Application of Monte Carlo Simulation to ^{134}Cs Standardization by Means of $4\pi\beta-\gamma$ Coincidence System

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***Abstract*—The methodology recently developed by the Nuclear Metrology Laboratory (LMN) from IPEN, in São Paulo, Brazil, for simulating all detection processes in a $4\pi(\beta,\mathrm{X})-\gamma$ coincidence system by means of the Monte Carlo technique is described. This procedure makes possible to predict the behavior of the observed activity as a function of the $4\pi\beta$ detector efficiency. The present paper describes its application to the standardization of a typical beta-gamma radionuclide, namely ^{134}Cs. In this approach, information contained in the decay scheme is used for determining the contribution of all radiations emitted by the selected radionuclide to the measured spectra of each detector. This simulation yields the shape of the coincidence spectrum, allowing the choice of suitable gamma-ray energies for which the activity can be obtained with maximum accuracy. The theoretical work applies the MCNP Monte Carlo code to a gas-flow proportional counter with 4π detection geometry, coupled to a pair of NaI(Tl) crystals. The calculated extrapolation curve showed good agreement when compared to experimental values obtained at the LMN of IPEN.**

***Index Terms*—Coincidence, Cs-134, Monte Carlo, simulation, standardization.**

I. Introduction

IN nuclear metrology, $4\pi\beta-\gamma$ coincidence has been considered for many years a primary technique for radionuclide standardization, due to high accuracy and because it depends only on observed quantities [1]–[10]. Usually, this kind of system makes use of a gas-flow or pressurized 4π proportional counter for charged particle detection, coupled to NaI(Tl) or HPGe gamma-ray spectrometers for gamma-ray detection. Alternatively, liquid scintillators are used instead of proportional counters.

The Nuclear Metrology Laboratory (LMN) of IPEN-CNEN/SP has developed several radionuclide standardization systems [10]–[14]. Presently, the LMN has two $4\pi\beta-\gamma$ coincidence systems composed of gas-flow or pressurized proportional counters [10] coupled to NaI(Tl) or HPGe detectors.

One difficulty encountered in $4\pi\beta-\gamma$ standardization is the necessary planning of experimental conditions in order to optimize measurements and therefore minimize uncertainty in the resulting activity. Usually, this value is obtained by the linear extrapolation method [3], changing the $4\pi\beta$ detector efficiency and extrapolating to 100% efficiency.

For simple decay schemes this procedure is straightforward and the results can be obtained in a fast and reliable way. However, for complex decay schemes this task becomes more difficult because several effects can occur, for instance: charged particle energy degradation; detection of different types of radiation by the same detector and overlapping of events produced by different detection mechanisms.

In literature, usually an analytical estimate is performed, taking into account the decay scheme information but without considering detailed detection characteristics for all radiations. The present approach is based on Monte Carlo simulation of all detection processes in a $4\pi\beta-\gamma$ coincidence system to allow predicting the extrapolation curve. The simulation is able to predict a detailed description of the functional relationship between the observed activity and the efficiency parameter, mainly in the region where the $4\pi(\beta,\mathrm{X})$ efficiency approaches 100%. This value is experimentally unreachable due to self absorption of low energy electrons in the radioactive source substrate. For simple decay schemes this relationship is approximately linear, but for complex decay schemes the behavior of this curve is difficult to predict.

The present paper describes the application of this methodology to the standardization of a typical beta-gamma radionuclide with complex decay, namely ^{134}Cs. In this approach, information contained in the decay scheme is used for determining the contribution of all radiations emitted by the selected radionuclide to the measured spectra of each detector. This simulation yields the shape of the coincidence spectrum, allowing the choice of suitable gamma-ray energy intervals for which the activity can be obtained with maximum accuracy. Detailed characteristics of the LMN coincidence system are taken into account in order to perform calculation as realistic as possible.

