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Study of a Monte Carlo rearrangement model for the activity determination of electron-capture nuclides by means of liquid scintillation counting

K. Kossert[a,*], A. Grau Carles[b]

[a]Department 6.1, Physikalisch-Technische Bundesanstalt, Bundesallee 100, D-38116 Braunschweig, Germany
[b]Instituto de Matemáticas y Física Fundamental (CSIC), Edif. 113b, Dcho. 211, C/Serrano 113b, 28006 Madrid, Spain

Abstract

A new Monte Carlo approach for the computation of the electron spectra of electron-capture nuclides is applied to obtain efficiencies in liquid scintillation counting for CIEMAT/NIST applications. The new method is applied to the radionuclides ^{109}Cd and ^{125}I by using a stochastic atomic rearrangement model, taking into account rearrangement processes including L-, M-, and N-subshells. The counting efficiencies were computed with the new code MICELLE which also comprises an approach for calculating the counting efficiency of a radionuclide in a gel phase sample. The calculated counting efficiencies are compared with experimental results.

Keywords: Liquid scintillation counting; CIEMAT/NIST method; Monte Carlo atomic rearrangement model; ^{109}Cd; ^{125}I

1. Introduction

In the past two decades many attempts have been made to extend the CIEMAT/NIST efficiency tracing method (Coursey et al., 1986; Grau Malonda, 1999) for the activity standardization of electron-capture (ec) nuclides. The method, which gives excellent results for beta emitters, requires the computation of electron spectra created in a liquid scintillation sample. For ec nuclides, the previous models compute the spectra applying simplified atomic rearrangement models, but comparison of the calculated efficiencies with experimental data revealed considerable discrepancies. In a recent attempt it has been shown that the efficiencies for low-Z ec nuclides computed applying the $KL_1L_2L_3M$ rearrangement model are in good agreement with experimental data, provided that the ejection of photoelectrons and subsequent rearrangement processes are taken into account in an adequate way (Grau Carles, 2006; Grau Carles et al., 2006; Kossert and Grau Carles, 2006).

Despite the improvements for low-Z ec nuclides, there are still large discrepancies of up to 8% between experimental data and the calculated counting efficiencies of ec nuclides with $Z>30$ when simplified rearrangement models are used (see, e.g., Günther, 2002; Pommé et al., 2005).

In this paper, we describe a new approach applying a stochastic method for the computation of the electron spectra. In this method almost all possible rearrangement processes including L-, M-, and N-subshells are simulated using Monte Carlo procedures. The model, which is utilized in the computer code MICELLE (Grau Carles, 2007) for ec, ec/gamma, and isomeric transitions with $30\leqslant Z\leqslant 54$ is applied here to the radionuclides ^{109}Cd and ^{125}I. The calculated counting efficiencies are compared with the experimental data obtained by liquid scintillation counting. For the measurements we used standard solutions, which were calibrated by other methods.

2. The stochastic rearrangement model

All previous approaches to computing the counting efficiency of ec nuclides according to the CIEMAT/NIST method are based on atomic rearrangement models which simplify the processes following ec events. These models as

*Corresponding author.
E-mail address: karsten.kossert@ptb.de (K. Kossert).

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well as their benefits and drawbacks have been discussed in other articles (e.g., Grau Carles, 2006; Kossert and Grau Carles, 2006). The simplification comprises the use of averaged energies and probabilities. Since the light produced by a charged particle interacting within a liquid scintillation sample is a non-linear function of the particle energy, E, such simplifications lead to discrepancies in experimental data. However, for low-Z ec nuclides, the $KL_1L_2L_3M$ atomic rearrangement model gives satisfactory results, provided that the interaction of X-rays via the photoelectric effect is taken into account in an adequate way (Grau Carles, 2006; Grau Carles et al., 2006; Kossert and Grau Carles, 2006).

For larger atomic numbers, i.e. $Z \geqslant 30$, the models fail and thus a more realistic model is required. In this work the program MICELLE, which applies a stochastic model, is used. This model is briefly described in the following. Further details can be found elsewhere (Grau Carles, 2007).

2.1. The stochastic atomic rearrangement model

The first step of the simulation of an ec decay comprises the computation of a 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 ec probabilities P_K, P_{L1}, P_{L2}, and P_{M+}, respectively. In the case of a K-capture, the next random number is used to decide which process follows. It might either be an X-ray emission, which transfers the vacancy to a higher shell, or an Auger electron that is ejected, in which case two new vacancies are created in the higher shell. The corresponding probabilities are the fluorescence yield ω_K and the Auger yield, $A_K = 1-\omega_K$. The model takes into account six types of K X-rays and 49 types of K-Auger electrons. In the case of L_1 and L_2 vacancies, the model distinguishes between Coster-Kronig electron emission, Auger electron emission and X-ray emission. The latter process again transfers the vacancy to a higher shell, whereas the electron ejection creates two new vacancies. L_3-subshell vacancies are treated in a similar way, but there are no Coster-Kronig transitions. The model considers 10 L_1L_2X, 10 L_1L_3X, and four L_2L_3X Coster-Kronig transitions as well as 40 L_iXY ($i = 1,2,3$; $X \geqslant M_1$) Auger transitions. In the case of M_i and N_i vacancies, only Auger and Coster-Kronig processes are simulated.

