

LSC Standardization of Multigamma Electron-Capture Radionuclides by the Efficiency Tracing Method

J.M. Los Arcos, A. Grau and E. Garcia-Toraño

INTRODUCTION

The basic problem in the LS standardization of radionuclide decay by electron capture (EC) processes is the accurate determination of the counting efficiency.¹⁻³ It must take into account all the energy absorbed in the scintillator by the different radiations emitted after the EC decay.

For pure EC nuclides it has been recently described an efficiency tracing method⁴⁻⁵ that predicts the LSC efficiency as a function of a figure of merit. It characterizes the scintillator-equipment system as a whole and does not depend on the radionuclide to be measured.

This method⁶ leans against an experimental quench curve of ³H standards and a model based on the X-rays and Auger-electrons emission. This includes the nonlinear response due to the X-ray escape and ionization quenching.

The efficiency evaluation is a little more difficult⁷⁻⁸ when the electron capture leaves the daughter nuclide in an excited level, which later may decay by a single or double gamma transition in coincidence, like it occurs in ⁵⁴Mn, ⁸⁵Sr, ¹²⁵I, ⁵⁷Co, and ⁹²Nb. For these nuclides, photon-scintillator interaction and eventually conversion electrons and the subsequent atomic rearrangement, must be considered in order to obtain an accurate value for the detection efficiency. The difficulty is even greater for radionuclides where pure EC is in competition with single or double gamma transitions, like ⁸⁸Y or ⁶⁵Zn.

Nevertheless, in all these cases the efficiency evaluation is straight forward because the total number of intermediate rearrangement pathways is kept to a reasonable value and several codes have been developed for toluene-based scintillators.

This is no longer valid when the daughter nuclide shows a complex gamma spectrum involving more than three excited levels, because the number of intermediate rearrangement steps increases very quickly.

An additional difficulty for EC nuclides arises from the photon-scintillator interaction. While most β - γ emitters are not very sensitive to the scintillator because of the intrinsically high β efficiency, EC nuclides show a rather significant efficiency dependence on the chemical composition of the scintillator, its intrinsic efficiency is usually lower and the Compton interactions greatly modify the spectrum of detected electrons. As a consequence, a more general evaluation model is needed to obtain realistic values in practical situations. Some special cases require complex decay schemes or nontoluene-based scintillation cocktails for sample preparation.

This chapter presents a new systematic procedure to overcome these problems. First, an in depth description of the models and its limitations will be developed. Then, the results for several scintillators will be discussed and compared to experimental measurements for ^{133}Ba in INSTAGEL dioxane-naphthalene scintillators.

COUNTING EFFICIENCY FOR SIMPLE EC NUCLIDES

In order to derive the partial efficiencies for some simple decay schemes it will be assumed that the liquid scintillation counting system is composed of two phototubes with the same gain, working in coincidence.

The counting efficiency can then be written as a function of the figure of merit as a sum of contributions for each atomic rearrangement:

$$[1 - \exp(-\sum_{\lambda} E_{\lambda} Q(E_{\lambda})/2M)]^2 \quad (1)$$

- where E_{λ} = the energy of any radiation absorbed in the scintillator after a single EC process,
 $Q(E_{\lambda})$ = the ionization quench correction,
 $E_{\lambda} \cdot Q(E_{\lambda})$ = the effective energy,
 M = the figure of merit.

The function $Q(E)$ can be evaluated with the Birks' semiempirical formula⁹

$$Q(E) = \frac{1}{E} \int_0^E \frac{dE}{1 + kB \frac{dE}{dx}} \quad (2)$$

A good approximation to equation 2, computed with recent values of dE/dx ^{10,11} and taking $KB = 0.0075 \text{ cm/MeV}$, is given by a rational function:

$$Q(E) = \frac{a_1 + a_2 \log E + a_3 \log^2 E}{1 + a_4 \log E + a_5 \log^2 E} \quad (3)$$

where $a_1 = 0.357478$
 $a_2 = 0.459577$
 $a_3 = 0.159905$
 $a_4 = 0.0977557$
 $a_7 = 0.215882$

Pure EC Nuclides

Assuming a K- and L-shell EC model and neglecting L-subshells, there are 21 different rearrangement pathways; the probabilities and effective energies are given in a previous work.^{5,6} An additional M-shell capture, with negligible effective energy, can be kept to combine EC with other subsequent processes in complex decay schemes.

