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# Improved method for the calculation of the counting efficiency of electron-capture nuclides in liquid scintillation samples

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ABSTRACT

The methods to compute the counting efficiency of electron-capture nuclides in liquid scintillation counting have been improved in several previous studies. The main improvements comprise a more realistic treatment of the ejection of photoelectrons and subsequent rearrangement processes in the atomic shell as well as a more detailed atomic rearrangement model. The latter was realized in the MICELLE code by means of a new stochastic approach. This new model was also developed to account for energy deposits within micelles.

The recent improvements have now been combined in an updated version of the MICELLE code, which also makes the computation of the counting efficiency of complex decay schemes possible.

In this paper, we describe and discuss recent extensions and improvements of the models and further corrections. The calculated counting efficiencies of selected radionuclides are compared with the experimental data obtained by liquid scintillation counting. For the measurements, we use standard solutions, which were calibrated by other methods.



## 1. Introduction

The computation of the liquid scintillation counting efficiencies for radionuclides decaying by β transitions can easily be performed by means of a free parameter model which forms the basis for two successful approaches: the CIEMAT/NIST and the TDCR methods (see, e.g., Broda et al. (2007) and references therein). Both methods require an accurate computation of the electron emission spectrum as well as the ionization quenching function to account for the nonlinear energy response of the scintillation mechanism.

The same is true for electron-capture nuclides, but it turns out that the pioneering models, as realized in the VIASKL (Los Arcos et al., 1987) or EMI (Grau Carles et al., 1994) codes, overestimated the counting efficiencies. Recent studies indicate that the reason for this discrepancy is an inadequate and oversimplified model of the atomic rearrangement processes, as well as the photoelectric interaction (Kossert and Grau Carles, 2006, 2008).

In this paper we discuss new approaches to apply a more realistic model and to improve the calculation precision of the electron spectra created due to electron-capture processes.

## 2. MICELLE 2

The electron emission spectrum created due to the decay via electron capture encompasses several contributions. The atomic rearrangement may lead to the ejection of Auger and Coster–Kronig electrons. After these ejections, new vacancies are created which may cause further emissions of electrons. In addition, vacancies in the atomic shell may lead to the ejection of X-rays which may then interact with other atoms in the liquid scintillation sample. For X-rays, which usually have energies of only a few keV up to a few tens of keV, the most dominant interaction process is the photoelectric effect, i.e. the energy of an X-ray is transferred to a photoelectron.

The first and pioneering approaches to calculate the counting efficiency of electron-capture nuclides are based on a simplified model, which reduces the atomic shells to the shells K, L and M, i.e. the L subshells and all higher shells are combined to form one L and one M shell, respectively. However, since the light produced by a charged particle interacting within a liquid scintillation sample is a nonlinear function of the particle energy, $E$, this simplification is not accurate enough. Indeed, the counting efficiency obtained when applying this simplified model, using averaged energies and probabilities shows large discrepancies with experimental data. A further simplification concerns the photoelectric interaction of X-rays since the models assume that all the photon energy is converted to the kinetic energy of the photoelectron. In contrast to this assumption, the binding energy has to be subtracted and consequently the overall counting

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efficiency is reduced. Moreover, the ejection of a photoelectron leads to vacancies in the atom that was involved in the interaction process. These atoms such as C, N, O, S, P, Cl and others are part of the sample, i.e. they are components of the scintillation cocktail, the radioactive solution (often mainly $H_2O$) and further ingredients (e.g. quenching agent). The newly created vacancies may lead to further electron and X-ray emissions which slightly increase the overall counting efficiency but do not fully compensate for the previously discussed binding-energy effect.

In an updated code of the MICELLE programme (Grau Carles, 2007), the stochastic atomic rearrangement model has been combined with a simplified approach to account for the photoelectric effect in a proper way. The specifics as well as recent extensions and improvements in the new MICELLE2 code are described in the following.

### 2.1. Stochastic atomic rearrangement model

As explained above, a more detailed atomic rearrangement model is required to compute a more accurate electron spectrum. Since the number of possible rearrangement processes grow rapidly with the increasing number of atomic shells that are taken into account, a stochastic model has been developed. In this model, the individual processes including the initial electron capture as well as subsequent rearrangement processes are simulated using random numbers and the corresponding probabilities. The electron capture creates a primary vacancy in the atomic shell of the daughter atom. To this end, a random number is generated to decide from which of the four shells K, $L_1$, $L_2$ or M an electron is captured. The probabilities are given by the corresponding fractional electron-capture probabilities $P_K$, $P_{L1}$, $P_{L2}$, and $P_{M+}$, respectively. After a K-capture, another random number is used to decide whether the following process is an X-ray emission with probability $\omega_K$ (fluorescence yield) or an Auger-electron ejection with probability $A_K = 1 - \omega_K$. Subsequent rearrangement processes are simulated in a similar manner as explained in previous publications (Grau Carles, 2007; Kossert and Grau Carles, 2008). The energies of all electrons in any individual simulated decay event are saved for the efficiency computation.