The primary vacancies can also be created due to an internal conversion process. In this case, the ejected conversion electron contributes to the counting efficiency. The simulation can be done in a way similar to that of ec processes, but instead of the fractional ec probabilities the probabilities for internal conversion are used. In the case of internal conversion the model also allows for primary vacancies due to the ejection of L_3 electrons.

The interaction of photons is simulated in the same way as in the EMILIA code (Grau Carles, 2006), but without the simulation of the rearrangement processes following the ejection of a photoelectron. Also, the binding energy of the photoelectron is not subtracted.

2.2. Ionization quenching

In this work, the ionization quenching function $Q(E)$ was computed by numerical integration of Birks's formula:

$$Q = \frac{1}{E}\int_0^E \frac{\mathrm{d}E}{1 + kB \cdot \mathrm{d}E/\mathrm{d}x}, \tag{1}$$

where kB is a constant and $\mathrm{d}E/\mathrm{d}X$ are the electron stopping powers calculated according to Rohrlich and Carlson (ICRU, 1984). The calculation method is part of the MICELLE code (option 4) and takes the atomic composition of the cocktail into account.

2.3. The micelle size effect

The new model also comprises the consideration of micelles in a 2-phase system with an organic scintillator and an aqueous sample. An energy deposit within a micelle does not produce any light and thus does not contribute to the counting efficiency. The micelles are assumed to be spherical volumes and the initial position of an emitted particle is randomly distributed within this volume. In gel samples, the micelle size as well as the reduction of the counting efficiency is quite large (Grau Carles, 2007). For the samples used in this work the micelle size effect is of minor importance.

3. Atomic and nuclear input data

3.1. ^{109}Cd

The radionuclide ^{109}Cd decays via ec to the excited level ^{109m}Ag which decays with a half-life of about 39.6 s and a transition energy of 88.0337(1) keV mainly by emission of conversion electrons (TdeR, 2006). The 88 keV gamma rays with an emission probability $P_\gamma = 0.03626(26)$ provide an important and frequently used calibration point in the low-energy region of gamma-ray spectrometers.

Fig. 1 shows the input file containing all atomic and nuclear data required for the calculation with MICELLE.

The $5/2^+ \rightarrow 7/2^+$ ec transition is of allowed nature and thus the fractional capture probabilities P_K, P_L, and P_{M+} were calculated by using the program EC-capture (Schönfeld, 1998). The fractional capture probabilities for L-subshells are estimated applying

$$P_{L1} = \frac{w_1 \cdot P_L}{w_1 + w_2}, \quad P_{L2} = \frac{w_2 \cdot P_L}{w_1 + w_2} \quad \text{and} \quad P_{L3} = 0 \tag{2}$$

with the weights, w_i ($i = 1,2$)

