Application of a Bayesian/generalised least-squares method to generate correlations between independent neutron fission yield data

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Abstract

Fission product yields are fundamental parameters for several nuclear engineering calculations and in particular for burn-up/activation problems. The impact of their uncertainties was widely studied in the past and evaluations were released, although still incomplete. Recently, the nuclear community expressed the need for full fission yield covariance matrices to produce inventory calculation results that take into account the complete uncertainty data.

In this work, we studied and applied a Bayesian/generalised least-squares method for covariance generation, and compared the generated uncertainties to the original data stored in the JEFF-3.1.2 library. Then, we focused on the effect of fission yield covariance information on fission pulse decay heat results for thermal fission of $^{235}$U. Calculations were carried out using different codes (ACAB and ALEPH-2) after introducing the new covariance values. Results were compared with those obtained with the uncertainty data currently provided by the library. The uncertainty quantification was performed with the Monte Carlo sampling technique. Indeed, correlations between fission yields strongly affect the statistics of decay heat.

Introduction

Nowadays, any engineering calculation performed in the nuclear field should be accompanied by an uncertainty analysis. In such an analysis, different sources of uncertainties are taken into account. Works such as those performed under the UAM project (Ivanov, et al., 2013) treat nuclear data as a source of uncertainty, in particular cross-section data for which uncertainties given in the form of covariance matrices are already provided in the major nuclear data libraries. Meanwhile, fission yield uncertainties were often neglected or treated shallowly, because their effects were considered of second order compared to cross-sections (Garcia-Herranz, et al., 2010).

However, the Working Party on International Nuclear Data Evaluation Co-operation (WPEC) – a party dedicated to assessing the needs of nuclear data improvement – raised new interest on fission yield data within its Subgroup 37 (SG37), with the goal of developing...
“Improved fission product yield evaluation methodologies” (Mills, 2013), not only in order to quantify the impact of such uncertainties, but also to provide a proper set of variances and correlation matrices.

In addition, fission yield data are of critical importance in decay heat applications (Katakura, 2012). The calculation of the decay heat and of its uncertainty has a deep impact on a series of industrial challenges like the design of emergency cooling systems, the design of transport waste casks and storage facilities or the cooling time that is needed before maintenance. The uncertainty on decay heat stems from the propagation of variance and covariance values of the nuclear data. Individual fission yield uncertainties, where no correlation is taken, are regarded as the main contributors to the fission pulse decay heat uncertainty (Diez, Cabellos and Martinez, 2011). However, the use of covariance data may have a huge impact on the final result.

The aim of this study was to analyse and apply a Bayesian/generalised least-squares (GLS) method for the generation of fission yield data covariances. Then, we assessed the impact of the correlations on fission pulse decay heat (FPDH) calculations for 235U thermal fission, after generating covariance matrices using the JEFF-3.1.2 library (Kellet, Bersillon and Mills, 2009). The uncertainty quantification (UQ) was performed with the Monte Carlo sampling method.

**Description of fission yields**

Fission yields (FY) characterise the probability of a particular nuclide or mass to be formed after fission. Accurate FY measurements and/or predictions, as well as the knowledge of the carried uncertainties, are essential to many applications in nuclear technology. The widely used general-purpose evaluated nuclear data library JEFF-3.1.2 provides these data in the ENDF-6 format (CSEWG, 2013) along with their uncertainties as standard deviation. To date, no correlation between FY is supplied in such a library, but several institutions/projects are devoting all their energies to developing methodologies to generate full covariance matrices.

There exist different definitions of FY, as outlined below.

The independent fission yield (IFY), \(y(A,Z,I)\), is defined as the number of atoms of nuclide with mass \(A\), charge \(Z\) and isomeric state \(I\) produced directly from one fission after the emission of prompt neutrons, but before the emission of delayed neutrons. Several coefficients need to be known for each fissioning system to calculate IFY, but even those chains with the highest coverage of measured data do not provide values for all the parameters. It is indeed necessary to resort to semi-empirical models and interpolation/extrapolation methods.