II. Methodology

A. Coincidence Equations

Fig. 1 shows ^{134}Cs decay scheme [15]. Several beta-ray branches populate the excited states of ^{134}Ba giving rise to a multitude of transitions associated to gamma-rays or internally converted electrons. An electron capture branch of very low intensity (0.0003%) populates an excited state of ^{134}Xe.

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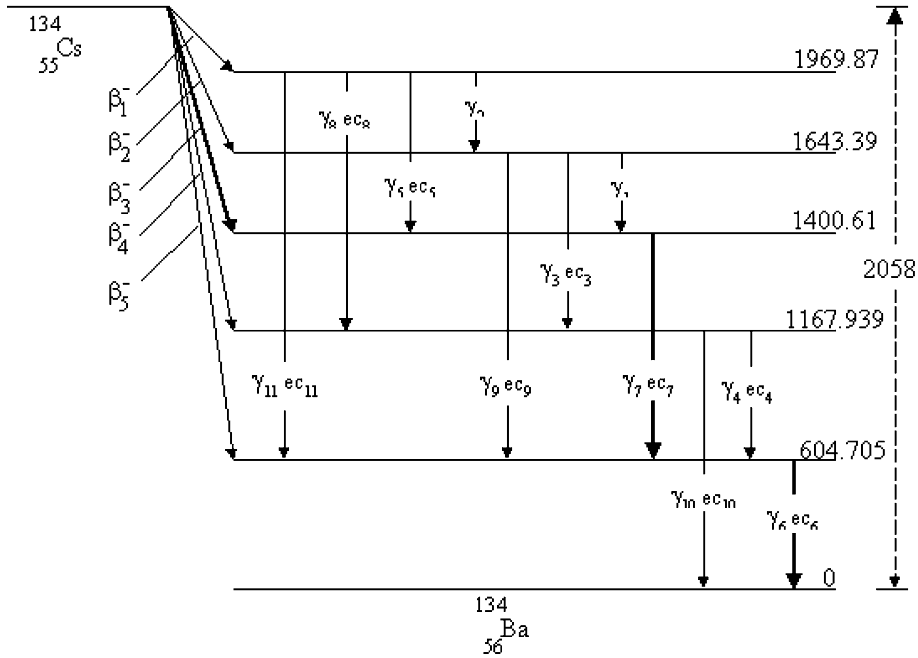

Fig. 1. Decay scheme of ^{134}Cs (electron capture branch omitted).

Neglecting this electron capture transition, the general coincidence equations applied to this radionuclide are

$$N_\beta = N_0 \sum_{i=1}^{m} a_i \cdot \left\{ \varepsilon_{\beta_i} + (1-\varepsilon_{\beta_i}) \sum_{j=1}^{n} b_{ij} \times \frac{\alpha_{ij}\left[\varepsilon_{CE_{ij}} + \left(1-\varepsilon_{CE_{ij}}\right)\varepsilon_{(X,A)_{ij}}\right] + \varepsilon_{\beta\gamma_{ij}}}{1+\alpha_{ij}} \right\} \quad (1)$$

$$N_\gamma = N_0 \sum_{i=1}^{m} a_i \sum_{j=1}^{n} b_{ij}\varepsilon_{\gamma_{ij}} \frac{1}{1+\alpha_{ij}} \quad (2)$$

$$N_c = N_0 \sum_{i}^{m} a_i \left[\varepsilon_{\beta_i} \sum_{j=1}^{n} b_{ij}\varepsilon_{\gamma_{ij}} \frac{1}{1+\alpha_{ij}} + (1-\varepsilon_{\beta_i}) \sum_{j=1}^{n} b_{ij}\varepsilon_{C_{ij}} \frac{1}{1+\alpha_{ij}} \right] \quad (3)$$

