



Generation of fission yield covariance matrices and its application in uncertainty analysis of decay heat

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Abstract

The uncertainties and covariance matrices of the fission yield are important in the uncertainty analysis of the decay heat. At present, there are no covariance matrices of fission yield given in the evaluated nuclear data library, although they provide uncertainties with good estimates. In this study, the generalized least squares (GLS) updating approach was adopted to evaluate the fission yield covariances with constraints from the basic physical conservation equation and chain yield data, using the nuclear data files from ENDF/B-VIII.0, JENDL-5, and JEFF-3.3. Based on the original and updated data, summation calculations were performed for the fission pulse decay heat of thermal neutron-induced fission of ^{235}U . The uncertainties of the decay heat were obtained using the generalized perturbation theory, including the uncertainties propagated from the fission yield, decay energy, decay constant, and branching ratio. The original uncorrelated yield data contributed to a $\sim 4\%$ uncertainty at all times, and dominated the decay heat uncertainty at cooling times longer than 100 s. With the generated covariance matrices, the uncertainty of the calculated decay heat was significantly reduced, and the decay energy data generally made a major contribution. The relative uncertainties at cooling time 0.1 s were $\sim 10\%$ for ENDF/B-VIII.0, JEFF-3.3, and $\sim 5\%$ for JENDL-5, and those at cooling time 10^5 s were approximately 1% for the three libraries. The influence of the GLS updating procedure on the contributions of important fission products to the decay heat and their sensitivity coefficients is also discussed.

Keywords Fission yield · Covariance matrix · Decay heat · Uncertainty analysis

1 Introduction

Uncertainty analysis is currently required in nuclear engineering research. As a major contributor to the energy released after a reactor shutdown, the decay heat of fission products is crucial in various aspects of nuclear engineering, such as reactor design, nuclear power plant safety analysis, and nuclear fuel waste management [1–3]. Significant effort has been devoted to the accurate estimation and uncertainty analysis of decay heat, which is typically performed within the framework of the summation method [4–8].

The summation calculations of the decay heat are dependent on the decay and fission yield data, which have been evaluated and compiled in sublibraries by nuclear data libraries such as ENDF/B [9], JENDL [10], and JEFF [11]. However, these libraries contained only the best estimates and uncertainties without covariances for the fission yield and branching ratio. In particular, the covariances of fission yields play an important role in the uncertainty analysis of fission products and can significantly reduce the impact of fission yield on the total uncertainty of the system [12–15].

Several efforts have been made to obtain fission yields using complete covariance matrices. A relatively simple procedure was provided by Katakura [4, 12], in which the chain yields were regarded as approximate mass distributions of independent fission yields and were used as a single constraint. The covariances given by this method exist only between fission products with the same mass number. Furthermore, constraints from experimental data and physical models were applied to generate optimized fission yield

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data and correlations between all isotopes [15–20]. Recently, different types of neural networks have been adopted to estimate fission yields and their uncertainties [21–26], in which complicated data relationships and the propagation of high-order errors can be handled naturally. At present, an international effort coordinated by IAEA is ongoing with the aim of updating and improving the fission yield of actinide nuclides [27].

In this study, we focused on the generation of fission yield covariances in which the constraints from the chain yield data and the physical conservation equation were considered. Covariance matrices are generated using the generalized least square (GLS) updating approach [28] and a slight adjustment is made to the fission yield data simultaneously. Fission pulse decay heat (FPDH) calculations were performed for thermal neutron-induced fission of ^{235}U , which is denoted by $^{235}\text{U}(n_{\text{th}}, f)$. The original data from ENDF/B-VIII.0, JENDL-5, and JEFF-3.3 [9–11] and the corresponding updated data were used. Uncertainty analysis was performed using the generalized perturbation theory [29, 30], including the uncertainties propagated from the fission yield and decay data.

The remainder of this paper is organized as follows: Theoretical methodologies for covariance generation, summation calculation, and generalized perturbation theory are described in Sect. 2. The calculated results and a discussion of the decay heat and its uncertainties are presented in Sect. 3. Finally, the conclusions are presented in Sect. 4.

2 Methodology

2.1 Independent fission yield data and covariance generation

The independent fission yield (IFY) $Y_I(A, Z, M)$ is required in decay heat calculations, which represents the probability of a particular nucleus with mass A , charge Z , and isomeric state M being produced directly from one fission after the emission of prompt neutrons and photons but before the emission of delayed neutrons. The most commonly used general-purpose nuclear data libraries: ENDF/B, JENDL, and JEFF, provide IFY data with uncertainties as standard deviations. ENDF/B-VIII.0 takes binary fission only, whereas JENDL-5 and JEFF-3.3 both include the light products of ternary fission. To date, no correlation between the fission yield has been reported in these libraries.

The IFY data in these libraries are generally provided by empirical models that are constrained by physical conditions and conservation equations, reflecting the general nature of a fissioning system. These properties should hold in the covariance generation for IFYs, corresponding to the constraint conditions in the GLS updating process. In the present work, the following constraints are employed:

(1) Mass and charge conservation

In a real fission event, the compound nucleus (CN) is generally split into two fission fragments in a process called binary fission. These fragments decay into fission products (FPs) by releasing neutrons and photons. Furthermore, ternary fission may occur with a low probability ($\sim 0.2\% - 0.5\%$), producing extra light-charged particles (LCP), such as protons, triton, and α particles [31]. Notably, light-charged particles were also included in the fission products in this study, and the yield data of LCPs and relatively heavy fission products would be updated together within the GLS updating process.

Conservation laws state that the mass and charge numbers of the compound nucleus must be conserved as follows:

$$\sum_i A_i Y_I(i) = \mathbf{A}^t \mathbf{Y}_I = A_{\text{CN}} - \bar{\nu}_p, \quad (1)$$

$$\sum_i Z_i Y_I(i) = \mathbf{Z}^t \mathbf{Y}_I = Z_{\text{CN}}, \quad (2)$$

where A_{CN} and Z_{CN} represent the mass and charge of the compound nucleus, respectively. The mass number, charge number, and isomeric state of i th FP are A_i , Z_i and M_i , respectively. The independent yield is $Y_I(i) = Y_I(A_i, Z_i, M_i)$. $\mathbf{A}$, $\mathbf{Z}$ and $\mathbf{Y}_I$ are the vectors containing the mass number, charge number, and IFY of each FP. The superscript t denotes the transposition of a vector or matrix. $\bar{\nu}_p$ is the average number of prompt neutrons. A_i ranges from 66 to 172 for IFYs in ENDF/B-VIII.0, because only binary fission was considered in this library. The range of A_i will be expanded for the yield data in JENDL-5 and JEFF-3.3 to include light-charged particles. Similar phenomena occur in the charge numbers of the fission products Z_i .

(2) Normalization of IFYs

The sum of IFYs for FPs except LCPs should be 2 by definition, that is

$$\sum_{i \notin \text{LCP}} Y_I(i) = \mathbf{H}_1^t \mathbf{Y}_I = 2. \quad (3)$$

The sensitivity vector $\mathbf{H}_1$ is a unit vector for ENDF/B-VIII.0, whereas it has zero elements for LCPs in JENDL-5 and JEFF-3.3.

(3) Normalization of heavier mass yields

For a set of IFY data, there should be a midpoint mass number A_{mid} such that the yields of FPs on either side of this midpoint are equal to one when the yield data of LCPs are

not counted. As there is a normalization constraint of IFYs to be 2, this condition can be expressed without repetition as

$$\sum_{A_i > A_{\text{mid}}} Y_I(i) = \mathbf{H}_2^t \mathbf{Y}_I = 1, \quad (4)$$

where $\mathbf{H}_2$ is a sensitivity vector with unit coefficients for products with mass numbers A greater than A_{mid} and zero elsewhere. In fact, Eq. (4) is an approximate condition because the mass number is an integer rather than a continuous number. Hence, A_{mid} is set to $(A_{\text{CN}} - \bar{\nu}_p)/2$.