Therefore, for pure EC nuclides, the total counting efficiency is:

$$\epsilon_o (M) = \sum_{\eta}^{22} W_n \left(1 - \exp(-E_n^{EC} / 2M) \right)^2 \tag{4}$$

where W_n is the probability of the n-th rearrangement pathway and E_n^{EC} is the effective energy of that pathway.

EC Nuclides with a Single Gamma Transition in Coincidence

The procedure to compute the efficiency can be easily extended to EC nuclides followed by a single gamma transition in coincidence, like ⁵⁴Mn or ⁵¹Cr.

In this case, three alternative processes can occur depending on whether the photon emitted escapes, is detected in the liquid scintillator, or transformed into a conversion electron ejected from an atomic shell.

Therefore, the efficiency can be written as:

$$\epsilon_1 (M) = \sum_{l=1}^3 \psi_{1l} \epsilon_{1l}(M) \tag{5}$$

where ψ_{1l} = the probability for the process mL,
 ϵ_{1l} = the partial efficiency for the process mL,
 $l = 1, 2, 3$, indicate the EC-photon escape, EC-photon detection, and EC-internal conversion processes, respectively.

The probabilities for each process are:

$$\begin{aligned} \psi_{11} &= P_g [1 - P_{in}] \\ \psi_{12} &= P_g P_{in} \\ \psi_{13} &= [1 - P_g] \end{aligned} \tag{6}$$

where P_g is the probability of photon emission in the gamma transition, and P_{in} is the photon-scintillator interaction probability.

Since the photon escape can be treated as a pure electron capture, its partial efficiency ϵ_{11} is evaluated, as was indicated previously in this section.

The photon-scintillator interaction gives rise to an electron absorption spectrum $N_c(E_c)$ which includes the photoelectric peak as well as the Compton electrons distributions so that the efficiency is:

$$\epsilon_{12}(M) = \int_0^{E_g} N_c(E_c) \sum_i^{22} W_i \left\{ 1 - \exp \left[- (E_i^{EC} + E_c Q(E_c) / 2M) \right] \right\}^2 dE_c \quad (7)$$

where E_g is the energy of the gamma transition.

The internal conversion process is similar to the EC process because both create a primary vacancy in an atomic shell that leads to the subsequent X-ray and Auger-electron emission. The difference lies in that for IC, instead of being absorbed by the nucleus like it happens in EC, an orbital electron is ejected away and contributes to the detection efficiency.

Neglecting IC in shells higher than M, there are 22 different atomic rearrangement pathways, and their weights are similar to those of the EC process, substituting the K-, L- conversion probabilities CK and CL for the capture probabilities PK, PL. The energies are also similar, adding the effective energy of conversion electrons. The partial efficiency can be written:

$$\epsilon_{13}(M) = \sum_{\gamma}^{22} V_j \sum_i^{22} W_i \left\{ 1 - \exp \left[- (E_i^{EC} + E_j^{IC}) / 2M \right] \right\}^2 \quad (8)$$

where V_j , E_j^{IC} are the probability and the effective energy of the j-th pathway in the IC process.

EC Nuclides with a Double Gamma Transition in Coincidence

When the EC decay is in coincidence with a double gamma cascade, there are nine possible rearrangement pathways according to the three different possibilities for each single gamma transition.