Therefore, (4), shown at the bottom of the page, where

N_β, N_γ, N_c beta, gamma, and coincidence counting rates, respectively;
N_0 the radioactive source disintegration rate;
a_i, b_{ij} the intensity per decay of the ith beta transition and relative intensity of the jth transition with respect to the ith transition;
n number of daughter transitions following the ith beta transition;
m number of beta transitions;
ε_{β_i} beta efficiency associated to ith beta transition;
$\varepsilon_{\gamma_{ij}}$, $\varepsilon_{\beta\gamma_{ij}}$ gamma detection efficiency and gamma efficiency of beta detector, respectively, associated to ijth transition;
$\varepsilon_{CE_{ij}}$, $\varepsilon_{(X,A)_{ij}}$ conversion electron detection efficiency and electron Auger or X-ray detection efficiency, respectively, associated to ijth transition, and
$\varepsilon_{C_{ij}}$, α_{ij} gamma–gamma coincidence detection efficiency and total internal conversion coefficient of the ijth transition.

A measure of the beta efficiency can be given by

$$\frac{N_c}{N_\gamma} = \frac{\sum_{i}^{m} a_i \left[\varepsilon_{\beta_i} \sum_{j=1}^{n} b_{ij}\varepsilon_{\gamma_{ij}} \frac{1}{1+\alpha_{ij}} + (1-\varepsilon_{\beta_i}) \sum_{j=1}^{n} b_{ij}\varepsilon_{C_{ij}} \frac{1}{1+\alpha_{ij}} \right]}{\sum_{i=1}^{m} a_i \sum_{j=1}^{n} b_{ij}\varepsilon_{\gamma_{ij}} \frac{1}{1+\alpha_{ij}}}. \quad (5)$$

In the special cases where the total energy absorption peak in the gamma channel can be selected as window for a single

$$\frac{N_\beta N_\gamma}{N_c} = \frac{N_0 \left\{ \sum_{i=1}^{m} a_i \left\{ \varepsilon_{\beta_i} + (1-\varepsilon_{\beta_i}) \cdot \sum_{j=1}^{n} b_{ij} \frac{\alpha_{ij}\left[\varepsilon_{CE_{ij}} + \left(1-\varepsilon_{CE_{ij}}\right)\varepsilon_{(X,A)_{ij}}\right] + \varepsilon_{\beta\gamma_{ij}}}{1+\alpha_{ij}} \right\} \sum_{i=1}^{m} a_i \sum_{j=1}^{n} b_{ij}\varepsilon_{\gamma_{ij}} \frac{1}{1+\alpha_{ij}} \right\}}{\sum_{i}^{m} a_i \left[\varepsilon_{\beta_i} \sum_{j=1}^{n} b_{ij}\varepsilon_{\gamma_{ij}} \frac{1}{1+\alpha_{ij}} + (1-\varepsilon_{\beta_i}) \sum_{j=1}^{n} b_{ij}\varepsilon_{C_{ij}} \frac{1}{1+\alpha_{ij}} \right]} \quad (4)$$