(4) Charge symmetry

In low-energy neutron-induced fission, FP has a low probability of releasing charged particles during its prompt de-excitation. Therefore, the charge distribution of IFYs should be strictly symmetrical if only binary fission occurs, whereas this symmetry is slightly broken by ternary fission. Following the procedure of Mills [32], the charge symmetry condition was applied to the charge number pairs $(Z, Z_{\text{CN}} - Z)$ with relatively large yields:

$$\sum_{Z_i=Z} Y_I(i) - \sum_{Z_i=Z_{\text{CN}}-Z} Y_I(i) = \mathbf{H}_3^t \mathbf{Y}_I = 0, \quad (5)$$

where $\mathbf{H}_3$ is a sensitive vector defined as follows:

$$\mathbf{H}_3(i) = \begin{cases} 1, & Z_i = Z \\ -1, & Z_i = Z_{\text{CN}} - Z \\ 0, & \text{else} \end{cases} \quad (6)$$

In practical calculations, this constraint is used when the charge yields $Y_I(Z)$ and $Y_I(Z_{\text{CN}} - Z)$ are greater than 1%. $Y_I(Z) = \sum_{Z_i=Z} Y_I$.

(5) Chain yield constraints:

The chain yield (ChFY), $Y_{\text{Ch}}(A)$, is defined as the sum of the cumulative yields of the last stable nuclei with the same mass number A and is much more precise than IFY experimental data. A strong negative correlation between different nuclides occurs if ChFY is used as a constraint in the evaluation of IFY.

As noted by England and Rider [33], the chain yields were evaluated after both prompt and delayed neutron emissions. Therefore, the pre-delayed neutron mass yields of IFYs must be corrected to post-delayed neutron chain yields. Based on this definition, ChFY can be calculated as follows:

$$\begin{aligned} Y_{\text{C}}(i) &= Y_I(i) + \sum_j b_{ij} Y_{\text{C}}(j), \\ Y_{\text{Ch}}(A) &= \sum_{A_i=A} d_i Y_{\text{C}}(i), \end{aligned} \quad (7)$$

where Y_{C} denotes cumulative yield. b_{ij} is the branching ratio at which the j th nucleus decays to the i th nucleus. $d_i = 1$ if the i th nucleus is stable, and $d_i = 0$ otherwise. Formally, Eq. (7) can be written in matrix form $\mathbf{Y}_{\text{Ch}} = \mathbf{D}^t \mathbf{Y}_I$ to describe the relationship between the chain yields and independent fission yields. $\mathbf{D}^t = \mathbf{d}^t[\mathbf{E} - \mathbf{b}]^{-1}$, $\mathbf{E}$ denotes the unit matrix. $\mathbf{d}$ and $\mathbf{b}$ are the vector and matrix filled with elements d_i and b_{ij} , respectively.

Currently, there are thousands of radionuclides in the evaluated nuclear data libraries, and their half-lives $T_{1/2}$ vary from $\sim 10^{-20}$ to 10^{20} . However, not all radionuclides should be considered in ChFY calculations. The chain yields are close to the mass distribution of the independent fission yields $Y_I(A) = \sum_{A_i=A} Y_I$ and their differences result from crossing mass chain decay processes in which the parent and daughter nuclides are in different mass chains. In the actual measurement of ChFY, the crossing mass chain decay pathway will have almost no contribution to the divergence between $Y_{\text{Ch}}(A)$ and $Y_I(A)$ if the half-life of the parent nuclide is too long. Hence, choosing a suitable cutoff value for the half-life T_{cut} is practical. The decay process of a radionuclide was considered only when its half-life was less than T_{cut} . Otherwise, the radionuclide is regarded as a stable nucleus. Notably, this half-life cutoff is only valid for ChFY calculations, which implies that the influence of the crossing mass chain decay processes of radionuclides with $T_{1/2} > T_{\text{cut}}$ on the chain yields is negligible. These relatively long-lived radionuclides can still influence the time evolution of the fission product composition and the true value of cumulative yields.

Figure 1 shows the calculated ChFYs for the thermal neutron-induced fission of ^{235}U using the fission yield and decay data in ENDF/B-VIII.0. When $T_{\text{cut}} = 0$, all decay processes were eliminated in ChFY calculations, and the result was equivalent to the mass distribution of the independent fission yields. With increasing T_{cut} , the calculated $Y_{\text{Ch}}(A)$ departs from $Y_I(A)$ gradually and converges at $T_{\text{cut}} = 1$ min. The relative divergence between the converged $Y_{\text{Ch}}(A)$ and $Y_I(A)$ can reach $\sim 10\%$, mainly located in the mass regions $A = 83\text{--}97$ and $135\text{--}140$. These differences were primarily attributed to the crossing mass chain decay processes of radionuclides with half-lives between 1 ms and 1 min. The decay processes of nuclides with $T_{1/2} > 1$ min hardly changed in the chain yields. Hence, $T_{\text{cut}} = 1$ min is adopted

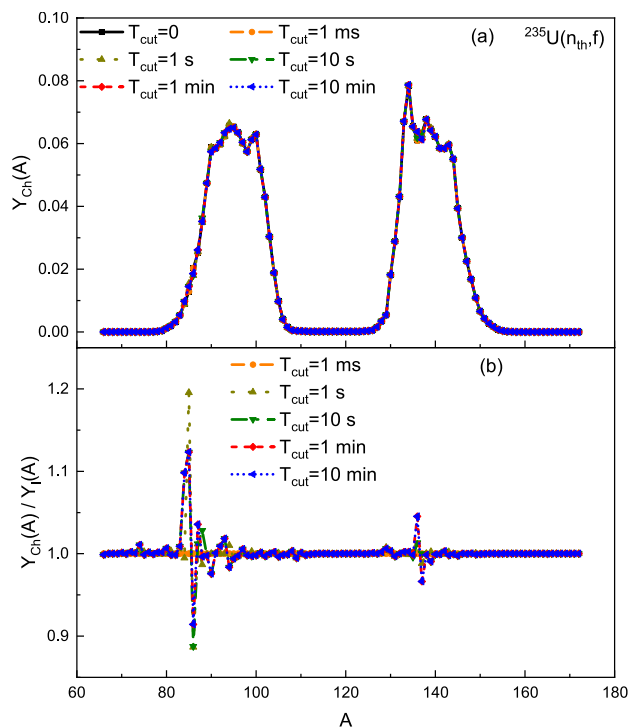


Fig. 1 (Color online) (a) Calculated chain yields for thermal neutron-induced fission of ^{235}U with different half-life cutoff T_{cut} , using the fission yield and decay data in ENDF/B-VIII.0. The result with $T_{\text{cut}} = 0$ is equivalent to the mass distribution of IFYs $Y_1(A)$. Solid, dashed, dotted, dash-dotted, short-dashed, and short-dotted lines represent the results calculated with $T_{\text{cut}} = 0, 1 \text{ ms}, 1 \text{ s}, 10 \text{ s}, 1 \text{ min},$ and 10 min , respectively. (b) The ratio of ChFY calculated with non-zero T_{cut} to $Y_1(A)$. See text for details

in the following calculations: This cutoff is consistent with the evaluation of England and Rider [33], in which the half-lives of the involved delayed neutron precursors were no longer than 1 min.

The generalized least squares method [16, 28] was adopted to estimate the covariance matrices of IFYs. For a certain fissioning system, the independent fission yields in the evaluated library are regarded as prior knowledge of the parameters $\theta_a = Y_1$, along with a diagonal prior covariance matrix V_a whose diagonal elements are taken from the corresponding variances. The constraints above can be summarized as the observation equation $\eta \sim y_a = S^t \cdot \theta_a$, where η is a vector and S denotes the design matrix. In the GLS approach, the prior parameters and covariance can be updated as follows:

$$\begin{aligned} \theta_{\text{upd}} &= \theta_a + V_a S^t [S^t V_a S + V]^{-1} (\eta - y_a), \\ V_{\text{upd}} &= V_a - V_a S^t [S^t V_a S + V]^{-1} S V_a, \end{aligned} \quad (8)$$

where θ_{upd} and V_{upd} are the posterior best estimates and covariance for IFYs, respectively. V denotes the covariance matrix of η .