The detection efficiency can be written as:

$$\epsilon_2(M) = \sum_{l=1}^3 \sum_{n=1}^3 \psi_{21n} \psi_{21n}(M) \quad (9)$$

where

$$\begin{aligned} \psi_{211} = & P_g(32) [1 - P_{in}(32)] P_g(21) [1 - P_{in}(21)] \\ & P_g(32) [1 - P_{in}(32)] P_g(21) P_{in}(21) \\ & P_g(32) [1 - P_{in}(32)] [1 - P_g(21)] \\ & P_g(32) P_{in}(32) P_g(21) [1 - P_{in}(21)] \\ & P_g(32) P_{in}(32) P_g(21) P_{in}(21) \\ & P_g(32) P_{in}(32) [1 - P_g(21)] \\ & [1 - P_g(32)] P_g(21) [1 - P_{in}(21)] \\ & [1 - P_g(32)] P_g(21) P_{in}(21) \\ \psi_{211} = & [1 - P_g(32)] [1 - P_g(21)] \end{aligned} \quad (10)$$

where

- $P_g(st)$ = the probability of photon emission for the gamma transition st ,
 $P_{in}(st)$ = the total probability of interaction for a photon emitted in the gamma transition st ,
 st = the gamma transition between levels s and t .

The respective efficiency for an electron capture-internal conversion and photon interaction pathway is given by:

$$\epsilon_{rst}(M) = \int_0^{E_g(st)} N_c \sum_{j=1}^{22} V_j \sum_{i=1}^{22} W_i \left\{ 1 - \exp \left[- (E_i^{EC} + E_j^{IC} + E_c Q(E_c)) / 2M \right] \right\}^2 \quad (11)$$

Similar expressions hold for other combinations of processes.

Three Level EC Nuclides

Any EC nuclide involving no more than three levels in the daughter nuclide, can be accommodated in the general decay scheme of Figure 1. Eventually, four different alternative paths can be followed after an EC occurs. Accordingly, the total counting efficiency can be written as:

$$\epsilon_T(M) = \sum_{l=1}^4 \psi_l \epsilon_l(M) \quad (12)$$

- where ψ_l = the probability of the l -th path,
 $\epsilon_l(M)$ = the partial efficiency for the l -th path,
 M = the figure of merit.

All four paths begin with an EC decay, the first one being a pure process that ends at the ground level. In the second and the third path, the EC is

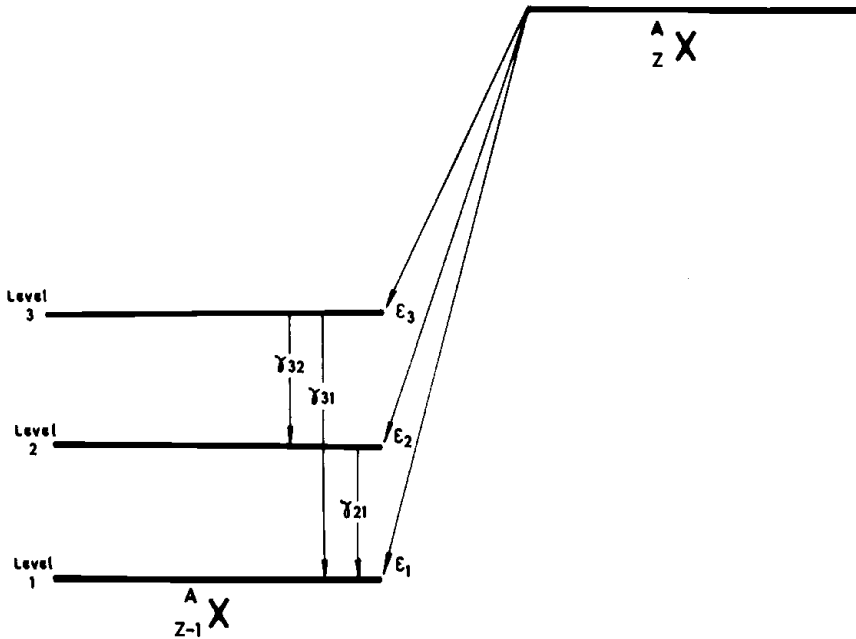


Figure 1. General decay scheme for 3 level EC radionuclides.

followed by a single gamma transition while in the fourth one it is accompanied by a double gamma cascade in coincidence. The probability of each path is:

$$\begin{aligned}
 \psi_1 &= P_{EC}(1) \\
 \psi_2 &= P_{EC}(2) \\
 \psi_3 &= P_{EC}(3) I_\gamma(31) \\
 \psi_4 &= P_{EC}(3) I_\gamma(32)
 \end{aligned} \tag{13}$$

where $P_{EC}(m)$ is the EC branching ratio to the m -th level in the daughter nuclide and $I_\gamma(mn)$ is the relative intensity of the gamma transition connecting the levels m - n in the daughter nuclide.

