



Parameters for Validating a Spectrometric System with Itinerant NaI(Tl) in the Activimeter's 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, knowledge 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



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.

1. INTRODUCTION

There is a pressing need to calibrate the activity meters 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 660 keV [5,6]. 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 NaI(Tl) 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 including 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 (661.7 keV). The activities on the reference date were 8 kBq - 45 kBq, punctiform 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 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 Na(Tl) roving 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).



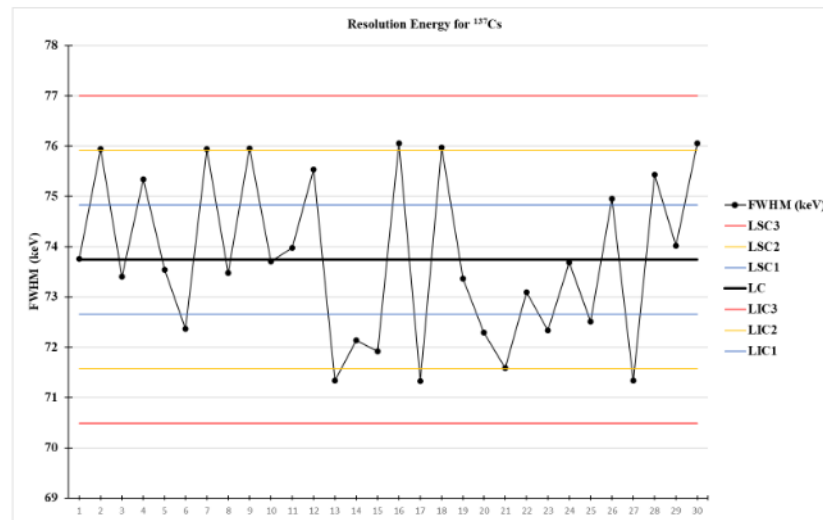
3. RESULTS AND DISCUSSIONS

The analysis of the results describes the behavior and performance of the proposed measurement system for each selected indicator parameter using statistical methods.

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 physicians [4]. Measurements were made at energies of 59.5 and 661.7 keV to check the resolution (FWHM) of the proposed system. 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. The range considered is within the specifications for NaI(Tl).

Figure 2 - FWHM behavior for a ^{137}Cs source.



3.2. Detection efficiency

The answer 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

3.3. 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]. Four different positions were taken for the set of ampoules and source holders with half-life radionuclides in ampoule geometry. 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.

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 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				

3.4. Effects of varying environmental conditions

Temperature, humidity and atmospheric pressure. These parameters were monitored for a certain period of time and, in accordance with the technical specifications contained in the equipment's operating manual, they remained strictly within the respective ranges specified by the manufacturer, demonstrating the robustness of this measuring system in the face of variations in environmental parameters.

3.5. 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 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.

3.6 - Linearity

A minimum of five sources with different activities distributed over the intended working range were used to construct the analytical curve. Figure 3 a and b show the results which show the ratio between the response of the measurement system as a function of the activity of the standard ^{137}Cs sources and the residue profile.