In this study, V is also a diagonal matrix with diagonal elements obtained from the variances of η . The uncertainties of $\bar{v}_p$ and chain yield were used directly for the mass conservation and chain yield constraints, and fairly small errors were set for the other constraints: 10^{-6} for normalization, 0.5% for charge symmetry, in the order of the probability of ternary fission, and 0.01% for the rest. Notably, this study focuses on fission yield covariance generation and its application in decay heat uncertainty analysis, rather than fission yield data evaluation. Hence, $\bar{v}_p$ and its uncertainty in each library were used in the GLS updating procedure for IFYs in the corresponding library. Similarly, the chain yield data of England and Rider [33] were used as constraints for IFYs in

Table 1 Impact of GLS updating procedure on the constraints for $^{235}\text{U}(n_{\text{th}}, f)$. ORI and UPD denote the results before and after the GLS updating process. The first four quantities are residual errors for the observation equation Eqs. (1), (2), (3) and (4) respectively. The data

$^{235}\text{U}(n_{\text{th}}, f)$	ENDF/B-VIII.0		JENDL-5		JEFF-3.3	
	ORI	UPD	ORI	UPD	ORI	UPD
$A_{\text{CN}} - \bar{v}_p - \sum AY_1$	$-1.42 \times 10^{-2} \pm 3.79$	$3.20 \times 10^{-3} \pm 7.48 \times 10^{-3}$	$-6.40 \times 10^{-3} \pm 3.01$	$-3.00 \times 10^{-3} \pm 6.13 \times 10^{-3}$	$1.00 \times 10^{-2} \pm 4.18$	$3.20 \times 10^{-3} \pm 1.14 \times 10^{-2}$
$Z_{\text{CN}} - \sum ZY_1$	$-5.31 \times 10^{-2} \pm 1.51$	$0 \pm 9.97 \times 10^{-5}$	$-5.45 \times 10^{-2} \pm 1.18$	$0 \pm 1.00 \times 10^{-4}$	$1.60 \times 10^{-4} \pm 1.64$	$0 \pm 1.00 \times 10^{-4}$
$2 - \sum_{i \notin \text{LCP}} Y_1$	$0 \pm 3.46 \times 10^{-2}$	$0 \pm 9.85 \times 10^{-7}$	$0 \pm 2.55 \times 10^{-2}$	$0 \pm 1.00 \times 10^{-6}$	$0 \pm 3.52 \times 10^{-2}$	$0 \pm 9.94 \times 10^{-7}$
$1 - \sum_{A_i > A_{\text{mid}}} Y_1$	$-7.40 \times 10^{-5} \pm 1.72 \times 10^{-2}$	$6.05 \times 10^{-5} \pm 9.55 \times 10^{-5}$	$1.32 \times 10^{-4} \pm 1.76 \times 10^{-2}$	$4.84 \times 10^{-5} \pm 9.41 \times 10^{-5}$	$0 \pm 2.48 \times 10^{-2}$	$0 \pm 9.85 \times 10^{-5}$
χ^2 for charge symmetry	2.23	1.50	10.36	0.80	0	0.01
χ^2 for chain yield	8.48	3.23	9.09	2.52	9.78	0.03

are given in the form $x \pm \delta x$, representing the calculated residual error and its uncertainty, respectively. χ^2 is given for charge symmetry and chain yield constraints. See text for details

ENDF/B-VIII.0, JENDL-5, whereas those of Nichols et al. [34] were used for IFYs in JEFF-3.3, as indicated in Refs. [9, 17, 35]. Uncertainties in branching ratios were not considered in the chain yield constraints.

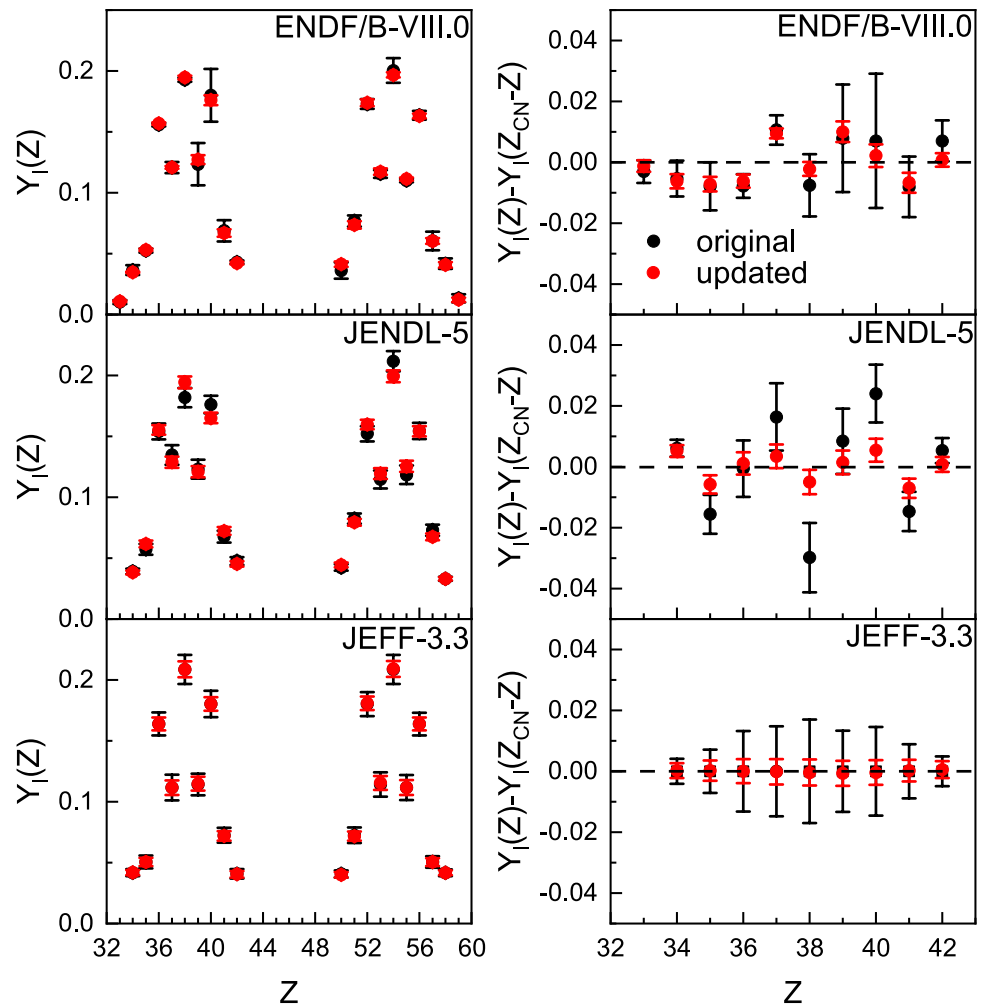
Table 1 lists the impact of GLS updating process on each constraint for the thermal neutron-induced fission of ^{235}U . The residual errors $x = \eta - S^t \cdot \theta$ and their uncertainties $\delta x = \sqrt{S^t \cdot V_\theta \cdot S}$ are calculated for the observation equations Eqs. (1), (2), (3), and (4), where V_θ denotes the covariance matrix of θ . The most significant effect is the reduction in the uncertainties of the residual errors by several orders of magnitude, particularly for the mass and charge conservation constraints. In most cases, the residual errors also decreased after GLS updating process.

For the charge symmetry and chain yield constraints, the quantity χ^2 was adopted to quantitatively assess the improvement, which is defined as

$$\chi^2 = \frac{1}{N} [\eta - S^t \cdot \theta]^t V^{-1} [\eta - S^t \cdot \theta], \quad (9)$$

where N is the length of the vector η . A strong reduction in χ^2 was observed in most cases, except for the charge symmetry constraint for IFY data in JEFF-3.3. This is because the independent yields in JEFF-3.3 [35] were adjusted to ensure the equality of the complement charge yields with higher precision than in the present work. However, this did not affect the effectiveness of GLS updating procedure. Figure 2 shows the original and updated complementary charge yields as well as $Y_I(Z) - Y_I(Z_{\text{CN}} - Z)$ for $^{235}\text{U}(n_{\text{th}}, f)$. For the fission yield data in ENDF/B-VIII.0 and JENDL-5, GLS updating procedure slightly adjusts IFYs to reduce the deviations between $Y_I(Z)$ and $Y_I(Z_{\text{CN}} - Z)$ and generates covariance matrix to decrease the uncertainties of $Y_I(Z)$ and $Y_I(Z) - Y_I(Z_{\text{CN}} - Z)$. For JEFF-3.3, there is a noticeable reduction on the uncertainties of $Y_I(Z)$ and $Y_I(Z) - Y_I(Z_{\text{CN}} - Z)$, while $Y_I(Z)$ themselves remain almost unchanged. Similarly, an improvement in agreement with

Fig. 2 (Color online) (Left panel) Comparison of original and updated charge yields $Y_I(Z)$ for $^{235}\text{U}(n_{\text{th}}, f)$. (Right panel) Comparison of $Y_I(Z) - Y_I(Z_{\text{CN}} - Z)$ calculated with original and updated yield data for $^{235}\text{U}(n_{\text{th}}, f)$. The black and red circles represent the results calculated with original and updated IFYs. The results are plotted only when $Y_I(Z)$ and $Y_I(Z_{\text{CN}} - Z)$ are both larger than 1%. See text for details