COUNTING EFFICIENCY FOR MULTIGAMMA EC NUCLIDES

Pathway Description

A direct evaluation of partial efficiency expressions for each alternative pathway leading to the ground level turns out to be impractical for EC nuclides involving more than three excited levels; the number of gamma cascade steps increases rapidly, as can be appreciated in Figure 2.

Instead, a different approach is preferred. One that sequentially simulates

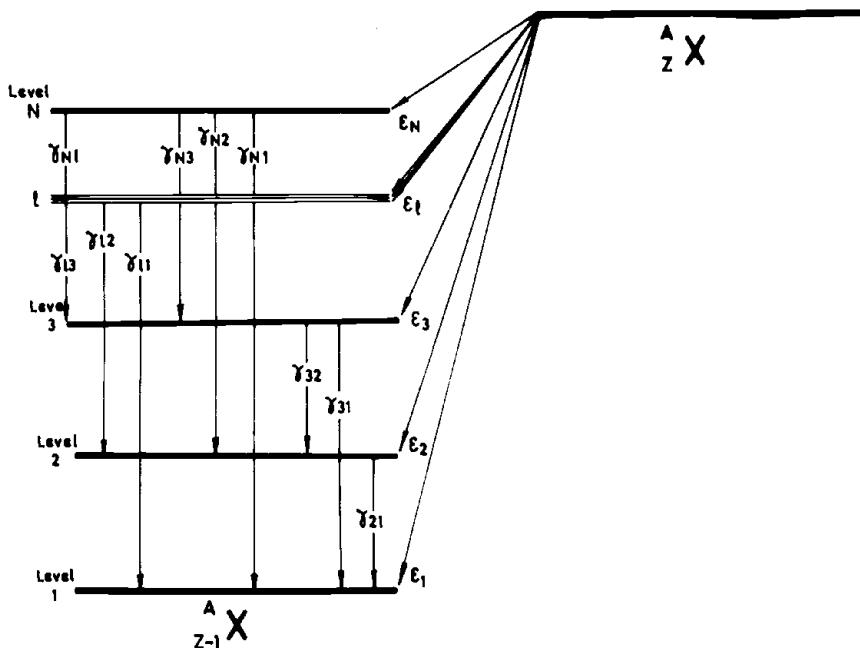


Figure 2. General decay scheme for multigamma EC radionuclides.

each pathway down to the ground level and systematically combines the different transition modes for each intermediate step, photon detection, photon escape, or internal conversion.

If a “route” is defined as a set of gamma transition steps following the EC process, the number of different routes starting at the n-th excited level is:

$$r = 2^{n-2} \tag{14}$$

Eventually, selection rules for gamma transitions apply and not all the routes are allowed.

After the starting level of a route has been fed by the EC, the number of different pathways associated to the j-th route is given by

$$P_j = S_j \tag{15}$$

where S_j is the number of steps which the route is made of.

A simple numbering algorithm for the k-th pathway allows all the possible routes and all different pathways in a route to be generated.

Efficiency Evaluation

Once the different modes in the k-th pathway starting at the n level have been stated, the total effective energy can be evaluated by adding the effective

energies of the S_j modes to the effective energy EEC_n of the EC process. The pathway probability is obtained as a product of the EC branching ratio and the probabilities of the S_j modes, so the partial efficiency for that pathway is:

$$\epsilon_{njK}(M) = P_{EC}(n) \prod_{l=1}^{S_j} P_{jkl} \left\{ 1 - \exp \left[- \frac{1}{2M} \left(E_n^{EC} + \sum_{l=1}^{S_j} E'_{njkl} \right) \right] \right\}^2 \quad (16)$$

where E'_{njkl} and P_{jkl} are the effective energy and probability of the l -th step in the K -th pathway of the j -th route starting at the n -th excited level, and have identical expressions to Equations 10 and 11.