The relative probabilities of the radiative (X-rays) and nonradiative transitions (Auger and Coster–Kronig electrons) were selected according to the following criteria: The radiative transitions were only constructed for the K shell and the three L subshells. The transitions from M and outer atomic shells were all considered as nonradiative. For the nonradiative transitions, the methodology followed by McGuire (1972, 1975) in his computed data tables was completely adopted. The probabilities were arranged according to the subshell in which the vacancy is filled, and no distinction was made between Auger and Coster–Kronig transitions for M and outer shells.

### 2.2. Stochastic approach for β branches

The new MICELLE2 code also computes the counting efficiency of $^3$H with the same analytical approach as in the previous EMILIA (Grau Carles, 2006) or MICELLE1 (Grau Carles, 2007) codes. In addition, a new technique has been implemented to compute the counting efficiency of β branches on the basis of a Monte Carlo approach. To this end, the β spectrum, $N(E)$, is first computed with the known analytical methods (Grau Malonda, 1999), taking into account the Fermi distribution and shape-factor functions. The analytical spectrum is normalized and split into 1000 energy bins. A probability is then assigned to each energy bin, $j$ ($j$=1,1000), according to

$$P_j = \sum_{i=1}^{j} N(E_i).$$

Due to the normalization we find $P_{1000}=1$ and $E_{1000}=E_{\beta,\max}$. A random number $N_r$ between 0 and 1 is then used to find the energy bin $j$ of the β electron. This is done in a simple loop searching for $j$ to fulfil $P_j < N_r < P_{j+1}$. Another random number is then utilized to determine the energy of the electron, assuming a uniform distribution within the energy bin. The same computational method is used for positron branches, but in addition, two 511 keV annihilation photons are computed as well.

With the new Monte Carlo method it is straightforward to combine a β branch with one or more γ transitions, and also nuclides decaying by electron capture as well as $\beta^-$ and/or $\beta^+$ (e.g. $^{64}$Cu, $^{65}$Zn, $^{88}$Y) can be computed with just one programme.

### 2.3. Ionization quenching

As in some previous works, the ionization quenching function $Q(E)$ was computed by numerical integration of Birks' formula using the electron stopping power calculation according to Rohrlich and Carlson (ICRU, 1984). The calculation takes the atomic composition of the cocktail into account. For electron energies below 1 keV we apply an empirical approach according to

$$\frac{dE}{dx} = \frac{dE}{dx}\Big|_{1\ \mathrm{keV}} \cdot E^{-1.1}.$$

### 2.4. The micelle effect

The MICELLE programme also includes routines to simulate the energy loss of electrons in micelles. The micelles are part of the 2-phase system with an organic scintillator and an aqueous sample which are only miscible due to the presence of an emulsifier. An energy deposit within a micelle does not produce any scintillation light and thus does not contribute to the counting efficiency. In the model, the micelles are assumed to be spherical volumes and the initial position of an emitted particle is randomly distributed within this volume. A general problem of the model is the lack of information on the size and the distribution of micelles. It can be assumed that the micelle size as well as the reduction of the counting efficiency is quite large (Grau Carles, 2007) when gel samples with a large amount of water are measured.

### 2.5. The photoelectric correction

The photoelectric correction (pc) (Grau Carles, 2006) has been implemented in a slightly simplified way. If a K X-ray interacts via the photoelectric effect, the atom $S$ (H, C, N, O, Na, P, S, Cl) that is involved in the interaction is determined, taking into account the corresponding cross sections. The binding energy, $E_K^S$ of an electron from the K shell is known. The programme then accounts for three contributions:

(i) the photoelectron with the reduced energy $E_{KX} - E_K^S$;
(ii) a KL1L1 Auger electron from the involved atom $S$ with the energy $E_{KL1L1}^S$;
(iii) an electron with the residual energy $E_K^S - E_{KL1L1}^S$.

Due to the latter electron the energy conservation is fulfilled.

### 2.6. The "Larkins correction"

If the "Larkins" correction is used, the energies of KL1L1 Auger electrons are corrected by the difference $\Delta E$ between $E_{KL1L1}$ as calculated from the electron binding energies and the semi-empirical Auger electron energy tabulated by Larkins (1977). For the KLX Auger electrons, $\Delta E/2$ is used as a correction term. No correction is applied to KXY Auger electrons. This effect was found to be large for low-Z ec nuclides (Kossert and Grau Carles, 2006), but for the nuclides $^{109}$Cd and $^{125}$I the correction is negligible due to the higher energies of K-Auger electrons.

