CHAPTER 26

On the Standardization of Beta-Gamma-Emitting Nuclides by Liquid Scintillation Counting

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INTRODUCTION

The $4\pi\beta$(LS) efficiency tracing method with ^3H has been successfully applied to the standardization of pure beta emitters.[1-5] In this method, a ^3H standard is used to calibrate the measuring system, and a combination of experimental measurements and theoretical computations standardizes the unknown nuclide. We present here the extension of this method to the case of any β-γ-emitting nuclide.

The basic diagram of the efficiency tracing method is shown in Figure 1 and a complete description can be found in previous papers.[1,2] A set of vials containing a ^3H standard is measured and a distribution of efficiency vs a quench parameter is obtained. On the other hand, the theoretical efficiency can be computed as a function of the figure of merit M, that we will define here as the energy in keV required to produce one photoelectron at the first dynode of the phototube.[4] The next step is obtaining the distribution of the figure of merit vs the quench parameter, which is independent of the radionuclide. The last distribution characterizes the measuring system. If we can compute the theoretical overall efficiency of the unknown nuclide vs the figure of merit, then we can calculate its activity from the precedent distributions.

COMPUTATION OF THE THEORETICAL COUNTING EFFICIENCY

In order to extend the method to any beta-gamma emitter, the counting efficiency must be computed. This implies the computation of all the possible ways the nuclide can decay to the ground state. There are three different processes that must be taken into account: beta emission, gamma emission, and electron conversion. First we will study the efficiency computation for these individual processes and then we will show how they can be combined to obtain the overall efficiency.

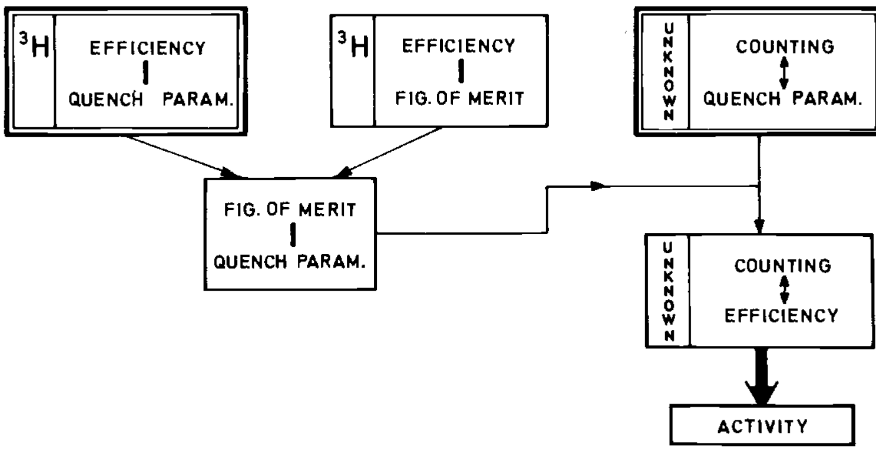

Figure 1. Block diagram of the efficiency tracing method with ^3H by LSC. Boxes drawn in thick lines correspond to experimental measurements.

Beta Efficiency

Taking into account shape factors, the beta spectra are calculated from the Fermi theory of beta decay. According to this theory, the number of particles of energy between E and E + dE is given by the equation:

$$\frac{dN(E)}{dE} = C(E)\ F_o(Z,E)\ L_o(Z,E)\ (E_m - E)^2\ (E + 1)\ p, \tag{1}$$

where E_m is the maximum kinetic beta particle energy in m_oc^2 units, and F_o and L_o are given in reference.[4] C(E) is the shape factor, and its value[6] is given in the Table 1. In the case E = 0, the equation is:

$$\frac{dN(E)}{dE} = 8\pi\ C(o)\ (2R)^{2\gamma-1}\ (\alpha Z)^{2\gamma-1}\ \frac{E^2\ L_m(Z_\delta o)}{|\Gamma(2\gamma + 1|^2} \tag{2}$$

where R is the nuclear radius of the residual nuclide and α is the fine structure constant. A more detailed description of the spectrum calculation can be found in References 1 and 4.