$$w_1 = q_{L1}^2 \beta_{L1}^2 B_{L1} \quad \text{and} \quad w_2 = q_{L2}^2 \beta_{L2}^2 B_{L2} \tag{3}$$

```
                    'Cd-109'

''                ATOMIC DATA

'Decay scheme (1-10)        :'     4
'Atomic number (daughter)   :'     47
'WK,WL1,WL2,WL3             :'     0.831, 0.016, 0.051, 0.052
'F12,F13,F23                :'     0.100, 0.590, 0.153

'PKL1L1,L2,L3,M1,M2,M3,M4,M5:'     0.0742,0.0861,0.1188,0.0251,0.0154,0.0213,0.0014,0.0015
'PKL1N1,N2,N4,O1,O2         :'     0.0048,0.0027,0.0037,0.0002,0.0000
'PKL2L2,L3,M1,M2,M3,M4,M5   :'     0.0123,0.2665,0.0122,0.0040,0.0399,0.0017,0.0057
'PKL2N1,N2,N4,O1,O2         :'     0.0023,0.0007,0.0073,0.0001,0.0000
'PKL3L3,M1,M2,M3,M4,M5      :'     0.1385,0.0167,0.0395,0.0425,0.0064,0.0065
'PKL3N1,N2,N4,O1,O2         :'     0.0031,0.0137,0.0015,0.0001,0.0000
'PKM1M1,M2,M3,N1,N2         :'     0.0021,0.0022,0.0030,0.0008,0.0009
'PKM2M3,N1,N2               :'     0.0060,0.0004,0.0010
'PKM3M3,M4,M5,N1,N2         :'     0.0033,0.0008,0.0008,0.0006,0.0021

'PL1L2M1,M2,M3,M4,M5        :'     0.0000,0.0000,0.0000,0.0000,0.0000
'PL1L2N1,N2,N4,O1,O2        :'     0.3215,0.4865,0.1787,0.0133,0.0000
'PL1L3M1,M2,M3,M4,M5        :'     0.0000,0.0000,0.0000,0.3205,0.5204
'PL1L3N1,N2,N4,O1,O2        :'     0.0439,0.0552,0.0583,0.0018,0.0000

'PL1M1M1,M2,M3,M4,M5        :'     0.0333,0.0657,0.1237,0.0964,0.1396
'PL1M1N1,N2,N4,O1,O2        :'     0.0109,0.0316,0.0239,0.0005,0.0000
'PL1M2M3,M4,M5,N1,N4,O1     :'     0.0028,0.0024,0.0346,0.0088,0.0030,0.0300
'PL1M3M3,M4,M5,N1,N2,N4,O1  :'     0.0000,0.0000,0.0191,0.0165,0.0008,0.0017,0.0007
'PL1M4M4,M5,N1,N2,N4,O1     :'     0.0051,0.1976,0.0124,0.0049,0.0164,0.0005
'PL1M5M5,N1,N2,N4,O1,O2     :'     0.0556,0.0179,0.0085,0.0240,0.0007,0.0000
'PL1N1N1,N2,N4,N2N4,O1,N4N4 :'     0.0009,0.0042,0.0031,0.0004,0.0001,0.0042

'PL2L3N1,N2,N4,O1,O2        :'     0.0907,0.4457,0.4601,0.0035,0.0000
'PL2M1M1,M2,M3,M4,M5,N2,O2  :'     0.0016,0.0362,0.0033,0.0025,0.0052,0.0048,0.0000
'PL2M2M2,M3,M4,M5           :'     0.0349,0.1124,0.0603,0.1059
'PL2M2N1,N2,N4,O1,O2        :'     0.0070,0.0286,0.0168,0.0000,0.0000
'PL2M3M3,M4,M5,N1,N2,N4,O2  :'     0.0030,0.0724,0.0098,0.0006,0.0148,0.0073,0.0000
'PL2M4M4,M5,N1,N2,N4,O2     :'     0.0667,0.2886,0.0005,0.0188,0.0388,0.0000
'PL2M5M5,N2,N4,O2           :'     0.0137,0.0132,0.0248,0.0000
'PL2N1N2,N2N2,N4,N4N4       :'     0.0009,0.0030,0.0020,0.0016

'PL3M1M1,M2,M3,M4,M5,N2     :'     0.0016,0.0012,0.0347,0.0034,0.0038,0.0041
'PL3M2M3,M4,M5,N2,N4        :'     0.0577,0.0039,0.0347,0.0071,0.0030
'PL3M3M3,M4,M5,N1           :'     0.0820,0.0814,0.1239,0.0067
'PL3M3N2,N4,O1,O2           :'     0.0093,0.0315,0.0120,0.0000
'PL3M4M4,M5,N1,N2,N4        :'     0.0117,0.2252,0.0006,0.0101,0.0196
'PL3M5M5,N1,N2,N4,O2        :'     0.1534,0.0007,0.0212,0.0465,0.0000
'PL3N1N2,N2N2,N4,N4N4       :'     0.0008,0.0012,0.0046,0.0028

'PM1M2N4,O1,O2,M3N2,N4,O1,O2:'     0.3809,0.0000,0.0000,0.4172,0.0947,0.0038,0.0000
'PM1M4N1,N4,M5N1,N2,N4,N1N4 :'     0.0000,0.0622,0.0413,0.0000,0.0000,0.0000

'PM2M3O1,O2,M4N1,N2,N4,O1   :'     0.0743,0.0000,0.1127,0.4029,0.0583,0.0050
'PM2M4O2,M5N1,N2,O2,N2N2,N4 :'     0.0000,0.0267,0.2344,0.0008,0.0418,0.0430

'PM3M4N1,N2,N4,M5N1,N2,N4   :'     0.0358,0.1851,0.0193,0.1539,0.1065,0.3962
'PM3M5O1,O2,N2N2,N4         :'     0.0062,0.0000,0.0238,0.0734
'PM4N1N1,N2,N4,N2N2         :'     0.0004,0.0197,0.1487,0.0011
'PM4N2N4,N4N4,O1,O2         :'     0.3827,0.4474,0.0000,0.0000

'PM5N1N4,N2N2,N4,N4N4,O1,O2 :'     0.1692,0.0185,0.3645,0.4478,0.0000,0.0000

'PN1N2O1,O2,N4O1,O2         :'     0.0000,0.0000,0.0000,0.0000
'PN2N4O1,O2,PN4O2O2         :'     0.0000,0.0000,0.0000

'PKL2,L3,M2,M3,M4,M5        :'     0.2910,0.5483,0.0469,0.0904,0.0007,0.0227
'PL1M2,M3,M4,M5             :'     0.3267,0.5073,0.0000,0.1660
'PL2M1,PL2M2,PL2M3,PL2M4,M5 :'     0.0354,0.0000,0.0000,0.9646,0.0000
'PL3M1,PL3M2,PL3M3,PL3M4,M5 :'     0.0314,0.0000,0.0000,0.0870,0.8816

'EK,EL1,EL2,EL3             :'      25514.0,  3805.8,  3523.7,  3351.1
'EM1,EM2,EM3,EM4,EM5        :'        717.5,   602.4,   571.4,   372.8,   366.7
'EN1,EN2,EN4,EO1,EO2        :'         95.2,    62.6,     3.3,     0.0,     0.0



''                NUCLEAR DATA
'PK,PL1,PL2,PM+             :'     0.8118,0.1467,0.0030,0.0385
'PGAM,EGAM (1)              :'     0.03626,88.034
'PIK,PIL1,PIL2,PIL3,PIM+(1) :'     0.4238,0.0336,0.1990,0.2314,0.1120
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Fig. 1. Input file with ^{109}Cd (Ag) nuclear and atomic data for the efficiency computation with MICELLE.

and $q_i = \Delta E - E_i$ the difference between the transition energy $\Delta E = Q^+ - E_f$ and the binding energies E_i (Browne and Firestone, 1986) of the captured electron in the daughter atom ($Z = 47$). The values $Q^+ = 213.8(27)$ keV and $E_f = 88.0337(1)$ keV were taken from TdeR (2006). The squared amplitudes of the bound-state electron radial

