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A self-consistent evaluation of ^{242}Cm alpha and gamma emission intensities

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HIGHLIGHTS

- Self-consistent evaluation of ^{242}Cm alpha and γ-ray emission intensities performed.
- Exact balance relationships were included in the evaluation procedure.
- Experimental covariances were taken into account.
- Uncertainties of evaluated intensities lower than ENDF/B-VII.1 and JEFF-3.1 values.
- Correlations between some of the evaluated intensities are significant.

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ABSTRACT

As a rule the evaluated decay data and their uncertainties reflect the current level of consistency of the experimental data as well as its shortage. However, if we assume *a priori* that the structure of the decay scheme is precisely established, the inclusion of the exact balance relationships in the evaluation procedure leads to lower uncertainties of the recommended data and strong correlations between some of the evaluated parameters. Such a self-consistent decay data evaluation was carried out using the ^{242}Cm alpha decay as an example.

1. Introduction

Modern evaluated nuclear data libraries include over ten sub-libraries (Chadwick et al., 2011). Three of them: neutron reactions, radioactive decay data and neutron-induced fission yields sub-libraries are of special importance for nuclear applications. In particular, the radioactive decay data are used in calculations of isotopic kinetics in a reactor core, neutron dosimetry, waste disposal, depletion and buildup studies, and decay heat calculations.

At present there exist only a few radioactive decay data libraries available for users: ENSDF (Tuli, 2001), ENDF/B-VII.1 (Chadwick et al., 2011), JEFF–3.1.1 (Kellett et al., 2009), or DDEP (Bé et al., 2013 and references therein). It should be noted that the ENDF/B-VII.1 radioactive decay data sub-library is mainly based on a translation of information from the ENSDF library into the ENDF-6 format (Trkov et al., 2011). The lists of radioactive nuclides presented in the libraries are very similar. On the whole the evaluated decay data most important for reactor applications (half-life, branching ratio, average energy of radiation) extracted from different libraries are consistent within the declared uncertainties (except for certain radionuclides).

At the same time there are two defects common to the libraries: (1) very often the evaluated decay data are not balanced due to a lack of experimental information or such a balance is accompanied by very large uncertainties, (2) covariance information for the evaluated data is absent as the commonly used formats currently have no way of storing this information. As is well-known most decay data measurements have been carried out using a relative method. When the uncertainty of the reference data used in the experiment predominates, the results of measurements are highly correlated and cannot be processed assuming statistical independence.

Besides, direct inclusion of balance relations in the evaluation procedure leads to a strong correlation between some resulting evaluated characteristics. For this reason the evaluated data reported without covariance information should be considered as incomplete.

Thus, preparation of this paper was basically motivated by a necessity for the generation of physically consistent evaluated decay data with complete covariance information. The statements of the Federal Programme for Special Purposes

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"Nuclear Power Technologies of New Generation from 2010 to 2015" launched by the Russian government in 2010 have provided an additional stimulus for development of advanced evaluated decay data. According to the Federal Programme new transport and inventory codes must be developed with the option of processing input evaluated covariance data.

In this paper we consider the ^{242}Cm alpha decay as an example, assuming that the structure of the decay scheme of ^{242}Cm is accurately known and use the following conservation laws: (1) a sum of the transition probabilities for particles and gamma quanta feeding any excited level of a daughter nuclide is equal to the sum of the transition probabilities for particles and gamma quanta depopulating the same level, (2) the sum of the transition probabilities for particles and gamma quanta feeding the ground state of a daughter nuclide is equal to 1. The least squares method with restrictions (LSMR) was used as the main mathematical instrument in this evaluation. The self-consistency of the ^{242}Cm evaluated data was provided by using an iterative evaluation procedure.

The paper is organized in the following way. In the second section a statistical model used for the analysis of the experimental data is described. In Section 3 the experimental data available for the evaluation are reviewed. The scheme of the calculations is presented in the fourth section. The results of the evaluation are given in Section 5. Finally, the conclusions are summarized in Section 6.

2. Statistical model

The experimental data of the alpha and gamma emission intensities have been analysed within the framework of the following model. It is assumed that the results of measurements y_i^k are a sum of a model function $f(E_i, \vec{\theta})$ and unbiased random experimental errors, ε_i^k

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

with m restrictions

$$H\vec{\theta} = \vec{d} \tag{2}$$

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

Each equation, in the system of Eq. (1), corresponds to one measurement of the alpha (gamma) emission intensity. The lower index, i, of the variables in the system (1) refers to the number of the alpha (gamma) transition; the upper index, k, to the number of the experiment; E_i^k is the energy of a given alpha (gamma) emission; $K(i)$ is a subset (of dimension n_i) of the indices from the set $(1, 2,\ldots, M)$; $n = \sum_{i=1}^{L} n_i$ the total number of measurements, $n > L$; and M is the total number of experiments. In Eq. (2) $\mathbf{H}$ is a known matrix of dimension $(m \times L)$, and $\vec{d}$ is a known vector of dimension m.

