^{226}Ra ACTIVITY STANDARDIZATION BY 4παβ(LS)-γ(NaI-Tl) ANTICOINCIDENCE COUNTING

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Abstract: From a commercial supplier a solution containing ^{226}Ra has been standardized at National Laboratory for Ionizing Radiation Metrology (LNMRI) for the first time using Liquid scintillation based measurement. The measurement methods are 4παβ-γ live-timed anticoincidence counting. The results obtained by anticoincidence counting were adopted as reference value and its combined uncertainty was 0.94 %. The agreement of this reference value with reference system with ionization chamber was 0.56 %. This standardization was made in order to reduce the uncertainty in ^{226}Ra measurement in Brazil.

Keywords: Anticoincidence Counting, primary standardization.

1. INTRODUCTION

^{226}Ra is a very complex gamma emitter with half-life of 1600 years decay for ^{222}Rn with half-life of 3.822 days, and gamma energy emission from 186 to 600 keV [1], the figure 1 shown the ^{226}Ra decay chain. In Brazil this radionuclide have been used as standard for environmental, water and food control. LNMRI supplies users with this standard solution and also in water matrix for comparison program in which take part 27 laboratories. The primary standardization of ^{226}Ra is very important also due its potential occurrence in drinking water. In order to perform the measurement of these radionuclide two independent methods were used: anticoincidence counting and Ionization chamber method. The results of activity concentration values presented very close agreement, within 0.60 % for a radionuclide with a very complex decay scheme. The difficult in the ^{226}Ra primary standardization result of decay scheme and also due ^{222}Rn. The gas presence in the source could in some cases produce stability in the source during the counting process.

2. METHODS

2.1 4πβ (LS) – γ (NaI (Tl)) live-timed Anticoincidence counting

The anticoincidence method is a complementary method of the coincidence method that initially was considered by Bryant[2] and later applied to the particular case of nuclides that present meta-stable levels emitting delayed gamma rays in the decay scheme Bryant [3] . Later on, Baerg al. [4] introduced the use of live-timed anticoincidence counting that eliminates the correction of dead-time using an extending dead-time device. In the present version LNMRI uses two modules of MTR2 in its anticoincidence system Bouchard, [5]. The description and operation of this anticoincidence system may be found in literature Bouchard [5], da Silva et al., [6], [7].

The anticoincidence set-up consists of a beta cell made of polyethylene painted inside with TiO_2 in order to reflect light and one glass vial containing the scintillation cocktail. The cell is coupled to a 10.2 cm x 10.2 cm (diameter x length) cylindrical crystal of NaI(Tl) for gamma counting events. The scintillations generated by beta events originating from the ^{226}Ra decay and its daughters are collected by two photomultipliers placed 180^{o} each other with respect to the cell. The gamma 839 keV of the ^{214}Pb was select as window and counted by NaI(Tl) crystal and also processed by the electronic chain of gamma channel.

The activity of a radionuclide can be determined using equation 1, which is the classic equation of the coincidence method differing only in that N_c, the coincidence count rate, that is determined for one given gamma window as the difference between the gamma rate and uncorrelated gamma rate:

$$A = \frac{N_\beta \, N_\gamma^w}{N_\gamma^w - {}^{i}N_\gamma^w} \qquad (1)$$

where, N_β is the count rate in the beta channel; N_γ^w is the count rate in the gamma window, and ${}^{i}N_\gamma^w$ is the uncorrelated gamma count rate.

The variation of $\varepsilon_{\beta\alpha}$ was carried out by electronic discrimination and ranged from 0.98 to 0.90. The activity per mass is determined by extrapolation for beta efficiency of 100 % where A (apparent activity) became A_o.

2.2 Source preparation

All counting samples for the primary methods were prepared using commercial LS cocktail (five in 3 mL Optiphase 'HiSafe' 3 (HS) manufactured by PerkinElmer, Inc. USA. The chemical composition of ^{226}Ra was in the form of a solution in 0.1 mol L^{-1} HCl and approximately 25 μg/g BaCl as inactive Carrie. The LS cocktail and masses ^{226}Ra are deposited in a NIST glass ampoule and flame sealed. The masses samples were determined in a Mettler Toledo XP 58 micro analytical balance and ranged from 20 to 25 mg. For background measurement two vials are prepared with same LS cocktails and volume. No relevant gamma impurities were observed by gamma spectrometry measurements using a germanium detector. For ionization chamber one sample was prepared in LNMRI standard ampoules mass around 2.6 grams.