wavefunctions ($\beta_i^2 B_i p_i^{2(k_i-1)}$) in Eq. (3) were taken from Browne and Firestone (1986), where $k_i = 1$ for L_1 and L_2 atomic shells.

Finally P_{M+} is given by

$$P_{M+} = 1 - P_K - P_L. \quad (4)$$

The fluorescence yield ω_K was taken from TdeR (2006), the L-subshell fluorescence yields ω_{Li} and Coster-Kronig yields f_{12}, f_{13} and f_{23} were taken from Browne and Firestone (1986).

The relative probabilities of K- and L- shell Auger and Coster-Kronig transitions were calculated using tabulated values from Chen et al. (1979). The relative probabilities of M-shell Auger and Coster-Kronig transitions were computed using the values from McGuire (1972).

The X-ray emission probabilities were taken from Browne and Firestone (1986). It is to be noted that all probabilities were normalized in subgroups, i.e. $\sum P_{KXY} = 1$, etc.

The energies of Auger electrons, Coster-Kronig electrons, and X-rays are directly calculated within the MICELLE code. For this calculation the electron binding energies, which were taken from Browne and Firestone (1986), are required.

3.2. ^{125}I

^{125}I disintegrates by ec via the excited 35.5 keV level of ^{125}Te to the ground state of ^{125}Te. The energy and emission probability of the γ-rays are $E_\gamma = 35.4919(5)$ keV and $P_\gamma = 0.0667(17)$, respectively (TdeR, 2006). The $5/2^+ \rightarrow 3/2^+$ ec transition is also of allowed nature, and consequently the fractional capture probabilities were obtained applying the same procedure as for ^{109}Cd.

The fluorescence yields, energies, and emission probabilities of X-rays as well as Auger and Coster-Kronig electrons were determined in the same way as for ^{109}Cd, too. The relative probabilities of M-shell Auger and Coster-Kronig transitions were determined by linear interpolation between the values for $Z = 50$ and 54 tabulated by McGuire (1972).

The input file containing all atomic and nuclear data required for the calculation of ^{125}I with the MICELLE code is shown in Fig. 2.

4. Experimental

The aim of the measurements was to determine the counting efficiency of the nuclide under study as a function of the quenching indicators *SQP*(*E*) and *tSIE*. 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 sample without active solution to measure the background counting rate, which was then subtracted. Nitromethane (CH_3NO_2) was used as a quenching agent. All samples were shaken and centrifuged and then measured in a Wallac 1414 Guardian™ liquid scintillation spectrometer with two photomultiplier tubes (PMTs). The coincidence resolving time of this counter was determined to be 24.9(10) ns. The quenching indicator *SQP*(*E*) was measured by means of an external source of ^{152}Eu. Additional measurements were carried out with a TriCarb® 2800 TR purchased at the end of 2006. In this counter the quenching indicator *tSIE* was measured by means of an external source of ^{133}Ba. The coincidence resolving time of this counter was adjusted to 25 ns.

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 with a relative standard uncertainty of $u(a)/a = 0.7\%$ by internal gas counting and was verified by other national metrology institutes within the scope of EUROMET and ICRM comparisons (Makepeace et al., 1994).

The ^{109}Cd solution used for the measurements had been standardized with a relative standard uncertainty of 0.27% by a combination of various methods without using emission probabilities (Kossert et al., 2006). The emission rate of conversion electrons had been measured by means of pressurized proportional counters as well as liquid scintillation spectrometry. The latter method is an easily applicable alternative method to the techniques based on free parameter models. In addition, the photon emission rate was measured with the aid of γ-ray spectrometers. An ampoule containing this calibrated ^{109}Cd solution was sent to the *Bureau International des Poids et Mesures* (BIPM) to include the activity result in the database of the international reference system (SIR). The result for the activity concentration is in good agreement with the key comparison reference value (Ratel et al., 2005).

The ^{125}I measurements were performed with several solutions that had been standardized by photon–photon coincidence counting using NaI detectors (Schrader and Walz, 1987; Schrader, 2006) and photon sum peak counting (Eldrige and Crowther, 1964) using an Si(Li) detector. One of these solutions was standardized with a relative standard uncertainty of 0.42% within the scope of an international comparison of activity measurements of an ^{125}I solution organized by the BIPM in 2004.

