memory and verified with each new measurement or when the equipment was restarted. Once established, this calibration enabled the execution of the remaining tests.

3.1. Energy resolution (FWHM)

Energy resolution, allows the detector to separate two neighboring photopeaks. This test must be carried out in the energy range of interest to the main gamma radiation emitter radiopharmaceuticals [4]. Measurements were made at energies of 59.5 and 661.6 keV to calibrate and check the resolution (FWHM) of the proposed system. Although tests were conducted for both standards, for illustration purposes and as it is used as a reference, only the energy resolution monitoring for ^{137}Cs was presented. Figure 2 shows the Control Chart, which demonstrates the good ability of the measurement system to discriminate the energies in the spectra obtained within the range considered. LSC, LC, and LIC indicate the upper, middle, and lower control limits, respectively. The numbers correspond to 1σ, 2σ and 3σ. The range considered is within the specifications for NaI(TI), between 6% and 13%, depending on the type and size of the crystal.

Figure 2: FWHM behavior for a 137Cs source. Values consistent with the manufacturer's specifications.



The efficiency calibration of the detection system was tested using the same standards. The response in a certain energy was obtained by dividing the net counts in each peak area by the certified activity value on the reference date. The activity values of the standard sources were derived from the primary and secondary systems available at LNMRI.The response to these results can be seen in table 1 where the adequacy in the Activity values was observed indicating consistency for the efficiencies as a function of energy. The total uncertainties are in percentages and Δ indicates the percentage deviation from the certified value.

Table 1: Values of activities in relation to a radioactive standard.

Radionuclide	Measurement 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

3.2. Reproducibility of sample positioning

This consists of the degree of agreement of the results obtained under different measurement conditions associated with the positioning of the sample [10]. The measurements were performed by rotating each tested source clockwise in 90° increments, resulting in averages calculated from of the mean of 10 readings.

Four different positions were taken for the set of ampoules and source holders. Ten measurements were taken at each position. The statistical test applied (ANOVA, F test) verified that the means obtained in each position are considered to be significantly equal, according to Tables 2 and 3. There was therefore no variation in the positioning of the samples for the ^{137}Cs liquid standard adopted, indicating a degree of agreement between the results obtained under different measurement conditions.

Table 2: Measurements obtained for the study of the positioning of the ^{137}Cs source.

Position	Counts	Mean	Variance
1	2322597	232259.7	1251298.90
2	2318030	231803.0	550089.78
3	2321625	232162.5	1130854.28
4	2319312	231931.2	1424076.62

Table 3: F-statistical test, analysis of variance indicating that the means obtained in table 2 are significantly equal.

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

Which: F is the F-statistic value, also known as the variance ratio; SQ (Sum of Squares)- refers to the sum of squared differences between the means or between the observed data and the predicted means; gl (Degrees of Freedom) - the number of degrees of freedom, related to the number of observations and groups in the study; MQ (Mean Squares)- obtained by dividing the sum of squares (SQ) by the degrees of freedom (gl); Value-P (p-value) - the probability value associated with the F-test. Indicates statistical significance; Fcritic (Critical F) - the cutoff value for the F-statistic, obtained from an F-distribution table.

3.3. Effects of varying environmental conditions

Temperature, humidity were monitored between May 23 and July 25, 2023. The temperature values ranged from 19 °C to 22 °C, while the humidity value was below 60%. Atmospheric pressure was taken at sea level. The environment in which the temperature and humidity measurements were taken had controlled parameters. Therefore, the values

obtained here are consistent with those specified by the manufacturer, which are: temperature – range: -10 to 50 °C; humidity – up to 85% in accordance with the technical specifications contained in the equipment's operating manual. For this validation to be suitable for the environment in which the mobile system is used, the minimum environmental conditions obtained here must be maintained.

3.4. Characteristic limits (AMD)

This corresponds to the lowest value of activity concentration that the sample must contain for the detection of a given radionuclide to be possible, depending on the characteristics of the measurement system and the analytical methods adopted [11]. The calculation for AMD depends on the background counts, the counting time for each standard, and the detection efficiency at each energy. Therefore, the counting time for the background was 2000 s, and the counting time for the standards was 200 s. The values presented for the background measurement correspond to 10 readings, with the average value taken as representative. The AMD here was determined for ^{241}Am and ^{137}Cs, whose values obtained were 4 and 9 Bq respectively on the reference date for ampoule geometry.

