2015 International Nuclear Atlantic Conference - INAC 2015
São Paulo, SP, Brazil, October 4-9, 2015
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
ISBN: 978-85-99141-06-9

STANDARDIZATION OF C-14 BY TRACING METHOD

Marina F. Koskinas, Maria Kuznetsova, Ione Yamazaki,
Franco Brancaccio, Mauro S. Dias

Laboratório de Metrologia Nuclear (LMN – Nuclear Metrology Laboratory)
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
yamazaki@ipen.br
fbrancac@ipen.br
msdias@ipen.br

ABSTRACT

The standardization of a ^{14}C radioactive solution by means of the efficiency tracing technique is described. The ^{14}C is a beta pure emitter with endpoint energy of 156 keV decaying to the ground state of ^{14}N. The activity measurement was performed in a $4\pi\beta-\gamma$ coincidence system, measuring the pure beta emitter mixed with a beta-gamma emitter, which provides the beta detection efficiency. The radionuclide ^{60}Co, which decays by beta particle followed by two gamma rays, was used as tracer and the efficiency was obtained by selecting the 1173 keV plus 1332 keV total energy absorption peak at the gamma channel. Known aliquots of the tracer, previously standardized by $4\pi\beta$ (PC)$-\gamma$ coincidence, were mixed with known aliquots of ^{14}C. The sources of ^{14}C + ^{60}Co were prepared by dropping known aliquots from each radioactive solution. The events were registered by a Software Coincidence System (SCS). The activity of the solution was determined by using the extrapolation technique, changing the beta efficiency by pulse height discrimination. In order to determine the final activity, a Monte Carlo simulation was used to calculate the extrapolation curve. All the uncertainties involved were treated rigorously, by means of the covariance analysis methodology. Measurements using a HIDEX, a commercial liquid scintillator system, were carried out and the results were compared with the tracing technique, showing a good agreement.

1. INTRODUCTION

C-14 decays with a half-life of 5700(30) y by pure beta emission, with end-point energy of 156.476 keV [1] The standardization of this radionuclide was performed in a $4\pi\beta-\gamma$ coincidence system, applying the tracing technique. It consists in measuring the beta pure emitter combined with a beta gamma emitter, called tracer, which provides the beta detection efficiency and was standardized previously [2]. The beta gamma emitter selected for this measurement was ^{60}Co, which decays by beta particle followed by two gamma rays of 1173 keV and 1332 keV respectively. Fig. 1 and 2 show the decay schemes of these two radionuclides.

The beta particles from both radionuclides were measured in a proportional counter with 4π geometry, and the gamma-rays from the tracer were measured in a NaI(Tl) crystal, selecting the two total energy absorption peaks of ^{60}Co.

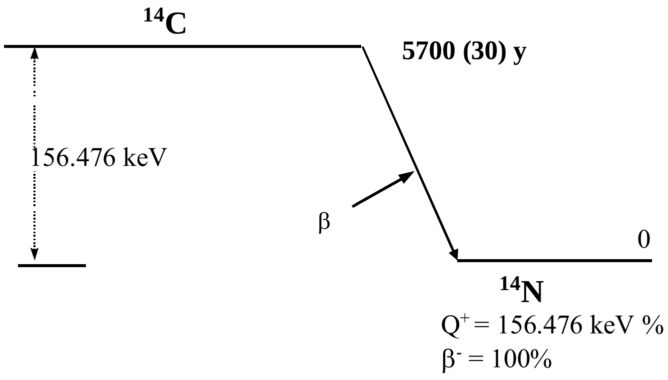

Figure 1: Decay scheme of ^{14}C [1].

