ICRM Gamma Spectrometry Working Group

True summing corrections

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Introduction

Summation effects play an important role in measurement of activity of radionuclides with a complex decay scheme by means of a spectrometric method. If simultaneous emission of photons occurs, areas of peaks must be corrected. In order to calculate the correction, we need to know the probability of the simultaneous emission of other gamma photons, x-ray and particles.

Coincidence probabilities were determined using the Monte Carlo method by CMI codes.

Values of total and peak efficiencies were calculated using MCNP code. A model of the detector (GMX15-PLUS-S), shielding and LNHB source was created.

Coincidence probabilities computation

The process of the radionuclide decay is simulated by the program based on the Monte Carlo method. Every history starts by alpha, electron, positron or EC decay to some level of the daughter nuclei according to decay intensities then goes on step by step by gamma transition or internal conversion to lower and lower levels up to the ground-state. The emission of gamma photons, beta, x-rays from EC decay or internal conversion is generated in single decay. Every photon in the one decay is recorded and accumulated together with its mutual coincidence and summing with other gamma, KX, LX, annihilation photons, beta particles.

The history is repeated many times.

Definitions, relations

The correction relations are as follows:

$$A_{corrected}=A_{exp} / K$$

$$K= K_{dec}* Kg_{amma} * K_{sumin}$$

K_{dec} denotes the summing-out effect with β or X-rays (from EC) emitted at the decay

K_{gamma} denotes the summing-out effect with other gamma and X-ray from internal conversion

K_{sumin} denotes the summing-in effect

Equations for K_{dec}

K_{dec} for beta decay:

Individual contribution of any beta component to summing-out effect equals

$$pGB*T(E_{bmax})$$

pGB: coinc. probability of γ photon with the β component

T(Ebmax): total efficiency for β component with E_{bmax}

Peak area is lowered by the sum of all individual contributions, because any component is in parallel cascades with photon, thus:

$$K_{dec} = 1-\Sigma(pGB*T(E_{bmax})).$$

K_{dec} for electron capture (EC)

Determination of K_{dec} for electron capture (EC) decay is more complicated.

The gamma photon is preceded by parallel cascade of the K_{α} ,K_{β}, LX-ray from direct capture and LX-ray emitted after Auger effect supplying K_{α} emission. (Any contribution of Auger electrons is neglected).

K_{α} and LX-ray are in a direct cascade. It is compensated by the term

$$pGK_{\alpha}*TK_{\alpha}*pLkask*TL$$

in the next equation for individual contribution D:

$D = pGK_\alpha * TK_\alpha * (1 - pLkask * TL) + pGK_\beta * TK_\beta + pGL * TL + pLAug * TL + pLkask * TL$

where

pGK_α: coincidence probability of γ photon with K_α X-ray from EC

pGK_β: coincidence probability of γ photon with K_β X-ray from EC

pGL: coincidence probability of γ photon with LX-ray from EC

pLAug: coincidence probability of γ photon with LX-ray after Auger effect supplying K_α emission

pLkask: coincidence probability of γ photon with LX-ray after K_α emission

T denotes total efficiency, K_α, K_β, L represent an X-ray energy

Net peak area is lowered by the sum of all individual contributions of decays to all levels, because any component is in a parallel cascade with photon, thus:

$$Kdec = 1 - \Sigma D$$

Equations for K_{gamma}

Summing-out effect due to coincidence with other γ photons, energy E1. Again, there is necessary to determine individual contribution D of all coincidence gamma photons.

The equation is very similar to previous equation, the term pGG*TE1 is added.

$$D = pGG*TE1+pGK_{\alpha}*TK_{\alpha}*(1-pLkask*TL)+pGK_{\beta}*TK_{\beta}+pGL*TL+pLAug*TL+pLkask*TL$$

pGG:	coinc. prob. of γ photon with gamma photon of energy E1
pGK_{α}:	coinc. prob. of γ photon with K_{α} X-ray from internal conversion
pGK_{β}:	coinc. prob. of γ photon with K_{β} X-ray from internal conversion
pGL:	coinc. prob. of γ photon with Lx-ray from internal conversion
pLAug:	coinc. prob. of γ photon with LX-ray after Auger effect supplying K_{α} emission
pLkask:	coinc. prob. of gamma photon with LX-ray after K_{α} emission
T :	total efficiency, E1, K_{α}, K_{β}, L represent γ or X-ray photon energy

Equations for γ photons with energy E0 should be created individually.