3.5. Linearity

In linearity, there are two variables to be correlated: the Activity quantity of the reference used and the response provided by the measurement system. In this way, it will always be possible to construct a graph that represents the quantity of interest on the x - axis (activity), and the response obtained from the measurement system on the y - axis (counting). Here a minimum of five sources with different activities distributed over the intended working range was used to construct the analytical curve. Figure 3 a and b below illustrates the ratio between the response of the measurement system as a function of the activity of the standard ^{137}Cs sources and the residual profile.

Figure 3(a): Response of the measurement system as a function of activity for standard ^{137}Cs sources.

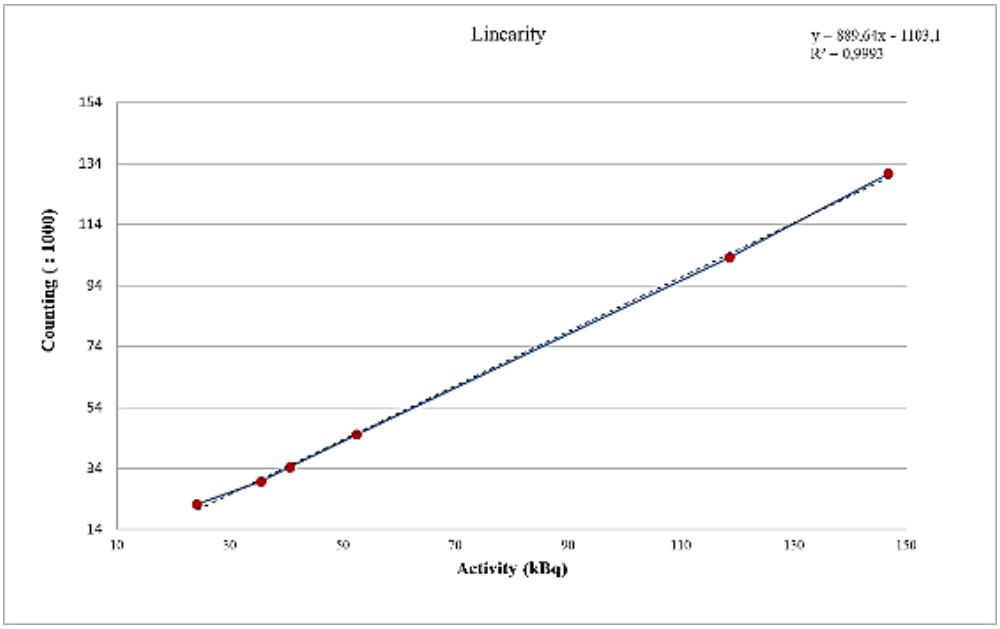
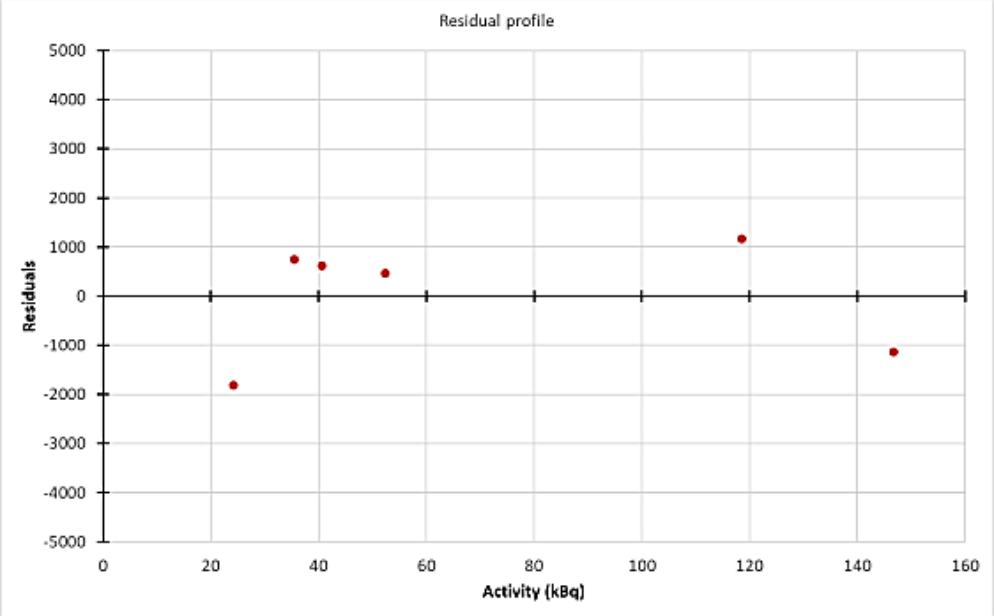


