Evaluation of the ^{239}Pu Prompt Fission Neutron Spectrum Induced by Neutrons of 500 keV and Associated Covariances

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Abstract

We present evaluations of the prompt fission neutron spectrum (PFNS) of ^{239}Pu induced by 500 keV neutrons, and associated covariances. In a previous evaluation by Talou et al. 2010, surprisingly low evaluated uncertainties were obtained, partly due to simplifying assumptions in the quantification of uncertainties from experiment and model. Therefore, special emphasis is placed here on a thorough uncertainty quantification of experimental data and of the Los Alamos model predicted values entering the evaluation. In addition, the Los Alamos model was extended and an evaluation technique was employed that takes into account the qualitative differences between normalized model predicted values and experimental shape data. These improvements lead to changes in the evaluated PFNS and overall larger evaluated uncertainties than in the previous work. However, these evaluated uncertainties are still smaller than those obtained in a statistical analysis using experimental information only, due to strong model correlations. Hence, suggestions to estimate model defect uncertainties are presented, which lead to more reasonable evaluated uncertainties. The calculated k_{eff} of selected criticality benchmarks obtained with these new evaluations agree with each other within their uncertainties despite the different approaches to estimate model defect uncertainties. The k_{eff} one standard deviations overlap with some of those obtained using ENDF/B-VII.1, albeit their mean values are further away from unity. Spectral indexes for the Jezebel critical assembly calculated with the newly evaluated PFNS agree with the experimental data for selected (n, γ) and (n,f) reactions, and show improvements for high-energy threshold (n,2n) reactions compared to ENDF/B-VII.1.

Keywords: Nuclear data evaluation, ^{239}Pu prompt fission neutron spectrum, uncertainty quantification

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1 Introduction

We present evaluations of the energy spectrum of prompt neutrons emitted in the fission process — the so-called prompt fission neutron spectrum (PFNS) — of ^{239}Pu induced by neutrons of 500 keV and associated covariances. The ^{239}Pu PFNS is a quantity of high interest for nuclear applications [1,2] and it is currently studied as part of an IAEA coordinated research project [3,4] as well as the international CIELO project [5]. The latter project aims to provide initially high-fidelity evaluated nuclear data for ^1H , ^{16}O , $^{235,238}\text{U}$ and ^{239}Pu . For this project, evaluated ^{239}Pu PFNS data and covariances are needed from thermal to 30 MeV. These evaluated data should include new experimental evidences [6,7] and improved modeling compared to the ENDF/B-VII.1 evaluation [8]. The work presented here is the first step towards this goal.

We chose to evaluate at an incident energy of 500 keV to compare to a previous evaluation [9] of the $^{239}\text{Pu}(n_{500\text{keV}},f)$ PFNS which provided covariances for ENDF/B-VII.1 and obtained very similar PFNS values compared to ENDF/B-VII.0. This earlier work was extended in Ref. [10] to suites of uranium and plutonium isotopes. Both evaluations include model information from the Los Alamos model (LAM) [11] as well as experimental knowledge. In both works, the model and experimental covariance matrices were estimated based on simplifying assumptions that were in part responsible for surprisingly low evaluated relative uncertainties [12], an order of magnitude lower than any experimental input uncertainty. The evaluated uncertainties of [9] were scaled a-posteriori by a factor to roughly correspond to the minimum experimental relative uncertainties.

In this paper, evaluated data and covariances are also obtained by a statistical analysis of experimental data from thermal to 500 keV incident neutron energies, LAM predicted values, and associated covariances. However, in order to obtain more reasonable evaluated covariances and data, (1) a detailed uncertainty quantification of experimental data has been undertaken and is briefly presented in Section 3.1, (2) uncertainties of more LAM parameters have been taken into account in Section 3.2.1, (3) the LAM has been extended following suggestions in [9] (Section 3.2), (4) an improved evaluation technique has been employed, as described in Section 2, taking into account the qualitative differences between model predictions and experimental data, and (5) model defect uncertainties have been considered in the prior covariances to weaken correlations computed using the LAM (Section 3.2.2). The evaluated PFNS and covariances are shown in comparison with ENDF/B-VII.1 data in Section 4.1. It is shown that the evaluated results obtained here are somewhat insensitive to the choice of the approach to estimate model defect uncertainties for the prior. The associated k_{eff} and spectral indexes for the Jezebel critical assembly differ little. The evaluated results are consistent with experimental data and uncertainties in Section 4. In addition, the impact of this new evaluation on selected critical assembly and reaction rates calculations is discussed in comparison to using ENDF/B-VII.1 and the experimental benchmarks.

