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The ENDF-6 Format for the Evaluated Covariances of Discrete Radiation Spectrum Data (Proposal)

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Contents

Modern radioactive decay data sublibraries

- ENDF/B-VII.1 (a copy of the ENSDF in the ENDF-6 format): 3817 materials (from neutron to roentgenium), stable and unstable nuclides, ground state and isomeric levels
- JEFF-3.1.1 : 3852 materials (from neutron to roentgenium), stable and unstable nuclides, ground state and isomeric levels, spectral data are given for 1521 nuclei
- EAF-99/DECAY : 1917 materials (from neutron to fermium), unstable nuclides, ground state and isomeric levels
- JENDL/FP-DD: 1229 materials (from vanadium to iterbium), unstable nuclides, ground state and isomeric levels

Modern radioactive decay data sublibraries (continued)

2 most complete sublibraries - ENDF/B-VII.1 and JEFF-3.1.1.

- The sublibraries have almost the same list of radionuclides

- On the whole, the evaluated data from the sublibraries are in agreement within declared uncertainties (except for single radionuclides).

Deficiencies, common for the sublibraries

- As a rule, the evaluated decay data is not balanced due to lack of experimental information or applying the physically inconsistent evaluation procedures
- The covariance information for the evaluated decay data is absent.

As known most of the decay data measurements are relative ones. When the accuracy of the reference data is poor the results of relative measurements correlate essentially and can not be processed in assumption of statistical independence as being made in considerable part of the evaluations.

What we mean under balanced decay scheme?

The balanced decay scheme must meet 2 conservation laws:

- A sum of the transition probabilities for particles and gamma quanta feeding any excited level of a daughter nuclide equals to a sum of the transition probabilities for particles and gamma quanta depopulating the level
- A sum of the transition probabilities for particles and gamma quanta feeding the ground state of a daughter nuclide equals to 1.

The inclusion of the balance relationships in the evaluation procedure must inevitably lead to lower uncertainties of the evaluated data and strong correlations between some of the evaluated parameters.

Thus, this study was motivated by a necessity of the generation of physically consistent evaluated decay data with complete covariance information. As an example, a decay of Cm-242 has been analyzed.

Decay of Cm-242 . Experimental data

- Cm-242 decays by alpha-emission to the ground state and 15 excited states of the Pu-238 which are depopulated by gamma emission
- With probability ~ 99.99% Cm-242 disintegrates to the ground and first excited level (44.08 keV) of Pu-238
- The gamma emission intensities were experimentally studied by Lederer, 1981. The relative intensities of 21 gamma rays were measured starting with the 336-keV γ-ray. The values were given relative to the intensity of the 561-keV γ-ray. The normalization factor P(γ 561keV) = 1.5 (4)·10–4

Decay of Cm-242 (continued)

Results of measurements of the ^{242}Cm alpha emission intensities (per 100 decays)

E_{α}, keV	Asaro 1953	Kondratev 1958	Dzhelepov 1963	Baranov 1966	Yang 1998
6113	73.7(5)	73.5(5)	74(2)	74.2(5)	74.08(7)
6069	26.3(5)	26.5(5)	26.0(9)	25.8(5)	25.92(6)
5969	0.035(2)	0.030(2)	0.035(2)	0.036(2)	
5816		0.0046(5)		0.0046	
5608				$2\cdot 10^{-5}$	
5518			$2.8(5)\cdot 10^{-4}$	$2.5(6)\cdot 10^{-4}$	
5187			$3.4(8)\cdot 10^{-5}$	$2.5(8)\cdot 10^{-5}$	

Statistical model

The measurements y_i^k are a sum of a model function $f(E_i,\vec{\theta})$ and unbiased random experimental errors, ε_i^k

$$y_i^k = f(E_i^k,\vec{\theta}) + \varepsilon_i^k, \qquad i = 1,\ldots,L, \qquad k \in K(i)$$

with m restrictions

$$\mathbf{H}\vec{\theta} = \vec{d}$$

imposed to the parameters $\theta_1,\ldots,\theta_L$ (alpha or gamma emission intensities) to be evaluated.