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

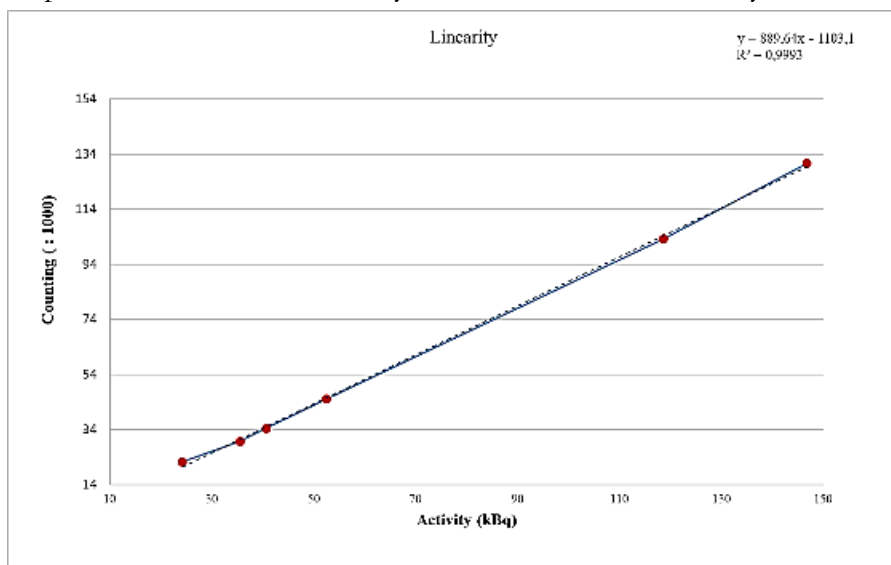
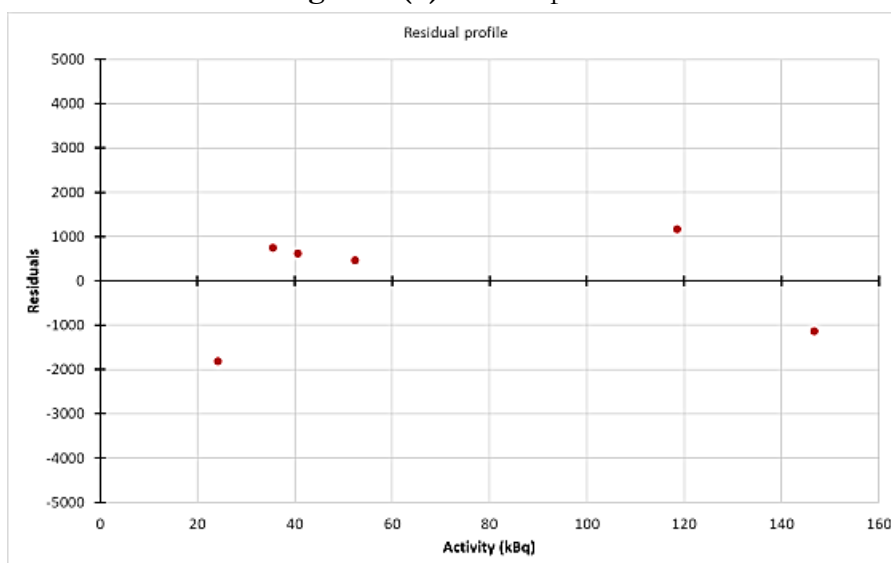


Figure 3 (b) - Waste profile



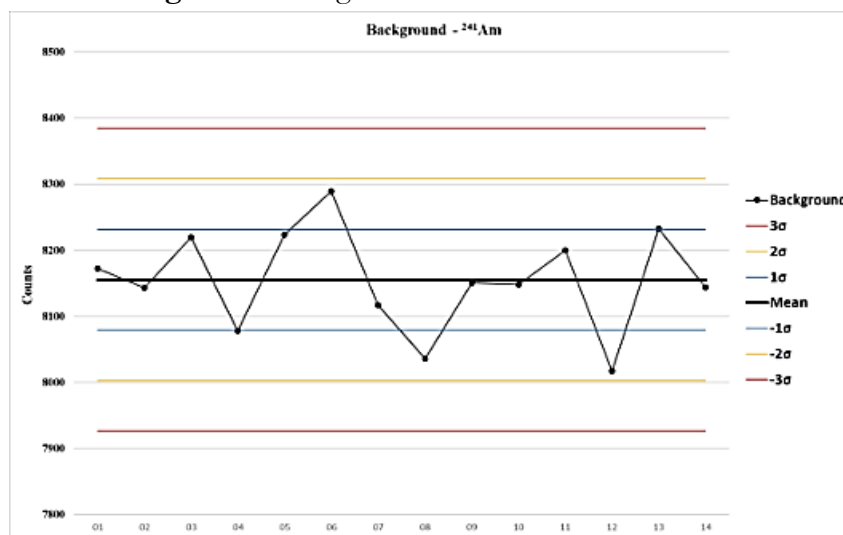
3.7 - Dead time

This is related to the accuracy of the correction technique, defined by the difference between real time and live time. The dead time for all the measurements carried out on this system did not exceed 3.9%, indicating that, according to Standard 20042, in energy calibration it should be less than 10%[4].

3.8. 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. 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 ²⁴¹Am



3.9. 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 answers 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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