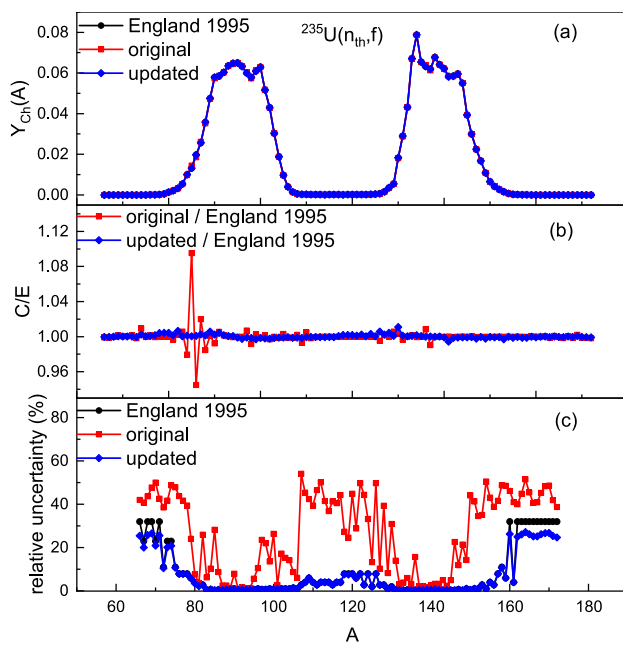


Fig. 3 (Color online) Impact of GLS updating procedure on chain yield for $^{235}\text{U}(n_{th}, f)$ based on the data file from ENDF/B-VIII.0. Evaluation data are taken from England and Rider [33]. (a) Comparison of original and updated chain yields as well as the evaluation values. (b) Ratio of calculated results to evaluation values (C/E). (c) Comparison of relative uncertainties

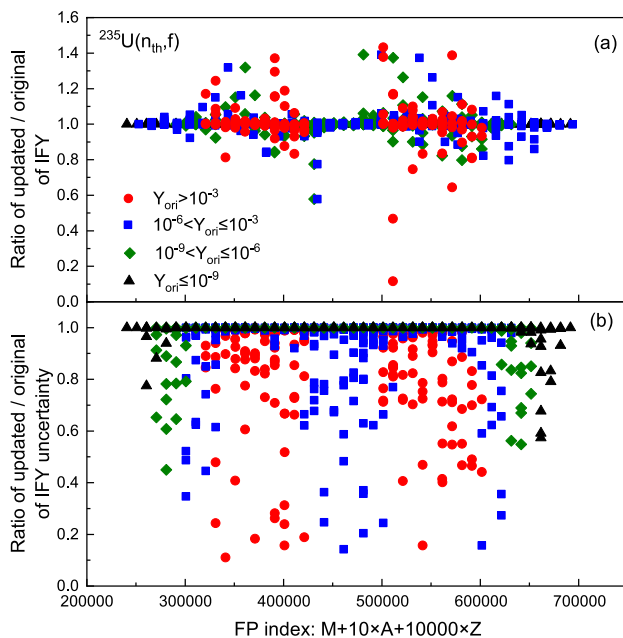


Fig. 4 (Color online) Ratio of updated to original data for (a) IFYs $Y_I(A, Z, M)$ and (b) their uncertainties for $^{235}\text{U}(n_{th}, f)$ based on the data file from ENDF/B-VIII.0. The values are denoted by circles, squares, rhombuses, and triangles when the original IFYs (Y_{ori}) are larger than 10^{-3} , in the regions 10^{-6} – 10^{-3} , 10^{-9} – 10^{-6} and smaller than 10^{-9} , respectively. See text for details

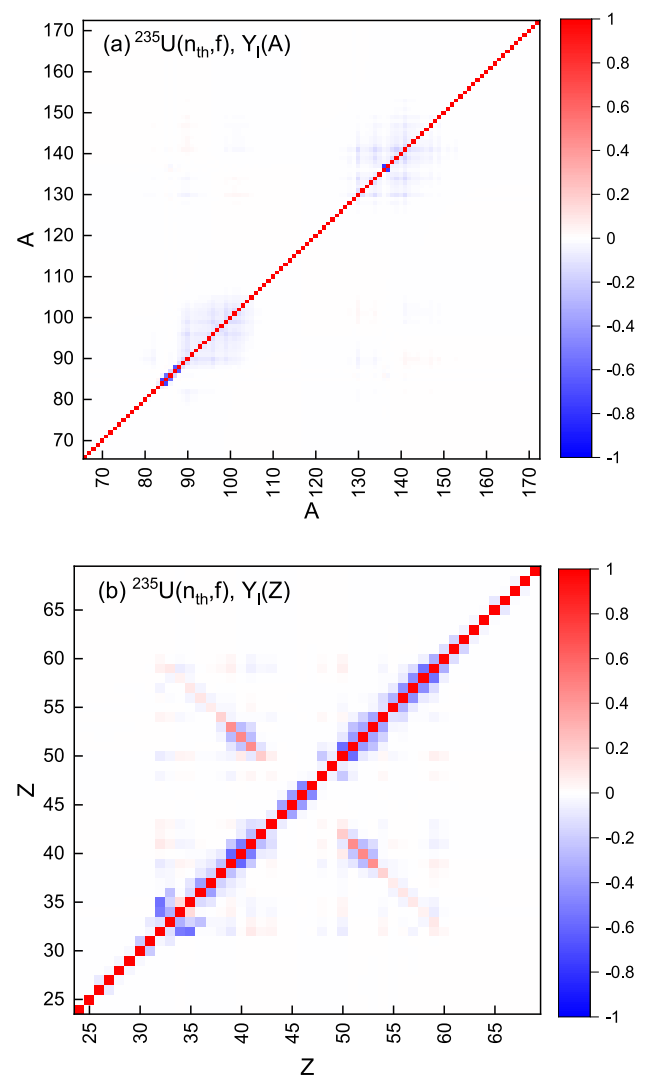


Fig. 5 (Color online) Correlation matrices for (a) $Y_I(A)$ and (b) $Y_I(Z)$ of updated IFYs based on the data file from ENDF/B-VIII.0 for $^{235}\text{U}(n_{th}, f)$

the chain yield data also occurs, along with a reduction in uncertainties, as presented in Fig. 3. Notably, the uncertainties in the calculated chain yields were reduced to no greater than those of the evaluation values in all mass regions after GLS updating process.

As shown in Eq. (8), GLS updating procedure not only generates the correlation of fission yields but also adjusts the yield data. A comparison of the original IFYs in ENDF/B-VIII.0 with updated IFYs is presented in Fig. 4 for thermal neutron-induced fission of ^{235}U . The adjustments are visible only when IFYs have high sensitivity to the constraint system. Strong reductions occur in the variances as they are removed from the diagonal part and reintroduced as correlations between IFYs.

Figure 5 presents the correlation matrices for $Y_1(A)$ and $Y_1(Z)$ of the updated IFYs based on the data file from ENDF/B-VIII.0. The correlation matrix $\bar{V}$ is connected to the covariance matrix V by:

$$\bar{V}_{ij} = \frac{V_{ij}}{\sqrt{V_{ii}V_{jj}}}. \quad (10)$$

It can be observed that there are mainly negative correlations between $Y_1(A)$ and the neighboring mass numbers, and the correlation becomes insignificant with an increase in the difference in mass. Relatively strong negative correlations were observed in the mass regions $A = 84-86$, $87-88$, and $136-137$, which were all located in the regions where the converged Y_{Ch} differed from $Y_1(A)$, as shown in Fig. 1(b). It can be inferred that these relatively strong negative correlations resulted from the inclusion of crossing mass chain decay processes in the chain yield constraint. In contrast to the mass yields, both positive and negative off-diagonal elements exist in the correlation matrix for $Y_1(Z)$, revealing a more complicated dependence structure for the charge yields.

2.2 Summation calculation and generalized perturbation theory

The fission pulse decay heat is the heat generated by radioactive decay after an instantaneous burst of fission occurring at time $t = 0$. FPDH can be calculated using the summation method as follows:

$$H(t) = \sum_i H_i = \sum_i \lambda_i \bar{E}_i N_i(t), \quad (11)$$

where λ_i , $\bar{E}_i$ and N_i are the decay constant, average released energy per decay, and number of i th fission products, respectively. The released decay energy $\bar{E}$ consists of contributions from the average energy released as the kinetic energy of light particle transitions $\bar{E}_{LP}$ (most frequently as β emissions), electromagnetic radiation $\bar{E}_{EM}$, and heavy particles $\bar{E}_{HP}$. The last term has a negligible contribution; therefore, only light particle and electromagnetic decay heat (H_{LP} and H_{EM}) calculations were performed in this study.

