Development of multi-system for measurement of radionuclide absolute activity

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Abstract: The development of a multi-system triple-to-double coincidence ratio (TDCR) and coincidence 4πβ-y methods, based on liquid scintillation to radionuclide standardization is presented in this work. The adjustments of multi-system were made using standards of ^{3}H and ^{14}C and ^{60}Co. The initial stage was performing measurements of pure beta emitters ^{3}H, ^{63}Ni, and ^{90}Sr^{90}Y standard solutions by TDCR. The results were consistent within the standard uncertainty. Measurements will be performed with a beta-gamma ^{60}Co in a comparison to the SIR / BIPM to assess the multi-system's performance.

Keywords: TDCR, coincidence, multi-systems, radionuclide standardization, radionuclide metrology.

1. INTRODUCTION

The current trend in the field of Radionuclide Metrology is to make use of multi-system based on liquid scintillation technique, which has the advantage of using the same information from radionuclide events together data acquisition systems based on a high-speed digitizers, where the A/D conversion is performed as close as possible to the output of the detector or preamplifier, in contrast to conventional analogue systems [1, 2].

There is an abundant literature on the application of TDCR and coincidence 4πβ-γ to standardization of radioactive sources. They are the absolute methods that have great versatility as may be applicable to all sources that decay by simultaneous emission (intervals less than 10^{-10} s) of two or more particles, such as β-γ, α-γ, X, e_A-γ and even pure beta emitters using a β-γ radionuclide as tracer [3, 4, 5, 6, 7]. They are reference methods to radionuclide standardizing after a large time of theoretical and experimental studies carried out by radionuclide laboratories of the National Metrology Institutes of the international BIPM network.

This work presents the development of a multi-system that uses the methods TDCR and coincidence 4πβ–γ based on liquid scintillation technique. However, in this first stage only will be made measurements TDCR. The basic process of liquid scintillation is converting the energy of the radionuclide decay-scheme into light photons. The scintillation process occurs when a radionuclide solution is dissolved in liquid scintillator cocktail and the energy of the particles is transferred to the chemical molecules, with consequent light emission [8].

The TDCR method is based on the free parameter model for the distribution of scintillation photons and their probabilities of detection in a counting system consisting of three photomultipliers [9]. The model takes into account the corrections for loss of linearity in the light emission by ionization quenching generated by the interaction of particles with the molecules of the chemical environment]. Birks [10] studied this physio-chemical process and developed an expression to represent the quenching effect correction.

Computational codes have been developed to evaluate the theoretical curve TDCR and efficiency in function of the free parameter for different quenching parameters (kB), from de data of the radionuclide nuclear decay-scheme, as well as, the interaction of the particles with scintillating cocktail and materials of the multi-system, considering its geometry, by use of Monte Carlo Simulation (Penelope 2008).

In coincidence $4\pi\beta$-γ method, the traditional analogue configuration consists of two detectors, an 4π gas flow proportional counter coupled to a scintillating NaI(Tl) crystal with their respective electronic chains, each responding exclusively to a type of particle emitted by the source. The signal originated by the events of the particles 1 and 2 are computed independently, therefore the system has a third channel capable of quantifying only the events that occurred simultaneously in the detector, that is coincident. Currently, liquid scintillation detectors replace the gas flow proportional counter.

2. EXPERIMENTAL PROCEDURE

2.1. Multi-system

The multi-system consists of a detection cell of Polyvinyl chloride (PVC). A glass vial with radionuclide solution dissolved in a liquid scintillator is placed in its center of the cell. Three photomultipliers are placed at 120 degrees from each other fixed in the cell, and a NaI(Tl) detector (BICRON). This system is placed inside an aluminum, copper, and lead shield. The signals obtained from photomultipliers (HAMAMATSU) are processed by electronic units and the acquisition and register the counts is made by LabVIEW computational code.

2.1.1 Characterization of multi-system

The geometry, dimensions, stochiometric cocktail composition, air and materials of the multi-system were applied to the Penelope 2008 to simulate the interaction particles process and obtaining of the data used in computational code to evaluate the efficiency.