### 2.7. Counting efficiencies

The counting efficiency for a system with two photomultiplier tubes is finally given by the arithmetic mean

$$\varepsilon_2(\lambda)=\sum_{i=1}^{N}\left\{1-\exp\left[\frac{-\sum_{l=1}^{M_i}E_{il}Q(E_{il})}{2\lambda}\right]\right\}^2/N,$$

where $N$ is the number of simulated events, $\lambda$ is the free parameter. The number of electrons, $M_i$, in a single decay event, $i$, may comprise several Auger electrons, Coster–Kronig electrons and electrons as a consequence of photon interaction. For β branches, each event comprises exactly one β particle plus additional electrons from potential γ transitions (if any).

In the most recent version, also the counting efficiencies for triple coincidences in a system with three photomultiplier tubes

$$\varepsilon_T(\lambda)=\sum_{i=1}^{N}\left\{1-\exp\left[\frac{-\sum_{l=1}^{M_i}E_{il}Q(E_{il})}{3\lambda}\right]\right\}^3/N$$

and the logical sum of double coincidences

$$\varepsilon_D(\lambda)=\sum_{i=1}^{N}\left\{3\left(1-\exp\left[\frac{-\sum_{l=1}^{M_i}E_{il}Q(E_{il})}{3\lambda}\right]\right)^2-2\left(1-\exp\left[\frac{-\sum_{l=1}^{M_i}E_{il}Q(E_{il})}{3\lambda}\right]\right)^3\right\}/N$$

are computed.

Thus, the ratio $\varepsilon_T(\lambda)/\varepsilon_D(\lambda)$ can be used for TDCR applications. When the micelle effect is taken into account, the individual electron energies $E_{il}$ are reduced by the energy deposit $\Delta E_{il}$ within a micelle (Grau Carles, 2007).

The present model does not take into account any asymmetry effects, i.e. the PMTs are assumed to have the same quantum efficiency.

Since both electron-capture and β branches can be combined with several γ branches (including conversion processes), the new programme is a versatile code that can even be used to treat radionuclides with complex decay schemes. A more complicated analytical approach to take several coincident branches into account (see, e.g., Oropesa Verdecia and Kossert, 2009) is not required.

## 3. Analysis

### 3.1. Nuclear and atomic input data

The computation with MICELLE2 requires atomic and nuclear input data. Since the atomic data can be used for several isotopes, the structure of the input data was changed in order to separate atomic and nuclear data. The atomic data were generated with the same procedures as discussed for $^{109}$Cd and $^{125}$I by Kossert and Grau Carles (2008). The data for K fluorescence yields were taken from the DDEP database (Bé et al., 2004). For nuclear decay data, such as the fractional electron-capture probabilities, as well as data for the β and γ transitions, we also used that database.

### 3.2. Experimental details

The aim of the measurements was to compare efficiency curves, i.e. the counting efficiency of the nuclide under study as a function of the counting efficiency of $^3$H. The computed efficiency curve can be obtained by linking them via the free parameter. The experimental efficiency curve can be obtained in a similar way using the quenching indicators *SQP*(*E*) and *tSIE* to link the data.

The samples were prepared with 15 mL Ultima Gold™ scintillator and 1 mL of distilled water in 20 mL low-potassium borosilicate glass vials. Each sample series comprised a background sample without an active solution. Nitromethane ($CH_3NO_2$) was used as a quenching agent. The samples were measured in a Wallac 1414 Guardian™ liquid scintillation spectrometer. The guard detector was switched off by disconnecting its high voltage supply. Additional measurements were carried out with a TriCarb® 2800 TR.

To determine the calibration curve—i.e. the counting efficiency of $^3$H as a function of the respective quenching indicator—amounts of a standard solution of $^3$H were used. The activity of the $^3$H standard solution was determined by internal gas counting and was verified by other national metrology institutes within the scope of EUROMET and ICRM comparisons (Makepeace et al., 1994).