The time evolution of $N(t)$ follows the Bateman's equation [36]:

$$\frac{dN_i}{dt} = -\lambda_i N_i + \sum_j \lambda_j b_{ji} N_j. \quad (12)$$

IFYs were used as the initial condition $N_i(t = 0)$. This ordinary differential equation is solved numerically using the algorithm proposed by Ladshaw et al. [37].

The uncertainty of FPDH was carried out with generalized perturbation theory [29, 30]. The variance of the decay heat V_H can be calculated using the sandwich formula as follows:

$$V_H = S_H^t V_\sigma S_H, \quad (13)$$

where S_H is the sensitivity vector of the decay heat with respect to the various nuclear data. The sensitivities of the decay heat to the decay constant and decay energy can be expressed analytically as

$$\begin{aligned} S_H(\lambda_i) &= \frac{\partial H}{\partial \lambda_i} = \bar{E}_i N_i(t), \\ S_H(\bar{E}_i) &= \frac{\partial H}{\partial \bar{E}_i} = \lambda_i N_i(t), \end{aligned} \quad (14)$$

whereas those of the branching ratio and IFY were calculated numerically.

V_σ is the covariance matrix of nuclear data. The covariance of the fission yield was generated in Sect. 2.1. V_σ of the branching ratio is meaningful for the different decay pathways of a single nuclide because of the normalization condition $\sum_j b_{ji} = 1$. For i th nucleus, the covariance matrix for the branching ratios is given by

$$\begin{aligned} V_{jj}^{(i)} &= \sigma_{ji}^2 \left(1 - \frac{\sigma_{ji}^2}{\sum_m \sigma_{mi}^2} \right), \\ V_{jk}^{(i)} &= -\frac{\sigma_{ji}^2 \sigma_{ki}^2}{\sum_m \sigma_{mi}^2}, k \neq j, \end{aligned} \quad (15)$$

where σ_{ji} is the uncertainty of branching ratio b_{ji} . The derivation of this expression is given in Appendix A. The uncertainty of the branching ratio was set to zero for radionuclides with only one decay pathway. V_σ of the decay constant and decay energy are diagonal matrices with diagonal elements taken from their variances. If the uncertainties of λ , $\bar{E}_{LP}$ and $\bar{E}_{EM}$ were not provided in the evaluated library, they were assumed to be 100%.

3 Results and discussions

For each evaluated nuclear data library, two sets of data were used for FPDH calculations.

Set-A: Original fission yield data with decay data.

Set-B: GLS-updates fission yield data with decay data.

No adjustments were made to the decay data in this study.

3.1 FPDH results

The light particles and electromagnetic components of FPDH for thermal neutron-induced fission of ^{235}U are

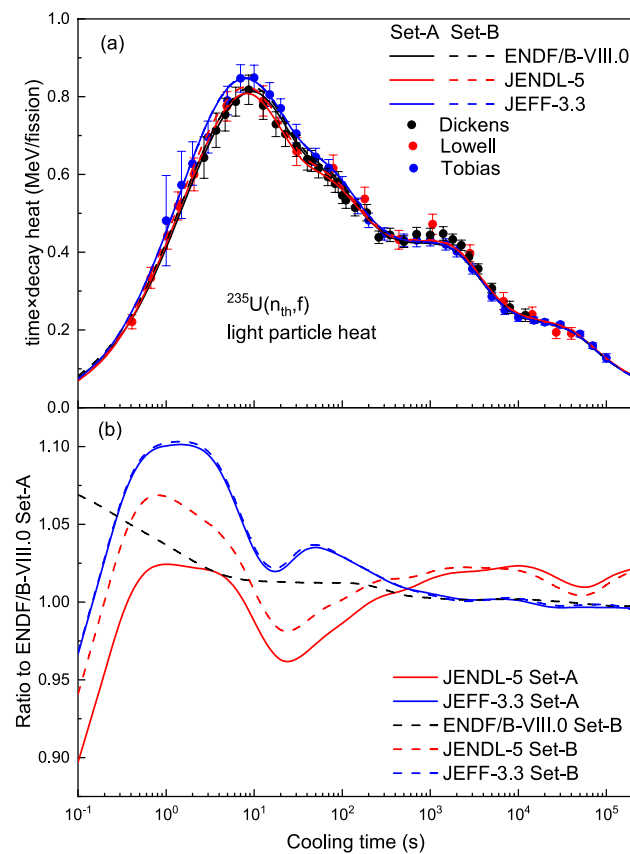


Fig. 6 (Color online) (a) Calculated light particle decay heat for $^{235}\text{U}(n_{\text{th}}, f)$ with Set-A and Set-B. Experimental data are taken from the CoNDERC database [38]. The solid and dashed lines represent the calculated results with Set-A and Set-B respectively. The calculated results based on ENDF/B-VIII.0, JENDL-5, JEFF-3.3 are denoted by black, red, and blue, lines respectively. (b) Ratio of calculated results to that obtained with ENDF/B-VIII.0 Set-A

presented in Figs. 6 and 7, respectively, which were calculated with Set-A and Set-B from ENDF/B-VIII.0, JENDL-5, JEFF-3.3. The experimental data were obtained from the IAEA CoNDERC database [38]. The resulting decay heat curves are in good agreement at cooling times above 1000 s, except that JEFF-3.3 provides a relatively large electromagnetic decay heat during 10^4 – 10^5 s. The discrepancies between the calculated results mainly occur within 1000 s and become significant at cooling times shorter than 1 s. In general, all the calculated results at cooling times below 1000 s are within the error range of the experimental data, except for the underestimation of the electromagnetic decay heat for JEFF-3.3 from 5 s to 50 s.

For both light particles and electromagnetic decay heats, the updated IFYs lead to an enhancement of up to $\sim 5\%$ at cooling times shorter than 1 s and increase the peaks of the curves by approximately 1–2% for ENDF/B-VIII.0 and JENDL-5, whereas the divergences between the calculated

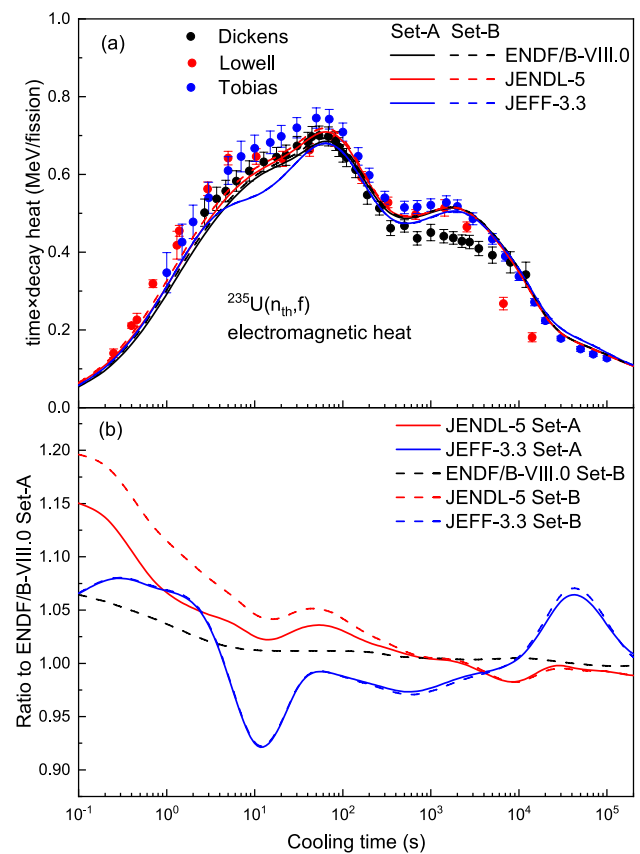


Fig. 7 (Color online) Same as Fig. 6 but for electromagnetic decay heat. Experimental data are taken from the CoNDERC database [38]

results with Set-A and Set-B are small for JEFF-3.3. These differences reveal that the update on IFYs themselves is moderate for ENDF/B-VIII.0 and JENDL-5 but negligible for JEFF-3.3.

