ABSOLUTE STANDARDIZATION OF $^{22}$Na, $^{65}$Zn and $^{60}$Co BY SUM-PEAK METHOD AND CRITICAL ANALYSIS OF INSTRUMENTATION

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Araújo M T F$^{1,2}$, Poledna R$^1$, da Silva R L$^1$, Delgado J U$^1$, de Almeida M C M$^1$, Lopes R T$^2$, Alfredo L F P$^{1,2}$, Veras E V$^1$, Rangel J$^1$, Cruz P A L$^1$, da Silva C J$^1$.

$^1$Laboratório Nacional de Metrologia das Radiações Ionizantes (LNMRI/IRD/CNEN), Av. Salvador Allende, s/n, Recreio, CEP 22780-160, Rio de Janeiro, Brazil, (miriamtaina@hotmail.com)

$^2$Laboratório de Instrumentação Nuclear (LIN/PEN/COPPE/UFRJ), Caixa Postal 68509, CEP 21945-970, Rio de Janeiro, Brazil, (miriamtaina@hotmail.com)

Abstract. The sum-peak method was used to obtain the absolute activity value for the $^{60}$Co, $^{65}$Zn and $^{22}$Na radioisotopes. Three HPGe detectors were used, being a planar, a coaxial and well type. The values of the activity obtained by sum-peak method are consistent, with values of uncertainty less 0.5% for all radioisotopes considering k = 1. LNMRI has introduced a new Calibration System that uses a HPGE type well detector. Thus, a comparative study was made between the electronics available at the time of conception of the method and the one currently used.

1. Introduction

The application of the sum-peak method to obtain the absolute value of activity has been reported in several publications made by LNMRI. The method can be employed in emitters that decay by electronic capture and β$^+$ emitters that emitting a subsequent γ-ray. Here was considered three radioisotopes of interest that meet the requirements for the application of the method: $^{22}$Na, $^{65}$Zn and $^{60}$Co.

The increasing use of radiopharmaceuticals composed of positron-emitting radionuclides in nuclear medicine has opened up recurrent discussions about their applications in Positron Emission Tomography (PET) imaging tests. In order to maintain the accuracy during the calibration procedure of the activity meters (activimeters) it is necessary to have a standardized source for this procedure to be performed. However, many difficulties end up appearing throughout the absolute standardization procedure of the sources due to the fact that most of the radionuclides used have short half-lives where many of them do not exceed 110 min [1]. The $^{22}$Na reveals itself as an important target for study since it has one of the longest half-lives (2.6029 years) between the postiron emitters. The $^{22}$Na presents no major difficulties for
the absolute calibration by the sum peak method because it has an interference-free spectrum. Its main characteristic is given by the decay $\beta^+$ which emits a subsequent gamma. $^{65}$Zn decays by electron capture and around 2.5% by $\beta^+$ through two branches with intensities of $\epsilon_1 = 0.5075$ and $\epsilon_2 = 0.4779$, respectively. The electron capture branches are followed by $\gamma$-ray emission mainly of 1115.5 keV (0.5075 intensity) and two others of negligible intensity (less than 0.001). This fact makes this radionuclide practically a monoenergetic $\gamma$-ray emitter and applied as a reference source in $\gamma$-ray spectrometry [2]. For $^{65}$Zn the sum peak method is easily applied since the planar type detector has a thin beryllium window which allows detection of the X-ray of interest of 8 keV.

$^{60}$Co is considered a perhaps the simplest nuclide exhibiting $\beta$-$\gamma$ cascade decay, and can be easily standardized by sum-peak method. Regarding the instrumentation used by Brinkman and his collaborators who proposed the methodology in 1962, some aspects need to be carefully observed. These aspects of greater importance reflect the current days where more powerful and modern instrumentation is available.

2. Experimental

Three type’s detectors of HPGe were used: a coaxial, a planar and a well-type with 20, 40 and 70% relative efficiency respectively. The well-type detector was submitted to an implantation and implementation procedure in the LNMRI and its performance regarding the accuracy and precision of results was expressed in comparison to the values obtained from the other two HPGe detectors of the laboratory. Each detector applied in the measurements has an appropriate electronics composed basically of the elements: High voltage supply, signal amplifier and a multichannel analyzer.