2 Evaluation and Uncertainty Quantification Methods

2.1 Qualitative Difference between Experimental Data and Model Values

The input quantities for the evaluation procedure are model calculations from the LAM [11], experimental PFNS, and their associated covariances. LAM predicted values and experimental data differ in so far as the former are inherently normalized and given at a defined outgoing neutron energy, while experimental data are not-normalized, bin-averaged quantities.

The LAM values are normalized such that the integral of LAM PFNS over the whole outgoing neutron energy range is unity. This normalization constraint agrees with the ENDF-6 format requirement on evaluated PFNS data [13],

$$\sum_{i=1}^m \psi(\mathcal{E}_i) \Delta \mathcal{E}_i = 1, \quad (1)$$

with the bin-averaged PFNS $\psi(\mathcal{E}_i)$ for a bin-averaged outgoing neutron energy $\mathcal{E}_i$ with bin-width $\Delta\mathcal{E}_i$. The sum over i covers an outgoing neutron energy range from 10^{-5} to approximately 30 MeV.

Due to the normalization constraint on the PFNS, a constraint is correspondingly imposed on the PFNS covariances $\text{Cov}(\psi(\mathcal{E}_i), \psi(\mathcal{E}_j))$, namely that their rows or columns sum to zero. This constraint is known as the ‘zero-sum-rule’. The ENDF-6 format requires that evaluated covariances also obey

$$\sum_i \text{Cov}(\psi(\mathcal{E}_i), \psi(\mathcal{E}_j)) / \psi(\mathcal{E}_j) < 10^{-5}, \quad (2)$$

making allowances for the finite computational accuracy.

The normalization constraint on the PFNS in Eq. (1) was introduced, because the physically observed prompt fission neutron yield is partitioned into two quantities: (1) The neutron multiplicity $\bar{\nu}$ gives the average number of neutrons emitted in the fission reaction. (2) The PFNS gives the probability distribution function for the neutrons to be emitted at a certain outgoing neutron energy. The neutron multiplicity is measured in independent measurements, typically using 4π detectors, with better accuracy and precision than in any PFNS measurement. Hence, $\bar{\nu}$ and PFNS are usually evaluated independently.

PFNS measurements cannot provide the total number of outgoing neutrons over all outgoing neutron energies to the same precision as dedicated measurements for $\bar{\nu}$. At low outgoing neutron energies up to 2 MeV, multiple scattered neutrons, and above 10 MeV, counting statistics limit the precision of the measured data substantially. Experimental data can therefore often not be normalized over the whole energy range and they are treated here as shape data in the evaluation procedure. The scaling of each experimental data set is chosen such that it is comparable with all other input data sets [14].

This qualitative difference of normalized model predicted values and not-normalized experimental data has a quantifiable impact on the evaluation. Preliminary results of a ^{235}U evaluation [15] showed that a bias is introduced in the evaluated results if the experimental data are not treated as shape data, and are not properly scaled with respect to all the other input data. Also, if the experimental data are not scaled prior to the evaluation procedure, the χ^2 of the evaluation input data can be very high, reflecting mainly the discrepancies of scaling rather than discrepancies in the shape. Here, experimental covariances do not satisfy the normalization condition, while LAM covariances do. In Refs. [12, 14], it is shown that the evaluated covariances do not differ if the evaluated covariances are normalized or if the normalization condition is introduced through normalized model covariances, apart from non-linear effects. We prefer to work with both not-normalized model and experimental covariances, and enforce the normalization condition as the last step of the evaluation. This process allows us on the one hand to study the impact of the normalization condition on the evaluated covariances, which usually leads to an overall reduction of uncertainties and on the other hand, to compare evaluated uncertainties using primarily experimental information without the influence of the normalization condition. This comparison is of importance to quantify whether the evaluated uncertainties using also model values and covariances are reasonable given experimental information.