The model function $f(E, \vec{\theta})$ has the form

$$f(E, \vec{\theta}) = \sum_{i=1}^{L} \theta_i \varphi_i(E) \tag{3}$$

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

$$\varphi_i(E_j^k) = \delta_{ij} = \begin{cases} 1 & i = j \\ 0 & i \neq j \end{cases} \tag{4}$$

The covariances of the experimental errors equal

$$\text{cov}(\varepsilon_i^k, \varepsilon_j^l) = V_{ij}^k \delta_{kl} \tag{5}$$

i.e. the measurements correlate only inside the experiment. According to eq. (5) the covariance matrix $\mathbf{V}$ of experimental errors consists of simple submatrices of smaller dimension: the covariance matrices $\mathbf{V}^{\boldsymbol{k}}$, related to individual experiments, are located on diagonal. The elements of the matrix $\mathbf{V}^{\boldsymbol{k}}$ are calculated as a sum of the covariances between the components of the total experimental errors

$$V_{ij}^k = \text{cov}(\varepsilon_i^k, \varepsilon_j^k) = \sum_l \text{cov}((\varepsilon_i^k)_l, (\varepsilon_j^k)_l) \tag{6}$$

on the basis of information given in publications. In Eq. (6) $(\varepsilon_i^k)_l$ is the lth component of the total experimental error. Very often one component of the total experimental error dominates over others. For example, in the case of γ-ray absolute intensity evaluation the uncertainty of the normalization factor dominates over other partial uncertainties.

After substitution of the expressions (3),(4) in the system of Eq. (1) the latter can be presented in a matrix form

$$\mathbf{X}\vec{\theta} + \vec{\varepsilon} = \vec{y} \tag{7}$$

where $\mathbf{X}$ is a matrix of sensitivity coefficients of the model function relative to the parameters

$$X_{ij} = \frac{\partial f(E_i, \vec{\theta})}{\partial \theta_j} \tag{8}$$

The solution $\hat{\vec{\theta}}$ of the system of Eq. (1) without restrictions (2) is calculated by minimizing the function

$$S^2(\vec{\theta}) = (\vec{y} - \mathbf{X}\vec{\theta})^T \mathbf{V}^{-1} (\vec{y} - \mathbf{X}\vec{\theta}) \tag{9}$$

where $\mathbf{V}^{-1}$ is the inverse matrix of $\mathbf{V}$, and superscript T denotes a vector transposition. The expressions for the vector of the evaluated parameters $\hat{\vec{\theta}}$ and its covariance matrix $\mathbf{W}$ are well known

$$\hat{\vec{\theta}} = (\mathbf{X}^T \mathbf{V}^{-1} \mathbf{X})^{-1} \mathbf{X}^T \mathbf{V}^{-1} \vec{y} \tag{10}$$

$$\mathbf{W} = (\mathbf{X}^T \mathbf{V}^{-1} \mathbf{X})^{-1} \tag{11}$$

Covariance matrix $\mathbf{R}$ of the errors of the evaluated values $\hat{y}_i$,

$$\hat{y}_i = f(E_i, \hat{\vec{\theta}}) \tag{12}$$

is given by

$$R = \text{XW}\mathbf{X}^T \tag{13}$$

The system of Eq. (1) with restrictions (2) is solved by the method of Lagrange multipliers. For a function to be minimized

$$\tilde{S}^2(\vec{\theta}, \vec{\lambda}) = (\vec{y} - \mathbf{X}\vec{\theta})^T \mathbf{V}^{-1} (\vec{y} - \mathbf{X}\vec{\theta}) + \vec{\lambda}^T (\mathbf{H}\vec{\theta} - \vec{d}) \tag{14}$$

where $\vec{\lambda}^T = (\lambda_1, \ldots, \lambda_m)$ are the Lagrange multipliers. The function (14) has a minimum at

$$\tilde{\vec{\theta}} = \hat{\vec{\theta}} + \mathbf{WH}^T (\mathbf{HWH}^T)^{-1} (\vec{d} - \mathbf{H}\hat{\vec{\theta}}) \tag{15}$$

$$\tilde{\vec{\lambda}} = -2(\vec{d}^T - \hat{\vec{\theta}}^T \mathbf{H}^T)(\mathbf{HWH}^T)^{-1} \tag{16}$$

The covariance matrix $\mathbf{U}$ of the vector $\tilde{\vec{\theta}}$ is calculated in the following way

$$U = W - W\mathbf{H^T}(\text{HW}\mathbf{H^T})^{-1}\mathbf{HW} \tag{17}$$

The random value $\tilde{S}^2(\vec{\theta}, \vec{\lambda})$ is distributed as χ^2 with $n - L + m$ degrees of freedom.

3. The experimental data

^{242}Cm decays by alpha-emissions to the ground state and 15 excited states of ^{238}Pu which are depopulated by γ-ray transitions. With a probability of ~99.99% ^{242}Cm disintegrates to the ground