Table 1. Values of the Prohibition Parameter (q is the Neutrino Momentum in m_oc^2 Units)

Prohibition	Value
Allowed and first forbidden	1
Unique-first forbidden and second forbidden	$p^2 + q^2$
Unique-second forbidden and third forbidden	$p^4 + \frac{10}{3}p^2 \cdot q^2 + q^4$
Unique-third forbidden and fourth forbidden	$p^6 + 7p^2q^2\ (p^2 + q^2) + q^6$

If the beta spectrum is normalized to be unity, the counting efficiency is given by the expression:

$$\epsilon = \int_{o}^{E_m} N(E)\left(1 - \exp\left[-\frac{E.Q(E)}{2.M}\right]\right)^2 dE \tag{3}$$

Here, Q(E) is the ionization quenching correction factor given by:

$$Q(E) = \frac{1}{E}\int_{o}^{E_m} \frac{dE}{1 + k.B\left(\frac{dE}{dx}\right)} \tag{4}$$

In this work, we used the following approximate equation[7]:

$$Q(E) = \frac{.357478 + .459577*t + .159905*t^2}{1 + .0977557*t + .215882*t^2} \quad \text{with } t = \log_{10}E \tag{5}$$

The expression E.Q(E) in Equation 3 represents the amount of energy converted in photons and if the figure of merit (M) is considered, the probability of nondetection will be given by:

$$\exp\left(-\frac{E.Q(E)}{M}\right) \tag{6}$$

Hence, for a system with two tubes working in coincidence mode, the probability of detection will be given by:

$$\left\{1 - \exp\left(-\frac{E.Q(E)}{2.M}\right)\right\}^2 \tag{7}$$

Gamma Efficiency

To compute the γ counting efficiency, the interaction probability and the Compton spectrum distribution must be obtained. This is carried out by means of a Monte Carlo simulation. The emission point of the photon is drawn according to the vial dimensions, then three direction cosines and a pathlength are drawn to define the arrival point for this step. The type of interaction is decided by a random process, taking into account the cross sections for photoelectric and Compton processes; the trajectory of the photon is modified accordingly. The process finishes when the photon escapes out of the vial, its energy drops below 10 eV, or if the interaction is photoelectric. In any case, the amount of energy lost by the photon in the scintillator is considered to build up the electron distribution. The interaction probability is also obtained from the spectrum. A complete description of the process is given in García-Toraño and

Grau (1987), and a typical spectrum obtained in the simulation is shown in Figure 2.[8]

If N(E) is the distribution of energy of the electrons, the counting efficiency can be obtained from the expression:

$$\epsilon = \int_{o}^{E_\gamma} N(E)\left(1 - \exp\left[-\frac{E.Q(E)}{2.M}\right]\right)^2 dE \tag{8}$$

where E_γ is the gamma-ray energy.

Internal Conversion

Internal conversion transitions produce vacancies in the atomic shells. To compute the effective energy converted into light, one must first calculate the probabilities and effective energies for the different processes involved. Only K,L, and M internal transitions are considered. The complete expressions for all the processes have been detailed in Grau (1982).[9]

The internal conversion processes contributes to the efficiency as a function $F(\phi_1, n_1, E_{ij}, E_e)$, where ϕ_1 and n_i are the probability and number of emitted particles in the i-atomic rearrangement, E_{ij} is the energy corresponding to the j-th particle and E_e is the converted electron energy.

Overall Efficiency

Consider the case of a radionuclide which decays by beta emission and is followed by a cascade of n gamma rays to the ground state of the daughter nuclide. Although this is a very simplified model of β-γ emitter, the expression that gives the counting efficiency is as complex as:

$$\epsilon = \int_{o}^{E_\beta}\int_{o}^{E_{\gamma 1}}\int_{o}^{E_{\gamma 2}}\cdots\int_{o}^{E_{\gamma n}} N(E)S_1(E_1)\,.\,.\,S_n(E_n)\left(1-e^{-E_\beta Q(E_\beta)-\sum E_{\gamma i}Q(E_{\gamma i})}\right)^2 dE_1.\,.dE_n \tag{9}$$

where

E_β = Maximum energy of the beta emitter
$E_{\gamma i}$ = Energy of the i-th gamma ray
N(E) = Beta spectrum
$S_i(E_i)$ = Spectrum of Compton and photoelectric electrons