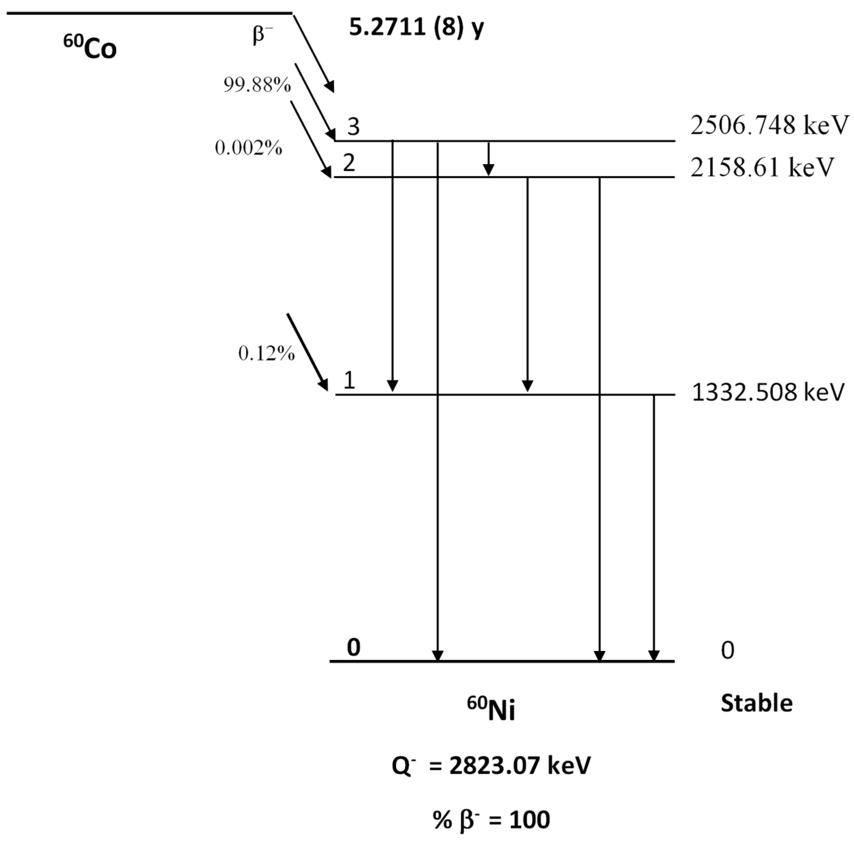

Figure 2: Decay scheme of ^{60} Co [3].

For ^{14}C activity measuring, the beta efficiency was changed by pulse height discrimination, by means of a Software Coincidence System (SCS) developed at the LMN (Nuclear Metrology Laboratory) of the IPEN-CNEN/SP [4]. The extrapolation curve was calculated by the Monte Carlo code ESQUEMA [5] and fitted to the experimental data, as described in sections 2.2 and 2.4.

2. EXPERIMENTAL METHOD

2.1. Source Preparation

The sources to be measured in the coincidence system were prepared by mixing known aliquots of tracer to known aliquots of the pure beta solution, which were then dropped 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 the film conductive. The sources were dried in desiccator and the mass determination has been performed using the pycnometer technique with a XP56 Mettler balance [6].

2.2. 4πβ–γ Coincidence Measurements

The 4πβ–γ coincidence system used for measuring the tracer solution and the beta pure plus 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 comprising both (1173 + 1332) keV total energy absorption peaks.

The coincidence equations applied to $^{14}C + ^{60}Co$ can be given by:

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

$$N_{\gamma} = N_{0tr}\varepsilon_{\gamma_{tr}} \frac{1}{1 + \alpha_{tr}} \tag{2}$$

$$N_{c} = N_{0tr}\varepsilon_{\beta tr}\varepsilon_{\gamma_{tr}} \frac{1}{1 + \alpha_{tr}} \tag{3}$$

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

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

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:

$$\frac{N_\beta N_\gamma}{N_c} - N_{0tr} = N_{0P}\frac{\varepsilon_{\beta P}}{\varepsilon_{\beta tr}} \tag{5}$$

When the beta pure and the tracer sources are combined into a single source, a relationship between the detection efficiencies can be represented by a function *F* of the tracer efficiency [7]. This relation can be defined by:

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

Or by a function G given by:

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

where $\varepsilon_{\beta tr}$ is given by the efficiency parameter N_c/N_γ.

Therefore, expression (5) can be rewritten as:

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

The function G' is usually represented by a polynomial, and goes to unity when $(1- N_c/N_\gamma)/\ N_c/N_\gamma$ goes to zero. In the present paper, this polynomial fitting has been replaced by a Monte Carlo calculation using code ESQUEMA [5].
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.

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

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 by the Monte Carlo code ESQUEMA, for unitary activity;

N_0 is the activity of the pure beta radioactive source;

V is the total covariance matrix, including both experimental and calculated uncertainties;

and T stands for matrix transposition.

2.3. Software Coincidence System (SCS)

The Software Coincidence System (SCS) used in the present measurements is based on the National Instruments PCI-6132 card, capable of the sampling of up to four independent analog inputs and the signals are processed by means of LabView Version 8.5 acquisition program. This system records the pulses from the amplifiers connected to each detector in the coincidence system (proportional counter and scintillator crystals) corresponding to the time of occurrence and pulse height.

The sources activities were calculated by means of the code SCTAC version 6 [8], which is a software developed at the LMN to calculate the activity based on the analysis of the files recorded on disk. These files store the nuclear pulses digitized by the SCS, corresponding to the beta, gamma and coincidence countings.