The general form of $K_{gamma}(E0)$ for several photons (energies E1, E2, E3, …) in direct cascade is:

$$\mathbf{K_{gamma}(E0) = (1- D(E1))*(1 - D(E2))*(1 - D(E3))* \ldots}$$

Technique can be called “multiplying“

The general form of $K_{gamma}(E0)$ for several photons (energies E4, E5, …) in parallel cascade is:

$$\mathbf{K_{gamma}(E0) = (1- D(E4)-D(E5) - \ldots)}$$

Technique can be called “adding“

Both techniques should be combined in case coincident parallel and direct cascades.

Example for 604 keV (^{134}Cs):

$$K_{gamma}(604)= (1-D(563)-D(796)-D(1039)-D(1365))*(1-D(475)-D(243)-D(802)-D(569)-D(327))$$

Technique can be called “realistic“

All equations for all photons are in tables hereinafter, pages from 18 to 20.

Equations for K_{sumin}

Summing-in effect to photon of energy E0 due to cross-over cascade of photons of energies E1 and E2 (E0 = E1 + E2) is expressed by the factor K_{sumin} after equation:

$$K_{sumin}(E0)=(1+Ps(E1,E2)*\varepsilon(E1)*\varepsilon(E2)/\varepsilon(E0))$$

Ps(E1,E2): summing probability of photon energies E1 and E2 to E0

ε (E1), ε (E2) and ε (E0): peak efficiencies for photon of energies E1, E2 and E0

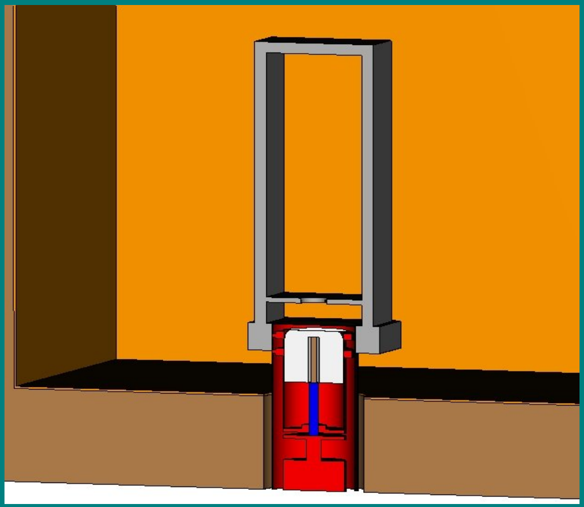
As many cascades Ea→Eb are summing to energy E0, as many terms Ps*ε (E1)*ε (E2)/ε (E0) in equation must be used. The values of the Ps are different for each cascade.

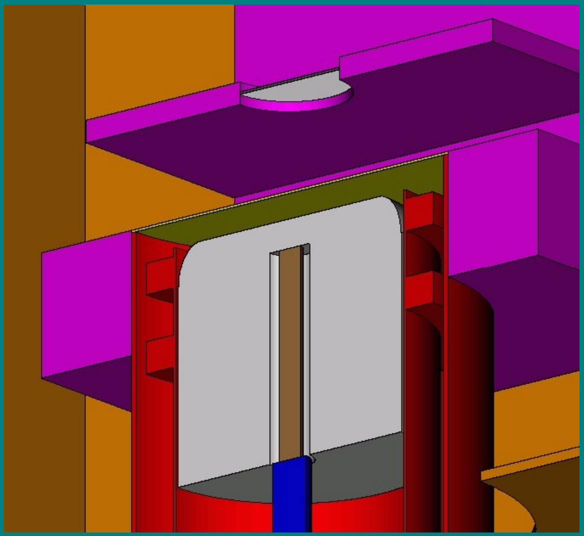
$$K_{sumin}(E0) = (1 + \Sigma Ps(Ea,Eb)*\varepsilon(Ea)*\varepsilon(Eb)/\varepsilon(E0))$$

Detector model for MCNP calculations

A special model was created for ICRM comparison. Construction information of the Detector GMXI5-PLUS-3, detector shielding, support and source were provided by LNHB.

The model is illustrated in the pictures.