In a real case, some of the gamma transitions could be converted, and the appropriate conversion coefficients would affect Equation 9, which should be split in all the possible combinations of gamma and electron conversion processes. The argument of the exponential should also be modified in accordance

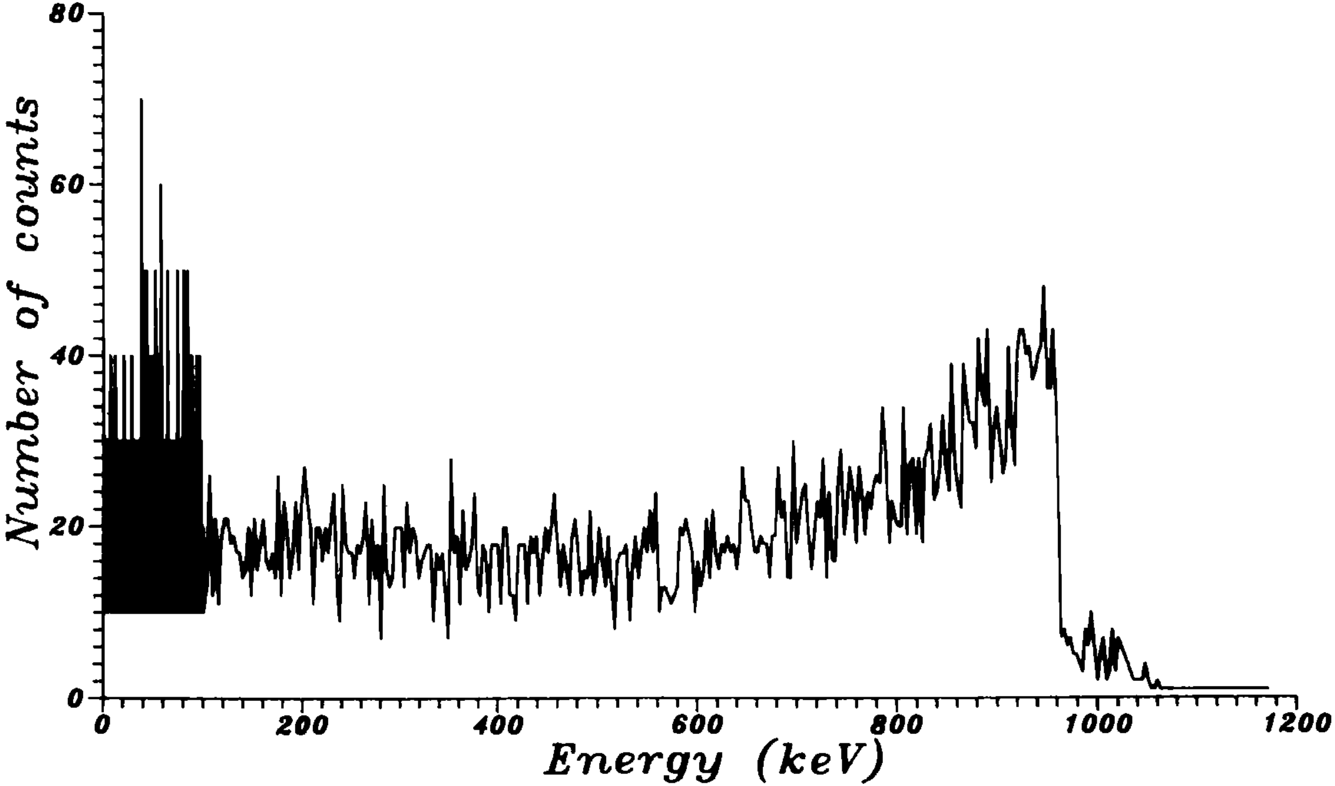

Figure 2. Spectrum of Compton electrons obtained in a typical simulation of the interaction between the 1.17 MeV γ ray of the ^{60}Co and the scintillator (dioxane-naphthalene). The volume of the vial is 10 ml and the radius is 1.25 cm.

with the internal conversion processes, and Equation 9 would become a sum of similar expressions, with the required normalizing factors.

