ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR – ABEN
2019 International Nuclear Atlantic Conference - INAC 2019
Santos, SP, Brazil, October 21-25, 2019

Agência Brasileira do ISBN
ISBN 978-85-99141-08-3

Preliminary measurements using a Triple to Double Coincidence Ratio (TDCR) Liquid Scintillator Counter System

Marina F. Koskinas[1], Maria Kuznetsova[1], Denise S. Moreira[1], Roberto M. Shoueri[1]
Thales S. L. Morais[1], Renato Semmler[1] and Mauro S. Dias[1]

Laboratório de Metrologia Nuclear (LMN – Nuclear Metrology Laboratory)
[1] Instituto de Pesquisas Energéticas e Nucleares (IPEN / CNEN - SP)
Av. Professor Lineu Prestes 2242
05508-000 São Paulo, SP
koskinas@ipen.br
marysmith@usp.br
denise.moreira@ipen.br
rmshoueri@yahoo.com.br
thales.morais@usp.br
renatosemmler@gmail.br
msdias@ipen.br

ABSTRACT

The preliminary measurements using a Triple to Double Coincidence Ratio (TDCR) Liquid Scintillator Counter System developed by the Nuclear Metrology Laboratory (LMN) at IPEN, is presented. The TDCR system makes use of three photomultipliers positioned at 120° relative angle, operating in coincidence. For this preliminary measurement, ^{14}C was selected to be standardized. This solution was previously calibrated by the efficiency tracing technique using a $4\pi(PC)\beta-\gamma$ coincidence system, employing ^{60}Co as a tracer. ^{14}C was chosen due to be a beta pure emitter with low end-point energy of 156 keV. The Software Coincidence System (SCS) developed by the LMN was used for both systems to register the events. MICELLE 2 code was used to calculate the theoretical TDCR efficiency. Measurements using HIDEX, a commercial liquid scintillator system, were also carried out and the results from the three methods were compared, showing a good agreement.

1. INTRODUCTION

Liquid scintillation is one of the most sensitive and versatile technique for measuring radioactivity, especially for measurement of alpha and beta emitters, presenting many advantages. Among them its high detection efficiency, since the radioactive solution is dissolved directly in the scintillator solution, simplicity in the preparation of samples and possibility to analyze, simultaneously, different radionuclides.

The preliminary measurements using a Triple to Double Coincidence Ratio (TDCR) Liquid Scintillator System developed by the Nuclear Metrology Laboratory (LMN) at IPEN, is presented. The TDCR system makes use of three photomultipliers positioned at a relative angle of 120°, operating in coincidence.

For this preliminary measurement, ^{14}C was selected to be standardized because it is a beta pure emitter with low end-point energy of 156 keV. It decays with a half-life of 5700(30) y [1].

The standardization of this radionuclide was performed by applying the tracing technique using a 4π(PC)β–γ coincidence system. It consisted in measuring the beta pure emitter combined with a beta gamma emitter (tracer), standardized previously, which provided the beta detection efficiency [2]. The tracer selected for this measurement was ^{60}Co, which decays by beta particle followed by two gamma rays of 1173 keV and 1332 keV, respectively. More details about this procedure have been presented elsewhere [3].

2. EXPERIMENTAL METHOD

2.1 Source preparation

The sources to be measured in the coincidence system were prepared by dropping known aliquots of tracer over known aliquots of pure beta solution, on a 40 μg cm^{-2} thick Collodion film. This film had been, previously, coated with a 20 μg cm^{-2} gold layer in order to turn it conductive. For the purpose of measurement in the liquid scintillator system, the commercial scintillating cocktail ULTIMA GOLD was used with a vial made of glass with low potassium content. The vial was filled with 15 mL of the scintillator cocktail and 1 mL of distilled H_2O. For the scintillator source, the same radioactive solution used for the tracer method, was poured into the vial with the cocktail. The mass determination was performed using the pycnometer technique with an XP56 Mettler balance.

2.2 TDCR method

The TDCR method used for activity determination is based on registering the double and triple coincidence events among the three photomultipliers.