The lower indices of the variables refer to number of the alpha (gamma) transition, the upper indices – to number of experiment,

E_i^k - known energies of alpha particles or gamma quanta,

K(i) is a subset (of dimension n_i) of the indices from the set (1, 2,…,*M*),

$n = \sum_{i=1}^{L} n_i$ - total number of measurements, $n > L$,

M - the number of experiments

$\mathbf{H}$ - a known matrix of dimension $(m \times L)$,

$\vec{d}$ - a known vector of dimension m.

Statistical model (continued)

A form of the model function $f(E,\vec{\theta})$ is directly induced by the problem

$$f(E,\vec{\theta}) = \sum_{i=1}^{L} \theta_i \varphi_i(E)$$

where functions $\varphi_1(E),\dots,\varphi_L(E)$ are defined on a discrete set of energies $E_1^k,\dots,E_L^k$ in following way

$$\varphi_i(E_j^k) = \delta_{ij} = \begin{cases} 1 & i = j \\ 0 & i \neq j \end{cases}$$

Statistical model (continued)

The measurements correlate only inside the experiment

$$cov(\varepsilon_i^k, \varepsilon_j^l) = V_{ij}^k \delta_{kl}$$

Correpondingly, the matrix V has a block structure.

The elements of the matrix $\mathbf{V}^k$ are calculated as a sum of the covariances between the components of the total experimental errors

$$V_{ij}^k = cov(\varepsilon_i^k, \varepsilon_j^k) = \sum_l cov((\varepsilon_i^k)_l, (\varepsilon_j^k)_l)$$

$(\varepsilon_i^k)_m$ - the m – th component of the total experimental error.

Basic equations

A functional to be minimized

$$\tilde{S}^2(\vec{\theta},\vec{\lambda}) = (\vec{y} - \mathbf{X}\vec{\theta})^T \mathbf{V}^{-1} (\vec{y} - \mathbf{X}\vec{\theta}) + \vec{\lambda}^T (\mathbf{H}\vec{\theta} - \vec{d}),$$

where $\vec{\lambda}^T = (\lambda_1, \ldots, \lambda_m)$ – the Lagrange multipliers,

X – the matrix of the sensitivities of the model function relative to the parameters.

The functional has minimum at

$$\tilde{\vec{\theta}} = \hat{\vec{\theta}} + \mathbf{W}\mathbf{H}^T (\mathbf{H}\mathbf{W}\mathbf{H}^T)^{-1} (\vec{d} - \mathbf{H}\hat{\vec{\theta}})$$

$\mathbf{W} = (\mathbf{X}^T \mathbf{V}^{-1} \mathbf{X})^{-1}$ – covariance matrix of evaluated parameters for the problem without restrictions.

The covariance matrix **U** of the vector $\tilde{\vec{\theta}}$ for the problem with restrictions is calculated in following way

$$\mathbf{U} = \mathbf{W} - \mathbf{W}\mathbf{H}^T (\mathbf{H}\mathbf{W}\mathbf{H}^T)^{-1} \mathbf{H}\mathbf{W}$$

Scheme of calculations

Two ideas underlie the evaluation procedure:

- it must be an iterative one to provide self-consistency of the evaluated data

- an order of data processing is important,

  because the experimental data on the alpha emission intensities are much more accurate (relative uncertainty of 0.1 – 5 %) compared to the measured gamma emission intensity (relative uncertainty of 20-50%)