In the following, the calculated results are compared in the residual plots (Figs. 1–8). In these plots the abscissa corresponds to the curves that were obtained by fitting the experimental data (see also Kossert and Grau Carles, 2006). It should be noted that the experimental data are sometimes based on only a few data points. This can cause problems for the fitting procedure, and thus the data in the residual plots may obtain strange structures

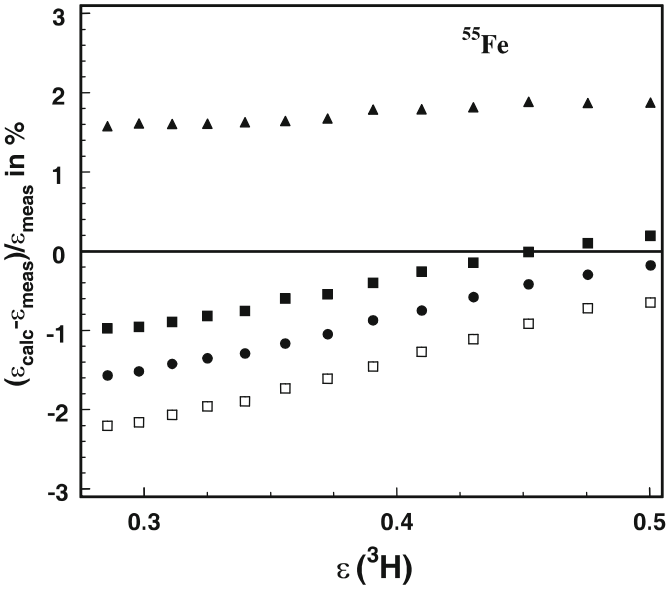


**Fig. 1.** Residual plot for the calculated counting efficiency of $^{55}$Fe vs. the $^3$H counting efficiency. The abscissa represents the counting efficiencies, which were obtained by fitting the experimental data. All calculations were conducted with $kB$=0.0075 cm/MeV. The calculations were performed without any correction (filled triangles), with the pc (filled squares), pc and Larkins correction (filled circles) and pc and micelle effect (open squares).

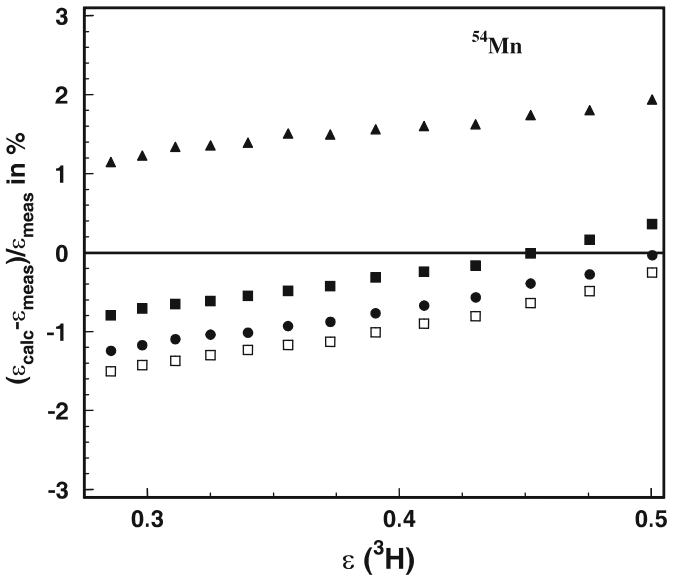


**Fig. 2.** Same as Fig. 1, but for $^{54}$Mn.

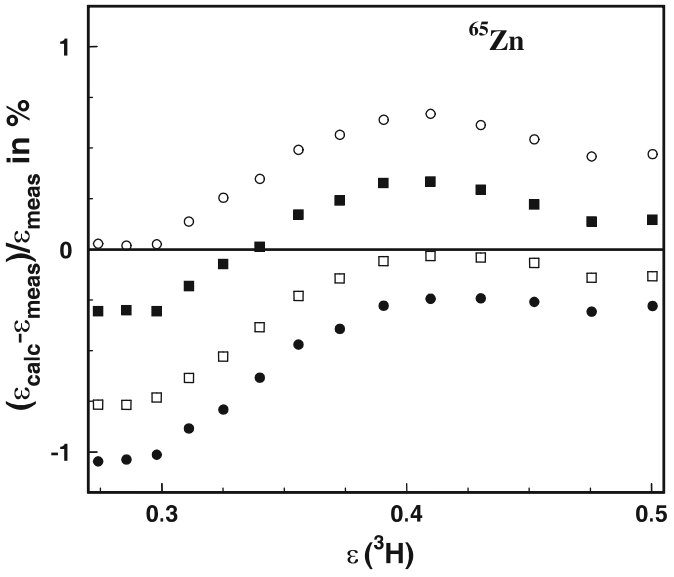


**Fig. 3.** Residual plot for the calculated counting efficiency of $^{65}$Zn vs. the $^3$H counting efficiency. All calculations were conducted with the pc and $kB$=0.0075 cm/MeV. The calculations were performed without any further correction (filled squares), the micelle effect (open squares) and a combination of the micelle effect and the Larkins correction (filled circles). The open circles were obtained without further corrections (as for filled squares) but with $\omega_K$=0.4411.