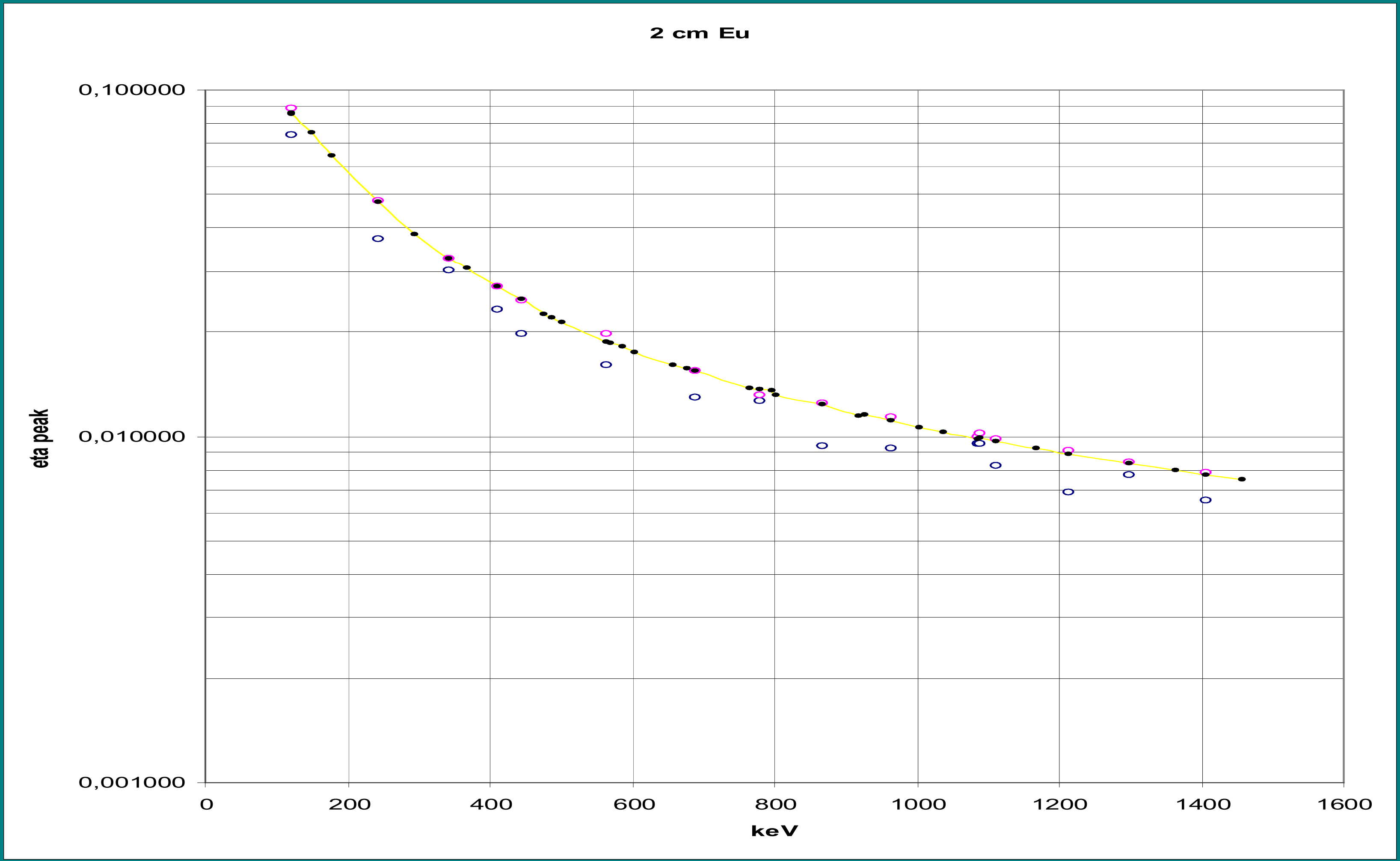


Detector model was used for peak and total efficiencies beta, gamma and x-ray of all three geometries.