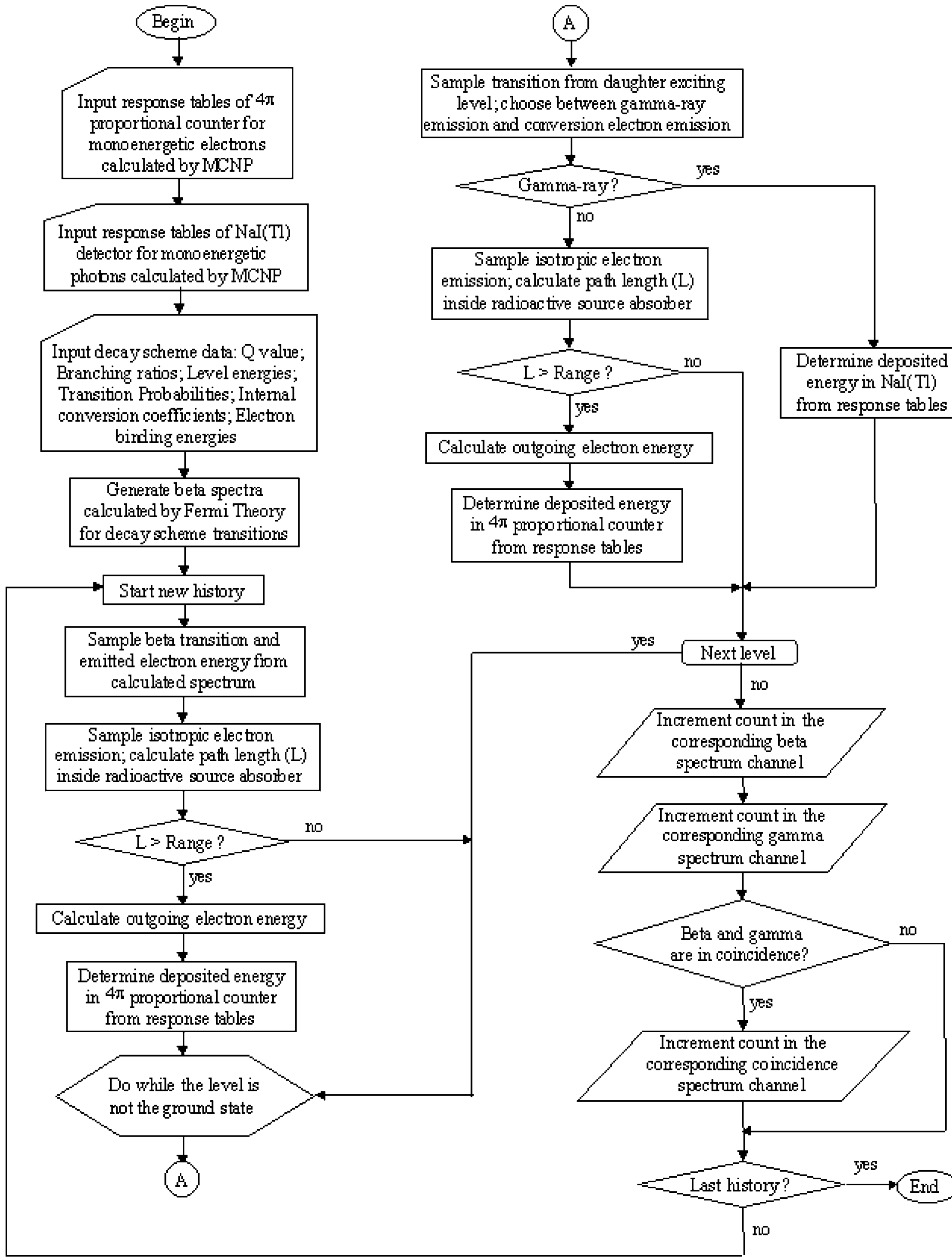

Fig. 2. Schematic diagram of code ESQUEMA.

gamma-ray line, gamma-gamma coincidences are eliminated and (4) and (5) can be simplified

$$\frac{N_\beta N_\gamma}{N_c} = N_0\left\{1+b\frac{(1-\varepsilon_\beta)}{\varepsilon_\beta}\frac{\alpha\left[\varepsilon_{CE}+(1-\varepsilon_{CE})\varepsilon_{(X,A)}\right]+\varepsilon_{\beta\gamma}}{1+\alpha}\right\} \tag{6}$$

and

$$\frac{N_c}{N_\gamma} = \varepsilon_{\beta\text{mean}} \tag{7}$$

where

$$\varepsilon_{\beta\text{mean}} = \sum_{i=1}^{m} a_i \varepsilon_{\beta i}.$$

As can be seen from (6), which corresponds to the simplified case, there is a linear dependence between $N_\beta N_\gamma/N_c$ and the efficiency parameter $(1-N_c/N_\gamma)/(N_c/N_\gamma)$. The corresponding slope is given by $\{\alpha[\varepsilon_{CE}+(1-\varepsilon_{CE})\varepsilon_{(X,A)}]+\varepsilon_{\beta\gamma}\}/(1+\alpha)$. For a complex decay scheme such as ^{134}Cs, the behavior of this curve is rather difficult to predict and depends critically on the gamma window chosen. The purpose of the present work is to simulate (4) and (5) by the Monte Carlo method for the case of ^{134}Cs and compare to experimental results. Furthermore, it was possible to predict the behavior of $N_\beta N_\gamma / N_c$ up to 100% efficiency.