Scheme of calculations: steps

1. The alpha transition probabilities for 7 transitions ((0,0) – (0,5), (0,8)) were evaluated by the LSMR on the basis of the experimental data presented in Table. The system of equations contained 21 equations (n = 21). The number of evaluated parameters – 7. The matrix **H** includes only one row

$$\theta_1^k + \theta_2^k + \theta_3^k + \theta_4^k + \theta_5^k + \theta_6^k + \theta_9^k = 100 - \hat{\theta}_7^{k-1} - \hat{\theta}_8^{k-1} - \sum_{i=10}^{16} \hat{\theta}_i^{k-1}$$

The values of the $\hat{\theta}_7^0$, $\hat{\theta}_8^0$, $\hat{\theta}_{10}^0$, ..., $\hat{\theta}_{16}^0$ are taken from the DDEP evaluation.

2. A new value N^k of the normalization factor for the experimental data of Lederer, 1981 was calculated from the balance relationship for fifth excited level of ^{238}Pu

$$\hat{\theta}_6^k + N^k I_{(8,5)}^k (1 + C_{t,(8,5)}) + N^k I_{(9,5)}^k (1 + C_{t,(9,5)}) =$$

$$N^k I_{(5,1)}^k (1 + C_{t,(5,1)}) + N^k I_{(5,0)}^k (1 + C_{t,(5,0)})$$

For a given index j, $C_{t,(j,l)}$ ($l < j$) is a total internal conversion coefficients (ICC) for the gamma transitions depopulating the level j,

Scheme of calculations: steps (continued)

3. The results of relative measurements (Lederer, 1981) were renormalized to the value N^k.

4. The gamma emission intensities were evaluated from th system of equations (1) on the basis of the experimental data c Lederer,1981 and alpha emission probabilities evaluated at step 1 The matrix **H** has 6 rows and 29 columns - 6 balanc relationships for the ground and 1-4,8 excited levels of the ^{238}P were used as restrictions:

$$\hat{\theta}_j^k = \sum_{l,l<j} I_{q(l)}^k (1 + C_{t,jl}) - \sum_{l,l>j} I_{q(l)}^k (1 + C_{t,lj}) \qquad j = 7,8,10\text{-}16$$

Scheme of calculations: steps (continued - 2)

5. New values $\hat{\theta}_j^{k+1}$, j = 7,8,10-16 of the alpha emission intensities were calculated as follows

$$\hat{\theta}_j^{k+1} = \sum_{l,l<j} \hat{I}^k_{q(l)}(1 + C_{t,jl}) - \sum_{l,l>j} \hat{I}^k_{q(l)}(1 + C_{t,lj}) \qquad j = 7,8,10\text{-}16$$

6. The steps 1-5 were repeated until the difference between tw successive estimates of the normalization factor was lower tha 0.01%.

The evaluated Cm-242 alpha emission intensities and their uncertainties (per 100 decays)

N	E_{α}, keV	this work	ENDF/B-VII.1	JEFF-3.1
0	6113	74.044(44)	74.080(70)	74,000(900)
1	6069	25.917(44)	25.920(60)	26,000(900)
2	5969	0.0340(10)	0.0350(20)	0.0350(10)
3	5816	0.00460(35)	0.00460(50)	0.00310(50)
4	5608	0.000020(5)	0.000020(0)	0.000020(10)
5	5518	0.000268(38)	0.000250(50)	0.00026(10)
6	5462	0.0000131(35)	0.0000126(24)	0.0000110(10)
7	5366	0.00000030(9)	0.00000022(3)	0.0000013(3)
8	5187	0.0000295(57)	0.0000360(70)	0.000053(9)
9	5166	0.00000117(30)	0.00000113(21)	0.0000018(3)
10	5146	0.00000171(50)	0.00000170(40)	0.00000170(40)
11	5111	0.00000021(11)	0.00000010(10)	
12	5101	0.0000038(11)	0.0000037(8)	0.0000036(5)
13	5005	0.00000033(10)	0.00000031(8)	0.00000032(6)
14	4904	0.00000052(52)	0.00000055(15)	0.00000054(8)
15	4869	0.00000054(17)	0.00000052(15)	0.00000045(12)