If we adopt a general scheme, other β emissions can feed the intermediate levels, and some additional gamma transitions will appear. The general expression for the efficiency then becomes a combination of expressions like Equation 9; for instance, in the case of ^{60}Co, 15 expressions must be computed. Its numerical evaluation is not possible without some kind of factorization; a typical Compton spectrum has about 2000 points and the multiple integral could take excessive time.

It may be seen that the integral (Equation 9) can be factorized in terms of some factors which only depend on the individual processes. We have developed a FORTRAN program which builds up all the possible decay ways, factorizes the integrals, and takes into account the normalizing factors (branching ratios, conversion coefficients, interaction probabilities, etc.).[10] It has been used in the efficiency calculations needed in this work.

EXPERIMENTAL RESULTS

The method described in this paper has been applied to the standardization of ^{60}Co. This nuclide, which decays by β-γ emission to ^{60}Ni is usually standardized by the method of $4\pi\beta$-γ coincidence-anticoincidence. A simplified decay scheme is shown in Figure 3, and some characteristic nuclear data are presented in Table 2.[11] Although there are other transitions, only the most significant have been considered in this study.

Materials

A standard solution of n-hexadecane ^{3}H from Amersham, U.K., was used as the reference for the efficiency tracing method. The radioactive concentration was 50 kBq/mL and the uncertainty, taken as the addition of both systematic and random components, was certified to 3%. The ^{60}Co was also from Amersham, and its chemical form was Cl_2Co, 0.1 *M*. The certified uncertainty was 0.5%.

The scintillation solution was formed by naphthalene 60 gr, PPO 4 gr, Dimetil POPOP 0.1 gr, methanol 100 mL, etilenglicol 20 mL and Dioxane until 1 L.

Sample Preparation and Equipment

Two sets of seven identical vials were prepared, one for the ^{3}H, the other for the ^{60}Co. Each vial contained 10 mL of the cocktail solution. In each set Cl_4C was added in 5 μL increments to obtain different quench parameters. The radionuclide solutions were added gravimetrically to the vials. The stability was studied over a period of one week and proved to be good.