3.2 Uncertainties of FPDH

The uncertainties in FPDH were propagated from those of the fission yield, decay energy, decay constant, and branching ratio, as formulated in Sect. 2.2. Computations were carried out with both Set-A and Set-B. To perform an uncertainty analysis with the Set-A data, a diagonal covariance matrix is given for the fission yield data, in which the diagonal elements are taken from their variances.

Figures 8 and 9 show the relative uncertainties of the light particles and electromagnetic decay heat for $^{235}\text{U}(n_{\text{th}}, f)$, including the contributions of each type of nuclear data. The uncorrelated yield data in Set-A provided a $\sim 4\%$ uncertainty in all cases, dominating the total uncertainty for cooling times longer than 100 s. In contrast, the contribution from the yield data is strongly reduced in Set-B and decreases gradually as time evolves, which is a result of the generated covariance matrix.

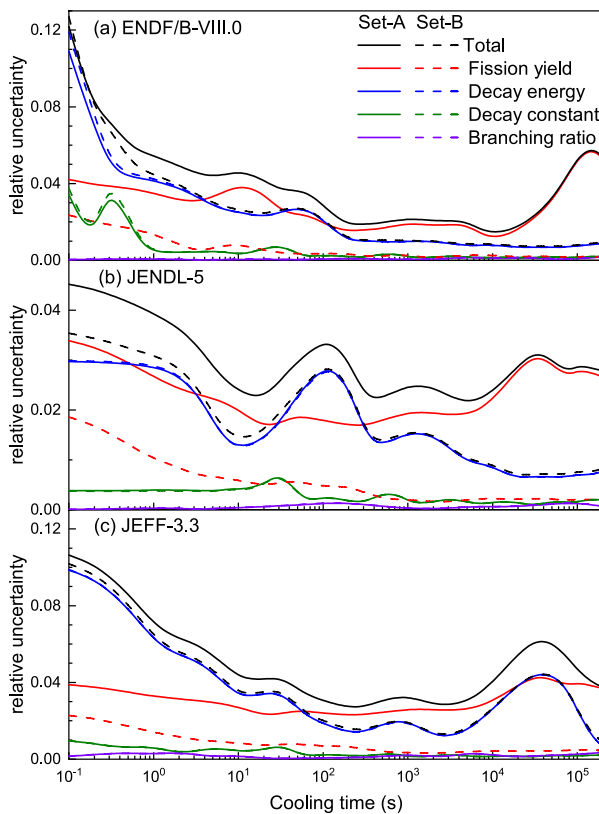


Fig. 8 (Color online) Relative uncertainties of light particle decay heat for $^{235}\text{U}(n_{\text{th}},f)$. The black lines denote the total uncertainty. The red, blue, green, and purple lines represent the contributions from fission yield, decay energy, decay constant, and branching ratio, respectively. The solid and dashed lines represent the results calculated with Set-A and Set-B

The uncertainties propagated from the decay energy, decay constant, and branching ratio are almost unchanged in the Set-A and Set-B results because the decay data have not been adjusted. In general, the decay energy was the primary contributor among the three types of decay data. Notably, the decay energy uncertainty obtained with JENDL-5 is much smaller than those obtained with ENDF/B-VIII.0 and JEFF-3.3 at cooling times shorter than 100 s. This is largely because JENDL-5 provides decay energy uncertainties for more radionuclides than ENDF/B-VIII.0 and JEFF-3.3. These radionuclides are primarily short-lived and release energy during early cooling. Therefore, the assumed 100% uncertainties were adopted less for JENDL-5 decay energy data, resulting in a smaller uncertainty.

With the generated covariance of the yield data, the uncertainties of the light particles and electromagnetic decay heat at cooling time 0.1 s are approximately 10% for ENDF/B-VIII.0 and JEFF-3.3, 5% for JENDL-5. For cooling times longer than 10^5 s, the uncertainties decreased to

approximately 1% for all three libraries. The main component of the uncertainty for the light particle decay heat is the decay energy uncertainty. This situation indicates that the electromagnetic decay heat uncertainty obtained with ENDF/B-VIII.0 and JEFF-3.3, while the uncertainties of yield data and decay energy dominate the total uncertainty of electromagnetic heat at cooling times shorter and longer than 50 s, respectively, for JENDL-5.

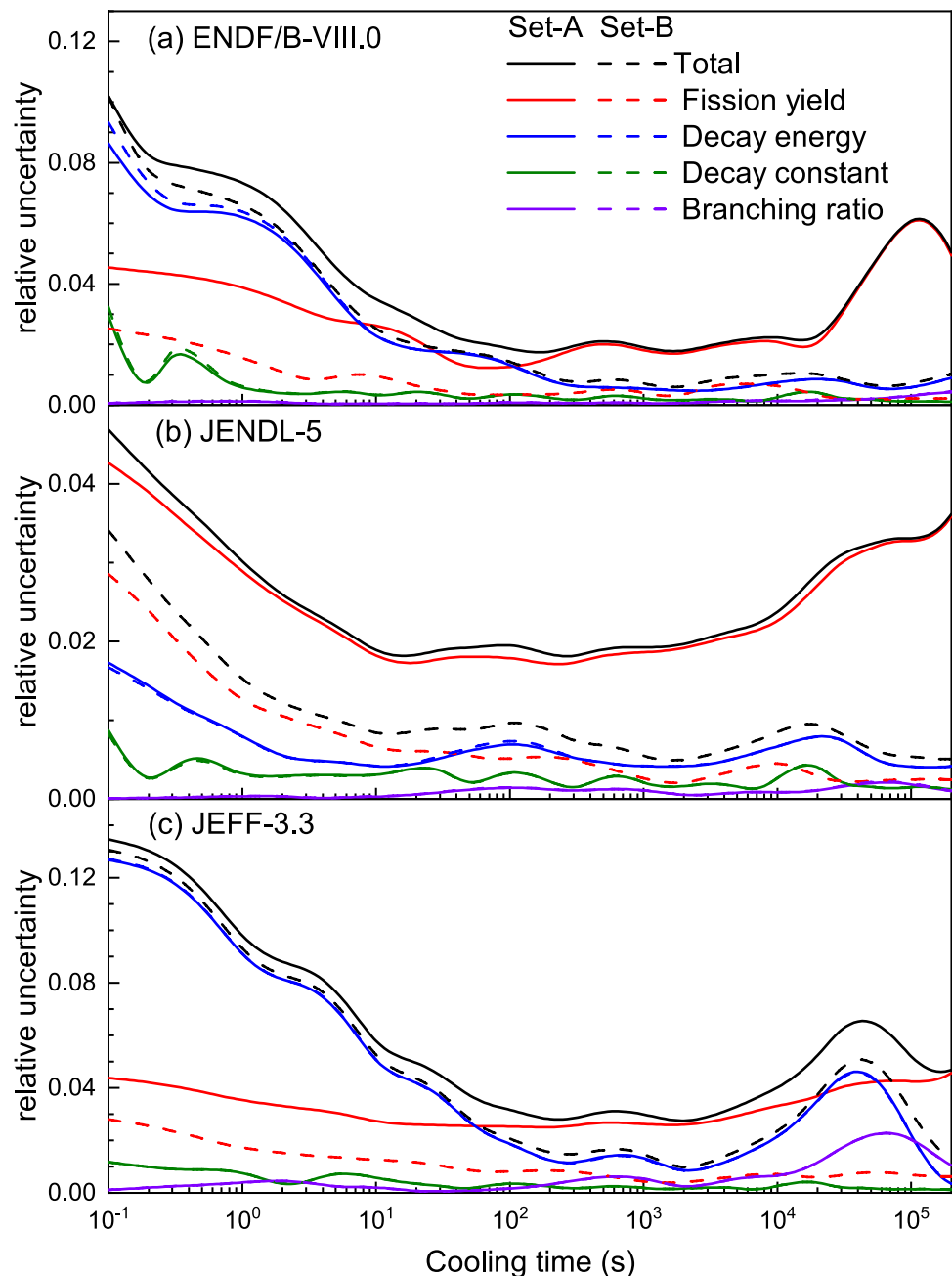
3.3 Contributions of important fission products and their sensitive coefficients

Because GLS updating procedure generates covariance and adjusts IFYs simultaneously, we are concerned about its influence on the contributions of important fission products to decay heat, as well as their sensitivity coefficients. The analysis was performed at a cooling time of 10 s, which is the position of the curve peak for the light particle heat, as shown in Fig. 6. Moreover, an underestimation was given by JEFF-3.3 for the electromagnetic heat around this time, as shown in Fig. 7.