2.1.2 Adjustment of multi-system and measurement of the radioactive sources

The multi-system was adjusted using ^{3}H, ^{14}C and ^{60}Co standards to perform the best measurement conditions of the multi-system. Three sources were prepared by adding an aliquot of ^{3}H standard solution in 15 mL of Ultima Gold cocktail in glass vials. Another's four sources of ^{63}Ni and ^{90}Sr^{90}Y standard solutions were prepared in 15 mL of HiSafe3 cocktail. These sources were measured by TDCR method to compare the results of activity with reference values.

The basic system uses three photomultipliers that provides three double coincidence placed 120 degrees one each other, and a time base. This way, it provides counts of the individual photomultipliers (A, B, C), logical sum (A + B + C), three double coincidences (AB, BC, AC), and a triple coincidence (ABC). The experimental TDCR is obtained by ratio of triple and double coincidence, which is interpolating in an efficiency curve TDCR versus efficiency for different quenching parameter (kB) from Birks equation. Then, the radionuclide activity is obtained by ratio between experimental double coincidence counts and efficiency.

The counting efficiency variation was performed by use of the filters of increasing optical density around sample glass vials for ^{3}H and ^{63}Ni and ^{90}Sr^{90}Y to evaluate the quenching parameter (kB).

3. RESULTS

The sources of pure beta emitters, ^{3}H (18,564 keV), ^{63}Ni (66.980 keV) and ^{90}Sr^{90}Y (545.9 and 2,278.7 keV), represent a large beta energy spectrum for the multi-system. A TDCR07 code [11] was used to evaluate the TDCR and efficiency theoretical curve in function of the free parameter, to permit the interpolating of the experimental TDCR and the calculate the activities. The double efficiency variation of sources is significative according kB value (0.007 and 0.020) in low and media energy: in order of 0.5818 to 0.5454 for ^{3}H and 0.8217 to 0.7975 for ^{63}Ni. In case of ^{90}Sr^{90}Y, a high energy beta emitter with a range of 0.9932 to 0.9916, the efficiency variation is not significant.

The figure 1 illustrates the procedure to obtain the values of quenching parameter for ^{3}H (kB = 0.016), ^{63}Ni (kB = 0.012), and ^{90}Sr^{90}Y (k B = 0.007) used to evaluate the respective activities.

The table 1 shows that results of the activities for the standard sources and table 2 the uncertainties components.

To compare the results of measurements and reference values, the activities and uncertainties of the standards and the measurements obtained were normalized to the respective standard reference values, according figure 2. The results obtained are in agreement within of the standard uncertainty.

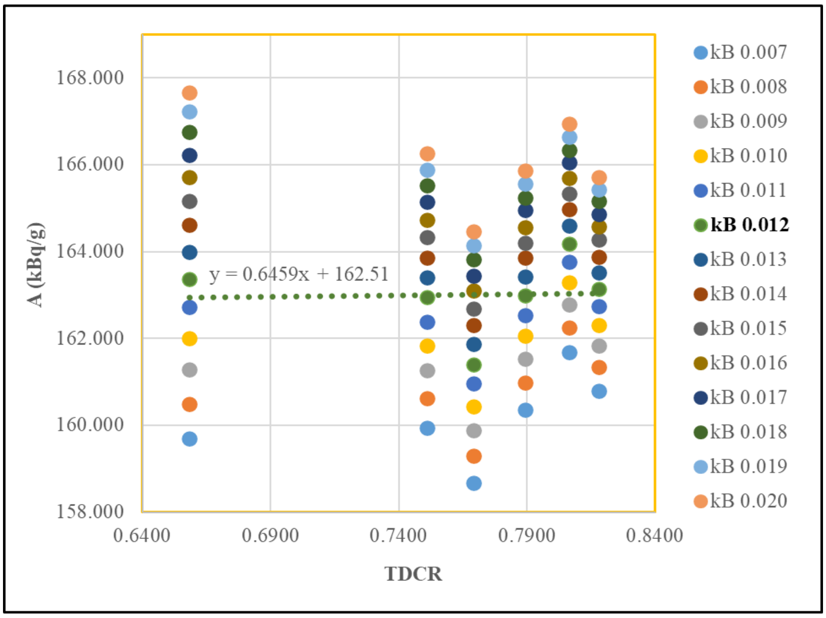

Figure 1. Experimental procedure to determine quenching (kB).

Table 1. Reference data and results obtained by TDCR.