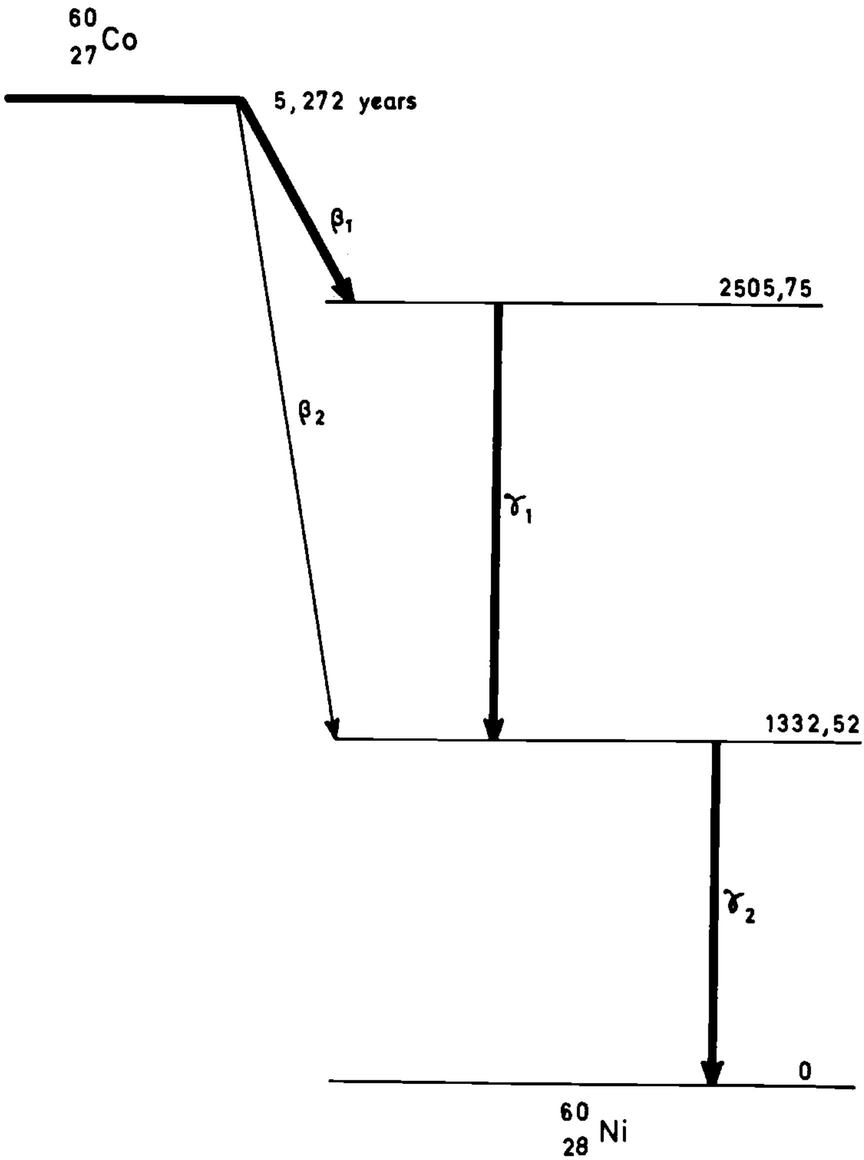

Figure 3. A simplified decay scheme of the ^{60}Co.[10]