A comparison of the 40 dominant contributors is shown in Fig. 10 for the light particle decay heat. These nuclides were sorted according to their contributions obtained using the ENDF/B-VIII.0 Set-A. In all cases, over 80% of the light particle decay heat was provided by these 40 nuclides at the cooling time of 10 s. In contrast to the reasonable agreement in H_{LP} , visible divergences occurred among the contributions of important fission products obtained using the different evaluated libraries. In particular, the contributions of ^{102}Nb , $^{96\text{m}}\text{Y}$, and $^{146\text{m}}\text{La}$ calculated using JENDL-5 were less than half of those calculated with ENDF/B-VIII.0 and JEFF-3.3, no matter the set of data used. In addition, the adjustment of IFYs has a weak influence on the contributions of ENDF/B-VIII.0 and JEFF-3.3, whereas the contributions of some specific nuclides are clearly affected by JENDL-5, such as ^{97}Y , ^{135}Te , $^{137,138}\text{I}$, and ^{89}Br .

More significant divergences were observed in the electromagnetic decay heat, as shown in Fig. 11. These 40 nuclides were the most significant contributors to the calculations using the ENDF/B-VIII.0 Set-A, and their total contribution accounted for over 75% of the electromagnetic decay heat in all cases. Notably, ^{98}Zr does not contribute to electromagnetic heat if the decay data of JENDL-5 or JEFF-3.3 are used, whereas it contributes 0.8% in the calculations with ENDF/B-VIII.0. This is because $\bar{E}_{\text{EM}}$ of ^{98}Zr is set to zero in JENDL-5 and JEFF-3.3, whereas it is set to 0.449 MeV in ENDF/B-VIII.0. The contributions calculated with JEFF-3.3 are lower than those obtained with the other two libraries for many

Fig. 9 (Color online) Same as Fig. 8 but for electromagnetic decay heat



nuclides, resulting in an underestimation of the electromagnetic heat around the cooling time 10 s. Comparing the results obtained with Set-A and Set-B, the influence of IFYs adjustment is only considerable for JENDL-5, such as ^{138}I , ^{136}Te , and ^{89}Br .

The relative sensitivity coefficient of the decay heat (H) to nuclear data (x) is defined as

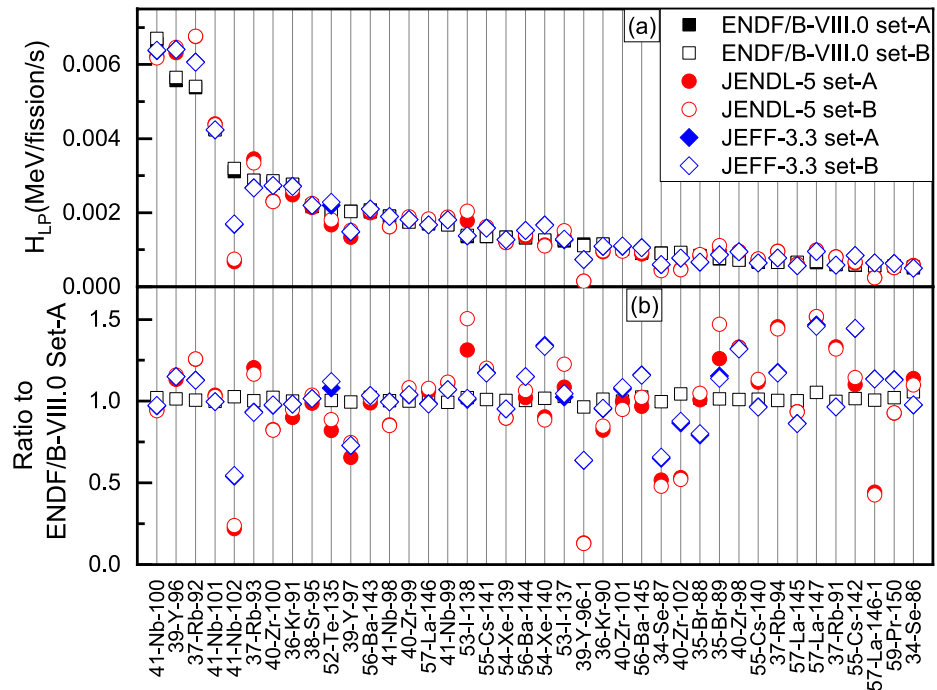
$$s_H(x) = \frac{\partial H/H}{\partial x/x}, \quad (16)$$

which is slightly different from S_H given in Eq. (14). This is discussed in Sect. 3.2, the decay heat uncertainties propagated from the decay data were almost unchanged in the Set-A and Set-B calculations. Therefore, we focused on the sensitivity coefficient relative to IFYs, that is, $s_H(Y_I)$.

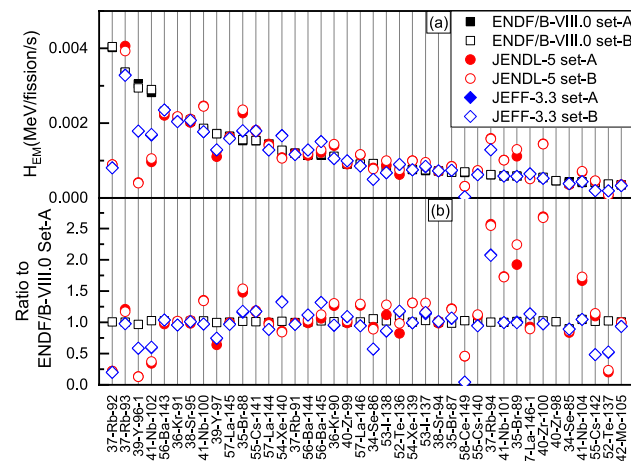
The compositions of $s_H(Y_I)$ are listed in Tables 2 and 3 for the important fission products presented in Figs. 10 and 11 respectively. The nuclide is omitted if its $s_H(Y_I)$ is smaller than 0.01 in all calculations. In general, ^{100}Zr and ^{93}Rb had the highest relative sensitivity coefficients for light particles and electromagnetic decay heat,

Fig. 10 (Color online)

Dominant contributors to light particle decay heat of $^{235}\text{U}(n_{\text{th}},f)$ at cooling time 10s. (a) Contributions of important fission products calculated with different sets of data. (b) Ratio of contribution of important fission product to that obtained with ENDF/B-VIII.0 Set-A. Nuclide is denoted in the form Z-[element name]-A or Z-[element name]-A-M if it is in the ground state or isomeric state (see text for details)



respectively, among the three evaluated libraries. The relative changes owing to the yield adjustment were smaller than 20% for $s_H(Y_i)$ for the two types of decay heat in most cases, revealing a weak influence. However, dramatic changes were observed in the relative sensitivity coefficients of the electromagnetic decay heat calculated using the JENDL-5. For example, the coefficient of ^{96m}Y is enhanced by approximately five times after GLS updating process, whereas those of ^{102}Nb and ^{146}La are reduced by approximately 95% and 65%, respectively.

**Fig. 11** (Color online) Same as Fig. 10 but for electromagnetic decay heat

4 Summary and conclusions

The generalized least squares updating approach was implemented in the covariance generation for $^{235}\text{U}(n_{\text{th}},f)$ independent fission yields in the widely used nuclear data libraries ENDF/B-VIII.0, JENDL-5, and JEFF-3.3, with constraints on the basic physical conservation equation and chain yield data. The variances of the original IFYs were strongly reduced and reintroduced as correlations between IFYs, which were mainly negative correlations. The yield data were also slightly adjusted during GLS updating process.

