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The TDCR Primary System for Radionuclide Standardization in LNMRI/IRD (Brazil)

R S Gomes[1], C J da Silva[1], R L da Silva[1], A L Ferreira Filho[1], J A Rangel[1], R Poledna[1] and J U Delgado[1].

[1] LNMRI - National Metrology Laboratory of Ionizing Radiation, IRD - Institute of Radioprotection and Dosimetry, Rio de Janeiro, Brazil.

regio.gomes@ird.gov.br

Abstract. In this article we will describe the radionuclide standardization system TDCR, Triple-to-Double Coincidence Ratio, of the *National Metrology Laboratory of Ionizing Radiation* and its stage of development to provide a method to standardize radionuclides difficult by other techniques. The TDCR is an absolute measurement method, therefore, it does not depend on a reference standard to determine the radioactive activity of the samples, initially developed to standardize radionuclides that decay by pure beta emission (electrons and positrons), such as ^{3}H, ^{14}C, ^{45}Ca and ^{63}Ni and electronic capture, such as the ^{55}Fe. However, it is possible to standardize alpha emitters, such as ^{241}Am, and other radionuclides with a complex decay scheme. The method uses three photomultiplier tubes aligned at an angle of 120° to each other, and double and triple coincidences between the three photomultiplier tubes are collected. The method is currently used in 26 radionuclide metrology laboratories around the world. In this article we also present the radionuclides that will be standardized by the method.

1. Introduction

Although the Liquid Scintillation Counting (LSC) technique for activity measurement was already known around 1950 and applied in several areas such as metrology, engineering, agronomy, arts, medicine, etc., radiation detectors based on this technique only became widespread from the 1980s.

The liquid scintillation measurement technique is one of the most sensitive and versatile for detecting and quantifying radioactivity, mainly for measuring alpha and beta emitters, offering many advantages. Among them, we can highlight its high detection efficiency, since the radioactive solution is dissolved directly in the scintillating solution [1], simplicity in sample preparation and ability to simultaneously analyze different radionuclides.

Liquid scintillators are usually composed of an organic solvent and one or more fluorescent solutes. In preparing samples for liquid scintillation counters, radioactive material is added to the scintillant cocktail. The energy emitted from the decay of the radionuclide is transferred to the solvent by ionization and excitation, and then transferred to the solute that will re-emit it as photons with wavelengths in the ultraviolet and violet range [2].

These photons, having an average wavelength characteristic of the solute, can be detected at the photocathode of one or more photomultipliers. After photon multiplication, the signal is sent to an amplifier and finally recorded in a counter or multichannel.

Measurements in a liquid scintillation counter are made by analyzing the energy spectrum. However, some interferences occur, which must be corrected, as they change the total efficiency by shifting the measured energy spectrum to lower values. This interference is called quenching, a term used to describe various physical or chemical processes that reduce the emission of light from the scintillator.

2. TDCR method

In 1979, Krzysztof Pochwalski, at the Institute of Nuclear Research, IBJ (Instytut Badań Jądrowych), in Warsaw, now Radioisotope Center POLATROM, developed a triple LSC detector with a coincidence counting system, and an innovative method of absolute activity measurement. for pure beta emitters. The method became known as Triple-to-Double Coincidence Ratio or, simply, TDCR [3].

In 1982, the implementation of the TDCR method at the Henri Becquerel National Laboratory (LNHB) in Saclay, France, enabled its development [4]. In this same laboratory, a dedicated data processing system was created, the innovative MAC3, praised for its real-time pulse counting solution [5]. Adding this to the improvement of the method by Ryszard Broda in 1988, which laid the theoretical foundations to calculate the detection efficiency, it was possible to measure the activity with only 1.0 % of uncertainty [6].

The TDCR method is an absolute measurement method, therefore, it does not depend on a reference standard to determine the radioactive activity of the samples, initially developed to standardize radionuclides that decay by pure beta emission (electrons and positrons), such as ^{3}H, ^{14}C, ^{45}Ca and ^{63}Ni and pure EC-emitters, such as ^{55}Fe. However, it is possible to standardize alpha emitters, such as ^{241}Am, and other radionuclides with a complex decay scheme [7]. In this method, the detection efficiency is calculated using a physical and statistical model of the photon distribution emitted by the source The method uses three photomultiplier tubes aligned at an angle of 120º to each other, and the double and triple coincidences between the three photomultiplier tubes are collected, according to the scheme in figure 1 [8].

