Monte Carlo Simulation of Activity Measurements by Means of $4\pi\beta - \gamma$ Coincidence System

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The methodology for simulating all detection processes in a $4\pi\beta-\gamma$ coincidence system by means of the Monte Carlo technique is described. The goal is to predict the behavior of the observed activity as a function of the $4\pi\beta$ detector efficiency. In this approach, the information contained in the decay scheme is used for determining the contribution of all radiations emitted by the selected radionuclide, to the measured spectra by each detector. This simulation yields the shape of the coincidence spectrum, allowing the choice of suitable gamma-ray windows for which the activity can be obtained with maximum accuracy. The simulation can predict a detailed description of the extrapolation curve, mainly in the region where the $4\pi\beta$ detector efficiency approaches 100%, which is experimentally unreachable due to self absorption of low energy electrons in the radioactive source substrate. The theoretical work is being developed with MCNP Monte Carlo code, applied to a gas-flow proportional counter of 4π geometry, coupled to a pair of NaI(TI) crystals. The calculated efficiencies are compared to experimental results. The extrapolation curve can be obtained by means of another Monte Carlo algorithm, being developed in the present work, to take into account fundamental characteristics of a complex decay scheme, including different types of radiation and transitions. The present paper shows preliminary calculated values obtained by the simulation and compared to predicted analytical values for a simple decay scheme.

1 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 uses 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 in place of proportional counters.

The Nuclear Metrology Laboratory (LMN) of IPEN-CNEN/SP has developed several radionuclide standardization systems [10,11,12,13,14]. Presently, the LMN has two $4\pi\beta-\gamma$ coincidence systems composed of gas-flow or pressurized proportional counters [10,11,12,13,14] coupled to NaI(Tl) or HPGe detectors. The type of gamma detector chosen depends on the readionuclide characteristics.

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 of the resulting activity. Usually, this value is obtained by the Linear Extrapolation Method [3], varying 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 process in a $4\pi\beta-\gamma$ coincidence system to allow predicting the extrapolation curve.

In this paper, a simple decay scheme is simulated, where the results can be easily predicted analytically. Detailed characteristics of the LMN coincidence system are taken into account in order to perform calculation as realistic as possible.

2 Methodology

2.1 Coincidence Equations

Figure 1 shows the hypothetical simple decay scheme assumed. A single 1.0 MeV beta ray is followed by a single 1.0 MeV gamma-ray. This gamma transition is partially internally converted, with a conversion coefficient (α) equals to 0.1. The Conversion electron energy was assumed to be 0.95 MeV. In this condition, the coincidence equations become:

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$$N_{\beta} = N_0 \epsilon_{\beta} + N_0 (1 - \epsilon_{\beta}) \frac{\alpha}{1 + \alpha} \epsilon_{CE} \tag{1}$$

$$N_{\gamma} = N_0 \epsilon_{\gamma} \frac{1}{1+\alpha} \tag{2}$$

$$N_C = N_0 \epsilon_\beta \epsilon_\gamma \frac{1}{1+\alpha} \tag{3}$$

Therefore,

$$\frac{N_{\beta}N_{\gamma}}{N_{C}} = N_{0}\left[1 + \frac{(1 - \epsilon_{\beta})}{\epsilon_{\beta}} \frac{\alpha}{1 + \alpha} \epsilon_{CE}\right] \tag{4}$$

where:

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

 N_0 is the radioactive source disintegration rate;

 ϵ_{β} and ϵ_{CE} are the beta and conversion electron detection efficiencies, respectively.

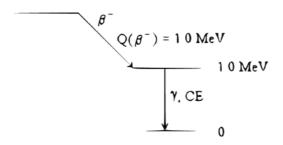


Figure 1. Decay scheme considered in the Monte Carlo simulation.

As can be seen in equation (4), as long as the conversion electron detection efficiency remains constant and close to 100%, there is a linear dependence between $N_{\beta}N_{\gamma}/N_{C}$ and the efficiency parameter $(1-\epsilon_{\beta})/\epsilon_{\beta}$, and the slope is given by $\alpha\epsilon_{CE}/(1+\alpha)$. The slope is dominated by the conversion coefficient. For the proposed simple decay scheme the expected slope is 0.0909.

2.2 Monte Carlo Simulation

The electron and the gamma-ray energy spectra were simulated by means of the MCNP code [16] using detailed design information on the LMN $4\pi(\beta,X)-\gamma$ coincidence system [10]. The beta spectrum shape was estimated by the Fermi Theory of β Decay [15], assuming an allowed transition of 1.0 MeV, maximum beta energy.