The original and updated IFYs as well as decay data were used in the uncertainty analyses of decay heat using the generalized perturbation theory, including the uncertainties propagated from the yield and decay data. Good agreement on the decay heat was achieved by the three libraries at cooling times longer than 1000 s. However, discrepancies were found within 1000 s, and a significant underestimation was caused by JEFF-3.3, for electromagnetic decay heat from 5 to 50 s. The adjustment of IFYs had a weak influence on the decay heat, whereas the generated correlation strongly reduced the contribution of the yield data to the decay heat uncertainty. Using the updated yield data with covariance matrices, the uncertainties of decay heat are $\sim 10\%$ for ENDF/B-VIII.0 and JEFF-3.3 and $\sim 5\%$ for JENDL-5 at cooling time 0.1 s, which are approximately 1% for the three libraries at 1×10^5 s, mainly coming from decay energy data. The

Table 2 Relative sensitivity coefficients of light particle decay heat to independent fission yield for important fission products of $^{235}\text{U}(\text{n}_{\text{th}}, \text{f})$ at cooling time 10 s. Coefficients are not presented if their values are both less than 0.01 in the analysis with Set-A and Set-B, and the nuclide is omitted if its $s_H(Y_i)$ is smaller than 0.01 in all cases. The rest are multiplied by 100 for ease of presentation. See text for details

Nuclide	ENDF/B-VIII.0		JENDL-5		JEFF-3.3	
	Set-A	Set-B	Set-A	Set-B	Set-A	Set-B
^{96}Y	-	-	2.02	1.73	-	-
^{92}Rb	3.64	3.59	4.81	4.41	3.58	3.57
^{101}Nb	1.70	1.67	1.78	1.89	1.37	1.34
^{93}Rb	2.98	2.94	3.88	3.64	2.50	2.48
^{100}Zr	10.13	9.78	9.40	9.06	9.01	9.01
^{91}Kr	3.48	3.43	3.46	3.58	3.36	3.35
^{95}Sr	2.31	2.32	2.31	2.38	2.25	2.24
^{135}Te	2.41	2.39	2.02	2.15	2.52	2.62
^{97}Y	1.21	1.17	-	-	-	-
^{143}Ba	1.81	1.82	1.89	1.84	1.70	1.70
^{99}Zr	2.27	2.20	2.45	2.27	2.27	2.25
^{146}La	-	-	1.25	1.18	-	-
^{138}I	1.59	1.59	2.11	2.38	1.48	1.49
^{141}Cs	1.15	1.15	1.36	1.39	1.11	1.10
^{139}Xe	1.43	1.41	1.32	1.26	1.32	1.32
^{144}Ba	1.81	1.79	1.85	1.85	1.85	1.85
^{140}Xe	1.80	1.80	1.57	1.49	2.24	2.23
^{137}I	1.36	1.38	1.58	1.74	1.31	1.34
$^{96\text{m}}\text{Y}$	1.42	1.35	-	-	-	-
^{90}Kr	1.36	1.35	1.17	1.16	1.26	1.26
^{101}Zr	4.29	4.23	4.62	4.27	4.48	4.51
^{145}Ba	1.45	1.47	1.44	1.49	1.57	1.57
^{87}Se	1.12	1.09	-	-	-	-
^{102}Zr	3.65	3.61	2.76	2.64	2.78	2.81
^{89}Br	-	-	1.21	1.39	-	-
^{98}Zr	1.48	1.28	1.39	1.20	1.65	1.63
^{94}Rb	-	-	1.27	1.23	-	-
^{147}La	-	-	1.11	1.07	-	-

influence of yield data adjustment on the decay heat contributions of important fission products and their sensitivity coefficients was also investigated. Evidently, the influence was small for ENDF/B-VIII.0 and JEFF-3.3, while noticeable changes occurred on some important fission products for JENDL-5.

The validity of the present fission yield covariance generation approach was justified, and the updated fission yield data are expected to be applicable in nuclear design and safety analysis together with their covariance matrices.

Covariance matrix for branching ratios

For a set of Gaussian random variables $\{x_1, x_2, \dots, x_n\}$ with mean values $\{\mu_1, \mu_2, \dots, \mu_n\}$ and variances $\{\sigma_1^2, \sigma_2^2, \dots, \sigma_n^2\}$, their joint distribution $f(x_1, x_2, \dots, x_n)$ is given as following if there is a summation constraint $x_1 + x_2 + \dots + x_n = x$,

Table 3 Same as Table 2 but for electromagnetic decay heat. See text for details

Nuclide	ENDF/B-VIII.0		JENDL-5		JEFF-3.3	
	Set-A	Set-B	Set-A	Set-B	Set-A	Set-B
⁹² Rb	3.77	3.72	-	-	-	-
⁹³ Rb	4.90	4.85	6.16	4.79	6.16	5.78
^{96m} Y	5.19	4.95	0.68	3.28	-	-
¹⁰² Nb	-	-	1.60	0.10	1.60	1.71
¹⁴³ Ba	2.78	2.80	2.76	2.93	2.76	2.69
⁹¹ Kr	4.27	4.22	4.22	4.48	4.22	4.36
⁹⁵ Sr	3.02	3.04	2.89	3.25	2.89	2.97
⁹⁷ Y	1.41	1.37	-	-	-	-
¹⁴⁵ La	1.50	1.46	1.36	1.07	1.36	1.25
⁸⁸ Br	2.06	2.04	3.28	2.61	3.28	3.38
¹⁴¹ Cs	1.79	1.79	2.02	1.92	2.02	2.07
¹⁴⁴ La	2.50	2.50	2.01	3.42	2.01	1.91
⁹¹ Rb	1.08	1.06	1.07	1.13	1.07	0.97
¹⁴⁴ Ba	3.25	3.22	3.13	3.61	3.13	3.14
¹⁴⁵ Ba	3.16	3.21	3.27	4.30	3.27	3.39
⁹⁰ Kr	1.85	1.84	2.29	1.92	2.29	2.27
⁹⁹ Zr	1.06	1.03	1.06	1.23	1.06	0.98
¹⁴⁶ La	-	-	1.06	0.37	1.06	1.00
⁸⁶ Se	1.18	1.17	1.65	1.15	1.65	1.56
¹³⁸ I	1.27	1.27	1.38	1.13	1.38	1.56
¹³⁶ Te	1.35	1.46	1.15	1.83	1.15	1.35
¹³⁹ Xe	1.11	1.09	1.45	1.20	1.45	1.38
¹³⁷ I	1.08	1.10	1.30	1.32	1.30	1.43
⁹⁴ Sr	-	-	0.83	1.02	-	-
⁸⁷ Br	-	-	1.07	1.02	1.07	1.06
¹⁴⁹ Ce	1.07	1.07	-	-	-	-
⁹⁴ Rb	1.28	1.27	2.80	2.35	2.80	2.72
⁸⁹ Br	1.02	1.02	1.92	1.05	1.92	2.21
^{146m} La	-	-	0.87	1.18	-	-
¹⁰⁰ Zr	3.53	3.41	5.74	3.51	5.74	5.53

$$f = N \int dx \delta(x_1 + x_2 + \dots + x_n - x) \times \exp \left[-\frac{(x - \mu)^2}{2\sigma^2} \right] \prod_{i=1}^n \exp \left[-\frac{(x_i - \mu_i)^2}{2\sigma_i^2} \right], \quad (\text{A.1})$$

where N is a normalization factor. The sum x is also a Gaussian random variable with a mean value μ and a variance σ^2 .

Owing to the summation constraint, the variance of x_i should be adjusted to:

$$\begin{aligned} \tilde{\sigma}_i^2 &= E(x_i^2) - E^2(x_i) \\ &= \int dx_1 dx_2 \dots dx_n x_i^2 f - \left[\int dx_1 dx_2 \dots dx_n x_i f \right]^2 \\ &= \sigma_i^2 \left[1 - \frac{\sigma_i^2}{\sigma^2 + r} \right], \quad r = \sum_{i=1}^n \sigma_i^2, \end{aligned} \quad (\text{A.2})$$

corresponds to the diagonal element of covariance matrix V_{ii} . The non-diagonal element V_{ij} is given by

$$\begin{aligned}
V_{ij} &= E(x_i x_j) - E(x_i)E(x_j) \\
&= \int dx_1 dx_2 \dots dx_n x_i x_j f \\
&\quad - \int dx_1 dx_2 \dots dx_n x_i f \times \int dx_1 dx_2 \dots dx_n x_j f \quad (\text{A.3}) \\
&= -\frac{\sigma_i^2 \sigma_j^2}{\sigma^2 + r}, i \neq j.
\end{aligned}$$

Because the branching ratios strictly satisfy the normalization condition $\sum_j b_{ji} = 1$, Eq. (A.2) and (A.3) should be used to generate a covariance matrix with $\sigma = 0$ as shown in Eq. (15), which implies that the Gaussian distribution of x approaches its limit and becomes a dirac delta distribution.

Author Contributions All authors contributed to the study conception and design. Material preparation, data collection, and analysis were performed by Wen-Di Chen, Tao Ye, and Hai-Rui Guo. The first draft of the manuscript was written by Wen-Di Chen, and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

Data Availability The data that support the findings of this study are openly available in Science Data Bank at <https://cstr.cn/31253.11.sciencedb.j00186.00958> and <https://doi.org/10.57760/sciencedb.j00186.00958>.

Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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