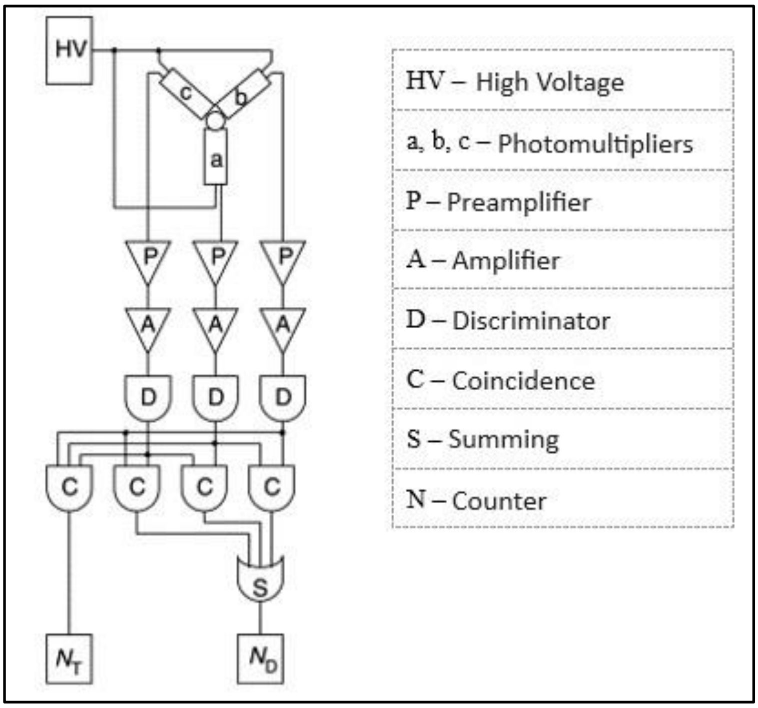

Figure 1. Schematic diagram of the TDCR system, with three photomultipliers a, b and c positioned at an angle of 120° between them and containing the flask with the radioactive sample in the center of the set.

The double count rates for the three pairs are N_{AB}, N_{BC}, and N_{AC} as these represent the logical sums of the double count rates. N_D and N_T represent the logical sums of the triple count rates, respectively. The arithmetic relationship between these counting rates is given by [9]:

$$N_D = N_{AB} + N_{BC} + N_{AC} - 2N_T \tag{1}$$

Double and triple match rates are defined as:

$$N_D = N_0\, \varepsilon_D \tag{2}$$

e

$$N_T = N_0\, \varepsilon_T \tag{3}$$

where ε_D and ε_T are the double and triple efficiencies and N_0 is the activity.

For many detected events, the ratio of triple match rates to N_T/N_D double match rate converges to the ratio of counting efficiencies, this rate is called TDCR efficiency:

$$N_T/N_D = \varepsilon_{TDCR} \tag{4}$$

Once the counting rates are obtained, the data can be analyzed by programs such as the TDCR07c code, from the LNHB, or the MICELLE2 code from the Physikalisch-Technische Bundesanstalt, PTB, Germany [10], obtaining a graph that relates the experimental value of the TDCR with counting efficiency, allowing the calculation of the activity of the radionuclide under study. This relationship differs greatly between radionuclides, as exemplified in figure 2 [7].

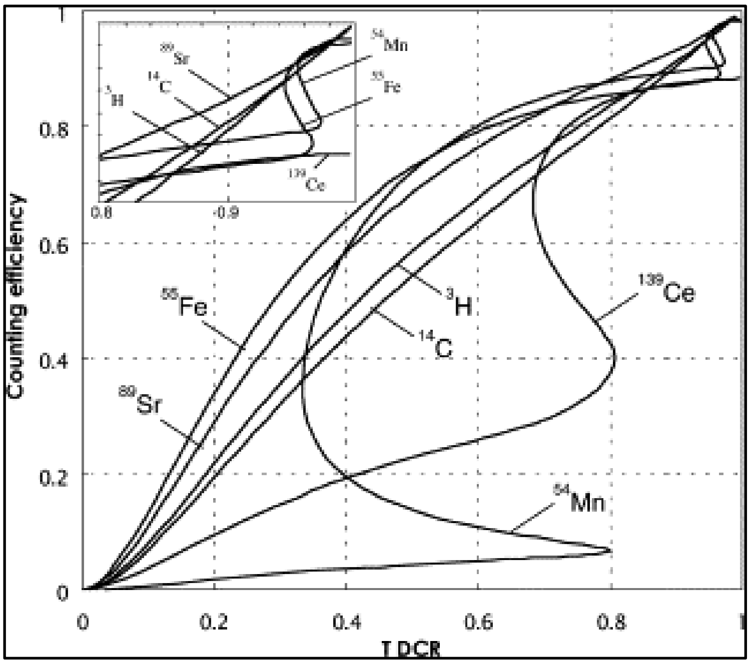

Figure 2. Counting efficiency as a function of TDCR for some beta-emitting radionuclides and electron capture.