No photon-emitting impurity was detected by γ-ray spectrometry in the ^{109}Cd and ^{125}I solutions used for this study. For quality assurance, the solution activities had also been measured by means of secondary standardization using 4π ionization chambers and a new technique using liquid scintillation counting as proposed by Kossert (2006).

5. Results and discussion

The electron spectra of ^{109}Cd and ^{125}I computed by using the MICELLE code and the input data described above are shown in Figs. 3 and 4.

Using the same procedures as described in a previous article (Kossert and Grau Carles, 2006), we obtain the

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                    'I-125'

''                ATOMIC DATA

'Decay scheme (1-10)        :'     3
'Atomic number (daughter)   :'     52
'WK,WL1,WL2,WL3             :'     0.875, 0.041, 0.074, 0.074
'F12,F13,F23                :'     0.180, 0.280, 0.155

'PKL1L1,L2,L3,M1,M2,M3,M4,M5:'     0.0785,0.0936,0.1167,0.0277,0.0176,0.0222,0.0015,0.0017
'PKL1N1,N2,N4,O1,O2         :'     0.0059,0.0035,0.0044,0.0008,0.0006
'PKL2L2,L3,M1,M2,M3,M4,M5   :'     0.0121,0.2548,0.0137,0.0041,0.0292,0.0007,0.0061
'PKL2N1,N2,N4,O1,O2         :'     0.0028,0.0008,0.0086,0.0004,0.0004
'PKL3L3,M1,M2,M3,M4,M5      :'     0.1296,0.0170,0.0397,0.0421,0.0066,0.0068
'PKL3N1,N2,N4,O1,O2         :'     0.0034,0.0156,0.0023,0.0005,0.0012
'PKM1M1,M2,M3,N1,N2         :'     0.0024,0.0026,0.0032,0.0010,0.0011
'PKM2M3,N1,N2               :'     0.0064,0.0005,0.0012
'PKM3M3,M4,M5,N1,N2         :'     0.0034,0.0009,0.0009,0.0007,0.0026

'PL1L2M1,M2,M3,M4,M5        :'     0.0000,0.0000,0.0000,0.0000,0.0000
'PL1L2N1,N2,N4,O1,O2        :'     0.2882,0.4586,0.1836,0.0379,0.0318
'PL1L3M1,M2,M3,M4,M5        :'     0.0000,0.0000,0.0000,0.0000,0.0000
'PL1L3N1,N2,N4,O1,O2        :'     0.2325,0.2973,0.4199,0.0307,0.0195

'PL1M1M1,M2,M3,M4,M5        :'     0.0315,0.0628,0.1165,0.0916,0.1317
'PL1M1N1,N2,N4,O1,O2        :'     0.0113,0.0343,0.0324,0.0015,0.0024
'PL1M2M3,M4,M5,N1,N4,O1     :'     0.0026,0.0026,0.0342,0.0091,0.0037,0.0012
'PL1M3M3,M4,M5,N1,N2,N4,O1  :'     0.0026,0.0244,0.0169,0.0168,0.0008,0.0021,0.0022
'PL1M4M4,M5,N1,N2,N4,O1     :'     0.0051,0.1883,0.0128,0.0052,0.0218,0.0017
'PL1M5M5,N1,N2,N4,O1,O2     :'     0.0531,0.0182,0.0093,0.0315,0.0024,0.0006
'PL1N1N1,N2,N4,N2N4,O1,N4N4 :'     0.0010,0.0050,0.0046,0.0006,0.0004,0.0050

'PL2L3N1,N2,N4,O1,O2        :'     0.0696,0.3901,0.4972,0.0086,0.0346
'PL2M1M1,M2,M3,M4,M5,N2,O2  :'     0.0015,0.0369,0.0034,0.0029,0.0066,0.0055,0.0005
'PL2M2M2,M3,M4,M5           :'     0.0362,0.0442,0.0621,0.1065
'PL2M2N1,N2,N4,O1,O2        :'     0.0079,0.0263,0.0345,0.0011,0.0023
'PL2M3M3,M4,M5,N1,N2,N4,O2  :'     0.0030,0.0733,0.0098,0.0007,0.0168,0.0104,0.0016
'PL2M4M4,M5,N1,N2,N4,O2     :'     0.0684,0.2922,0.0006,0.0216,0.0559,0.0015
'PL2M5M5,N2,N4,O2           :'     0.0139,0.0148,0.0347,0.0013
'PL2N1N2,N2N2,N4,N4N4       :'     0.0011,0.0039,0.0032,0.0027

'PL3M1M1,M2,M3,M4,M5,N2     :'     0.0015,0.0010,0.0323,0.0027,0.0037,0.0043
'PL3M2M3,M4,M5,N2,N4        :'     0.0540,0.0036,0.0321,0.0074,0.0038
'PL3M3M3,M4,M5,N1           :'     0.0759,0.0771,0.1164,0.0068
'PL3M3N2,N4,O1,O2           :'     0.0099,0.0355,0.0171,0.0023
'PL3M4M4,M5,N1,N2,N4        :'     0.0113,0.2161,0.0006,0.0106,0.0262
'PL3M5M5,N1,N2,N4,O2        :'     0.1470,0.0007,0.0224,0.0625,0.0014
'PL3N1N2,N2N2,N4,N4N4       :'     0.0009,0.0014,0.0064,0.0052

'PM1M2N4,O1,O2,M3N2,N4,O1,O2:'     0.2663,0.0085,0.0166,0.1394,0.3378,0.0132,0.0242
'PM1M4N1,N4,M5N1,N2,N4,N1N4 :'     0.0637,0.0589,0.0494,0.0000,0.0264,0.0000

'PM2M3O1,O2,M4N1,N2,N4,O1   :'     0.0499,0.0152,0.1102,0.1321,0.3324,0.0187
'PM2M4O2,M5N1,N2,O2,N2N2,N4 :'     0.0300,0.0276,0.1916,0.0112,0.0277,0.0533

'PM3M4N1,N2,N4,M5N1,N2,N4   :'     0.0312,0.1506,0.0239,0.1196,0.4146,0.1294
'PM3M5O1,O2,N2N2,N4         :'     0.0207,0.0386,0.0000,0.0714
'PM4N1N1,N2,N4,N2N2         :'     0.0001,0.0091,0.0985,0.0028
'PM4N2N4,N4N4,O1,O2         :'     0.3153,0.5300,0.0164,0.0279

'PM5N1N4,N2N2,N4,N4N4,O1,O2 :'     0.0924,0.0152,0.1019,0.7445,0.0171,0.0288

'PN1N2O1,O2,N4O1,O2         :'     0.0000,0.0000,0.0000,0.0000
'PN2N4O1,O2,PN4O2O2         :'     0.0000,0.0000,0.0000

'PKL2,L3,M2,M3,M4,M5        :'     0.2898,0.5355,0.0494,0.0952,0.0009,0.0293
'PL1M2,M3,M4,M5             :'     0.3053,0.5044,0.0000,0.1903
'PL2M1,PL2M2,PL2M3,PL2M4,M5 :'     0.0277,0.0000,0.0000,0.8623,0.1100
'PL3M1,PL3M2,PL3M3,PL3M4,M5 :'     0.0300,0.0000,0.0000,0.0838,0.8861

'EK,EL1,EL2,EL3             :'      31813.8,  4939.2,  4612.0,  4341.4
'EM1,EM2,EM3,EM4,EM5        :'       1006.0,   869.7,   818.7,   582.5,   572.1
'EN1,EN2,EN4,EO1,EO2        :'        168.3,   110.2,    39.8,    11.6,     2.3


 ''                NUCLEAR DATA
'PK,PL1,PL2,PM              :'     0.8007,0.1522,0.0039,0.0432
'PGAM,EGAM (1)              :'     0.0667,35.4919
'PIK,PIL1,PIL2,PIL3,PIM (1) :'     0.8557,0.1027,0.0095,0.0038,0.0283
```