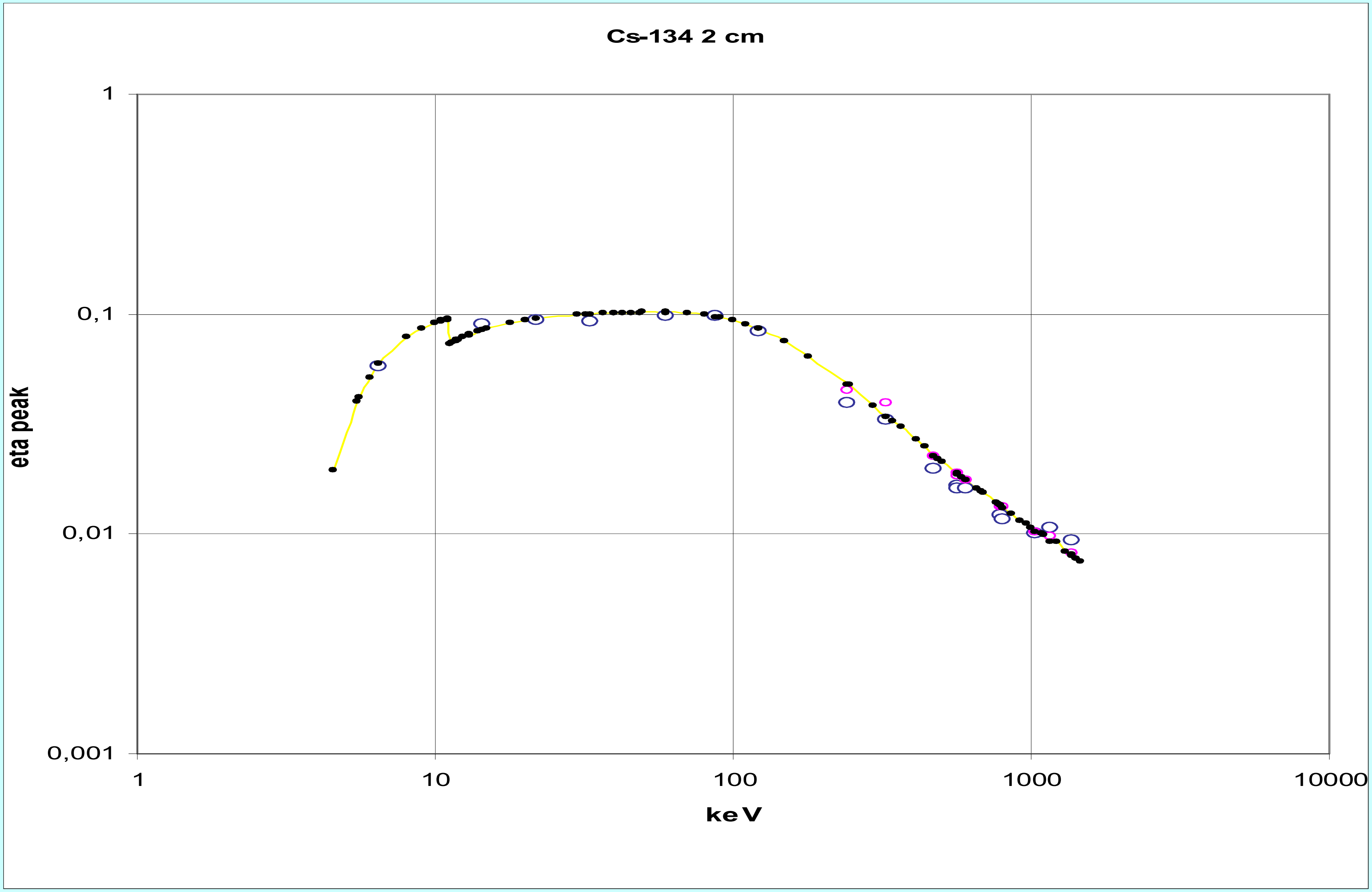
Total and peak efficiencies

Corrected experimental peak eff. values and calculated MCNP-values for geometry “2cm” are presented in following pictures.

Eu-152, **black**: calculated; **blue**: exp uncorrected; **pink**: exp corrected



Cs-134 , peak eff.: **black**: calculated; **blue**: exp uncorrected; **pink**: exp corrected



Some low energy experimental points were added to the previous picture for ^{134}Cs:

