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

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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 and ^{14}C was selected to be standardized. This solution was previously calibrated by the efficiency tracing technique using a 4π(PC)β–γ coincidence system, employing ^{60}Co as a tracer. In order to determine the final activity, a Monte Carlo simulation was used to generate the extrapolation curve. 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.

Keywords: **Carbon-14, Liquid scintillation, Tracer technique, TDCR.**

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1. INTRODUCTION

Liquid scintillation is one of the most sensitive and versatile techniques for measuring radioactivity, especially for alpha and beta emitters, with several advantages. Among them are the high detection efficiency, because the radioactive solution is dissolved directly in the scintillator solution, simplicity in the sample preparation and the possibility of analyzing different radionuclides, simultaneously.

Preliminary measurements using a Triple to Double Coincidence Ratio (TDCR) liquid scintillator system, developed by the Nuclear Metrology Laboratory (LMN) at IPEN, are 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 pure beta emitter with low end-point energy of 156 keV. It decays with a half-life of 5700(30) y [1].

In order to evaluate the results of these preliminary measurementss, the standardization of the ^{14}C solution was also performed by applying the efficiency tracing technique using a $4\pi(PC)\beta-\gamma$ coincidence system. It consists of measuring the pure beta emitter mixed with a previously standardized beta gamma emitter (tracer), which provides 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. The extrapolation technique [3] was applied, changing the beta efficiency by pulse height discrimination.

The data acquisition for both systems was performed by means of a Software Coincidence System (SCS) developed at the LMN (Nuclear Metrology Laboratory) of the IPEN-CNEN/SP [4].

2. MATERIALS AND METHODS

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.

The measurement in the liquid scintillator system was performed by using the commercial scintillating cocktail ULTIMA GOLD, with a vial made of glass with low potassium content. The vial was filled with 15 mL of 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. 4πβ–γ Coincidence Measurement

The 4πβ–γ coincidence system used for measuring the tracer solution and the pure beta mixed with the tracer solution, consisted of a proportional counter with 4π geometry filled with 0.1 MPa P10 gas mixture, coupled to a pair of 76 mm x 76 mm NaI(Tl) crystals. The calibrations of tracer and tracer plus pure beta were performed by selecting a γ–ray discrimination window including both (1173 + 1332) keV total absorption peaks.

The coincidence equations applied to ^{14}C +^{60}Co are given [5] by:

$$\boldsymbol{N_{\beta} = \left\{ N_{0tr}\left[\varepsilon_{\beta tr} + \left(1-\varepsilon_{\beta_{tr}}\right)\frac{\alpha_{tr}\varepsilon_{ce_{tr}} + \varepsilon_{\beta\gamma_{tr}}}{1+\alpha_{tr}} \right] \right\} + N_{0p}\varepsilon_{\beta p}} \quad \textbf{(1)}$$

$$\boldsymbol{N_{\gamma} = N_{0tr}\varepsilon_{\gamma_{tr}}\frac{1}{1+\alpha_{tr}}} \quad \textbf{(2)}$$

$$\boldsymbol{N_{c} = N_{0tr}\varepsilon_{\beta tr}\varepsilon_{\gamma_{tr}}\frac{1}{1+\alpha_{tr}}} \quad \textbf{(3)}$$

Equations (1), (2) and (3) lead to:

$$\frac{N_{\beta}N_{\gamma}}{N_c} = N_{0tr}\left\{1+\frac{\left(1-\varepsilon_{\beta tr}\right)}{\varepsilon_{\beta tr}}\frac{(\alpha_{tr}\,\varepsilon_{ce_{tr}}+\varepsilon_{\beta\gamma_{tr}})}{1+\alpha_{tr}}\right\}+N_{0p}\frac{\varepsilon_{\beta p}}{\varepsilon_{\beta tr}} \qquad \textbf{(4)}$$

Where:

N_{β}, N_{γ} and N_c are the beta, gamma and coincidence counting rates, respectively;

N_{0tr} is the tracer disintegration rate;

N_{0p} is the pure beta disintegration rate;

$\varepsilon_{\beta tr}$ is the tracer beta detection efficiency;

$\varepsilon_{\beta p}$ is the pure beta detection efficiency;

$\varepsilon_{\gamma tr}$ is the gamma detection efficiency;

$\varepsilon_{\beta\gamma tr}$ is the tracer gamma detection efficiency for the beta detector;