From this information, it is possible to calculate the beta and gamma spectra, and all other necessary data to calculate the extrapolation curve. This software allows selection of dead time, resolution time as well as beta and gamma windows. In this way, a complete extrapolation curve can be achieved from a single measurement.

2.4. Monte Carlo Simulation

Monte Carlo code ESQUEMA [5] is a code developed at LMN to calculate the extrapolation curve in the $4\pi\beta-\gamma$ coincidence experiment. This code makes use of decay scheme parameters, system geometry and radioactive source characteristics. In this way, all detection processes in the coincidence system are simulated, predicting the behavior of the extrapolation curve by the Monte Carlo technique. The detector response curves are obtained by means of the radiation transport code MCNPX [9].

In the case of the tracing technique, the decay scheme of the tracer (^{60}Co) used in the simulation, is modified, including the decay scheme of ^{14}C. As a result, the whole coincidence experiment can be simulated. In this way, equation (8) could be reproduced by calculation as a function of the beta efficiency, for the selected gamma-ray windows yielding the extrapolation curve for each experimental condition. In the present experiment, the beta efficiency was varied by pulse height discrimination.

2.5. Liquid Scintillator Counting System

Measurements of the ^{14}C solution were carried out using a commercial liquid scintillator system, called HIDEX 300 SL, which consist of three photomultipliers operated in coincidence.

This system is based on the Triple to Double Coincidence Ratio (TDCR) method [10], giving the TDCR efficiency, which is the ratio of the efficiency from the measured ratio of double, and triple coincidence counting rates. The activity is given directly by the system and is the ratio of counting to TDCR efficiency.

For this measurement, three distinct liquid scintillators were used: Ultima Gold, Insta-Gel Plus and OptiPhase Hisafe 3. The samples for liquid scintillation counting were prepared in 20 mL borosilicate glass vials with low potassium concentration. The volume of each liquid scintillator was 15 mL, determined by mean of a calibrated pipette. Known aliquots of the radioactive material were determined by the pycnometer technique [6].

For the present measurements, six vials of ^{14}C with different quenching amounts of nitromethane were prepared for each liquid scintillator in order to compare their performance.

3. RESULTS

3.1. $4\pi\beta-\gamma$ Measurements

Fig. 3 shows the Monte Carlo simulation of $(N_\beta N_\gamma/N_c - No_{tr})$ as a function of the parameter $(1-N_c/N_\gamma)/N_c/N_\gamma$ for ^{14}C, is shown as black dots. The experimental values for different radioactive sources are represented as red, blue and yellow dots. The curve was normalized to the ^{14}C activity: N_0.