Point sources were prepared with the aid of a pycnometer, depositing drops of radionuclide solution in a polystyrene film, with a thickness of 0.05 mm, set in one acrylic ring. The ring has an external diameter of 15 mm, inner diameter of 4 mm and a thickness of 1 mm. Once dried, the sources were covered with the same polystyrene film [3]. The energies of 1173 keV and 1332 keV were used for a $^{60}$Co measurement.

The 8 keV X-ray and the 1115 keV gamma ray were selected to standardize the $^{65}$Zn and $^{22}$Na were selected as energies of 511 keV and 1274 keV. The measurement of sources was provided on various positions to applied sum-peak method. The counting time of each sample varied by about 24 h for each radionuclide and the dead-time was below 4% for all measurements. The control of radionuclidic impurities was also done and the presence of other radioisotopes in the samples analyzed was not verified.

3. Methodology

3.1 Sum-peak method

For all isotopes, the situation is simple, since there are only two lines for analysis. In this case we used two lines corresponding to X-ray and $\gamma$-ray or $\gamma$-$\gamma$ interaction. The activity was obtained by the classical formula given by Brinkman et al. (1963).

$$N_0 = N_T + \frac{N_1N_2}{N_{12}}$$ 

(1)
Where, $N_T$ are the counts under the total spectrum, $N_1$ is the x-ray count ($K_\alpha+K_\beta$) or the first $\gamma$, $N_2$ is the less peak count and $N_{12}$ is the sum-peak count. The sum-peaks are characterized for each radioisotope as being: $^{60}\text{Co}$: $(1173 + 1232 = 2405 \text{ keV})$; $^{22}\text{Na}$: $(511 + 511 = 1022 \text{ keV})$; $\gamma$-ray = $1274 \text{ keV}$; $511 + 1274 = 1785 \text{ keV}$; $1022 + 1274 = 2296 \text{ keV}$; $65\text{Zn}$: $(1115 + 8 = 1123\text{keV})$.

### 3.2 Efficiency Curve

The Efficiency Curve method consists of a methodology to obtain the photopeak efficiency for spectrometry system. The efficiency of photopeak is defined as the ratio between the rate of radiation detected by the system and the rate emitted by the source. With the use of calibrated standard sources of known energies and intensities, using the same detection system and the same counting geometry, an energy efficiency curve is obtained through the relation:

$$\varepsilon = \frac{S}{T \cdot A \cdot K \cdot P_\gamma}$$

Where $S$ is energy photopeak area of a standard source at $(s^{-1})$, $T$ is the count time $(s)$, $A$ is activity of standard source at $(s^{-1})$, $K$ is decay factor of the radionuclide standard source and $P_\gamma$ is a probability of emission for the gamma radiation of photopeak in $(s^{-1})$.

The efficiency curve was obtained using various radionuclidic standardized sources such as: $^{166m}\text{Ho}$, $^{152}\text{Eu}$, $^{133}\text{Ba}$, $^{137}\text{Cs}$, $^{134}\text{Cs}$, $^{241}\text{Am}$, $^{54}\text{Mn}$, $^{65}\text{Zn}$ totaling 59 energy points and the graphic can be observed in Figure 1. The HPGe spectrometric system for detector 40% has been calibrated in efficiency through the use of standard point sources. The range of energy was established between 48 keV and 1427 keV originally. Then a cut was made considering only the energies above 100 keV since the low energy region was not necessary for the purpose of the calibration.

![Efficiency curve](image)

Figure 1. Efficiency curve obtained (linear – long scale) for the spectrometry system using the radionuclidic standards in point source geometry: $^{166m}\text{Ho}$, $^{152}\text{Eu}$, $^{133}\text{Ba}$, $^{137}\text{Cs}$, $^{134}\text{Cs}$, $^{241}\text{Am}$, $^{54}\text{Mn}$, $^{65}\text{Zn}$. 