B. Experimental Setup

The standardization of ^{134}Cs was performed by means of a conventional $4\pi(\mathrm{PC})\beta - \gamma$ coincidence system [16], consisting of 4π proportional counter filled with P-10 gas mixture at 0.1 MPa, coupled to a pair of $3'' \times 3''$ NaI(Tl) crystals. A detailed description of this system is given elsewhere [10]. All pulses above noise threshold (0.7 keV) were measured in the beta channel. The gamma-rays were gated at two discrimination windows, covering the total absorption peaks of 600 and 800 keV, respectively.

The radioactive sources to be measured in the $4\pi(\mathrm{PC})\beta - \gamma$ system were prepared by dropping known aliquots of the ^{134}Cs solution on 20 μg.cm^{-2} thick Collodion film. This film had been previously coated with a 10 μg.cm^{-2} thick gold layer on each side, in order to render the film conductive. A seeding agent (Ludox) was used for improving the deposit uniformity and the sources were dried in warm (40 °C) nitrogen jet flow. The accurate source mass determination was performed using a Mettler 5SA balance by the pycnometer technique [4]. The variation in the efficiency was achieved by placing external Collodion or aluminum absorbers over or under the radioactive sources.

C. Monte Carlo Simulation

The electron and the gamma-ray detection spectra were determined as a function of the energy by means of the MCNP4C code [17] using detailed design information on the LMN $4\pi(\beta, \mathrm{X}) - \gamma$ coincidence system [10]. A Monte Carlo code called ESQUEMA was written in order to follow the decay scheme, from the precursor nucleus to the ground state of the daughter nucleus. A flow chart of this code is shown in Fig. 2. A set of response tables for monoenergetic electrons (4π proportional counter) and photons (NaI(Tl) crystals) were prepared beforehand by means of code MCNP4C. These tables, together as decay scheme data were used as input for code ESQUEMA. The beta spectrum shape was estimated by the Fermi Theory of β Decay [18] applied to allowed transitions. The code generated tables of beta spectra for all transitions from the decay scheme. A probability function was derived from the beta spectrum shape, and the emitted beta energy was sampled using a random number. Following the decay scheme, the choice between the gamma or conversion electron transition was made by sampling another random number. When a gamma transition was detected in coincidence with a beta event, a count in the coincidence spectrum was registered. If the conversion electron was detected in coincidence with the beta particle, a count was registered in the beta spectrum corresponding to the sum of both energies. The present version of the code allows selection by changing the cutoff of deposited energy in the $4\pi\beta$ detector or by applying absorbers on the radioactive source substrate. In this way it was possible to simulate variation in beta efficiency.

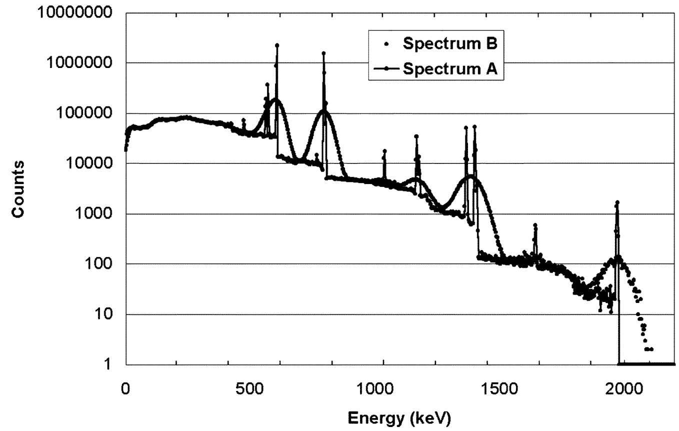

Fig. 3. Gamma ray spectra from ^{134}Cs simulated by Monte Carlo. Spectrum A is without resolution effects. Spectrum B includes resolution matching experimental spectrum at 661.6 keV.