Fig. 2. Input file with ^{125}I (Te) nuclear and atomic data for the efficiency computation with MICELLE.

experimental counting efficiency $\varepsilon_{\text{nuclide}}$ of the nuclide under study as a function of the counting efficiency $\varepsilon_{\text{tracer}}$ of the tracer. The corresponding polynomial coefficients as well as reference counting efficiencies are listed in Table 1. Such experimentally determined efficiency curves can then be compared with the data calculated with the MICELLE code.

The calculated results are compared with the experimentally determined curves in the residual plots in Figs. 5 and 6. The horizontal zero line corresponds to the curves which were obtained by fitting the experimental data. We show only calculated results in a region in which no extrapolation of the experimental data was required. The calculations were performed with the ionization quenching option 4 in the code for a mixture of 15 mL Ultima Gold^{TM} and

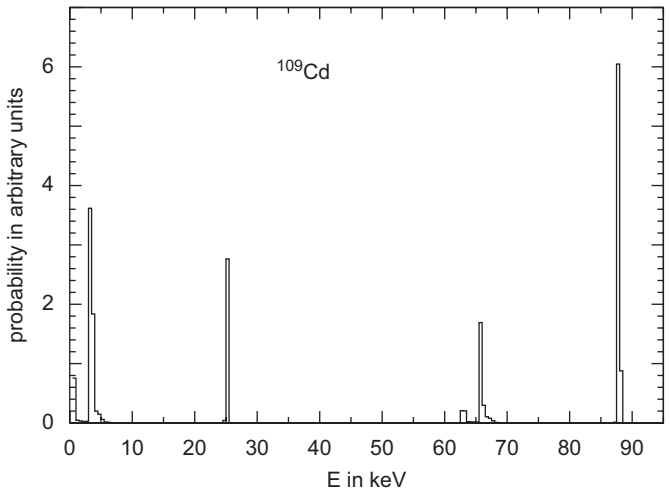

Fig. 3. Computed spectrum of the emitted total electron energy per decay of ^{109}Cd. The decay of the isomer ^{109m}Ag is treated as an independent decay and included in the spectrum.

