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Development of multi-systems for measurement of radionuclide absolute activity

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ABSTRACT

The development of a multi-systems triple-to-double coincidence ratio (TDCR) and coincidence $4\pi\beta$–γ methods, based on liquid scintillation to radionuclide standardization is presented in this work. The adjustments of multi-systems 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.**

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1. INTRODUCTION

The current trend in the field of Radionuclide Metrology is to make use of multi-systems 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, eA-γ and even pure beta emitters using a β-γ radionuclide as tracer [3-7]. 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 the multi-systems that uses TDCR and Coincidence 4πβ−γ methods based on liquid scintillation technique. However, in this first stage, only the measurements on the TDCR will be performed. 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 versus 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 ionizing radiation with scintillating cocktail and materials of the multi-system, considering its geometry, by use of Monte Carlo Simulation (PENELOPE 2008) [11]. This is important to assess the absorption or escape of the ionizing radiation energy by scintillating liquid as a function of detection geometry.

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. MATERIALS AND METHODS

2.1. Multi-system

The multi-systems consist 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 of the counts is made by LabVIEW computational code.

2.1.1 Characterization of multi-system

The geometry, dimensions, stoichiometric cocktail composition, air and materials of the multi-systems were applied Monte Carlo Simulation (PENELOPE 2008) to simulate the interaction process of the ionizing radiation and obtaining of the data used in computational code to evaluate the efficiency. it is important to assess the absorption or escape of radiation energy by scintillating liquid as a function of detection geometry.

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

The multi-systems were 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 3H 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 coincidences 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 between the 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 from interpolating.

The counting efficiency variation was performed by use of the grey 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).

According to Broda [12], the fluorescence yield of the scintillator is given by Birks expression,

$$L(E) = \eta_0 \int_0^E \frac{dE}{1 + kB\,\frac{dE}{dx}} = \eta_0 E Q(E)$$

and

$$Q(E) = \frac{1}{E}\int_0^E \frac{dE}{1 + kB\,\frac{dE}{dx}}$$

where η_0 is the scintillation efficiency (figure of merit or number of fluorescence photons emitted per unit of energy); Q(E) is the ionization quenching function; kB is the ionization quenching parameter (Birks parameter) in units of $cm.MeV^{-1}$; and dE/dx is the stopping power of the incident particle (defined as the average energy dissipated by the ionizing radiation per unit path length of travel in a medium).

Applying the Poisson statistical distribution, for pure beta-emitters, the ratio between triple and double coincidences (TDCR) to the system with equals three photomultipliers is,

$$\frac{\varepsilon_T}{\varepsilon_D} = \frac{T}{D} = TDCR = \frac{\int_0^{E_{max}} S(E)\left(1-e^{-\frac{EQ(E)}{3\lambda}}\right)^3 dE}{\int_0^{E_{max}} S(E)\left(3\left(1-e^{-\frac{EQ(E)}{3\lambda}}\right)^2 - 2\left(1-e^{-\frac{EQ(E)}{3\lambda}}\right)^3\right) dE}$$

where $\int_0^{E_{max}} S(E)\, dE$ is the Fermi expression to the beta particle and (λ) is the free parameter.

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-systems.

A TDCR07 code [13] was used to evaluate the TDCR versus efficiency theoretical curve in function of the free parameter, to permit the interpolating of the experimental TDCR and to determine the activities.

3. RESULTS AND DISCUSSION

The double efficiency variation of sources is significant according kB value (0.007 and 0.020 cm.MeV-^{1}) 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 cm.MeV-^{1}), ^{63}Ni (kB = 0.012 cm.MeV-^{1}), and $^{90}Sr^{90}Y$ (kB = 0.007 cm.MeV-^{1}) used to evaluate the respective activities. The table 1 shows the results of the activities for the standard sources. The TDCR07 code was run from the atomic and nuclear radionuclide data, the free parameter model using Poisson statistical distribution, (PENELOPE 2008) to characterize the ionizing radiation interacting in liquid scintillation components according to detection geometry of the TDCR system and the quenching correction from Birks equation. The code provides the TDCR versus double theoretical efficiency curve for kB values (quenching factor). The activity is obtained by ratio between experimental value of the double coincidence counts and theoretical double efficiency obtained by interpolating of the experimental TDCR into TDCR versus double efficiency curve from TDCR07 code, and radionuclide solution mass, to the kB value determined by use of grey filters.

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 standard uncertainties are obtained by quadratic sum of the type A and B uncertainty components, according to the Guide to the expression of Uncertainties in Measurements [14].

The results obtained in table 1 and 2 are in agreement within of the standard uncertainty. However, the TDCR system needs to some adjust because the result obtained to the ^{3}H is reasonable kB value lower than 0.015 cm.MeV^{-1} and the best activity result in comparison to the activity of its respective standard solution, as presented in figure 2.