ε_{cetr} is the tracer conversion electron detection efficiency;

α_{tr} is the tracer total internal conversion coefficient;

Since the tracer disintegration rate is previously known, equation (4) becomes

$$\frac{N_{\beta}N_{\gamma}}{N_c}-N_{0tr} = N_{0P}\frac{\varepsilon_{\beta P}}{\varepsilon_{\beta tr}} \qquad \textbf{(5)}$$

When the pure beta and the tracer are combined in a single source, a relationship between the detection efficiencies can be represented by a function ***F*** of the tracer efficiency [6].This relation can be defined by

$$\varepsilon_{\beta p} = F\left(1-\varepsilon_{\beta tr}\right) \qquad \textbf{(6)}$$

Or by a function G given by

$$\varepsilon_{\beta p} = G\left(\frac{1-\varepsilon_{\beta tr}}{\varepsilon_{\beta tr}}\right) \qquad \textbf{(7)}$$

where$\varepsilon_{\beta tr}$ is approximately equal to the efficiency parameter N_c/N_{γ}.

Expression (5) can be rewritten as equation (8):

$$\frac{N_{\beta}N_{\gamma}}{N_c}-N_{0tr} = N_{0p}\left(1+G'\left(\frac{1-N_c/N_{\gamma}}{N_c/N_{\gamma}}\right)\right) \qquad \textbf{(8)}$$

The function 1+ $\boldsymbol{G'}$ is usually represented by a polynomial, and goes to unity when $(\boldsymbol{1- N_c/N_\gamma})/ \boldsymbol{N_c/N_\gamma}$ goes to zero.

In the present paper, this polynomial fitting has been replaced by a Monte Carlo calculation using ESQUEMA code [7] for unitary activity and fitted to the experimental data by least squares methodology applying covariance calculation. This code uses the decay scheme parameters, the system geometry and the source characteristics, in order to simulate all detection processes in the coincidence system. The response functions of all detectors were previously calculated by the Monte Carlo MCNPX code [8], for electrons and photons in a wide range of energies. In the case of NaI(Tl) photons, the energy range was from 10 keV to 4 MeV; for electrons in the 4π proportional counter, the energy range was from 100 eV to 3 MeV.

The final activity was calculated by Least Square fitting combining experimental and simulated values of $\left(\frac{N_\beta N_\gamma}{N_c} - N_{0tr}\right)$ for the selected gamma-ray window. The result was obtained by minimizing the following Chi-Squared value

$$\chi^2 = (\vec{y}_{exp} - N_0\,\vec{y}_{MC})^T V^{-1} (\vec{y}_{exp} - N_0\,\vec{y}_{MC}) \qquad \textbf{(9)}$$

where:

$\vec{y}_{exp}$ is the experimental vector of $\left(\frac{N_\beta N_\gamma}{N_c} - N_{0tr}\right)_{exp}$,

$\vec{y}_{MC}$ is the $\left(\frac{N_\beta N_\gamma}{N_c} - N_{0tr}\right)_{MC}$ vector calculated from the Monte Carlo for unitary activity;

$\boldsymbol{N_0}$ is the activity of the radioactive source;

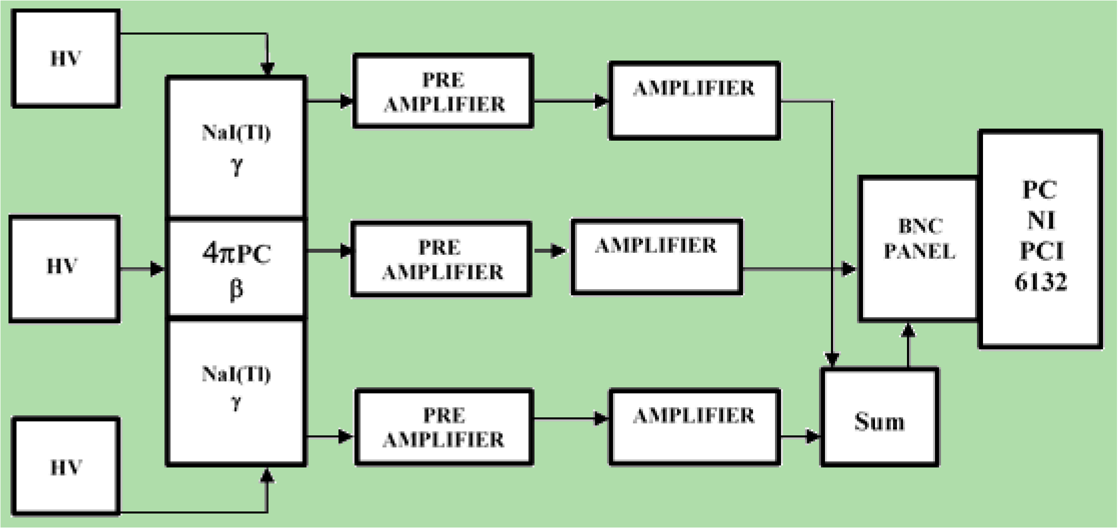
$\boldsymbol{V}$ is the total covariance matrix, including both experimental and calculated uncertainties; and $\boldsymbol{T}$ stands formatrix transposition.