The second qualitative difference stems from the fact that in a PFNS experiment, neutrons emitted from the fission target are counted for certain time-of-flight (TOF) bins which are then converted into counts per outgoing neutron energy (i.e., bin-averaged). Contrary to experimental bin-averaged PFNS, models like the LAM predict a PFNS at a certain outgoing neutron energy (i.e., point-wise). This qualitative difference might yield — depending on the binning size and especially at high outgoing neutron energy — values different than the corresponding bin-averaged values. This issue is discussed in Appendix B of [11]. Hence, model values need to be converted to bin-averaged values to resolve this qualitative difference.

The difference between bin-averaged PFNS and PFNS for one specific outgoing neutron energy is pointed out in this paper by using the variables ϕ and $\mathcal{E}$, respectively for bin-averaged PFNS and bin-averaged outgoing neutron energies. The variable N is used for the PFNS at one specific outgoing neutron energy E in the laboratory frame. The variable ψ is used for normalized, bin-averaged PFNS

obeying Eq. (1), while ϕ is not-normalized. The superscripts y^M , y^x and y^{post} of a variable y are used to distinguish between model, experimental, and evaluated quantities. Vectors of y are indicated with $\underline{y}$, while matrices $\mathbf{y}$ are given in bold and ϕ_i is used as a short notation for $\phi(\mathcal{E}_i)$.

2.2 Experimental Covariance Estimation

2.2.1 Data Preparation

Experimental data are interpreted as shape data and they need to be scaled with a constant factor for the evaluation process such that they are in reasonable agreement with model values.¹ The original experimental data $\tilde{\phi}^x$ are scaled by²,

$$\phi^x(\mathcal{E}_i) = \tilde{\phi}^x(\mathcal{E}_i) \frac{\sum_j \phi^M(\mathcal{E}_j) \Delta \mathcal{E}_j}{\sum_j \tilde{\phi}^x(\mathcal{E}_j) \Delta \mathcal{E}_j}, \quad (3)$$

where the index j runs over all bin-averaged outgoing neutron energies of one specific experimental data set where $\tilde{\phi}^x$ is assumed to be well-known. Equation (3) leads to a different scaling factor for each experimental data set depending on the initial scale chosen for this experiment. The variable $\phi^M(\mathcal{E}_j)$ corresponds to bin-averaged model PFNS values from the LAM.

Concerns were raised whether the use of model values for the scaling of experimental shape PFNS might introduce a bias in the evaluation. If we rescale the experimental data only with respect to each other and then as a whole to the model data, the evaluated mean values change less than 0.4% up to 10 MeV and the evaluated relative uncertainties less than 0.06%. Of course, it is understood that we use for Eq. (3) model values which are in statistical agreement with experimental data.

2.2.2 Covariance Estimation

Experimental covariance matrices are estimated as recommended e.g. in Ref. [16] and references therein by partitioning the uncertainties according to their respective source — for instance due to counting statistics, background correction, detector efficiency determination — and estimating the correlation matrices associated with these partial uncertainties. In general, it is more transparent to estimate the correlations for well-known partial uncertainties than for total uncertainties containing many uncertainty sources [16]. The total experimental covariance matrices $\text{Cov}^x(\phi_i^x, \phi_j^x)$ are estimated by summing up the partial uncertainty contributions:

$$\text{Cov}^x(\phi_i^x, \phi_j^x) = \phi_i^x \phi_j^x \sum_k \delta \phi_i^k \delta \phi_j^k \text{Cor}^k(\phi_i^x, \phi_j^x). \quad (4)$$

The index k runs over all possible partial uncertainty contributions with relative uncertainty $\delta \phi^k$ and correlation matrix element $\text{Cor}^k(\phi_i^x, \phi_j^x)$. Depending on how the neutron detector efficiency was determined, additional uncertainties of a reference data set such as the $^{252}\text{Cf(sf)}$ PFNS and the calibration measurement might apply. A description of typical experimental uncertainty sources can be found in [16, 17].