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4. Results and discussion

The results obtained for the three radioisotopes are shown to be coherent and with reduced uncertainty values and compatible with those obtained by the $4\pi\beta$-$\gamma$ system. Regarding instrumentation, it should be noted that at the time the standardization methodology known as sum-peak method was developed, the idealizers had a multichannel analyzer with 100-channel RIDL to measure $N_1$, $N_2$ and $N_{12}$ while $N_T$ was obtained by the intermediary of counter. In their study, there is no approach of the dead time parameter of the system since they did not have this function. The modern spectrometer with 16,000 channels has a dead-time management system and simultaneously provides $N_T$, $N_1$, $N_2$, and $N_{12}$ values, as well as its uncertainties. Table 1 shows the mean activity values for the three radioisotopes of interest and Table 2 presents an example of uncertainty values for $^{60}$Co, which is also valid for the other isotopes.

| Table 1. Activity values for $^{22}$Na (Sum-peak for detector wheel-type) |
| Source | Activity (kBq) | Uncertainties (k = 1) | $U_a$% | $U_b$% |
| 06S16 | 1.188 | 0.03 0.23 |
| 07S16 | 2.418 | 0.02 0.23 |
| 08S16 | 1.470 | 0.02 0.23 |
| 09S16 | 3.364 | 0.02 0.23 |
| 10S16 | 1.951 | 0.26 0.34 |

| Table 2. Activity values for $^{22}$Na (Efficiency Curve for detector 40%) |
| Source | Activity (kBq) | Uncertainties (k = 1) | $U_a$% | $U_b$% |
| 06S16 | 1.317 | 0.16 0.3 |
| 07S16 | 2.563 | 0.05 0.3 |
| 08S16 | 1.546 | 0.15 0.3 |
| 09S16 | 3.373 | 0.10 0.3 |
| 10S16 | 1.838 | 0.13 0.3 |

| Table 3. Activity values for $^{60}$Co (Sum-peak for detector wheel-type) |
| Source | Activity (kBq) | Uncertainties (k = 1) | $U_a$% | $U_b$% |
| 44S16 | 4.421 | 0.03 0.23 |
| 45S16 | 3.578 | 0.04 0.23 |
| 46S16 | 2.975 | 0.04 0.23 |

| Table 4. Activity values for $^{60}$Co (Sum-peak for detector 20%) |
| Source | Activity (kBq) | Uncertainties (k = 1) | $U_a$% | $U_b$% |
| 44S16 | 4.343 | 0.03 0.23 |
| 45S16 | 3.539 | 0.04 0.23 |
| 46S16 | 2.929 | 0.04 0.23 |

| Table 5. Activity values for $^{65}$Zn (Sum-peak for detector 20%) |
| Source | Activity (kBq) | Uncertainties (k = 1) | $U_a$% | $U_b$% |
| 22S16 | 3.186 | 0.25 0.34 |
| 23S16 | 2.959 | 0.26 0.34 |
| 24S16 | 3.572 | 0.26 0.34 |
| 25S16 | 2.675 | 0.17 0.34 |
| 26S16 | 2.570 | 0.74 0.34 |

| Table 6. Activity values for $^{66}$Zn (Efficiency curve for detector 40%) |
| Source | Activity (kBq) | Uncertainties (k = 1) | $U_a$% | $U_b$% |
| 22S16 | 2.887 | 0.27 0.7 |
| 23S16 | 2.315 | 0.46 0.7 |
| 24S16 | 3.723 | 0.18 0.7 |
| 25S16 | * | * |
| 26S16 | 2.320 | 0.31 0.7 |
Table 7. $^{60}$Co uncertainty of activity measured by sum peak method.

| Uncertainty Components       | A (%) | B (%) |
|------------------------------|-------|-------|
| Live time                    | -     | 0.01  |
| Weight                       | -     | 0.20  |
| Half life                    | -     | 0.10  |
| Background                   | -     | 0.02  |
| Decay                        | -     | 0.01  |
| Counting statistic           |       | 0.03  |
| **Combined uncertainty (k = 1)** |       | 0.23  |

5. Conclusion

The objective of this work was to do an exercise in the use of the well-type HPGe detector that was implanted in the LNMRI recently. The sum-peak method made it possible to obtain values of activity with uncertainties of 0.23% for $^{60}$Co, 0.23% for $^{22}$Na and 0.26% for $^{65}$Zn. In addition to being fast, this method made it possible to obtain good values so much for the activity as to the precise determination of emission probabilities for the radionuclides analyzed.

6. References
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