IMPLEMENTATION OF A SECONDARY STANDARDIZATION SYSTEM APPLYING A WELL-TYPE IONIZATION CHAMBER

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ABSTRACT

Radioactive materials are applied in industry, medicine, radiation protection and environmental control. In many of these applications the necessary accuracy must be lower than 2\%. The most used primary calibration methods of radioactive solutions are the coincidence, anti-coincidence and sum-peak [1]. Usually the standardization using primary methodologies requires long counting and analysing times. Additionally, complex sample preparation techniques are also necessary. An alternative and efficient methodology is working with a stable standardization system that allows registering the results of the primary standardization and not introducing significant uncertainties in the results [2]. Among the systems, which are used in the secondary standardization and presents a high stability is the well-type ionisation chamber connected to a digital electrometer. Recently, the Laboratório Nacional de Metrologia das Radiações Ionizantes of Instituto de Radioproteção e Dosimetria, LNMRI/IRD, acquired an ionization chamber model IG 11 manufactured by Centronic and connected it to a Keithley model 6517A electrometer.

In order to verify the stability of this system, measurements with a long life radionuclide (\textsuperscript{226}Ra) were performed during an observation period of 280 days where a stability of 0.4\% was observed. This system allows calibrating \textsuperscript{241}Am, \textsuperscript{239}Am, \textsuperscript{133}Ba, \textsuperscript{137}Cs, \textsuperscript{134}Cs, \textsuperscript{57}Co, \textsuperscript{152}Eu, \textsuperscript{67}Ga, \textsuperscript{131}I, and other solutions with a maximum standard uncertainty of 2.0\%.

This work has an objective to demonstrate the calibration process of this secondary system, the determination of uncertainties of each radionuclide and the metrological traceability of the participation in the key-comparisons organized by Bureau International des Poids and Mesures (BIPM).

1. INTRODUCTION

The ionization chambers are relatively simple to operate and give measurement results with good reproducibility and accuracy.

A calibrated re-entrant ionization chamber working in current mode is suitable for measuring the activity of a radioactive source that contains a single or more photon-emitting radionuclide.

A re-entrant or $4\pi\gamma$ ionization chamber can be calibrated in terms of activity by appropriate radioactive standard sources, also called activity standards. Radioactivity standards are prepared and calibrated mainly in national standards laboratories that are responsible for
radionuclide metrology [1]. Other radioactivity standards are traceable to national standards, which have been the subject of international comparisons or have been compared with the International Reference System (SIR). The SIR is maintained at the BIPM and takes the form of an ionization chamber measuring system for comparing the activity values of radioactive solutions [2]. The LNMRI has been participated in key comparisons organized by BIPM. The activity of a radioactivity standard is measured by direct or absolute methods. The absolute method used at LNMRI is the coincidence method, anti-coincidence, sum-peak and liquid scintillation.

The stability of an ionization chamber measuring system is usually verified by repeated measurements of a reference source of long-lived radionuclide such as \(^{226}\text{Ra}\) in equilibrium with its daughters. Sample, background and reference source measurements are alternated in a cycle, which checks the stability of the equipment and the reproducibility of the measurement conditions.

2. METHODOLOGY

The reference system with ionisation chamber is compound by the following equipments: well-type ionisation chamber model IG11 manufactured by Centronic; Keithley electrometer model 6517A that allows the programming for the data acquisition in automatic way. Radioactive solutions standardized by absolute methods such as \(4\pi\beta-\gamma\) coincidence counting has been used for calibration factors determination. The solutions previously standardized and with metrological traceability established by means of the key-comparisons organized by BIPM have been used to preparing 5ml standard glass ampoule with masses of about 2.60 g solution. With these solutions the calibration factors of the ionisation chamber were determined [3]. Equation 1 allows obtaining the calibration factor, whose dimension is the ratio electrical current/activity (A/Bq) [3]. A stability study of the system was carried out in the first step taking as reference the ionising current yielded by a \(^{226}\text{Ra}\) source. This radionuclide was used due to its long half-life of 1600y [4].

The methodology used for the verification of the stability of the system was:
- 6 measurements of background (BG) with count time of 600 s in the scale of 20 nC.
- 6 measurements of the source of \(^{226}\text{Ra}\) with count time of 120 s in the scale of 20 nC.

The measurements are carried out twice, totalising twelve (12) measurements of \(^{226}\text{Ra}\) and twelve (12) measurements of BG, in each day.

The data collected in the measurements are launched in a worksheet of excel for accomplishment of the calculations of the current produced by the source of \(^{226}\text{Ra}\).

After verification, the determination of the calibration factors of the following radionuclides was carried out: \(^{241}\text{Am}, \ ^{243}\text{Am}, \ ^{134}\text{Cs}, \ ^{131}\text{I}, \ ^{57}\text{Co}, \ ^{60}\text{Co}, \ ^{133}\text{Ba}\) and \(^{137}\text{Cs}\).

The methodology used for the determination of the calibration factor was:
- methodology of the stability of the system;
- 2 sequences of 21 measurements for each ampoule with count times varying from 60 to 3000s, in 20 or 200 nC scale, depending on the source activity.
The data collected in the measurements are launched in a spreadsheet of Excel for accomplishment of the calculations of the calibration factor, as well as the uncertainties associated.

Internal comparisons using different measuring systems were used in order to achieve higher trueness in the measurements.