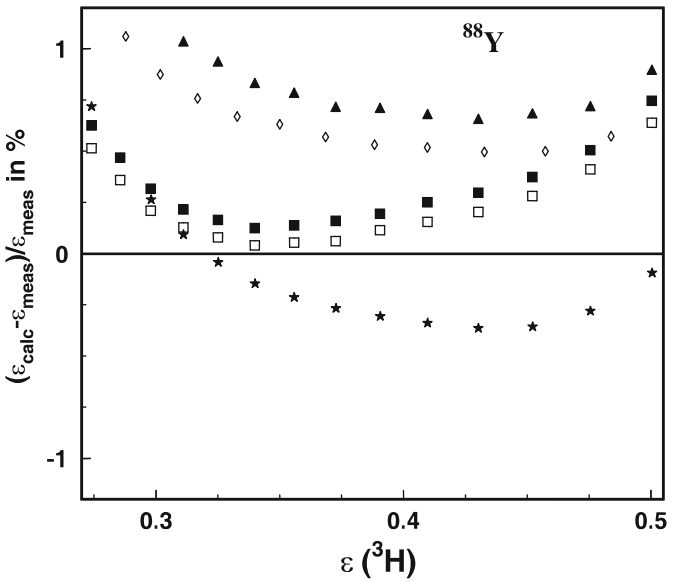


**Fig. 4.** Residual plot for the calculated counting efficiency of $^{88}$Y vs. the $^3$H counting efficiency. The calculations were done with $kB$=0.0075 cm/MeV and without any correction (filled triangles), with the pc (filled squares) and pc and micelle effect (open squares). When only the pc is applied and $P_K$ and $P_L$ as computed with *logft* the stars are obtained. The open diamonds were obtained applying only the pc (as for filled squares) but with $kB$=0.0110 cm/MeV.

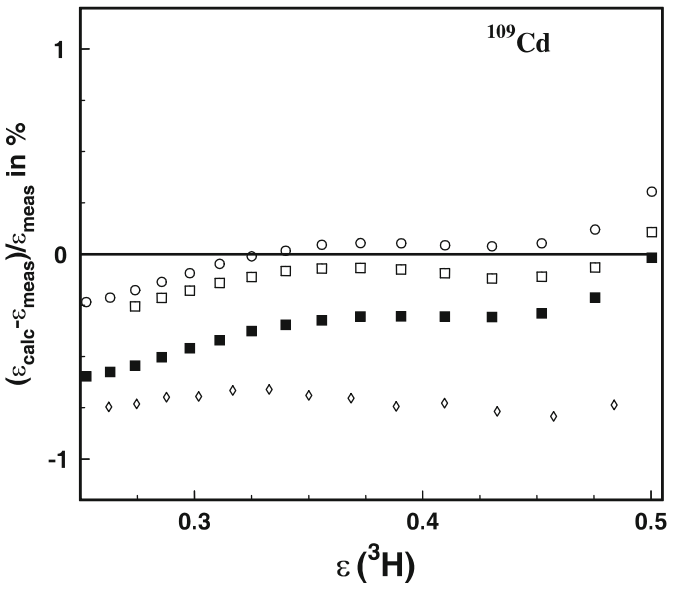


**Fig. 5.** Residual plot for the calculated counting efficiency of $^{109}$Cd vs. the $^3$H counting efficiency. The calculations were done with the pc and $kB$=0.0075 cm/MeV (filled squares) or $kB$=0.0110 cm/MeV (open diamonds). The open circles were calculated with $kB$=0.0075 cm/MeV and the pc but with $\omega_K$=0.8219. The open squares were obtained with these same conditions but applying the micelle effect.

(e.g. waves) that are not necessarily due to the theoretical computation of the counting efficiencies.

If the micelle correction is used, a spherical micelle with a radius of 4 nm and a randomly distributed initial position of the electrons are assumed. The choice of 4 nm for the micelle radius is based on reports from other authors, e.g. a mean value of 4.4 nm is obtained from the size range reported by Rodríguez et al. (1998). With this assumption the micelle effect for $^3$H is about 0.1%. However, the micelle effect for the tracer has not been taken into account in this analysis. The maximum β energy for $^3$H (tritiated water) was assumed to be 18.58(2) keV, as in previous studies.