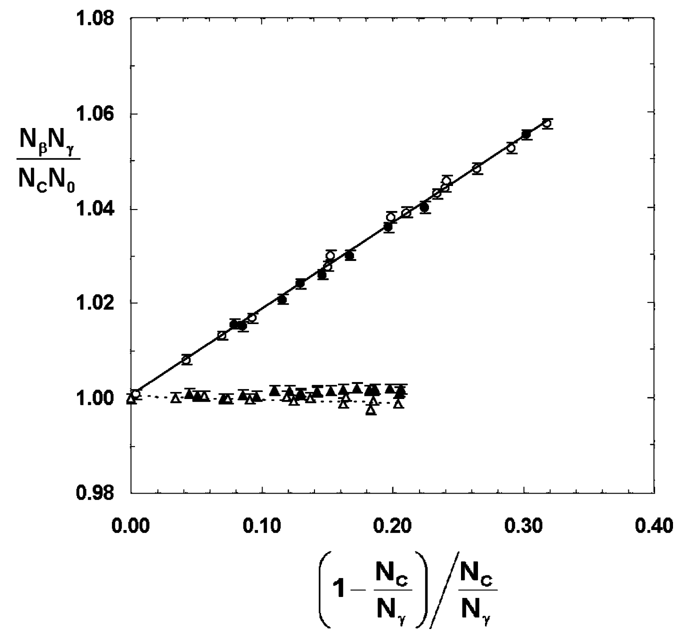

Fig. 4. Behavior of $N_\beta N_\gamma / N_c N_0$ as a function of efficiency parameter $(1 - N_c/N_\gamma)/(N_c/N_\gamma)$. Upper points correspond to 600 keV gamma window. Lower points correspond to 800 keV gamma window. Full symbols—experimental results; open symbols—Monte Carlo results.

III. Results and Discussion

Fig. 3 shows two gamma spectra from the NaI(Tl) simulated by Monte Carlo according to the geometry of $4\pi\beta(\mathrm{PC}) - \gamma$ system. Spectrum A does not incorporate resolution effects and shows the contribution of all gamma lines. Spectrum B incorporates NaI(Tl) resolution matching the experimental spectrum at 661.6 keV which corresponds to ^{137}Cs gamma energy. The resolution at other energies follows the 1/E law [19].

As can be seen, the window of spectrum B at 600 keV, besides the main gamma line of 604.69 (98.21%), incorporates two other gamma lines, namely: 563.23 keV (8.44%) and 569.32 keV (15.54%), as well as Compton events from 795.84 and 801.93 keV transitions.

Fig. 4 shows the behavior of $N_\beta N_\gamma / N_c$ as a function of efficiency parameter $(1 - N_c/N_\gamma)/(N_c/N_\gamma)$. The circles correspond to 600 keV gamma window and the triangles to 800 keV gamma window. The closed circles and triangles correspond to experimental results obtained by placing external absorbers above and below the radioactive sources. The open circles and triangles correspond to values predicted by the Monte Carlo method.

There is good agreement between the experimental slope (0.1819(30)) and the calculated one (0.1783(24)) at 600 keV gamma window. The explanation for this high slope may be contribution of 569.32 and 801.93 keV transitions to 600 keV gamma window. These transitions come from a low energy beta transition (88.5 keV maximum energy) which is absorbed more strongly than the main beta ray (657.8 keV maximum energy). The window at 800 keV is much less affected by the low energy beta branch and the resulting slope is small. The experimental slope at 800 keV resulted (0.0099(19)) and the Monte Carlo result was $(-0.0041(36))$. The cause of this small difference is being investigated. However, the effect in the extrapolated activity is less than 0.1% for beta efficiencies higher than 94%.

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