The correlation matrix of the evaluated Cm-242 alpha emission intensities (in percent)

	0	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
0	100															
1	-99.9	100														
2	0	0	100													
3	0	0	0	100												
4	0	0	0	0	100											
5	0	0	0	0	0	100										
6	0	0	0	0	0	99	100									
7	0	0	0	0	0	90	89	100								
8	0	0	0	0	0	0	0	0	100							
9	0	0	0	0	0	97	97	88	0	100						
10	0	0	0	0	0	90	90	81	0	88	100					
11	0	0	0	0	0	50	49	45	0	49	45	100				
12	0	0	0	0	0	94	93	85	0	92	85	47	100			
13	0	0	0	0	0	86	86	78	0	84	78	43	82	100		
14	99.9	-99.9	-8	-8	-7	0	0	0	-7	0	0	0	0	0	100	
15	0	0	0	0	0	82	82	74	0	80	75	41	78	71	0	100

The evaluated Cm-242 gamma emission intensities and their uncertainties (per 100 decays)

N	E_γ, кэВ	this work	ENDF/B-VII.1	JEFF-3.1
1	44.08	0.03294(6)	0.03294(8)	0.03227(860)
2	101.92	0.00250(7)	0.00259(13)	0.00241(65)
3	157.42	0.00145(11)	0.00144(16)	0.00096(30)
4	210.2	0.0000117(29)	0.00001170(2)	0.0000116(67)
5	336.36	0.0000007(3)	0.0000007(3)	0.0000007(3)
6	357.64	0.000000045(9)	0.000000045(9)	0.000000585(250)
7	459.8	0.000000059(30)	0.000000060(30)	0.000000057(28)
8	515.25	0.0000046(13)	0.0000045(12)	0.0000045(12)
9	561.02	0.000156(42)	0.000150(40)	0.000150(40)
10	605.04	0.000109(29)	0.000100(30)	0.000105(28)
11	617.20	0.0000082(22)	0.0000081(21)	0.0000012(4)
12	617.22	0.00000017(5)	-	0.0000068(18)
13	837.01	0.00000019(6)	0.00000019(6)	0.00000019(6)
14	882.63	0.000000067(15)	0.000000067(15)	0.000000060(22)
15	897.33	0.000022(6)	0.000022(6)	0.000022(6)
16	918.7	0.00000056(16)	0.00000054(15)	0.00000054(15)
17	938.91	0.00000018(6)	0.00000018(6)	0.00000018(6)
18	962.8	0.00000055(15)	0.00000053(15)	0.00000053(15)
19	974.5	0.00000020(11)	0.00000010(10)	-
20	979.8	0.00000027(9)	0.00000026(8)	0.00000026(8)
21	983.0	0.00000052(19)	0.00000050(18)	0.00000050(18)
22	984.5	0.0000020(6)	0.0000020(6)	0.0000020(6)
23	1028.5	0.0000016(5)	0.0000016(5)	0.0000016(4)
24	1081.7	0.000000052(21)	0.000000050(20)	0.000000050(20)
25	1118.3	0.00000017(9)	0.00000017(9)	0.00000017(9)
26	1184.6	0.00000052(15)	0.00000050(15)	0.00000050(14)
27	1220.2	0.00000029(9)	0.00000028(9)	0.00000028(9)

A block of the correlation matrix with considerable correlations between the evaluated Cm-242 alpha emission intensities (in %)

	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22
7	100															
8	52	100														
9	53	97	100													
10	53	96	99	100												
11	53	96	99	98	100											
12	53	96	99	98	98	100										
13	46	83	85	85	85	84	100									
14	52	94	97	96	96	95	83	100								
15	51	92	95	95	95	94	82	92	100							
16	45	81	84	84	83	83	72	81	80	100						
17	51	92	95	95	94	94	81	92	91	80	100					
18	26	48	50	50	49	49	43	48	48	42	47	100				
19	45	81	84	83	83	83	72	81	80	70	80	42	100			
20	39	72	74	73	73	73	63	71	70	62	70	37	62	100		
21	46	84	87	86	86	85	74	84	83	73	82	43	73	64	100	
22	50	91	94	94	93	93	80	91	90	79	90	47	79	70	82	100