Counting efficiency can be varied by voltage discrimination methods, addition of nitromethane or by using filters in the radioactive sample scintillation vials.

At the 21st edition of the *International Conference on Radionuclide Metrology*, ICRM in 2017, the main congress in radionuclide metrology, papers were presented, showing the continuous improvement of the TDCR method, and revealing that 26 metrology institutes in different countries have adopted the method [11 -16].

3. The experimental setup

The experimental setup of the TDCR measurement system consists of a shield, still under construction, where the measurement cell will be housed, a rack with two NIM racks, where the electronic modules, necessary for the acquisition and processing of signals, were installed, and a computer connected to the equipment through the GPIB interface, where the data acquisition program is executed.

3.1. Shielding

The shielding, which is under construction by SECAME/IRD, IRD's Mechanical and Electronic Engineering Sector, will have a metal structure, measuring 101 x 70 cm and 160 cm high, according to the schematic design in figure 3.

The shielding will use lead blocks measuring 10 x 10 x 5 cm, with a mass of 5.66 kg each block. The estimated number of lead blocks needed for the shield is 187 blocks and the estimated total mass of the shield will be 1,100 kg = 1.1 tonnes.

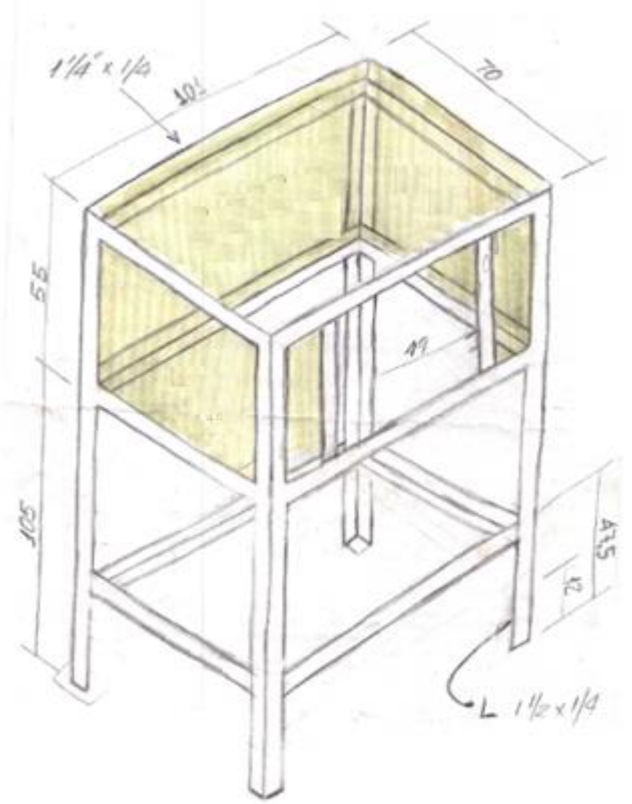

Figure 3. TDCR shielding schematic diagram. Designer: Fabricio Oliveira (SECAME/IRD).

3.2. *Measuring cell*

The measuring cell (optical chamber) of the TDCR system and its schematic diagram can be seen in figure 4. The photomultiplier tubes make an angle of 120° between them. The sample contained in a liquid scintillation vial with the scintillant cocktail and the radionuclide is positioned in the center of the array.