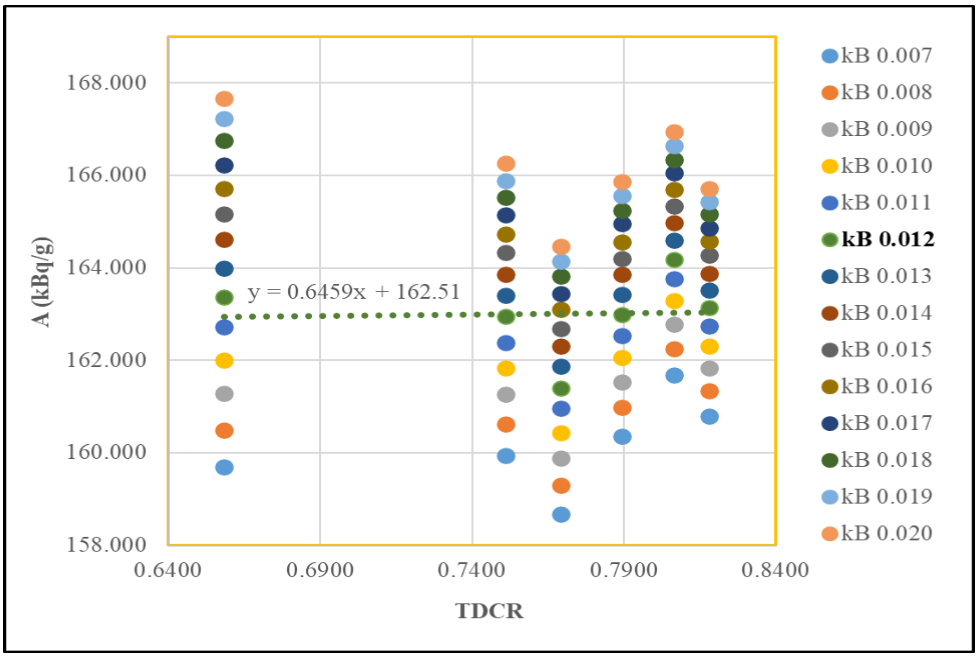

Figure 1: Procedure to determine quenching (kB) by using grey filters.

Table 1: Reference data and results of radionuclide solution activities 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

Table 2: Uncertainty budget of measurements of the standards solutions by TDCR.

Nuclide	**Weight**	**Counting statistics**	**Quenching (kB)**	**U (k = 1)**	**U (k = 2)**
Unc. Type	**B**	**A**	**A**		
^{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

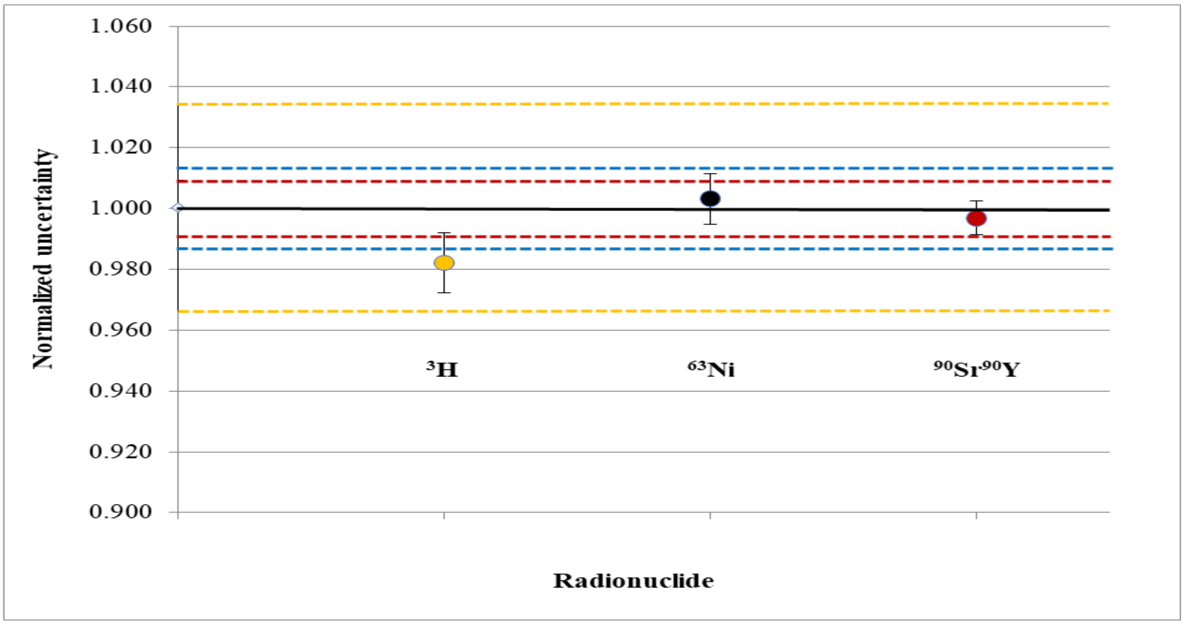

Figure 2: Normalized uncertainty of radionuclide standard solutions and results obtained by TDCR.

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-systems. 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 $4\pi\beta$-γ. The advantage of the multisystem is that both primary methods use the primary event produced by the interaction of the ionizing radiation in the scintillating liquid, which allows a better comparison and reliability of the results.

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