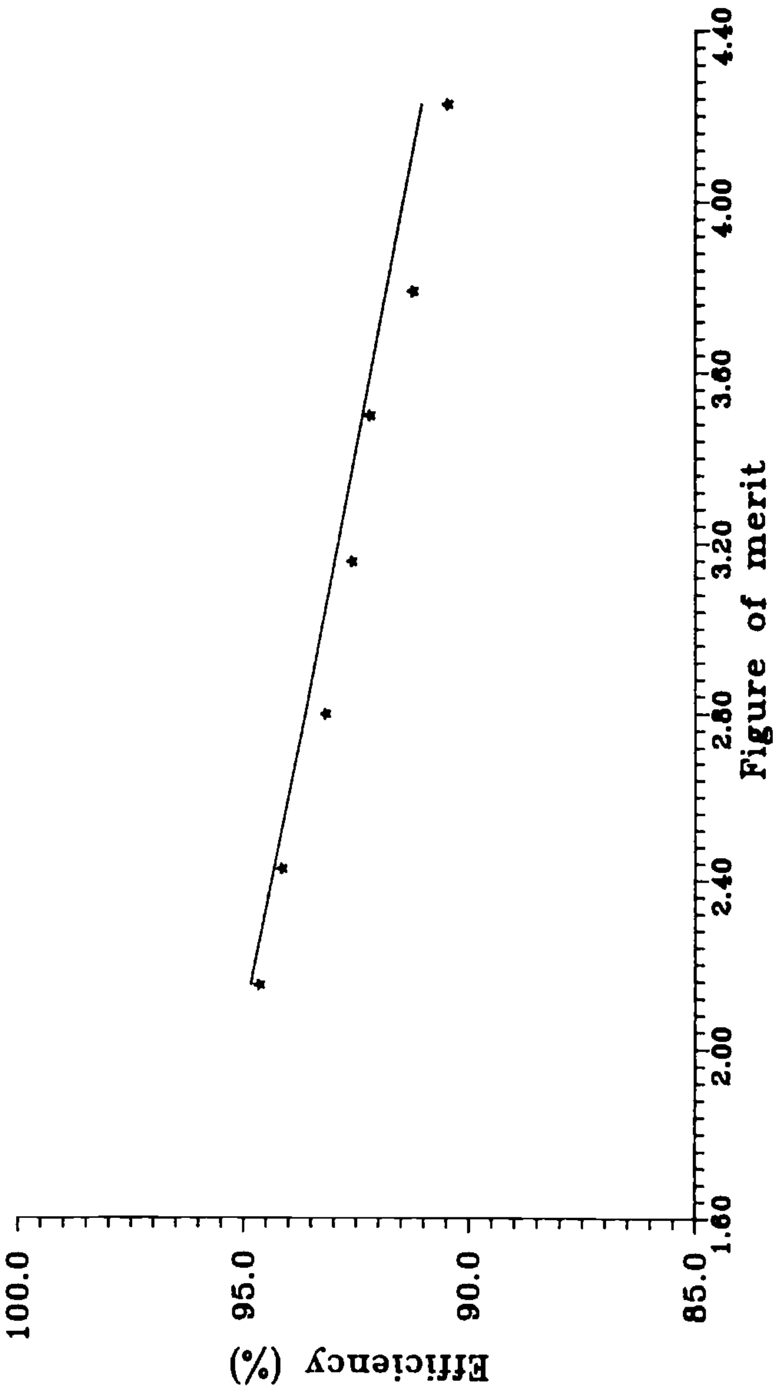

Figure 4. Measured (single points) and computed (full line) efficiencies in the standardization of ^{60}Co, as a function of the figure of merit.

Table 2. Selected Nuclear Data[10] from ^{60}Co

Transition	Energy (keV)	Intensity (%)
β^1	317.9 (0.3)	99.89 (0.06)
β^2	1491.1 (0.3)	0.10 (0.02)
γ^1	1173.22 (0.05)	99.89 (0.06)
γ^2	1332.51 (0.05)	99.993 (0.002)

Table 3. Measured and Computed Efficiencies for ^{60}Co

Sample	Quench Parameter	Figure of Merit	Measured Efficiency	Computed Efficiency	Diff (%)
1	344	2.1492	94.6194	94.8144	−0.20
2	327	2.4262	94.1109	94.2893	−0.19
3	306	2.7979	93.1427	93.5946	−0.48
4	289	3.1562	92.5480	92.9340	−0.41
5	275	3.4985	92.1405	92.3104	−0.18
6	262	3.7898	91.1859	91.7846	−0.66
7	249	4.2260	90.4283	91.0053	−0.63
					Average −0.4%

Measurements were carried out with a LKB RackBeta liquid scintillation counter, which was connected on-line to a personal computer.

Results

We present in Table 3 and Figure 4 the results for the measured and computed efficiencies. In the same table, values are also given for the quench parameter and the figure of merit. The differences between experimental and computed[11] efficiencies, also shown in the table, vary between 0.18 and 0.66%, with an average value of 0.4%.

The uncertainties estimated for the method are shown in Table 4. The most important are due to the counting statistics and the quench parameter determination; the contribution of the 3% uncertainty in the ^{3}H standard leads to only a 0.1% in the efficiency of ^{60}Co. We also considered the component due to the nuclear data, and finally, we numerically estimated the influence of the Monte

Table 4. Estimated Uncertainties in the Standardization of ^{60}Co by the LSC Efficiency Tracing Method with ^{3}H

Source of Uncertainty	Uncertainty (%)
Liquid scintillation counting of ^{3}H	0.2
Liquid scintillation counting of ^{60}Co	0.17
Quench parameter determination	0.1
Sample preparation	0.1
Nuclear data	0.09
Monte Carlo simulation	0.06
^{3}H standard	0.1
Combined uncertainty	0.33%
Overall uncertainty (three times the combined uncertainty)	0.99%

Carlo simulation as 0.06. The combined uncertainty resulted in 0.332% and for the overall uncertainty, taken as three times the combined, we found 0.99%. These values are in good agreement with the average differences found in Table 3, taking into account the uncertainties in the ^{60}Co standard.

In conclusion, a method based on the 4πLS efficiency tracing with ^{3}H has been developed which allows the standardization of β-γ emitters; the application to the case of ^{60}Co has given results which agree well with the values obtained for other methods.

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