2.2.1 4πβ–γ coincidence measurement system

The 4πβ–γ Software Coincidence System (SCS) diagram is presented in Figure 1. In this system, the signals from the 4πβ amplifier (ORTEC Model 572) are sent to the BCN panel. The

NaI(Tl) amplifiers (ORTEC Model 572) signals are added in the SUM module (ORTEC Model 533) and sent to the BCN panel, which is connected to the NI PCI6132.

Figure 1: *Diagram of 4πβ–γ SCS.*



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2.3. 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 the equations (10) and (11) respectively:

$$N_D = N_0 \varepsilon_D \tag{10}$$

$$N_T = N_0 \varepsilon_T \tag{11}$$

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 [9].

The arithmetic relationships among these rates [10,11] are given by equations (12) and (13):

$$N_D = N_{AB} + N_{BC} + N_{AC} - 2N_{ABC} \tag{12}$$

$$N_T = N_{ABC} \tag{13}$$

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 to the double coincidence counting gives a value ***R***, which is called: experimental TDCR efficiency.

$$\boldsymbol{N_{ABC}/N_D = \varepsilon_{TDCR}} \quad \textbf{(14)}$$

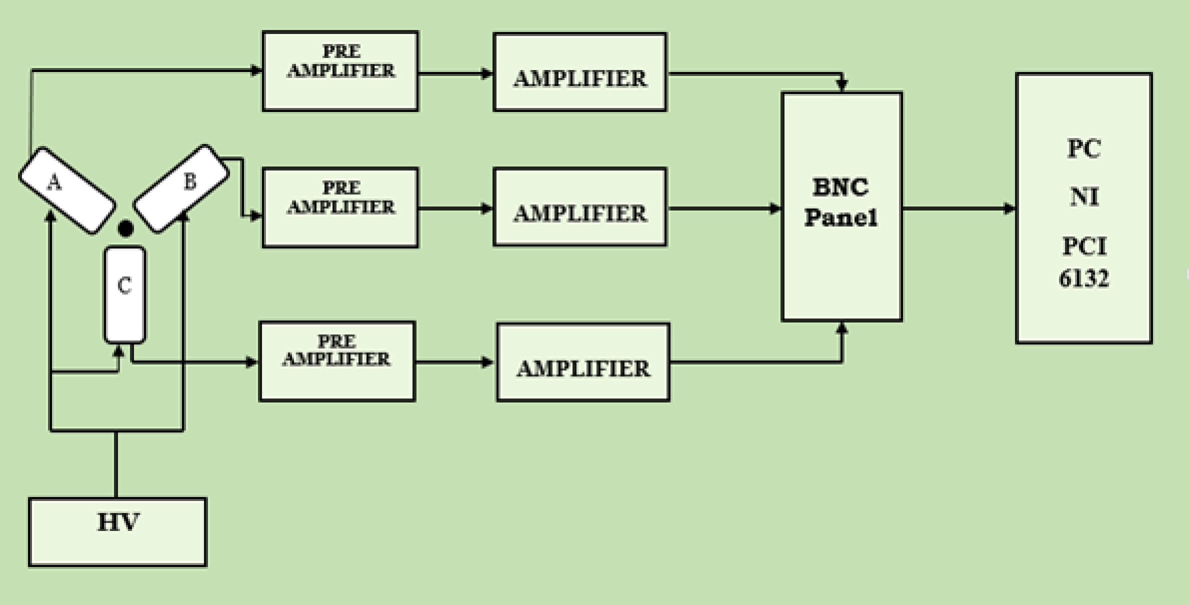
$$\boldsymbol{N_T = N_{ABC}} \quad \textbf{(15)}$$

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 MICELLE2 code [12], developed by Kossert and Grau Charles, that considers the characteristics of the radionuclide disintegration scheme and the specifications of the chosen liquid scintillator.