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 probability function was derived from the beta spectrum shape, and the emitted beta energy was sorted by a random number. Following the decay scheme, the choice between the gamma or conversion electron transition was made by sorting 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 is detected in coincidence with the beta particle, a count

is registered in the beta spectrum corresponding to the sum of both energies. The present version of the code allows selecting cutoff energies for both the emitted beta particle energy and/or the deposited energy in the $4\pi\beta-\gamma$ detector. In this way it was possible to simulate variation in the beta efficiency.

3 Results

Figure 2 shows the extrapolation curve predicted by the Monte Carlo method. Curve A corresponds to variation in the beta efficiency by changing the cutoff energy for emitted beta particles and curve B to variation in the detected beta energy, respectively. Both curves agree in the high efficiency region and a least square fit yielded the expected slope (0.0909) within the statistical uncertainty for both curves. As the abscissa goes above 1.0 (beta efficiencies below 50%), curve B decreases, indicating that the conversion electron efficiency is falling to values below 100%. Although the conversion electron has a high energy, approaching the maximum beta energy, its deposited energy is very small due to the track length in the detector which is much smaller that the electron range. Therefore, the deposited energy spectrum of the conversion electron overlaps in a great extent with the deposited energy spectrum of the beta ray. For this reason, the conversion electron efficiency decreases with the descriminator lower level.

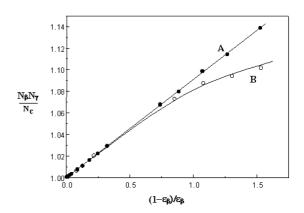


Figure 2. Extrapolation curve simulated by Monte Carlo. Curves A and B correspond to variation in beta efficiency by changing the emitted and detected electron energy, respectively.

Improvements in the code are underway in order to achieve better theoretical estimates and allow comparison with extrapolation curves obtained experimentally. These improvements include better gamma-ray energy resolution and simulated geometry in closer resemblance to the experimental conditions.

References

- [1] A. P. Baerg, Metrologia, 2, 23 (1966).
- [2] A. P. Baerg, Metrologia, 3, 105 (1967).
- [3] A. P. Baerg, Nuclear Instruments and Methods **112**, 143 (1973).
- [4] P. J. Campion, Int. J. Appl. Radiat. Iso. 4, 232 (1959).
- [5] A. Gandy, Int. Journ. Appl. Radiat. Isot. 11, 75 (1961).
- [6] A. Gandy, Int. Journ. Appi. Radiat. Isot. 13, 501 (1962).
- [7] H. Houthermans, M. Miguel, Int. J. Appl. Radiat. Isot. 13, 137 (1962).
- [8] Y. Kawada, Extended applications and improvement of the $4\pi\beta-\gamma$ coincidence method in the standardization of radionuclides. Res. of ETL. Japan, ETL-730, 1972.
- [9] J. G. V. Taylor, Canad. Journ. Phys. 40, 383 (1962).
- [10] L. P. Moura, "Generalized coincidence method for absolute activity measurements of radionuclides". Application for determining the internal conversion coefficient of Tl-203 279keV transition. PhD Thesis, University of Campinas, 1969 (in Portuguese).

- [11] M. S. Dias, "Calibration of a Well-Type Ionization Chamber System for Radionuclide Standardization". MSc. Thesis, University of São Paulo, 1978 (in Portuguese).
- [12] M. F. Koskinas, "Desenvolvimento de um sistema de coincidência para a medida absoluta da atividade de radionuclídeos empregando detectores de barreira de superfície". Tese de Doutorado apresentada ao Instituto de Pesquisas Energéticas e Nucleares, 1988.
- [13] W. O. Lavras, M. F. Koskinas, M. S. Dias, and K. A. Fonseca, "Primary Standardization of ⁵¹Cr Radioactive Solution". IRPA, 2000 (CDROM).
- [14] A. M. Baccarelli, M. S. Dias, and M. F. Koskinas, Appl. Radiat. Isot. 58, 239 (2003).
- [15] R. D. Evans, *The Atomic Nucleus*, McGraw-Hill Book Company, p.548, 1955.
- [16] ORNL, Monte Carlo N-Particle Transport Code System, MCNP4C, RSICC Computer Code Collection, Oak Ridge National Laboratory. Report CCC-700, 2001.