The cumulative fission yield (CFY) \(C(A,Z,I)\) is the total number of atoms of nuclide with mass number \(A\), charge \(Z\) and isomeric state \(I\) produced over all time after one single fission. That is, the total number of atoms of that nuclide generated both through one single direct fission and radioactive decay of all the precursors. CFY have a strong relationship with fission products decay chains, which means that they can be calculated from IFY and decay data branching fractions using the so-called “Q-matrix” approach (James, Mills and Weaver, 1991), represented by Eq. (1):

\[
C_j = \sum_i Q_{ij} y_i
\]  

where \(Q_{ij}\) are the decay branching ratios from isotope \(i\) to \(j\). Therefore, the Q-matrix is the matrix of the decay branching ratios that steers fission products toward stable nuclides.

The chain fission yield (ChFY) \(Ch(A)\) is defined as the sum of cumulative yields of the last stable or long-lived chain members with same mass \(A\) and is obtained in classical mass spectrometric measurements of long-lived and stable end products of mass chains.
The term "chain yield" has been commonly used to describe both the sum of cumulative yields of the last stable or long-lived chain members, and the isobaric sum of independent yields [mass fission yields (MFY) $Y(A)$]:

$$Y(A) = \sum_{i} Y_{i}$$

(2)

The two definitions, even if slightly, may differ by a few per cent as the second does not include the contribution of delayed-neutron emission (Mills, 1995). However, for the purpose of this work, ChFY and MFY are treated indistinctly.

Figure 1 shows that the evaluated MFY (or ChFY) stored in the libraries are in good agreement with those calculated with Eq. (2). However, the associated uncertainties show discrepancies. Uncertainties on the total MFY are calculated by means of simple propagation through the sum of IFY, assuming no correlation between different IFY belonging to the same chain [Eq. (3)]:

$$\Delta Y(A) = \sqrt{\sum_{i} (\Delta Y_{i})^2}$$

(3)

The gap between calculated and evaluated MFY uncertainties may arise from the lack of correlations between IFY, highlighted in Eq. (3). Since a similar behaviour also occurred in the calculation of CFY uncertainties, the implementation of a covariance matrix generation method appears as an urgent need.

**Figure 1: Evaluated and calculated mass yield distribution and uncertainties for $^{235}$U thermal fission products from the JEFF-3.1.2 database**
Bayesian/generalised least-squares method

Great efforts have been committed to developing methodologies for correlation generation (full covariance matrices) for IFY data. Katakura (2012), followed by Kawano and Chadwick (2013), proposed a Bayesian/generalised least-squares (GLS) method, where the IFY covariance matrix is updated with information on the chain yields. These proposals, together with a variation in which the IFY covariance matrix is updated with CFY uncertainties, are described and reported hereunder.

The Bayesian/generalised least-squares (GLS) method is an adjustment technique which states that the information on some prior system parameters can be improved with the addition of new knowledge — new data — (e.g. experimental or evaluated response values $\eta$), for which relationships between data and parameters are established [Eq. (4)]. These relationships, or constraints, must be linearised in the form:

$$y - y_a = S(\theta - \theta_a)$$

(4)

where $\theta$ are the parameters of the system, $\theta_a$ the prior estimates of $\theta$, $y$ the responses of the constraining equation, $y_a$ the responses of the constraining equation to the prior estimates $\theta_a$ and $S$ are the sensitivity coefficients of the response $y - y_a$ to the parameters $\theta - \theta_a$. It was assumed that no correlation existed between the prior and the new information. Then, further information $\eta$ could be introduced in order to derive refined values for the parameters $\theta$, with all the available uncertainty information properly incorporated into the formalism. The updating process is represented by Eqs. (5) and (6):

$$V_a = V_a - V_a S (S V_a S + V)^{-1} V_a$$

(5)

$$V = V - V S V (S V_a S + V)^{-1} S V_a$$

(6)

where $V_a$ is the covariance matrix of the prior estimates of the parameters $\theta_a$, $V$ is the covariance matrix of the introduced data fitting the constraining system $\eta$, and $V_s$ is the updated covariance matrix of the system parameters ($\theta$). Superscript $t$ refers to the transpose of a vector or a matrix.