2.1. Calculation of the Calibration Factor

The mathematical expression used for the calculation of the calibration factor of the source is presented below:

\[
F = \frac{I_A - I_B}{A \cdot m} \cdot f_n
\]

where:
F – calibration factor in A/Bq,
m – mass of the solution in g (differential weighing),
\(I_A\) – current produced by the source,
\(I_B\) – current of background (BG),
f\(_n\) – normalization factor, and
A – activity in Bq/g.

The normalization factor is the ratio between the ion current of a \(^{226}\)Ra source measured in a reference date and the ion current of the same \(^{226}\)Ra source in the measurement date of the source.

2.1.1. Results

The results of currents produced in the measurements of the sample of \(^{226}\)Ra (\(I_{Ra}\)) are presented in Figure 1.

![Figure 1. Stability of the Calibration System](image-url)
Through the Fig.1, a fluctuation of the chart control produced for the source of $^{226}\text{Ra}$ of up to 0.4% can be observed.

Table 1 presents the calibration factors determined in the ionization chamber system. The presented uncertainties have a coverage factor of $k=2$. It can be perceived that only the radionuclides of difficult standardization or lower energies, as for example, the radionuclides $^{241}\text{Am}$, $^{243}\text{Am}$, $^{152}\text{Eu}$, and $^{67}\text{Ga}$ presented larger uncertainties than 1.6%.

Table 1. Calibration Factors

<table>
<thead>
<tr>
<th>Radionuclides</th>
<th>Calibration Factor (A/Bq)$^{-17}$</th>
<th>$U_a$ (%)</th>
<th>$U_b$ (%)</th>
<th>$U(%)$ k=2</th>
<th>Traceability</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{241}\text{Am}$</td>
<td>0.3983</td>
<td>0.55</td>
<td>0.38</td>
<td>1.3</td>
<td>BIPM.RI(II)</td>
</tr>
<tr>
<td>$^{243}\text{Am}$</td>
<td>1.9067</td>
<td>0.42</td>
<td>0.71</td>
<td>1.6</td>
<td>LNMRI</td>
</tr>
<tr>
<td>$^{133}\text{Ba}$</td>
<td>1.9032</td>
<td>0.13</td>
<td>0.19</td>
<td>0.46</td>
<td>BIPM.RI(II)</td>
</tr>
<tr>
<td>$^{57}\text{Co}$</td>
<td>9.8840</td>
<td>0.12</td>
<td>0.18</td>
<td>0.43</td>
<td>BIPM.RI(II)</td>
</tr>
<tr>
<td>$^{60}\text{Co}$</td>
<td>8.7802</td>
<td>0.09</td>
<td>0.15</td>
<td>0.35</td>
<td>BIPM.RI(II)</td>
</tr>
<tr>
<td>$^{134}\text{Cs}$</td>
<td>6.3046</td>
<td>0.29</td>
<td>0.19</td>
<td>0.69</td>
<td>BIPM.RI(II)</td>
</tr>
<tr>
<td>$^{137}\text{Cs}$</td>
<td>2.2986</td>
<td>0.23</td>
<td>0.27</td>
<td>0.59</td>
<td>LNMRI</td>
</tr>
<tr>
<td>$^{131}\text{I}$</td>
<td>1.6612</td>
<td>0.16</td>
<td>0.34</td>
<td>0.75</td>
<td>BIPM.RI(II)</td>
</tr>
<tr>
<td>$^{152}\text{Eu}$</td>
<td>4.6665</td>
<td>0.49</td>
<td>0.55</td>
<td>1.5</td>
<td>BIPM.RI(II)</td>
</tr>
<tr>
<td>$^{67}\text{Ga}$</td>
<td>0.9867</td>
<td>0.47</td>
<td>0.41</td>
<td>1.2</td>
<td>LNMRI</td>
</tr>
</tbody>
</table>

Table 2 presents the results of the comparisons organized internally in the LNMRI laboratory.

Table 2. Results of internal comparisons

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Activity (kBq/g) – IG11 3*</th>
<th>Activity (kBq/g) – IG12 **</th>
<th>Deviation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{133}\text{Ba}$</td>
<td>15.808 ± 0.237***</td>
<td>15.763 ± 0.299</td>
<td>0.28</td>
</tr>
<tr>
<td>$^{152}\text{Eu}$</td>
<td>11.251 ± 0.203</td>
<td>11.245 ± 0.304</td>
<td>0.05</td>
</tr>
<tr>
<td>$^{60}\text{Co}$</td>
<td>11.982 ± 0.073</td>
<td>12.096 ± 0.096</td>
<td>0.90</td>
</tr>
<tr>
<td>$^{137}\text{Cs}$</td>
<td>12.428 ± 0.124</td>
<td>12.572 ± 0.339</td>
<td>1.09</td>
</tr>
</tbody>
</table>

*IG11 3 – new calibration system in implantation; **IG12 – old calibration system already implanted; ***k=2
The results show the coherence of the measurements carried out in the two systems.

3. CONCLUSIONS

Based on the obtained data experimentally, it can say that the ionization chamber system complies the requirements of a detection system with metrological characteristics. A better dimensional tolerance of the holder that is responsible to set up the standard sources reduced the data deviation obtained using the $^{226}$Ra source.

As suggestion, an accompanying of the stability of the system must be carried through in order to analyse the reasons of the fluctuation of the chain trying to reduce this value to 0.1%.

REFERENCES