The double and triple coincidence rates are given by:

$$\boldsymbol{N_D = N_0 \varepsilon_D} \qquad (1)$$

$$\boldsymbol{N_T = N_0 \varepsilon_T} \qquad (2)$$

where ε_D and ε_T are the double and triple counting efficiencies, respectively, and N_0 is the activity, considering a certain value of the ionization-quenching parameter kB [4].

The arithmetic relationships among these rates [5, 6] are given by:

$$\boldsymbol{N_D = N_{AB} + N_{BC} + N_{AC} - 2N_{ABC}} \qquad (3)$$

$$\boldsymbol{N_T = N_{ABC}} \qquad (4)$$

Where:

N_{ABC} is the triple coincidence count rates for the three photomultipliers A, B e C;

N_{AB} is the double coincidence count rates for the photomultipliers A and B;

N_{AC} is the double coincidence count rates for the photomultipliers A and C and

N_{BC} is the double coincidence count rates for the photomultipliers B and C.

The ratio of the triple counting rate and the double counting give a value R, which is called experimental TDCR efficiency.

$$N_{ABC}/N_D = \varepsilon_{TDCR} \tag{5}$$

However, to find the source activity it is necessary to calculate the true double efficiency, and the true triple coincidence for different kB (ionization-quenching parameter) values between 0.007 to 0.05 cm/MeV, using theoretical computer calculation. In this paper, it was used code MICELLE2 [7], developed by Kossert and Grau Charles, that considers the characteristics of the radionuclide disintegration scheme and the specifications of the chosen liquid scintillator.

2.3 TDCR measurement system

The Triple to Double Coincidence Ratio (TDCR) system diagram is shown in Fig. 1. This system is composed by three similar photo multipliers Hamamatsu model R329-02 coupled to three bases with preamplifiers Ortec model 270; the preamplifier outputs are connected to three spectroscopy amplifiers Ortec model 572.

The data acquisition was performed by means of a Software Coincidence System (SCS), composed by a BNC-2110 connection panel that receives the amplifiers signals. This panel is connected via 184749-01 cable to a PCI-6132 card (inserted in a Desktop Computer – PCI slot), which is capable of the sampling of up to four independent analog inputs. The signals are processed by means of LabView Version 8.5 acquisition program. This system (SCS) was developed at the LMN (Nuclear Metrology Laboratory) of the IPEN-CNEN/SP [8].

All data analysis are performed offline after the data acquisition is completed, by means of a computer code, developed at the LMN, called LSCALC01G [9], which allows the calculation of the double and triple coincidences. This code performs spectra discrimination, as well as definition of the dead time and the resolution time as input parameters.