Since some of the radionuclides in this study also have $\beta^-$ or $\beta^+$ branches, we first checked the computation of β emitters. The counting efficiencies of $^3$H computed with the stochastic approach agree well with the analytical approach, provided that a large number of events are simulated. A difference of $< 0.1\%$ was found when $4 \times 10^5$ events were simulated. As expected, the agreement between the two approaches is even better for high-energy β emitters. For $^{89}$Sr the difference between the stochastic approach and established analytical calculations are negligible, provided that the same parameters for the ionization quenching are used.

### 3.3. $^{54}$Mn and $^{55}$Fe

In a previous study, a reasonable agreement of the analytically determined counting efficiencies of $^{54}$Mn and $^{55}$Fe with

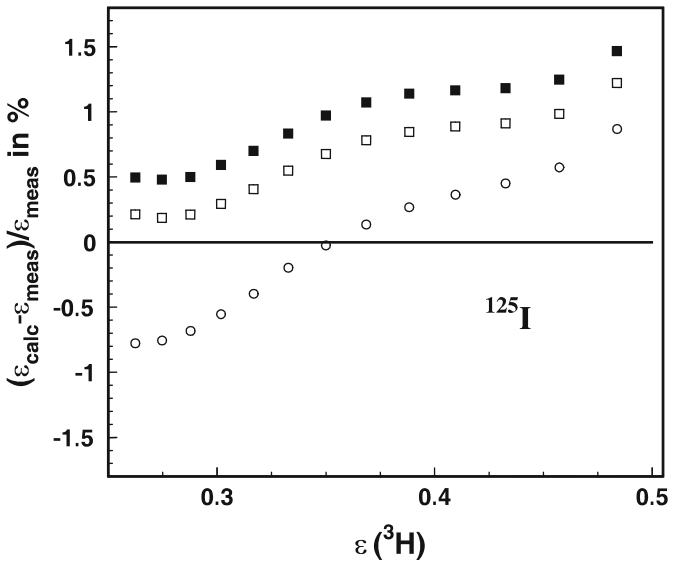


**Fig. 6.** Residual plot for the calculated counting efficiency of $^{125}$I vs. the $^3$H counting efficiency. The calculations were conducted with $kB$=0.0110 cm/MeV and the pc (filled squares) or the pc and the micelle effect (open squares). The open circles were obtained with the pc, the micelle effect and $\omega_K$=0.883.

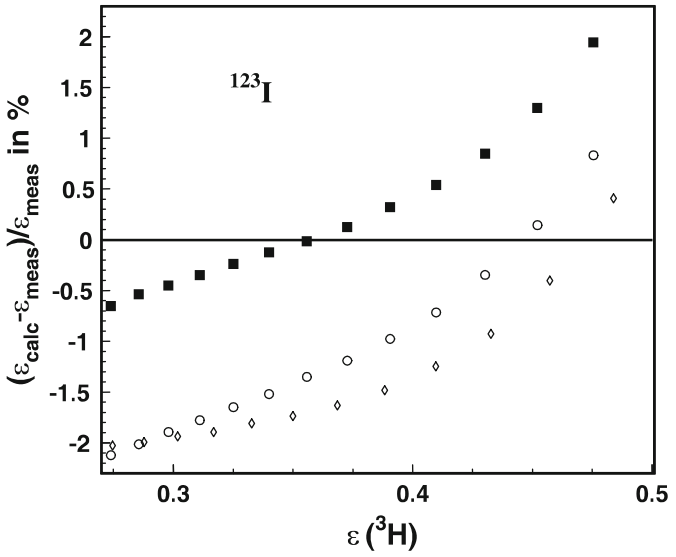


**Fig. 7.** Residual plot for the calculated counting efficiency of $^{123}$I vs. the $^3$H counting efficiency. The calculations were conducted with $kB$=0.0075 cm/MeV and the pc, with $\omega_K$=0.875 (filled squares) or $\omega_K$=0.883 (open circles). The open diamonds were obtained with $\omega_K$=0.875 and $kB$=0.0110 cm/MeV.

experimental data was found when the KL1L2L3M atomic rearrangement model, in combination with the photoelectric correction and an ionization quenching constant of $kB$=0.0075 cm/MeV, were used (Kossert and Grau Carles, 2006).

Some results of the calculations of $^{55}$Fe with MICELLE2 are shown in Fig. 1. All calculations were performed with $kB$=0.0075 cm/MeV. The effect of the pc is about 2% and, thus, similar to that of in the EMILIA code (Kossert and Grau Carles, 2006). The filled triangles, which were obtained without the pc, clearly overestimate the counting efficiency. A reasonable agreement is found with the pc (filled squares). The Larkins correction (filled circles) reduces the counting efficiencies by about half a percent. An even larger reduction is found for the micelle effect (open squares). This is mainly due to low-energy L-Auger electrons which lose a considerable part of their energy within the micelles.