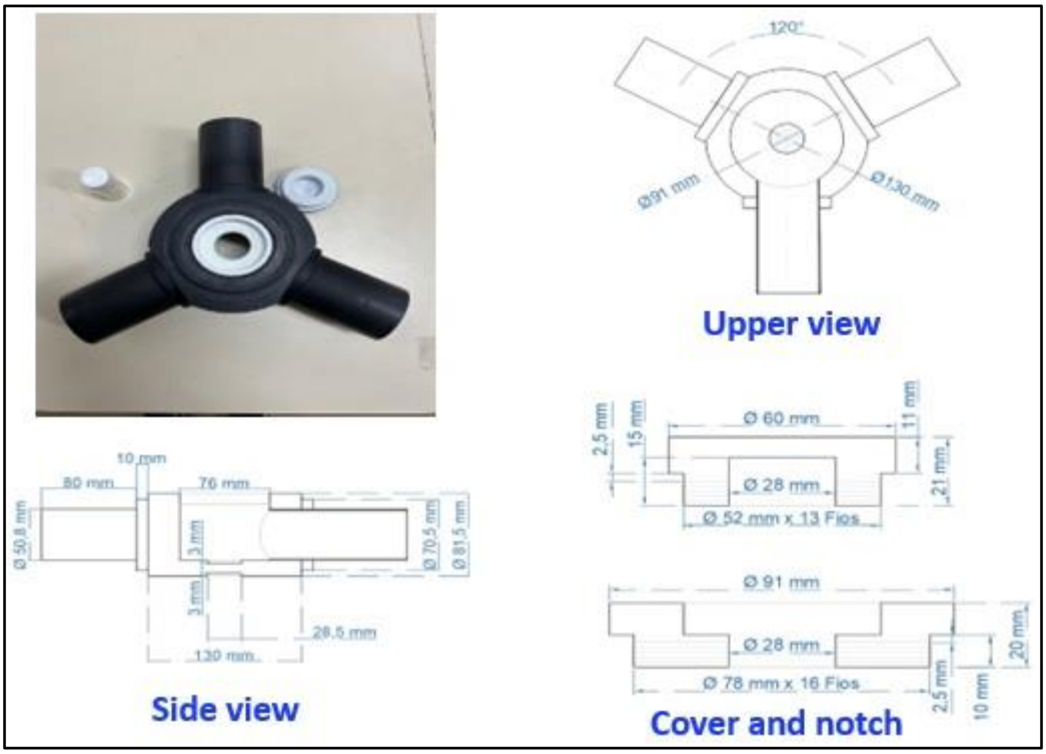

Figure 4. Schematic diagram of the TDCR measuring cell (Designers: Fabrício Oliveira - SECAME/IRD and Felipe Santos).

3.3. *Electronic modules*

- Three Hamamatsu model R331-05 photomultiplier tubes;
- Three bases for ORTEC 265A 21-pin photomultiplier tubes;
- Two high voltage sources of 3 kV ORTEC 556;
- Three preamplifiers ORTEC 113 for photomultiplier tubes;
- NIM ORTEC 4001A rack with ORTEC 4002D continuous voltage source
- Tennelec TC 410A pulse delay generator;
- ORTEC 9309-4 Quad Fast Amplifier;

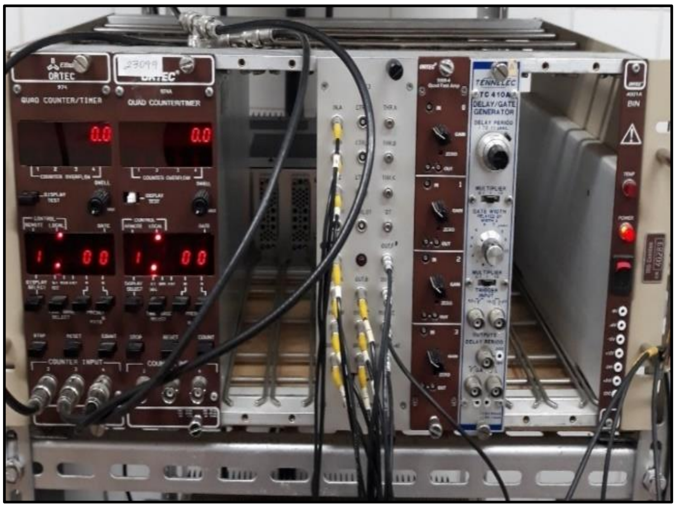

Figure 5. ORTEC NIM rack and electronic modules.

- ➢ MAC3 double and triple coincidence module (BNM-CEA-LPRI);
- ➢ Three ORTEC 974 quad/counters.

The electronic modules seen in figure 5 are connected to the Dell Optiplex 7040 computer, via serial ports and GPIB interface for the data acquisition system.

3.4. Data acquisition system

The TDCR data acquisition system was developed using the National Instruments LabVIEW graphical language. The counts acquired from measurements of radioactive samples are stored in a MySQL database and in a Microsoft Excel spreadsheet for verification and validation of information. The data acquisition system runs on a Dell Optiplex 7040 computer.