6.5 keV (from sum peaks of Co-57),

14.3 keV (from sum peaks of Y-88),

22 keV (KX Cd-109),

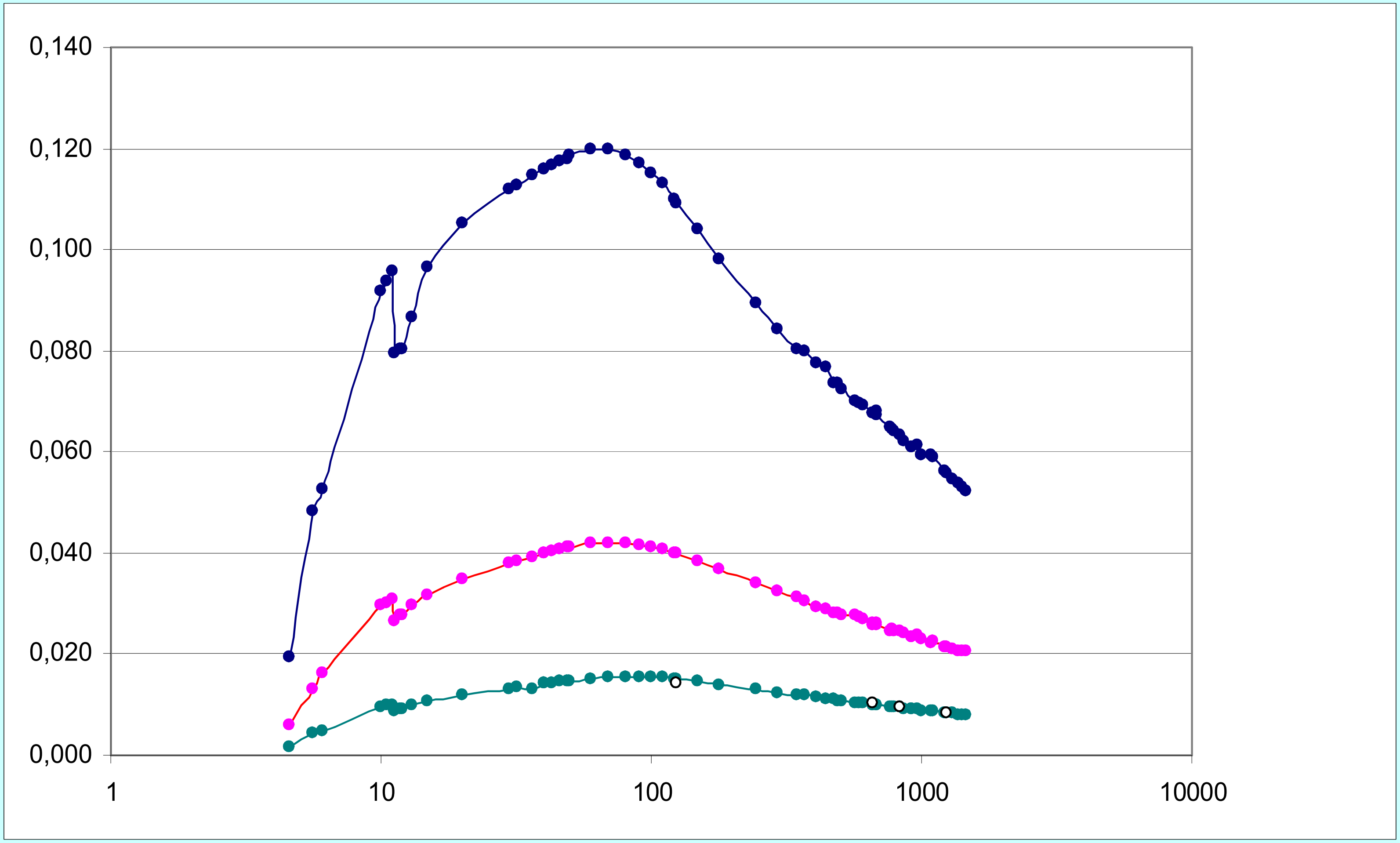
33.3 keV (from sum peaks of Ce-139),

59.6 keV (Am-241),

88 keV (Cd-109)

Total efficiencies for photons were calculated for geometries "2 cm", "5 cm" and "10 cm".

Green: “10 cm” with several experimental values (125; 661; 834 and 1250 keV)
Pink: “5 cm”
Blue: “2 cm”



List of K_{gamma} equations .

There were only strong parallel cascade taken into account. All remaining coincidence photons not explicitly mentioned in the equation are considered as direct cascade. They are include in the term π(1-Drest) as the product of individual contributions (1-D).

EuSm	121.78	(1-D(244)-D(688)-D(841)-D(919)-D(964)-D(1112)-D(1249)-D(1408)-D(1457)-D(1528))*π(1-Drest)
EuSm	244.7	(1-D(121)*(1-D(444)-D(656)-D(674)-D(719)-D(867)-D(926)-D(1005)-D(1212))* π(1-Drest)
EuSm	688.67	product of all (1-D)
EuSm	443.969	product of all (1-D)
EuSm	1085.85	(1-D(444)-D(564))* π(1-Drest)
EuSm	964.07	(1-D(444)-D(564))* π(1-Drest)
EuSm	1112.09	product of all (1-D)
EuSm	867.38	product of all (1-D)
EuSm	1408.03	(1-D(121))*(1-D(239))
EuSm	443.957	(1-D(121))*(1-D(239))*(1-D(964)-D(1085))*(1-D(239))* π(1-Drest)
EuSm	1212.88	product of all (1-D)
EuSm	564,06	(1-D(1085)-D(964)-D(719))* π(1-Drest)