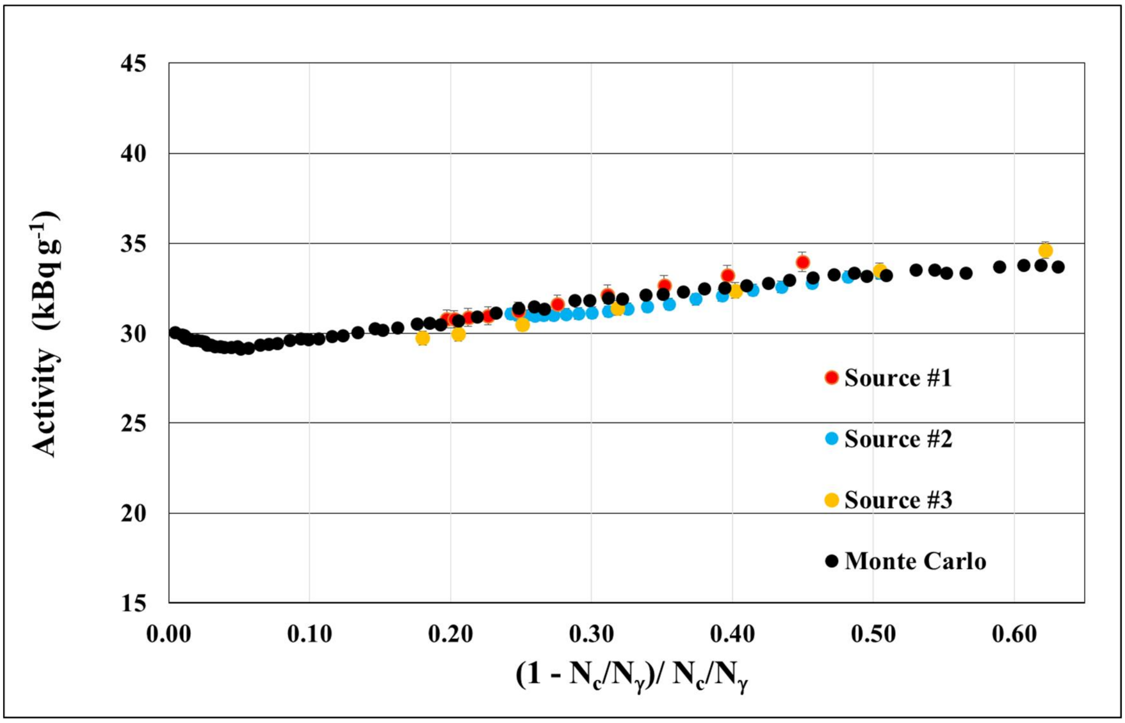

Figure 3: Normalized Monte Carlo simulation of predicted $\left(\frac{N_\beta N_\gamma}{N_c} - N_{0tr}\right)$ as a function of the parameter $(1 - N_c/N_\gamma)/(N_c/N_\gamma)$ for ^{14}C (black dots). Red, blue and yellow dots correspond to experimental data.

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

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

Source	Activity kBq g^{-1}
1	30.4 (4)
2	30.4 (5)
3	29.9 (3)
Average	30.2 (2)

3.2. Liquid Scintillator Measurements

The average activities obtained from the three liquid scintillators for the six vail are presented in table 2, the uncertainty presented is the statistical counting; as can be seen the activities are in good agreement, showing that the liquid scintillators used are equivalents for the measurement of ^{14}C; the final activity obtained was 30.4 (3) kBq g^{-1} in good agreement with the final value obtained by means of the tracer technique method presented in Table 1.

Table 2: Average activities from the three liquid scintillators. The uncertainties correspond to one standard deviation (u=1).

Liquid scintillator	Activity kBq g^{-1}
Ultima Gold	**30.3 (3)**
Insta-Gel Plus	**30.3 (3)**
OptiPhase Hisafe 3	**30.5 (5)**
Average	**30.4 (3)**

4. CONCLUSIONS

The Monte Carlo simulation code ESQUEMA was developed by the LMN for the efficiency extrapolation curve simulation, taking disintegration scheme, detection geometry and material details into account. Code ESQUEMA was successful applied combined with experimental measurements using the 4π(PC) $\beta-\gamma$ software coincidence system; demonstrating its feasibility for the standardization of ^{14}C. The results are in good agreement with those obtained using the commercial liquid scintillator system (HIDEX), showing that such system presents good results and it is suitable for routine calibrations.

ACKNOWLEDGMENTS

The authors are grateful to the National Council for Scientific and Technological Development (CNPq) for partial support to the present research project.

REFERENCES

1. M.-M. Bé, et al., C-14 Accessed at http://www.nucleide.org/DDEP_WG/DDEPdata.htm. (2015).
2. M.F. Koskinas, C. A. Pires, I. M. Yamazaki, E.A. Silva and M.S. Dias, "Standardization of ^{55}Fe by tracing method," *Appl. Radiat. Isot,* **66,** pp.733-736 (2008).
3. M.-M. Bé, et al., Co-60 Accessed at http://www.nucleide.org/DDEP_WG/DDEPdata.htm. (2015).
4. F. Toledo, ***ñ****Development of an electronic system with simultaneous recording of amplitude and time of occurrence of the pulses applied to the method of coincidence 4πβ–γ***ò**. Master Thesis, IPEN-CNEN/SP (2008).
5. M.S. Dias, M. N. Takeda, M.F. Koskinas, "Application of Monte Carlo simulation to the prediction of extrapolation curves in the coincidence technique," *Appl. Radiat. Isot,* **64,** pp.1186-1192 (2006).
6. P. J. Campion, *Procedures for accurately diluting and dispensing radioactive solutions,* Bureau International des Poids et Mesures, Monographie BIPM – 1 (1975).

7. M. S. Dias, M. F. Koskinas, “Standardization of a Tl-204 radioactive solution,” *Appl. Radiat. and Isot.*, **58**, pp. 235-238 (2003).
8. M. S. Dias, SCTAC 6 – User´s Manual. Internal Report, Nuclear Metrology Laboratory, IPEN-CNEN/SP (2014).
9. ORNL, *Monte Carlo N-Particle Transport Code System, MCNP5*, RSICC Computer Code Collection, Oak Ridge National Laboratory (2006).
10. R. Broda, P. Cassette, K. Kossert, “Radionuclide metrology using liquid scintillation counting” *Metrologia,* **44**, pp S36–S52 (2007).