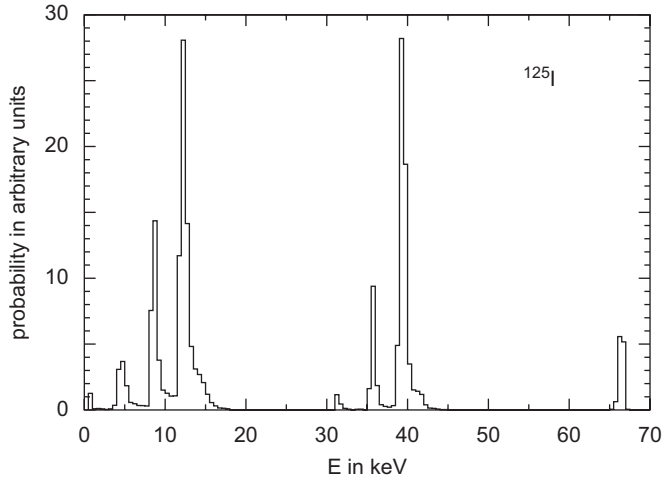

Fig. 4. Computed spectrum of the emitted total electron energy per decay of ^{125}I.

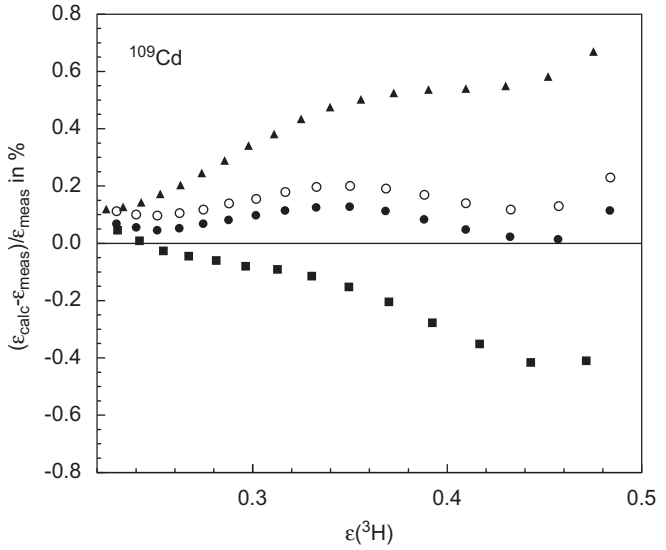

Fig. 5. Residual plot for the counting efficiency of ^{109}Cd, calculated by using the stochastic atomic rearrangement model vs. the ^{3}H counting efficiency. The calculations were done for $kB = 0.0075$ cm/MeV (triangles), $kB = 0.0110$ cm/MeV (circles), and $kB = 0.0150$ cm/MeV (squares). The calculation with $kB = 0.0110$ cm/MeV was repeated without using the micelle size correction (open circles).

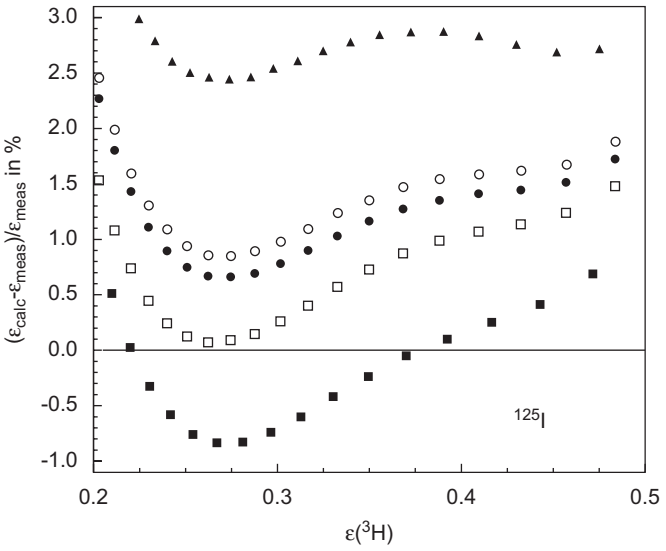

Fig. 6. Residual plot for the counting efficiency of ^{125}I, calculated by using the stochastic atomic rearrangement model vs. the ^{3}H counting efficiency. The calculations were done for $kB = 0.0075$ cm/MeV (triangles), $kB = 0.0110$ cm/MeV (circles), and $kB = 0.0150$ cm/MeV (filled squares). The calculation with $kB = 0.0110$ cm/MeV was repeated without using the micelle size correction (open circles). The open squares were calculated with the same conditions as the filled circles ($kB = 0.0110$ cm/MeV) but with $\omega_K = 0.883$ (Hubbell et al., 1994).