EuGd	344,28	(1-D(271)-D(411)-D(586)-D(765)-D(779)-D(1089)-D(1299)* π(1-Drest)
EuGd	411,12	(1-D(344))*(1-D(368)-D(527)-D(678)-D(794)-D(937))* π(1-Drest)
EuGd	778.9	(1-D(344))*(1-D(482)-D(520))
EuGd	1089.74	(1-D(344))*(1-D(209))
EuGd	1299,11	(1-D(344))

Cs-134	604.69	(1-D(563)-D(796)-D(1039)-D(1365))*(1-D(475)-D(243)-D(802)-D(569)-D(327))
Cs-134	1167,92	(1-D(475)-D(802))*(1-D(327))
Cs-134	563.23	(1-D(604))*(1-D(475)-D(802))*(1-D(327))
Cs-134	795.84	(1-D(604))*(1-D(243)-D(569))*(1-D(327))
Cs-134	1038.55	(1-D(604))*(1-D(327))
Cs-134	475.32	(1-D(604))*(1-D(1168)-D(563))*(1-D(327))
Cs-134	242.71	(1-D(604))*(1-D(796))*(1-D(327))
Cs-134	1365.16	(1-D(604))
Cs-134	801.93	(1-D(604))*(1-D(1168)-D(563))
Cs-134	569.32	(1-D(604))*(1-D(796))
Cs-134	326.61	(1-D(604))*(1-D(1168))*(1-D(563))*(1-D(796))*(1-D(1039)-D(475)-D(243))

Are equations for K_{gamma} correct?

Test of K_{gamma} equations correctness

The difference among three computation techniques (adding, realistic and multiplying) is small for detection system with total efficiencies up to 12 %.

The difference is notable if different three methods are applied to a hypothetical detection system with total efficiency for photons (γ, X-ray) equal 1 and for Auger and conversion electrons 0. Well-type Ge detector approaches such hypothetical detector type. The results for the three different methods are shown in the next tables.

Cs-134			
E0	adding	realistic	multiplying
242,71	-1,0035	0,0000	0,0000
326,71	-1,5019	0,00009	0,01121
475,32	-0,8281	0,00018	0,02546
563,23	-0,9946	0,00001	0,00013
569,32	-0,9985	0,00000	0,00000
604,69	-0,2433	0,00048	0,08618
795,84	-0,1800	0,00083	0,00083
801,93	-0,8227	0,00018	0,02566
1038,55	-0,0042	0,00101	0,00101
1167,92	0,00454	0,00529	0,1294
1365,16	0,00092	0,00092	0,00092

EuGd			
E0	adding	realistic	multiplying
344,28	0,2207	0,2779	0,3886
411,12	-0,6050	0,0020	0,0025
778,90	0,0013	0,0053	0,0053
1089,74	0,0032	0,0055	0,0055
1299,11	0,0053	0,0053	0,0053

EuSm			
E0	adding	realistic	multiplying
121,78	-0,1519	0,0277	0,2721
244,71	-0,7694	0,0179	0,0587
443,96	-0,4948	0,0063	0,1255
564,06	-0,4941	0,0015	0,1255
688,67	0,0099	0,1591	0,1591
867,38	-0,8218	0,0034	0,0034
964,07	0,0548	0,0316	0,1658
1085,85	0,8636	0,8643	0,8663
1112,09	0,1590	0,1849	0,1849
1212,88	-0,7889	0,0036	0,0036
1408,03	0,1913	0,1916	0,1916

Comment:

Adding method overestimates most of cases. Negative values have no physical meaning.

Multiplying method is correct for direct cascades without paralel cascades. The correction is underestimated.

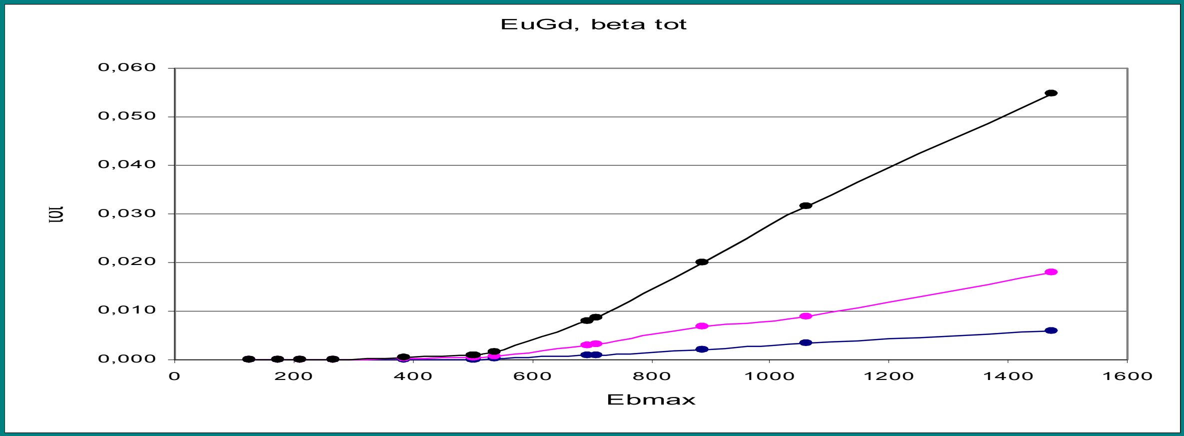
Realistic method gives value K_{gamma} near zero, small positive correction corresponds to non-photon process (internal conversion at M,N,O levels, Auger emission not followed by photon emission).