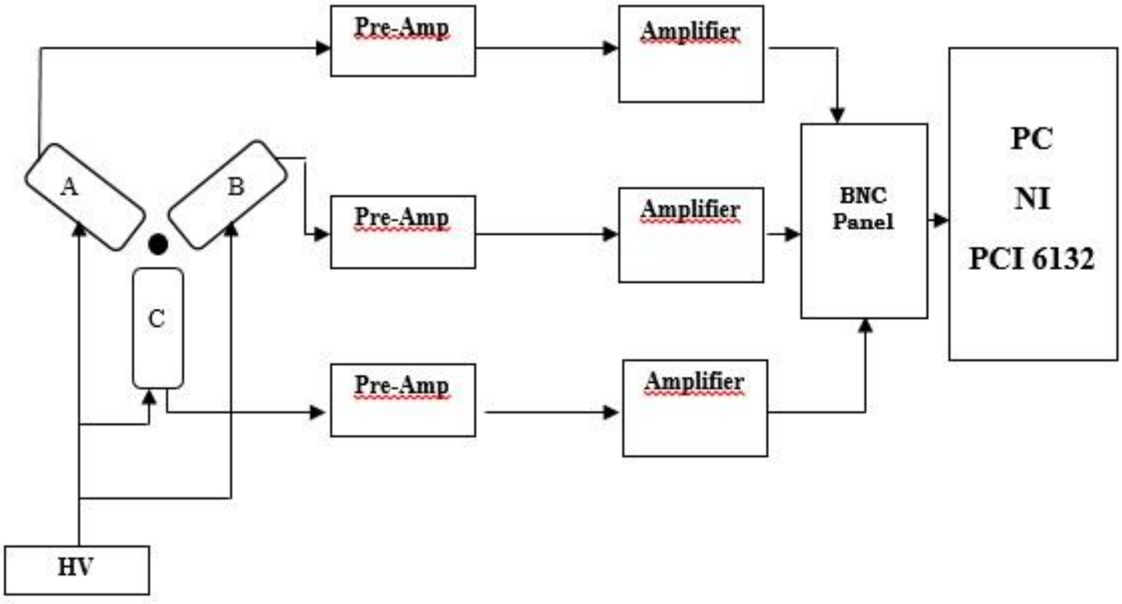

Figure 1: TDCR Electronic diagram

3. RESULTS AND DISCUSSION

Fig. 2 shows the double coincidence efficiency versus TDCR obtained with the MICELLE2 for kb 0.0075 cm/MeV and 0.012 cm/MeV ionization quenching parameter for ^{14}C. To determine the double coincidence efficiency corresponding to the experimental TDCR efficiency, the curve was fitted to 4th degree a polynomial for kB 0.0075 and to a 3th degree polynomial for kB 0.012.

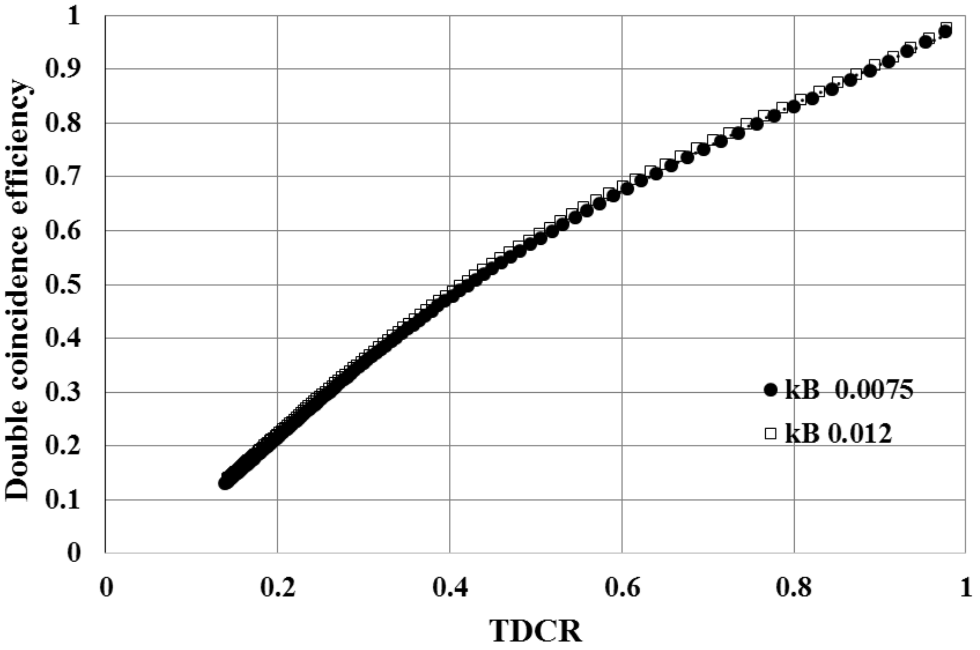

Figure 2: Double coincidence efficiency versus TDCR obtained with the MICELLE2 for kB 0.0075 cm/MeV and 0.012 cm/MeV ionization quenching parameter for ^{14}C.

Table 1 shows the activities for two kB values of the ^{14}C source measured, experimental TDCR efficiency and the corresponding double efficiency obtained by code MICELLE2, performed with four different resolution times. The ^{14}C source used was previously standardized in 4πβ(PC)–γ coincidence system by tracing method.

Table 1: ^{14}C activity obtained with TDCR system.