SUMMARY AND RECOMMENDATIONS

1. On the whole, the recommended values from this work is consistent with the ENDF/B-VII.1 and JEFF-3.1 evaluations within declared uncertainties. The gamma emission intensities starting from 515 keV are higher the ENDF/B-VII.1 and JEFF-3.1 values by 4.2 % approximately due to the correction of the renormalization factor defined in (Lederer, 1981).

2. The main difference between our and the ENDF/B-VII.1 and JEFF-3.1 evaluations is related to the uncertainty information. The inclusion of balance relationships in the scheme of calculations lead inevitably to lower uncertainties of the evaluated data and strong correlations between some of the evaluated parameters. For this reason, the alpha and gamma emission intensities evaluated in this work are strongly correlated unlike the ENDF/B-VII.1 and JEFF-3.1 recommended values and have lower (by 25 – 50 %) uncertainties for most intense transitions. The confidence regions for strongly correlated evaluated values are completely distinct from the regions which correspond to the uncorrelated data. Thus, the uncertainty information presented in the ENDF/B-VII.1 and JEFF-3.1 evaluations is incomplete and essentially distorted.

SUMMARY AND RECOMMENDATIONS (CONTINUED)

3. The available formats of decay data representation – ENSDF, ENDF-6 – must be revised to provide a possibility for representation of evaluated covariance data for discrete radiation decay spectrum.

4. The results of this work are in favor of reevaluation of the decay data for other actinides.

The ENDF-6 format for the evaluated covariances of discrete radiation spectrum data (proposal)

The following changes could be made in the structure of a subsection of the section MT = 457 (MF = 8)

[MAT,8,457/ 0.0, STYP, LCON, LCOV, 6, NER/

FD, ΔFD, ERAV, ΔERAV, FC, ΔFC] LIST

[MAT,8,457/ER(1),ΔER(1),0,0,NT,0/

RTYP(1), TYPE(1), RI(1), ΔRI(1), RIS(1), ΔRIS(1),

RICC(1), ΔRICC(1),RICK(1),ΔRICK(1),RICL(1),ΔRICL(1)] LIST

……………………………………………………………………………..

[MAT,8,457/ ER(NER), ΔER(NER),0,0,NT,0/

RTYP(NER),TYPE(NER),RI(NER),ΔRI(NER),RIS(NER),ΔRIS(NER),

RICC(NER),ΔRICC(NER),RICK(NER),ΔRICK(NER),

RICL(NER),ΔRICL(NER)] LIST

(omit LIST-record if LCON = 1)

[MAT,8,457/ RTYP, 0.0, 0, **0**, NR, NP/ EINT/ RP(E)] **TAB1-record**

(omit **TAB1-record** if **LCOV = 0, 2**)

The ENDF-6 format for the evaluated covariances of discrete radiation spectrum data (continued)

[MAT,8,457/ 0.0, 0.0, 0, LB=2, 2*NPP, NPP/ (E(K),F(K))] LIST

(omit **LIST-record** if **LCOV = 0, 2**)

[MAT,8,457/ 0.0, 0.0, 0, LB=5, NT, NE/ (E(K),F(K,J))] LIST

(omit **LIST-record** if **LCOV = 0,1**)

where F(K,J) – the elements of relative covariance matrix for evaluated intensities of discrete spectrum

Possible values for flag LCOV:

LCOV = 0, no covariance data given
LCOV = 1, covariance data for continuum spectrum are given
LCOV = 2, covariance data for discrete spectrum are given