Photons 344, 688, 1085, 1112 and 1408 keV have high value of K_{gamma}, because they are emitted in significant cases without coincidence with other photons.

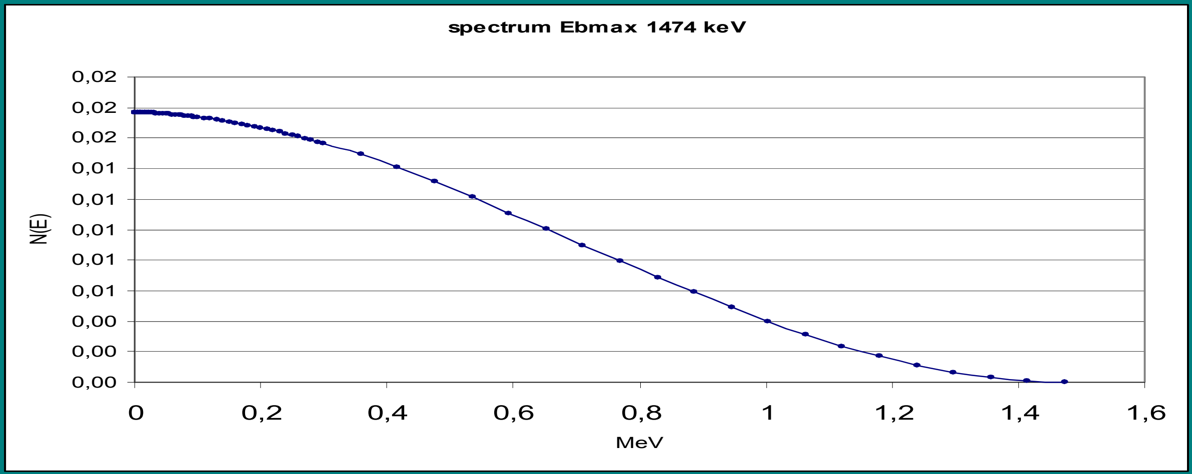
Other results

Total efficiencies for beta radiation were calculated using MCNP for geometries "2 cm", "5 cm" and "10 cm" . Efficiencies were calculated for real form of spectra, not only for Ebmax or mean beta energy.