2.3.1. TDCR measurement system

The Triple to Double Coincidence Ratio (TDCR) system diagram is shown in Figure 2. This system is composed of three similar photomultipliers Hamamatsu model R329-02, identified as A, B and C in the diagram, coupled to three bases with preamplifiers Ortec model 270; the preamplifier outputs are connected to three spectroscopy amplifiers Ortec model 572.

Figure 2: *TDCR Electronic diagram.*



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2.4. Software Coincidence System (SCS)

The data acquisition for both methods was performed by means of a Software Coincidence System (SCS), composed of a National Instruments (NI) BNC-2110 connection panel that receives the amplifiers signals. This panel is connected via NI-184749-01 cable to a NI-PCI-6132 card (inserted in a Desktop Computer – PCI slot), which is capable of the sampling of up to four independent analog inputs [13]. 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.

The SCS system, when connected in the $4\pi\beta-\gamma$ system, records the amplitude of the pulses from each detector amplifier of the coincidence system (proportional counter and scintillator crystals) and the corresponding occurrence time of these pulses.

For the 4πβ–γ system, the data analysis was performed by a code developed at the LMN, called SCTAC version 6.0 [14], which allows the calculation of beta and gamma spectra and pulse rates. This code also performs beta discrimination, selection of gamma window as well as definition of dead time and the resolution time by software. The final output is the source activity.

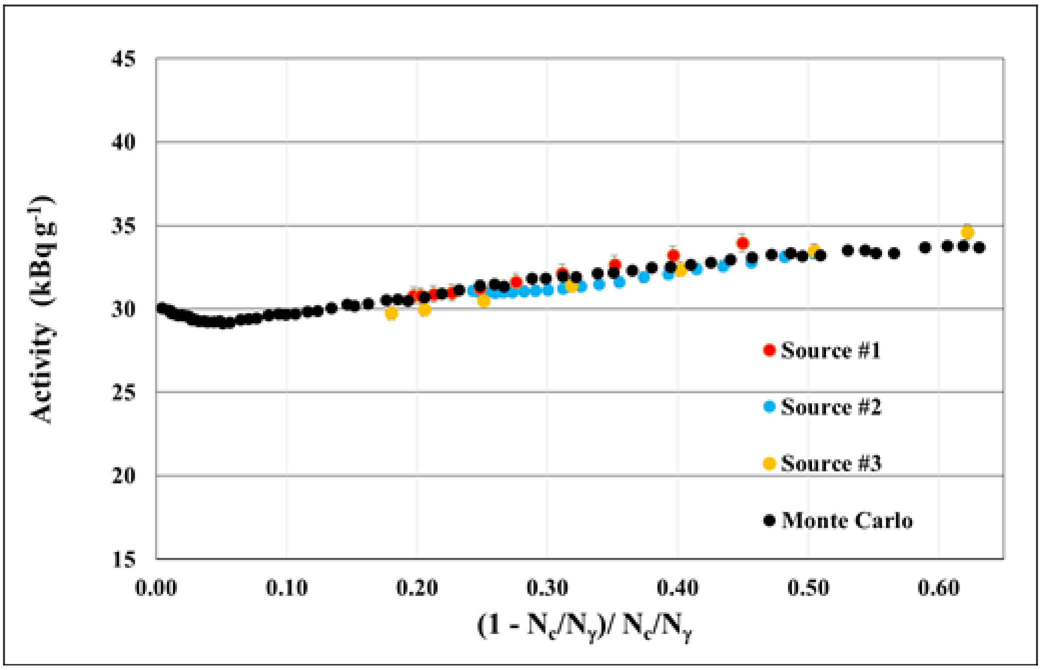
For the TDCR system, the data analysis was also performed offline, after the data acquisition was completed, by means of another computer code, developed at the LMN, called LSCALC01G [15], which allows the calculation of the double and triple coincidences. This code performs spectra discrimination, as well as definition of dead time and resolution time as input parameters.