It has already been proposed to update the covariance matrix with the evaluated MFY variance information. In such a case, $S$ becomes the array of sensitivity coefficients of MFY to IFY, $\eta$ the evaluated MFY introduced in the system and $y_a$ the MFY calculated with the prior IFY ($\theta_a$) in Eq. (4), that is, summing up all the yields belonging to the same chain. $V_a$ and $V$ are the variance matrices of the prior $\theta_a$ and of the experimental MFY, respectively.

A new proposal introduced in this current paper and developed by the authors is the use of the CFY evaluated data to update the variance matrix of the IFY. Then, $\eta$ becomes the evaluated CFY with variance matrix $V$, $y_a$ is the array of CFY calculated with Eq. (4), where $\theta_a$ is the vector of prior IFY and $V_a$ its variance matrix. Here, the Q-matrix equation [Eq. (1)] is the linear constraining system and $S$ are the sensitivity coefficients of IFY to CFY.

Uncertainty quantification on FPDH calculations

FY data and uncertainties have a major role in fission pulse decay heat (FPDH) calculations and correlations may significantly influence the decay heat uncertainty. In such cases, uncertainty quantification (UQ) studies come to hand to assess this impact. Here, we generated covariance data by means of the Bayesian/GLS method previously
described, using uncertainties from the JEFF-3.1.2 database. The UQ on the FPDH calculation for $^{235}\text{U}$ thermal fission follows.

The fission pulse decay heat is the heat generated by the radioactive decay after a single atom of a specific material fissions. Accurate calculations of such values assume a capital importance in reactor operation strategies as the residual heat, which inevitably follows the reactor shutdown, is one of the most important parameters for reactor safety. FPDH was calculated for a thermal fission event of $^{235}\text{U}$: radioactive decay and fission yield data were taken from the library, whereas cross-sections do not take part in this kind of calculation. The time evolution of radioactive material subject to pure decay is described by the system of ordinary differential equations (ODE) in Eq. (7):

$$\frac{dN_i}{dt} = -\lambda_i N_i + \sum_j \lambda_j \beta_{ji} N_j$$  \hspace{1cm} i = 1, \ldots, M \tag{7}$$

where $\lambda_i$ are the decay constants, $N_i$ the concentrations of isotopes involved in the calculation, $\beta_{ji}$ is the branching ratio which indicates the decay mode and the fraction of decays that converts isotope $j$ into $i$, and $M$ is a finite integer, that is, the size of the system. The initial composition, $N(t = 0)$, is the same FY distribution given in the library for $^{235}\text{U}$ thermal fission.

The decay heat is calculated with Eq. (8), with the isotopic inventory being followed throughout the whole cooling time:

$$DH = \sum_i DH_i = \sum_i \lambda_i N_i \left( \sum_j \beta_{ji} E_j \right)$$  \hspace{1cm} (8)$$

Here, $E_j$ is the average released energy for the corresponding decay model given by the $\beta_{ji}$ branching ratio.

Two codes were used to perform these calculations:

- ACAB (Sanz, Cabellos and Garcia-Herranz, 2008) is an activation/transmutation code that solves the general nuclear transmutation chains for multi-dimensional neutron flux distributions. Its ODE solver is based upon the ORIGEN algorithm (Isotalo and Aarnio, 2011), which uses a truncated Taylor series expansion of the exponential matrix, from which all the short-lived nuclides are removed and handled separately with the Gauss-Seidel iterative method under an assumption of secular equilibrium.

- ALEPH (Van den Eynde, et al., 2013) is a general-purpose burn-up code created and developed at the SCK•CEN Belgian Nuclear Research Centre. Its new release ALEPH-2 has decay heat and FPDH calculation capabilities and resorts to the highly accurate Runge-Kutta method RADAU5 (Hairer and Wanner, 1980), as an inherent routine, to solve the system of depletion equations.

We carried out a Monte Carlo sampling to quantify the impact of IFY uncertainties on FPDH. One thousand i.i.d. random samples from normal probability density functions (PDF) granted the convergence of the mean and standard deviation of the IFY. Best-estimate and uncertainty values stored in JEFF-3.1.2 represented the mean and standard deviation of the PDF, respectively. Since small IFY generally carry high uncertainties, the random sampling of a negative value is likely to happen. When it occurred, such random samples were set to zero. Then, a small bias could be expected from the use of truncated normal PDF, still negligible for decay heat calculation purposes.