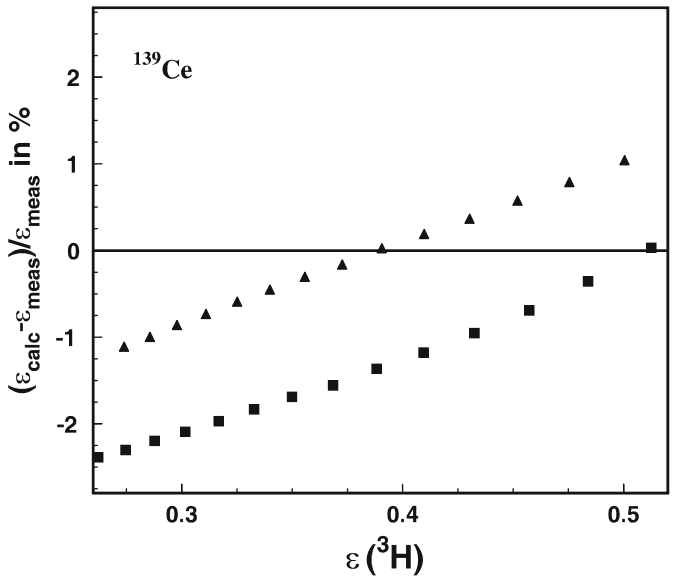


**Fig. 8.** Residual plot for the calculated counting efficiency of $^{139}$Ce vs. the $^3$H counting efficiency. The calculations were done with the pc and $kB$=0.0075 cm/MeV (filled triangles) and $kB$=0.0110 cm/MeV (filled squares).

Similar findings are obtained for $^{54}$Mn (Fig. 2). Again the best agreement is obtained with the pc, but both the Larkins correction and the micelle effect reduce the computed efficiencies.

For both radionuclides, a reduction of the $kB$ value increases the computed residual counting efficiency, whereas a higher $kB$ value causes an opposite effect. The large model dependence can be considerably reduced when $^{55}$Fe is measured with $^{54}$Mn as the tracer nuclide (Günther, 1998; Kossert and Grau Carles, 2006).

### 3.4. $^{65}$Zn and $^{88}$Y

The radionuclide $^{65}$Zn has already been tested with EMILIA applying the KL1L2L3M atomic rearrangement model in combination with the pc. The efficiencies with that approach were overestimated by about 1%.

Fig. 3 shows the residual plot with some results obtained with the new MICELLE2 programme. All calculations are done with $kB$=0.0075 cm/MeV. A larger $kB$ value reduces the results and causes a trend in the data. The pc is—as also for the other low-Z electron-capture nuclides—again very important and was used for all calculations. A good agreement is obtained when no further correction is applied (filled squares), however, the micelle correction (open squares) reduces the results considerably. The influence of the Larkins correction is lower; a combination of the micelle correction and the Larkins correction is shown by the filled circles.

The open circles are obtained with the pc but without any further correction (as for filled squares) but with $\omega_K$=0.4411 (Hubbell et al., 1994).

A new study was made with $^{88}$Y (Fig. 4). The dependence on the $kB$ parameter is lower than for low-Z electron-capture nuclides, but an increase in the $kB$ value now increases the calculated counting efficiency. The fractional electron-capture probabilities for the branch to the 1836 keV level of $^{88}$Sr were calculated according to the procedures from Bambynek et al. (1977) (see also Browne and Firestone, 1986). The resulting $P_K$=0.8745 is in contradiction to the much lower value $P_K$=0.65 from Schönfeld (Bé et al., 2004). With the latter value, the overall counting efficiency is reduced by about 1.3%. A good agreement is found with the capture probabilities computed with the *logft*

programme, i.e. $P_K$=0.8726, $P_L$=0.1046 (NNDC, 2001). Also, the dependence on the fluorescence yield is large. Using $\omega_K$=0.690 (Hubbell et al., 1994) instead of 0.696 increases the results by about 0.5%.

### 3.5. $^{109}$Cd, $^{125}$I and $^{123}$I

The two radionuclides $^{109}$Cd and $^{125}$I were already studied with the first version of the MICELLE programme (Kossert and Grau Carles, 2008). For both nuclides the best agreement was found with $kB$=0.0110 cm/MeV. The new MICELLE2 code takes more transitions into account, and thus a change in the results is expected. Indeed, the counting efficiencies are somewhat lower. For $^{109}$Cd a better agreement with the experimental data is now found with $kb$=0.0075 cm/MeV (Fig. 5). The agreement is also better when the fluorescence yield $\omega_K$=0.8219 from Hubbell et al. (1994) is used.