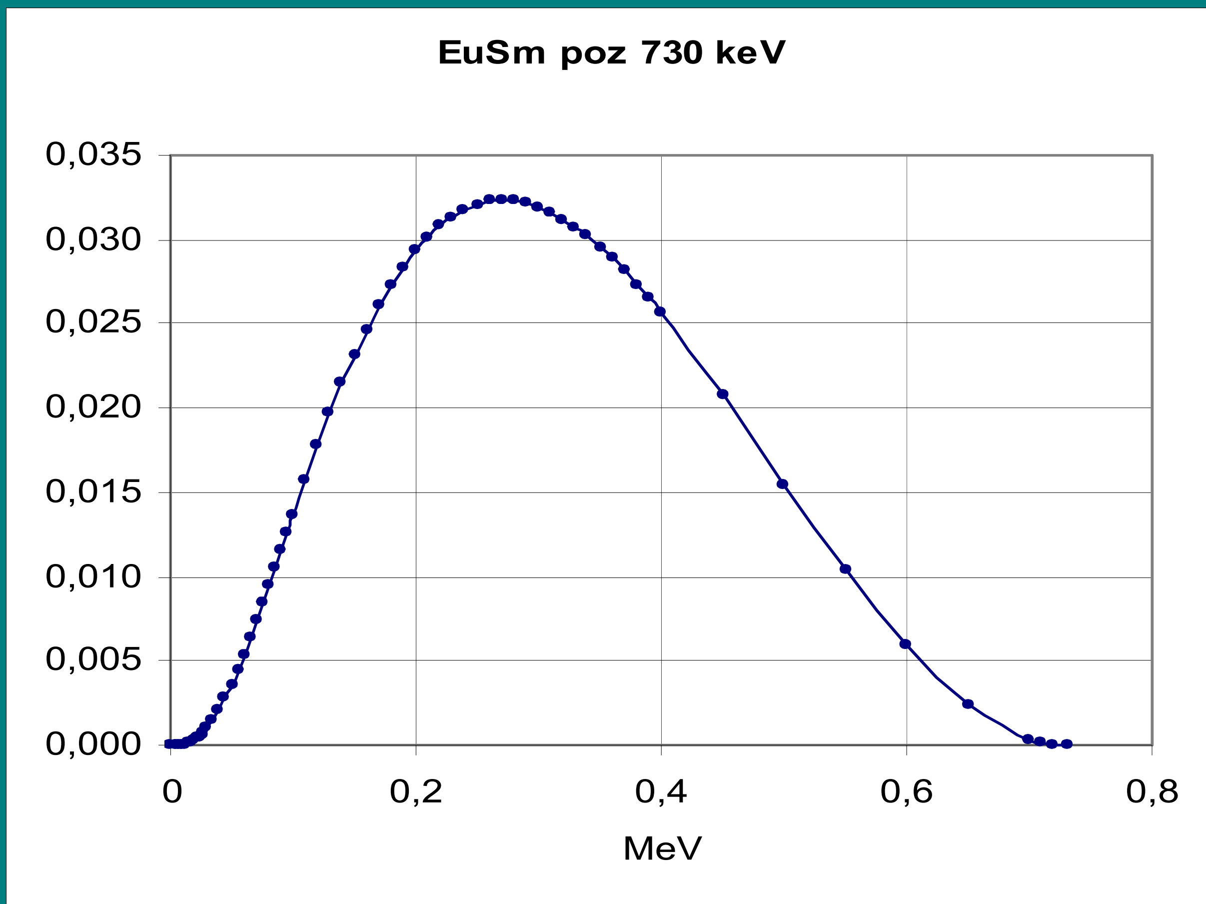
The values for all beta components of Eu-Gd are shown in the next picture.



- **Example of beta spectra in Eu – Gd decay with Ebmax=1474 keV**



Example of positron spectra in Eu- Sm decay with Ebmax=730 keV



Uncertainties

Uncertainties type A of coincidence probabilities from Monte Carlo method are less than 0.0005 (k=1). They are lesser than uncertainties of photon intensities, conversion coefficients, branching ratios and other parameters coming into calculation.

Uncertainties type B of Kgamma result from equations presented in the table 6 incorrectness.

Parallelism or directness of weak cascades with intensity less than 0.1% in Eu decay was omitted and all of them were suggested as direct.

Equations for Cs-134 are exact.

Equations for Kdec and Ksumin are exact.

Estimation: Uncertainties of correction factors presented in the tables 7 and 8 are on a level of uncertainties of main gamma photons and branching decay ratios.

Uncertainty “A” of the coincidence probabilities from Monte Carlo method are smaller than 0,0005 (k=1) in all cases. They are less significant than uncertainties of intensities of photons, conversion coefficients, branching ratios and other parameters coming into calculation.

One component of the Kgamma uncertainty “B” results from incorrectness of equations presented in table 6. Parallelism or directness of weak cascades with intensity less than 0,1% in Eu decay was omitted and all of them were suggested as direct.

Equations for K_{gamma} of ^{134}Cs are exact.
Equations for K_{dec} and K_{sumin} are exact.

Another component of the Kgamma uncertainty “B” results from calculated peak and total efficiencies.

Peak efficiency: Deviations of calculated and experimental values of peak efficiency are smaller than 1 % for all geometries and main lines (see pictures in section 5).

Total efficiency: the comparison experiment-calculation was performed only for geometry “10 cm”. Confidential experimental points (^{57}Co, ^{137}Cs, ^{54}Mn, ^{60}Co) vary from calculated values 3 % at most. We suppose the maximum uncertainty of total efficiency 3 % for all geometries and typical uncertainty 1 % due to facts:

The substantial part of total efficiency is a contribution of radiation scattered from surround materials. It is highest for geometry “10 cm”, for closer geometries is the contribution smaller.

Deviations of calculated and experimental peak efficiency values are smaller than 1 % for all geometries, so the calculated total efficiencies have the same uncertainty.

Estimation: uncertainty of all components (nuclear and atomic data, peak and total efficiencies etc.) propagates to the uncertainty of corrections factors presented in tables 7 and 8 on the level 0,004 (k=1).