3. RESULTS AND DISCUSSION

3.1. 4πβ–γ Measurements

Figure 3 shows the Monte Carlo simulation of the predicted $(N_{\beta}N_{\gamma}/N_c – No_{tr})$ curve determined by ESQUEMA code, as a function of inefficiency parameter $(1-N_c/N_{\gamma})/N_c/N_{\gamma}$ for ^{14}C, shown as black dots. The experimental values for different radioactive sources are represented as red, blue, and yellow dots, respectively. The curve was normalized to the final average activity.

Figure 3: *Normalized Monte Carlo simulation of predicted* $\left(\frac{N_{\beta}N_{\gamma}}{N_c} - N_{0tr}\right)$ *as a function of efficiency parameter* $(1 - N_c/N_{\gamma})/(N_c/N_{\gamma})$ *for* ^{14}C *(black dots). Red, blue, and yellow dots correspond to experimental data.*



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The source activities were determined by equation (9) for each radioactive source. In Table 1 the results are presented.

Table 1: Activity values determined by equation (9), for each radioactive source. The uncertainties correspond to one standard deviation (k=1).

Source	**Activity** **kBq g^{-1}**
1	30.40 (40)
2	30.40 (50)
3	29.90 (30)
Average	30.20 (20)

3.2. TDCR results

Figure 3 shows the double coincidence efficiency versus TDCR obtained with the MICELLE2 code 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 3rd degree polynomial for kB 0.012.

Figure 3: *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.*

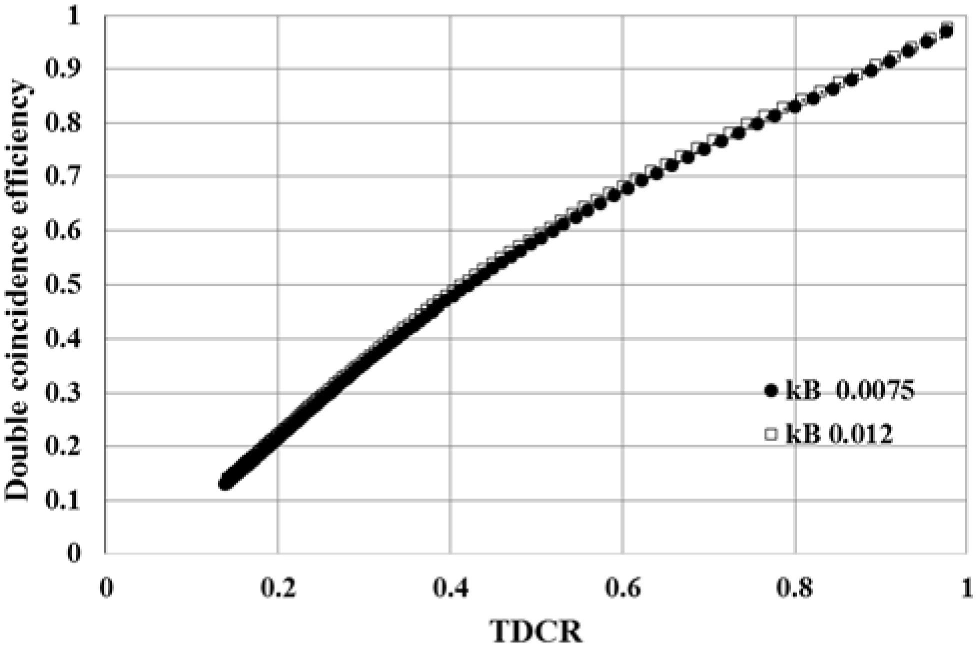

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Table 2 shows the activities for two kB values of the ^{14}C source measured, experimental TDCR efficiency and the corresponding double efficiency obtained by code MICELLE2, performed with four different resolution times.

Table 2: ^{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* (kBq g^{-1})**	**ε_D**	**Activity* (kBq g^{-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 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 3, 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 3: 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 primary standardization of ^{14}C by means of the tracer method in a 4πβ–γ coincidence counting system, associated with a Software Coincidence System (SCS) was performed. The Monte Carlo simulation ESQUEMA code was successful applied combined with experimental measurements demonstrating its feasibility for the standardization of ^{14}C. The preliminary results of the measurement of ^{14}C using the TDCR system developed by the LMN 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 implemented.

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