For each draw, a full FPDH calculation was performed and 1 000 different response functions were retrieved. Then, through a statistical analysis of the response functions, it was possible to gather information such as PDF, mean and standard deviation of the FPDH.
Results and discussion

Plots and results of FPDH calculations for $^{235}$U thermal fission with UQ are presented here. Monte Carlo sampling is performed with 1 000 samples to guarantee the convergence of IFY means and standard deviations. Full simulations are carried out using both ACAB and ALEPH codes, making sure to keep full consistency on all the data treated in the system. A list of the performed calculation is presented as follows:

I. Total FPDH calculation with variance matrix without correlations (JEFF-3.1.2 + no corr.).

II. Total FPDH calculation with correlation matrix generated with Bayesian/GLS method and mass chain yield information [JEFF-3.1.2 + COV(ChFY)].

III. Total FPDH calculation with correlation matrix generated with Bayesian/GLS method and cumulative yield information [JEFF-3.1.2 + COV(CFY)].

IV. Total FPDH calculation with correlation matrix generated with Bayesian/GLS method and cumulative yield information; we allowed only for the diagonal terms of the matrix [JEFF-3.1.2 + COV(CFY), only diag.].

To guarantee coherence in the calculations, all the simulated values have been compared with Tobias’ (1989) compiled data.

As expected, for pure decay systems, the discrepancy related to the use of different codes – ACAB or ALEPH – is negligible in FPDH calculations.

Results obtained with data from the JEFF-3.1.2 library are reported in Figure 2. Uncertainties, in the form of relative standard deviations, are plotted as functions of the decay time and sketched together with Tobias’ experimental uncertainties (black line). The red curve (squares) is calculated using non-correlated fission yields (I). The blue curve (circles) corresponds to calculation II, where information on evaluated mass fission yields is introduced from the IAEA (1974). The orange line (diamonds) shows results for calculation III.

The full covariance matrices for IFY contribute to strongly reducing the uncertainty on decay heat. A glance at Eq. (6) immediately explains this behaviour; evaluated uncertainties in the right-hand term of the equation cut down the diagonal terms of the FY covariance matrix. This effect is more enhanced when the new data introduced in the system carry uncertainties that are smaller than the prior parameters. This happens because the piece of information introduced in the system in order to update the prior IFY data is more powerful when the variance of the new data is much smaller than that of the calculated observables, so that the evaluated new data are dominant. Strong variance reductions also occur when the uncertainty of a single parameter (IFY) has a very high sensitivity to the constraining system in Eq. (4). Hence, most of the uncertainty is removed from the diagonal to be reintroduced as negative correlation between IFY. Small adjusted uncertainties of IFY inevitably contribute to reduce the uncertainty on FPDH, as well. However, reduced IFY variances are not the only contributors to this effect.

FPDH in the purple line (triangles) is obtained using the Bayesian/GLS method with CFY evaluated data, this time with off-diagonal terms of the IFY covariance matrix cancelled out (IV); only the contribution of the main diagonal is taken into account. The purple curve has a shape similar to the red line, that is, when IFY are not correlated, but the uncertainty suffers a sharp reduction.

However, such a reduction is not sufficient to justify the uncertainty values obtained with IV, which means that not only adjusted diagonal terms (variances), but also correlations between IFY affect the uncertainty of the decay heat introducing negative contributions.
Conclusion

Fission yields are catalogued amongst the sources of uncertainties in the widely-used nuclear data library JEFF-3.1.2, however the lack of correlations impacts on the reliability and consistency of the library itself and its use for practical applications. A Bayesian/generalised least-squares method was applied to generate covariance matrices for independent fission yields. We performed our study on the $^{235}$U thermal IFY stored in the JEFF-3.1.2 database, introducing evaluated CFY and MFY data. Then, we tested the effect of such correlations on a fission pulse decay heat problem.

The implementation of Bayesian/generalised least-squares method provided negative correlations between IFY and simultaneously reduced the IFY variance values. Correlations on IFY levelled off the discrepancies on evaluated and calculated fission yields, thus justifying the covariance generation methodology. The uncertainty quantification of FPDH results proved that the use of non-correlated IFY overestimates the decay heat uncertainty.

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