3.5. Nuclear parameters database

The nuclear parameters of the radionuclides will be stored in a MySQL database modeled using the entity relationship model [17-18]. The modeling has 28 tables. In figure 6 we can see some of the tables.

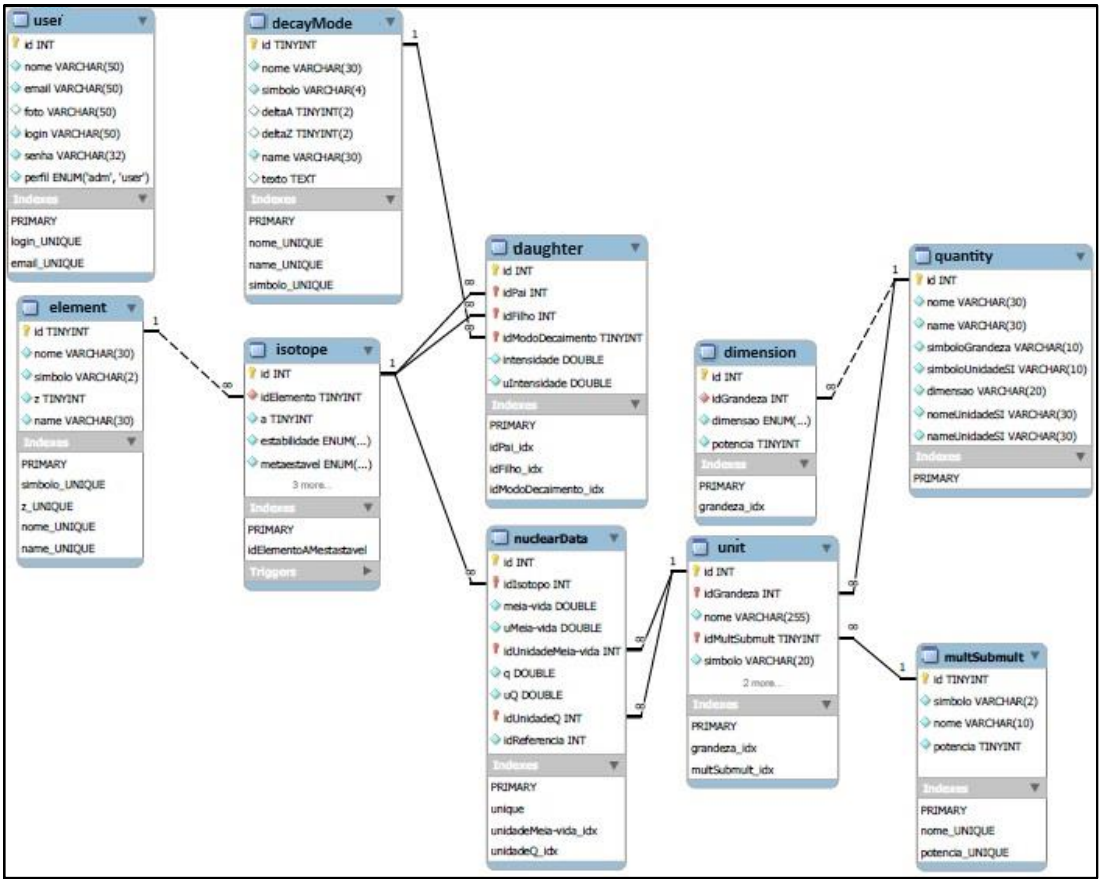

Figure 6. Nuclear parameter database modeling.

3.6. Data analysis system

The TDCR data analysis system is being developed using the C++ programming language, using the object-oriented programming technique [19].

4. Final considerations

The TDCR method was implemented in 2013 in a way that is not fully operational in the LNMRI/IRD [20]. Upon completion of this work, the TDCR method will be definitively implemented, fully operational and inserted in Quality Management, under the ISO/IEC 17.025 standard, at the Radionuclide Metrology Laboratory of the National Laboratory of Metrology of Ionizing Radiations at IRD.

In the present work, radionuclides will be standardized: ^{14}C, ^{3}H, ^{63}Ni, ^{55}Fe, ^{129}I and ^{56}Mn. The radiopharmaceuticals: ^{90}Y, ^{123}I, ^{153}Sm and ^{18}F.

The method will be validated through measurements in the primary LNMRI/IRD coincidence and anti-coincidence methods.

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