Figure 3(b): Residual profile



3.6. Dead-time

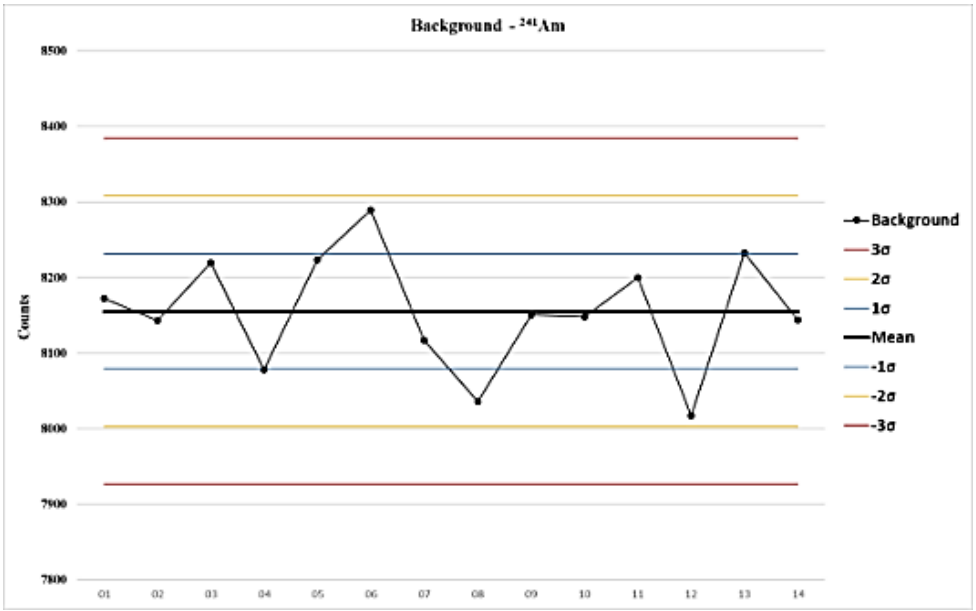
This is related to the accuracy of the correction technique, defined by the difference between real time and live time. The dead time in all the measurements carried out on this

system did not exceed 3.9%, thus conforming to with Standard ISO/20042, which requires a value of less than 10% [4].

3.7. Background radiation

Background radiation takes into account not only the events in the spectrum that form a smooth curve on which the photopeaks are superimposed, but also the contribution of environmental radiation external to the detection system. The background measurements were made using empty vials. Figure 4 shows the results of the control chart for individual values and the moving amplitude. The data obtained for background radiation shows that the points remained between the upper and lower control limits.

Figure 4: Background control charts for ^{241}Am



3.8. Robustness

The tests were conducted and documented and small changes in experimental conditions were evaluated in relation to the parameters defined above. The set of the response obtained showed greater confidence in using the proposed method.

4. CONCLUSIONS

The control parameters adopted here proved to be suitable for evaluating performance in terms of characterizing the proposed detection system. According to the monitoring carried out on the performance parameters, via statistical tests, control charts, comparison of detection efficiencies and monitoring of environmental conditions, they proved to be compatible, not only in terms of reliability, but also the robustness of the data obtained for the system presented. In short, this phase of characterizing the spectrometric arrangement has demonstrated the potential use and suitability of the proposed system to provide future in situ calibration of the activimeters that monitor the main radiopharmaceuticals used in nuclear medicine services.

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