The partial efficiencies are accumulated as a function of the figure of merit during the pathway generation so that at the end, the total counting efficiency is available for each figure of merit previously stated.

RESULTS AND DISCUSSION

A computer code, CEGAN, has been written to carry out all the efficiency computations described in the previous section for any nuclide included in the general decay scheme of Figure 2. It allows specification of the vial geometry and the chemical composition of scintillator.

These parameters concern the Monte Carlo computation of the photon interactions and the subsequent Compton electron distribution.¹²

A single database file for atomic and nuclear parameters makes it easy to specify any nuclide of interest in terms of capture probabilities, branching ratios, allowed gamma transitions, internal conversion coefficients, electron binding energies, fluorescence yields, and X-ray and Auger electron energies and intensities.

The standardization procedure has been tested with ^{133}Ba , which has the decay scheme¹³ shown in Figure 3. $^{133}\text{BaCl}_2$ samples were incorporated into 10 mL of INSTAGEL and dioxane-naphtalene, and a set of quenched samples was measured in an LS counter, LKB 1219 RackBeta spectral. A set of ^3H standards was used to obtain the quench calibration curve.

Figure 4 shows the efficiency values vs the figure of merit for both scintillators. The discrepancy between experimental and computed values is lower than 0.8%, with figures of merit in the range 1.5 to 4 for dioxane and 1.0 to 3.0 for INSTAGEL. An important prediction of the model is the efficiency dependence (several per cent) on the scintillator being used. The differences between some commonly used scintillators can be seen in Figure 5.

The pair INSTAGEL/HISAFE II as well as the PCS/READYSOLV can not be resolved from the point of view of its efficiency. Also, there is a very small difference between the two pairs.

Finally, it has to be noted that the efficiency estimations depend strongly on the uncertainty of the nuclear constants, especially the K-shell electron capture probability which transmits almost all uncertainty to the efficiency value.