Table 1
Polynomial coefficients for the calculation of the efficiency curves which were determined by experiment and reference counting efficiency ε_{ref} for a sample with a corresponding ^{3}H counting efficiency of 0.5

Nuclide	p_0	p_1	p_2	p_3	p_4	ε_{ref}
^{109}Cd	0.873020	3.807226	−14.368940	25.798410	−16.710510	1.3648
^{125}I	−0.1123076	6.719414	−24.24317	44.09311	−29.89470	0.8298

1 mL water. The counting efficiency of ^{3}H was computed with routines implemented in the MICELLE code using $E_{max}(^{3}H) = 18.58$ keV.

Fig. 5 shows the results for ^{109}Cd with various values for the constant kB. Using $kB = 0.0075$ cm/MeV (triangles) the data show a positive slope, whereas for $kB = 0.0150$ cm/MeV (squares) a negative slope is observed. The best agreement with the experimental data is found using $kB = 0.0110$ cm/MeV as proposed by Broda et al. (2002). Indications of a similar kB value were also found by Kossert (2003) when analyzing experimental data of the β emitter ^{87}Rb. The calculations of the open circles in Fig. 5 were done without using the micelle size effect. For all other data the micelle size effect was taken into account assuming a radius $r = 4$ nm of the micelle sphere. However, the comparison of the calculation with (filled circles) and without the micelle effect (open circles) shows a difference of about 0.1% or less.

The dependence on the kB value is larger for ^{125}I (see Fig. 6) than for ^{109}Cd. This is basically due to the isomeric transition in the decay scheme of ^{109}Cd which emits conversion electrons with relative high energy (see also Fig. 3). The counting efficiency of these electrons is very close to 1 and does not depend on the kB value. The best agreement between calculated efficiencies and the experimental data of ^{125}I is obtained when using $kB = 0.0150$ cm/MeV. When switching off the micelle size effect correction, the computed counting efficiency increases by about 0.2% or less. The results obtained using $kB = 0.0110$ cm/MeV also agree better with the experimental data when the fluorescence yield $\omega_K = 0.883$ from Hubbell et al. (1994) is used.

In addition to the results shown in Figs. 5 and 6, many other effects were studied. The outcomes of some of these tests are summarized in the following.

The calculations were repeated using $E_{max}(^{3}H) = 18.60$ keV instead of $E_{max}(^{3}H) = 18.58$ keV, giving very similar results.

Another check was made changing the energies of KLL Auger electrons. The energies were corrected by the difference ΔE between $E_{KL_1L_1}$ 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 for KXY Auger electrons. This effect was found to be very important for low-Z ec nuclides (Kossert and Grau Carles, 2006) but for ^{109}Cd and ^{125}I the correction is negligible due to the higher energies of K-Auger electrons. A similar argument holds for the explanation that the photoelectric correction (Grau Carles, 2006) is of minor importance for ^{109}Cd and ^{125}I.

In the case of ^{109}Cd, the calculations were also repeated using $P_\gamma = 0.03663(33)$ (Kossert et al., 2006) instead of $P_\gamma = 0.03626(26)$. The difference was also found to be negligible.

6. Conclusion and outlook

The analysis presented in this work shows that the counting efficiencies of ^{109}Cd obtained by using the stochastic atomic rearrangement model are in good agreement with experimental data when a value of $kB = 0.0110$ cm/MeV is used for the computation of the ionization quenching function. Using the new model, activity standardizations with estimated relative standard uncertainties of the order of 0.5% for ^{109}Cd are possible.

For ^{125}I the agreement between calculated results and experimental data is also considerably better than with previous simplified rearrangement models. However, the analysis revealed a large dependence on the value of kB.

For activity standardization the most dominant uncertainty contributions are expected to be due to the uncertainties assigned to the fluorescence yield ω_K, the fractional capture probability P_K (see also Günther, 2002), the kB value as well as the uncertainty assigned to the ^{3}H-tracer activity. A full uncertainty analysis should be done for activity standardization, but this is not the subject of this work.

Despite the improvements achieved with the more realistic stochastic atomic rearrangement model, further ec radionuclides, such as ^{88}Y, ^{99m}Tc, ^{111}In, ^{123}I, ^{124}I, and ^{139}Ce should be investigated. In principle, the model can also be applied to low-Z ec nuclides, but it has been shown in previous works that the photoelectric correction is then indispensable (see, e.g., Kossert and Grau Carles, 2006). Thus, we propose a combination of the atomic rearrangement model realized in the MICELLE code with a possibility of computing the photoelectric correction as implemented in the EMILIA code.

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