3. EXPERIMENTAL PROCEDURE

3.1 $4\pi\beta$ (LS) – γ (NaI (Tl)) Anticoincidence counting with live-time

All four LS sources were counted for determining the activity of the master solution. In this present anticoincidence system the MTR2 modules allow operating with dead time values from 25 to 200 μs. In the measurements of ^{226}Ra it was used a minimum dead time of 50 μs for both beta and gamma channels. The LS beta efficiency, ε_β was varied using electronic discrimination and extrapolated for $\varepsilon_\beta = 1$. The discrimination level was varied from 0.1 V to 1.0 V, with step of 0.1 V.

Each experimental point is the mean value of 5 measurements with time counting interval of 120 s, for each source 1 extrapolation was performed. For each discrimination level were made the background measurement with vial blank containing 10 ml LS cocktails.

The LNMRI/IRD algorithm for this activity determination take in account the live time correction, background and decay correction[6], it was validate in 2005 by comparison with LNHB program and have agreement better than 0.05 % for ^{67}Ga measurement[7].

The NaI(Tl) was gated from 700 keV to 900 keV region using a single channel analyzer. The LS beta efficiency, $\varepsilon_{\beta\alpha}$ was varied between 0.98 and 0.90 by electronic discrimination.

4. MEASUREMENT OF IMPURITIES

No gamma impurities above the detection limits of our Ge detector spectrometer for ^{226}Ra solution, in this solution we cannot found also ^{210}Pb. The measurement was performed with 4 hours of measurements.

5. RESULTS AND DISCUSSION

The sources were prepared in April 2014, and the measurement was performed in October 2014. The minimum time for the sources achieve the equilibrium of the decay chain is 40 days due half-life of ^{222}Rn. The activity concentration value for ^{226}Ra was 714.066 kBq/g in 2014/10/01 12:00 UTC determined by anticoincidence counting. In Table 1 are presented the results of two methods which agree within a standard deviation, LNMRI adopted as reference value the results obtained by anticoincidence counting. ^{226}Ra was standardized by primary method at LNMRI/IRD it this is an effort to offer primary standards with minor uncertainties in order to guarantee the quality of the medicines made in Brazil. This radionuclide is widely used in reference systems with ionization chamber to verify their performance and stability [8], [9].

Tabela 1. Activity comparison by two methods

M. Method	Activity (Bq/g)	(%)
Anticoincidence	714066	0.0
Ionization Chamber	709674	0.60

Tabela 2. Uncertaity Anticoincidence Counting

Uncertainty due	Type	(%)
Mass	B	0.05
Live time	B	0.01
Background	B	0.06
Extrapolation	A	0.38
Counting statistics	A	0.22
Half -life	B	< 0.01
C. uncertainty		0.45

Tabela 3. Uncertaity Ionization Chamber

Uncertainty due to	Type	(%)
Mass	B	0.05
Timing	B	0.05
226Ra standard	B	0.90
226Ra Current	A	0.15
Counting statistics	A	0.22
Half -life	B	< 0.01
C. uncertainty		0.94

The tables 2 and 3 uncertainties values of these measurement methods are presented and they are lower than 1.0 %. The uncertainty in extrapolation curve and also statistic counting are the main sources of uncertainty in anticoincidence measurement method. The problems related to primary standardization of ^{226}Ra are mainly due the very complex decay scheme and also because ^{222}Rn is a gas that could cause the stability in the sources during standardization.

6. CONCLUSIONS

A solution with ^{226}Ra free of impurity has been calibrated at LNMRI/IRD for the first time with a combined uncertainty of 0.45 % working with Live-timed anticoincidence counting. The results obtained for ^{226}Ra activity per mass agree with value obtained by ionization chamber system 0.60 %.

This standard solution is in LNMRI/IRD stock of standard and cold be used in the future studies. The LNMRI could also supplier users with ^{226}Ra standard with uncertainty lower than 1.0 % it is a good value for a radionuclide with a very complex decay scheme.

7. REFERÊNCIAS

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