	Standard Solution			**Measurements**		
Nuclide	**Reference date**	**Activity (kBq/g)**	**Uncertainty (k = 2)**	**Activity (kBq/g)**	**Uncertainty U (k = 2)**	**Difference (%)**
^{3}H	02/06/20	95.398	3.40	93.699	1.00	1.78
^{63}Ni	12/18/19	163.495	1.30	164.021	0.84	0.32
$^{90}Sr^{90}Y$	06/03/20	500.006	0.92	498.451	0.56	0.31

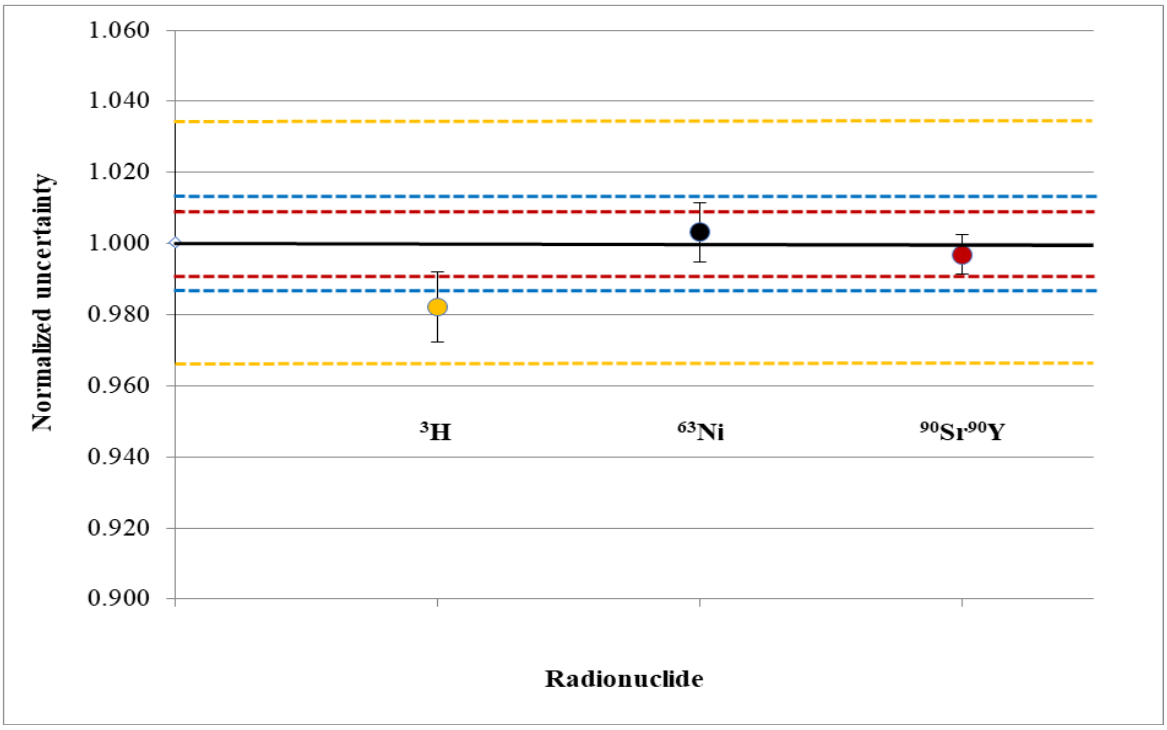

Figure 2. Normalized uncertainty of radionuclide standard solutions and results obtained by TDCR.

Table 2. Uncertainty budget of the measurement of the standards solutions by TDCR method.

Nuclide/Comp.	**Weight**	**Counting statistics**	**Activity (quenching kB)**	**U (k = 1)**	**U (k = 2)**
^{3}H	0.05	0.30	0.40	0.50	1.00
^{63}Ni	0.05	0.21	0.36	0.42	0.84
$^{90}Sr^{90}Y$	0.05	0.19	0.20	0.28	0.56

4. CONCLUSION

The status of the development of the multi-system that uses TDCR and coincidence methods based on liquid scintillation, especially in the face of consistent measurement results obtained to ^{3}H, ^{63}Ni, and ^{90}Sr^{90}Y performed by TDCR, opens a good prospect of progress towards digital multi-system. Measurements will be performed with a beta-gamma ^{60}Co in a comparison to the SIR / BIPM to assess the multi-system's performance in TDCR and coincidence 4πβ-γ**.**

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