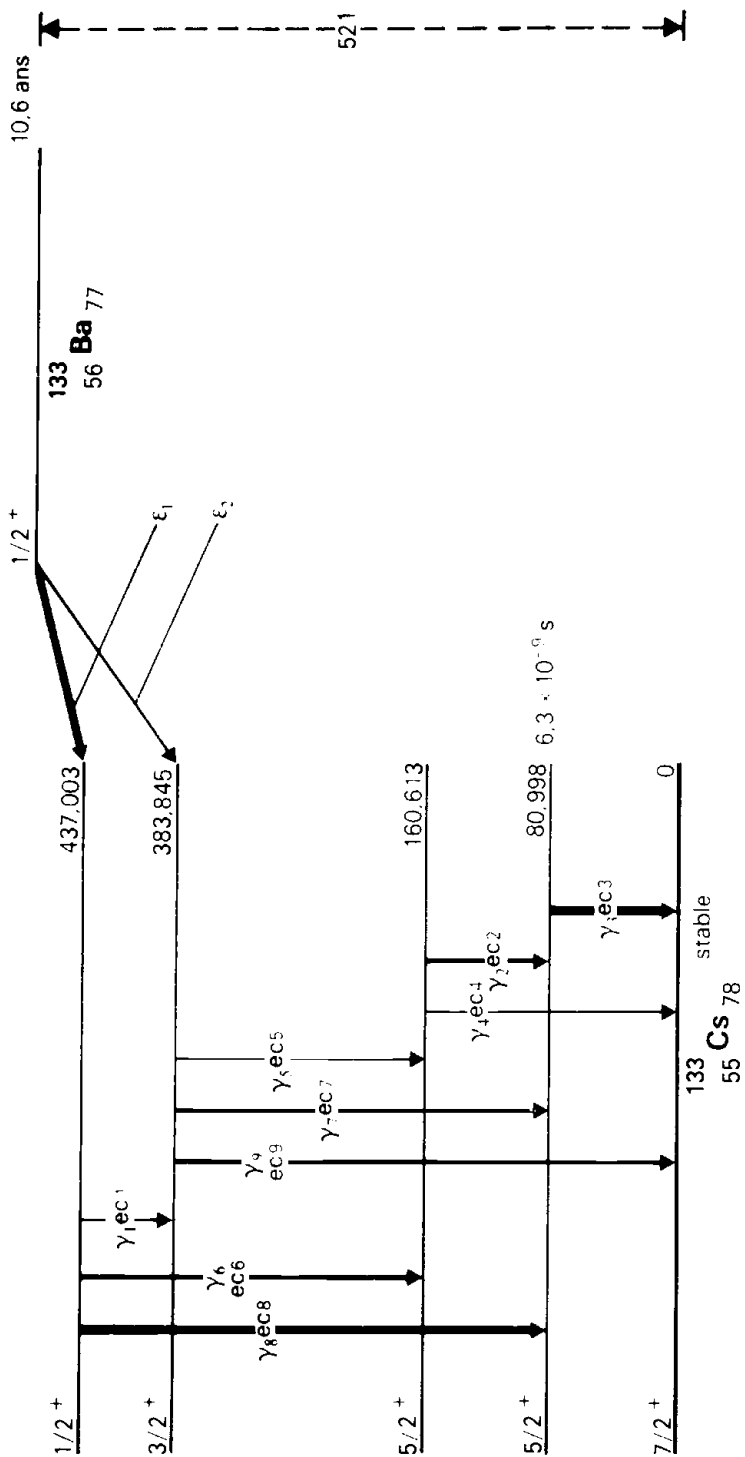


Figure 3. Rearrangement pathways in ^{133}Ba . F: photon detection; G: photon escape; C: internal conversion.

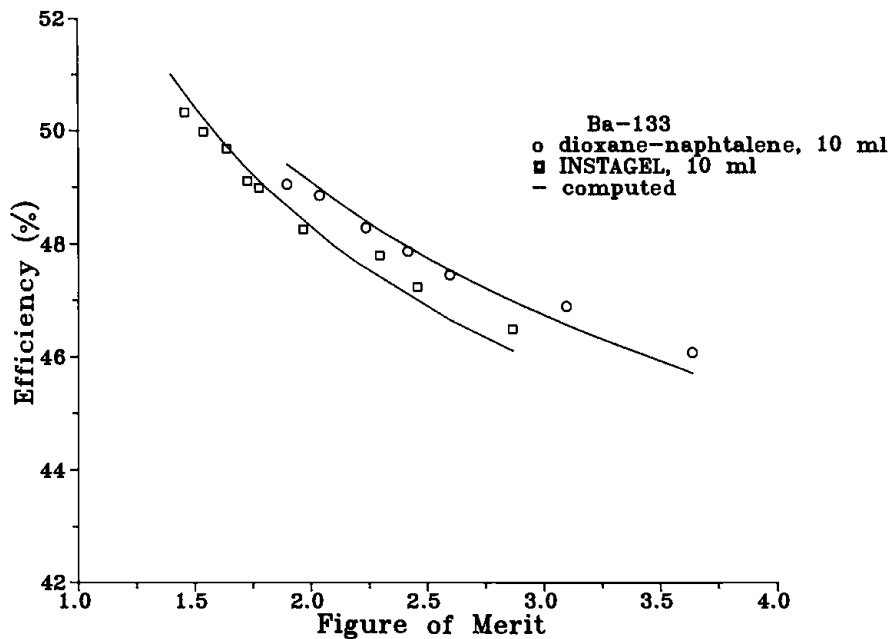


Figure 4. Efficiency vs figure of merit for ^{133}Ba .