Ionization parameter		kB =0.0075 cm/MeV		kB =0.012 cm/MeV	
Resolution time μs	ε_{TDCR}* (%)	ε_D (%)	Activity* (Bq mg^{-1})	ε_D (%)	Activity* (Bq mg^{-1})
3.0	0.575(3)	0.660	30.38 (28)	0.650	30.83(28)
1.2	0.562(3)	0.648	30.93(29)	0.638	31.41(29)
0.6	0.542(3)	0.629	30.19(29)	0.619	30.68(29)
0.3	0.514(3)	0.603	30.14(29)	0.593	30.61(30)

*Absolute uncertainty in the last digits is shown in parentheses.

The TDCR values for the four resolution times applied varied from 0.514 for the 0.3 μs to 0.575 for 3.0 μs. This variation may indicate that when the resolution time is low, real coincidences may be lost, therefore the final activity considered it was the one obtained using the highest double efficiency.

Measurements using a HIDEX 300SL system, which is a commercial liquid scintillator counting system that uses the TDCR method, were also carried out.

In Table 2, the activity obtained by the double efficiency considering kB = 0.0075 with resolution time = 3.0 μs is compared with the results obtained with 4πβ(PC)–γ tracer method and with the HIDEX 300SL system. As can be seen, the results from the three methods are in good agreement within the uncertainties.

Table 2: Activity comparison among the three standardization methods.

System	Activity ($kBq.g^{-1}$)
HIDEX 300SL	30.30 (30)
4πβ–γ Tracer method	30.20 (20)
TDCR	30.38 (28)

4. CONCLUSION

The preliminary results of the measurement of ^{14}C presented a quite good agreement with the tracer method activity that is an absolute method. However, the TDCR efficiency was much lower than expected, indicating that improvements in the TDCR system need to be performed.

ACKNOWLEDGMENTS

The authors are indebted to the National Council for Scientific and Technological Development (CNPq), from Brazil, for partial support to the present research work.

REFERENCES

1. M.-M. Bé, V. Chisté, V. Chechev, LNHB – Laboratoire National Henri Becquerel Recommended Data – *Table de Radionucléides* – ^{14}C, Accessed at http://www.nucleide.org/DDEP_WG/DDEPdata.htm. 2019.
2. M. S. Dias, M. F. Koskinas, "Standardization of a ^{204}Tl radioactive solution," *Applied Radiation And Isotopes*, **58** pp. 235-238 (2003).
3. M. F. Koskinas, M. Kuznetsova, I. M. Yamazaki, F. Brancaccio, M. S. Dias "Standardization of C-14 by tracing method." In *International Nuclear Atlantic Conference*, São Paulo, Brazil, 4-9 October, in CDROM (2015).
4. R. A. Broda, P. Cassette, K. Kossert, Radionuclide metrology using liquid scintillation counting," *Metrologia*, **44**, S36-S52, (2007).
5. R. A. Broda, "A review of the triple-to-double coincidence ratio (TDCR) method for standardization radionuclides," *Applied Radiation and Isotopes* **58**, pp.585-594 (2003).
6. Annunziata, M. L. *Handbook of Radioactivity Analysis*. 3a Ed., Academic Press, (2012).
7. K. Kossert, A. Grau Carles, "Improved method for the calculation of the counting efficiency of electron-capture nuclides in liquid scintillation samples." *Appl.Radiat.Isot.* **68**, pp. 1482-1488 (2010).
8. F. Toledo, F. Brancaccio, A. Lapolli, M. S. Dias. "Design of Electronic System with Simultaneous Registering of Pulse and Event Time Applied to $4\pi\beta-\gamma$ Coincidence Method." *Proceedings of the IEEE 2008 Nuclear Science Symposium, Medical Imaging Conference and 16th Room Temperature Semiconductor Detector Workshop* Dresden, Germany 19 - 25 October in CDROM, (2008).
9. D. S. Moreira. LSCALC01G – User´s Manual. Internal Report, Nuclear Metrology Laboratory, IPEN-CNEN/SP, 2019.