For $^{125}$I (Fig. 6), the agreement is somewhat better than the previous version. Again it must be emphasized that the results also depend on the input data, for example the agreement with the experimental data is much improved when the fluorescence yield $\omega_K$=0.883 from Hubbell et al. (1994) is used.

A residual plot for $^{123}$I is shown in Fig. 7. The agreement with experimental data is better when $kB$=0.0075 cm/MeV is used. In contrast to $^{125}$I the agreement is better when the fluorescence yield $\omega_K$=0.875 is used. The trend in the plot indicates that the model and/or the input data are not accurate enough.

For this group of radionuclides, the Larkins correction as well as the pc are of minor importance, whereas the micelle effect is in the order of 0.1% for $^{109}$Cd and about 0.2% for the iodine isotopes.

### 3.6. $^{64}$Cu

The MICELLE2 code was also used to compute the counting efficiencies of $^{64}$Cu which comprises a $\beta^-$, a $\beta^+$ and two electron-capture branches. The agreement with an analytical approach was better than 0.05% and the LSC result for the activity was in excellent agreement with the outcome from 4πβ(PC)-γ coincidence counting. More details about the standardization of $^{64}$Cu are presented elsewhere in these proceedings (Wanke et al., 2009).

### 3.7. $^{139}$Ce

The results for $^{139}$Ce are shown in Fig. 8. For $kB$=0.0075 cm/MeV the agreement with experimental data is satisfactory, but a trend in the data is a hint at a deficient model and/or a problem with the input data. The trend remains when other $kB$ values are used.

### 3.8. TDCR

The new programme was also used to compute the counting efficiencies for TDCR applications. For pure beta emitters such as $^{89}$Sr the Monte Carlo results were in good agreement with an analytical approach.

For $^{55}$Fe, calculations were made to check the importance of the pc. In contrast to the CIEMAT/NIST calculations, the pc is of minor importance for TDCR, since it causes a reduction in the counting efficiencies $\varepsilon_D$ and $\varepsilon_T$. As a consequence almost the same efficiency value would be extracted with and without pc for a given TDCR value. However, a test for other nuclides is still outstanding. In addition, a comparison of the new atomic rearrangement with the KLM model would be interesting.

## 4. Conclusion and outlook

*Atomic rearrangement model*: The new, more detailed consideration of the atomic rearrangement process has a large influence on the computation results. For most electron-capture nuclides the deviations from experimental data are $<2\%$. This is a considerable improvement compared to differences of up to 8% for the KLM model. The analysis also shows that even the KL1L2L3M atomic rearrangement is not accurate enough when more atomic shells are involved.

The results for $^{139}$Ce show the limits of the model for higher atomic numbers. However, the MICELLE2 code is the first CIEMAT/NIST programme that yields reasonable results for electron-capture nuclides with atomic numbers of up to about 58.

*Larkins correction*: The Larkins correction was used for previous studies, applying the less sophisticated models. In this work, it is shown that this correction is not required. It even worsens the results for low-Z electron-capture nuclides. For higher atomic numbers, the Larkins correction has a negligible influence.

*Photoelectric correction*: The importance of the pc for low-Z electron-capture nuclides was confirmed for CIEMAT/NIST calculations. For higher atomic numbers the correction has a lower impact. For $^{55}$Fe the pc is almost negligible when the TDCR method is used.

*Micelle effect*: The micelle effect also has a high influence on the results of low-Z electron-capture nuclides and apparently impairs the results when accounted for. The effect requires more research work. A promising approach for such a study could be to adapt and extend the model for plastic scintillation beads. If such plastic spheres are embedded in an aqueous radioactive solution, the situation would be effectively the inverse to the micelles in an organic system.

*Ionization quenching*: It is important to note that the counting efficiency of low-energy β emitters and electron-capture nuclides often strongly depends on the ionization quenching function. A general problem is the poor knowledge of the electron stopping powers below 1 keV and the large dependence on the choice of the parameter $kB$. The fact that the choice of the $kB$ constant depends on the nuclide indicates a problem with the parameterization used. The authors are aware of these difficulties and more research is required in this field. A promising approach is the experimental determination of the nonlinear response function of the scintillator by means of a TDCR-Compton spectrometer (e.g. Cassette and Do, 2008). However, it is not yet known whether the ionization quenching mechanisms are also valid for coincident multi-electron emissions as in the case of electron-capture nuclides (Grau Malonda and Grau Carles, 2008).

All in all, the analysis of this work clearly indicates the advantages of the stochastic approach and the improvements due to a better and more detailed atomic rearrangement model.

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## Discussion:

Due to technical problems (unclear records) the transcription can not be published.