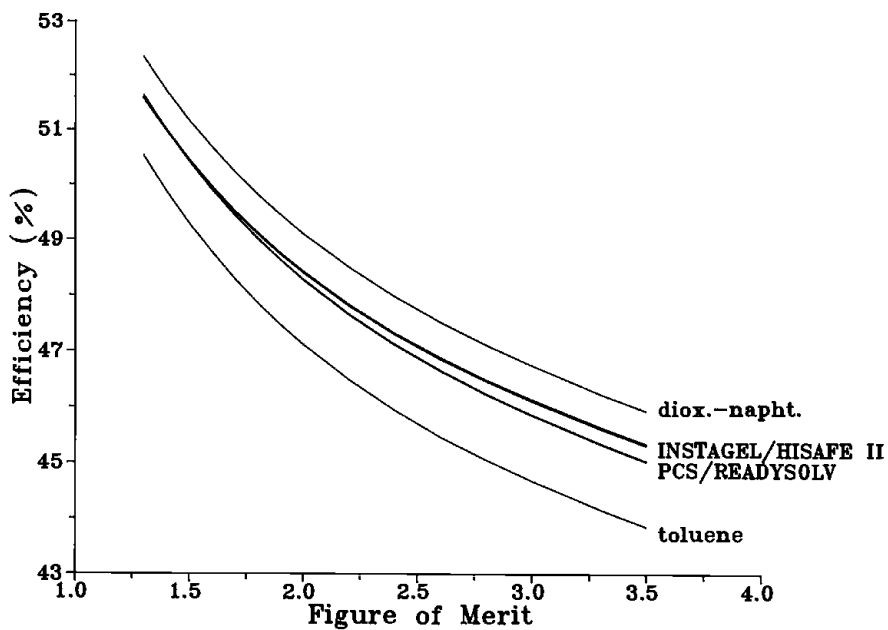


Figure 5. Influence of the scintillator composition on the ^{133}Ba efficiency.

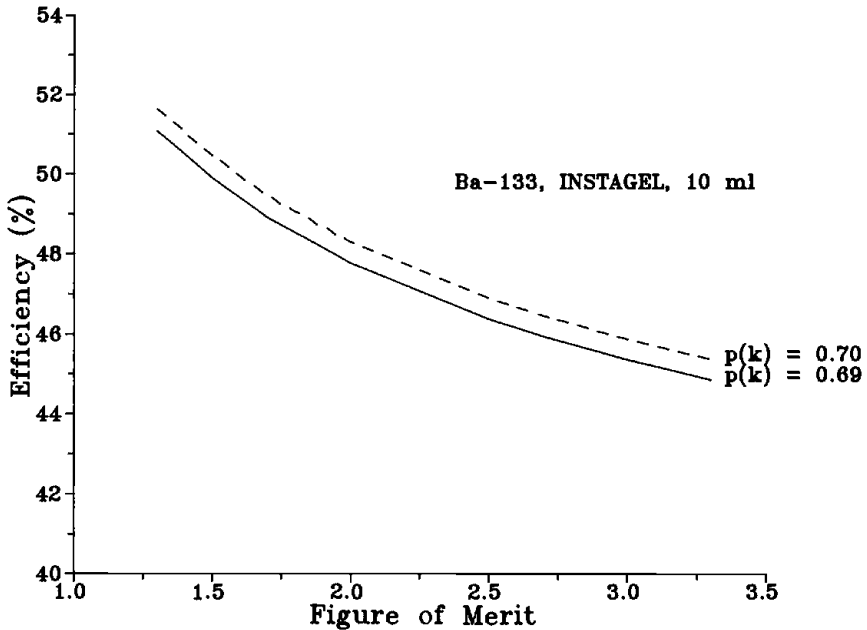


Figure 6. Influence of nuclear constants on the ^{133}Ba efficiency in INSTAGEL. -: $p(k) = 0.69$; -: $p(k) = 0.70$.

Figure 6 shows the influence of an apparently small variation of the K-shell EC probability on the efficiency values in INSTAGEL, as computed by the code.

CONCLUSIONS

The LSC standardization of EC nuclides by the efficiency tracing method has been extended to complex decay schemes with an arbitrary number of coincident gamma emissions. It was done by means of an efficiencies model which takes into account the X-ray and Auger electrons, as well as the interaction of photons with the scintillator, eventually the conversion electrons emitted.

A computer code that performs the pathway evaluations has been prepared and applied to the decay of ^{133}Ba . It shows a good agreement, within 0.8% with experimental measurements and in the range of figures of merit 1 to 4.

A warning should be made against the unconcerned use of nuclear constant values that could transmit big errors on the efficiency evaluation. Also some predictions of the model about the behaviour of several commonly used scintillators have been analyzed.

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