Experimental covariances between different experiments were estimated by comparing the sources of the uncertainty contributions. If the partial uncertainties have a common origin (for instance a common reference data set used to determine the detector efficiency), a non-zero correlation between this partial uncertainty source of the two experiments arises. The covariances between uncertainties

¹The experimental data will be published on <https://www-nds.iaea.org/pfns/>.

²The chosen scaling process is only one of many reasonable possibilities to align model values and experimental data such that the χ^2 of the input data only reflects discrepancies in the shape and not the scaling of the data. A detailed discussion of different scaling techniques is the subject of a separate INDC report [14].

of different experiments are obtained by adding up the covariances of common uncertainty sources in analogy to Eq. (4).

As the original experimental data were rescaled for the evaluation process in Eq. (3), the experimental covariances also need to be rescaled. This rescaling is already implicitly done in Eq. (4) because the already rescaled experimental PFNS ϕ_i^x are used.

2.3 Model Covariance Estimation

2.3.1 Data Preparation

The LAM gives normalized PFNS values $N^M(E)$ for one specific outgoing neutron energy E which need to be transformed to bin-averaged PFNS values for the evaluation procedure. The bin-averaged PFNS values ϕ_i^M for a specific parameter vector $\underline{p}$ and the bin-averaged energy $\mathcal{E}_i^M$ are calculated by a numerical integral of

$$\phi_i^M(\underline{p}) = \int_{\Delta\mathcal{E}_i} N^M(E) dE / \Delta\mathcal{E}_i, \quad \mathcal{E}_i^M = \int_{\Delta\mathcal{E}_i} E N^M(E) dE / \int_{\Delta\mathcal{E}_i} N^M(E) dE, \quad (5)$$

where $\Delta\mathcal{E}_i$ is the energy bin-width. The bin-averaged model values ϕ_i^M are properly scaled with respect to experimental PFNS ϕ_i^x by Eq. (3). Thus the associated model covariances are also implicitly correctly scaled.

2.3.2 Covariance Estimation

The model covariances $\text{Cov}^M(\phi_i^M, \phi_j^M)$ are assumed to consist of two terms:

$$\text{Cov}^M(\phi_i^M, \phi_j^M) = \frac{1}{K} \sum_{k=1}^K \left(\phi_i^M(\underline{p}_k) - \phi_i^M \right) \left(\phi_j^M(\underline{p}_k) - \phi_j^M \right) + \text{Cov}^{MD}(\phi_i^M, \phi_j^M). \quad (6)$$

The first term on the right-hand side accounts for uncertainties in the model parameters $\underline{p}$, the second term is the model defect covariance matrix. As a first step, a total number of K vectors $\underline{p}_k$ of model parameters are obtained by randomly sampling from a Gaussian distribution with an initial model vector $\underline{p}^0$ and relative uncertainties $\delta\underline{p}^0$. Then, the PFNS $\phi_i^M(\underline{p}_k)$ are calculated by the LAM using $\underline{p}_k$. The model mean values ϕ_i^M are also calculated from the $\phi_i^M(\underline{p}_k)$,

$$\phi_i^M = \langle \phi_i^M(\underline{p}) \rangle_{\underline{p}_k} = \frac{1}{K} \sum_{k=1}^K \phi_i^M(\underline{p}_k). \quad (7)$$

Both, ϕ^M and the first term of Cov^M are calculated in logarithmic space to avoid an artificial bias because of the different orders of magnitude $\phi_i^M(\underline{p}_k)$ can assume for different $\underline{p}_k$ at high outgoing neutron energies. The evaluated results change less than 0.5% up to 7 MeV due to this transformation. The evaluation itself is also performed in logarithmic space because of the several orders of magnitude covered by the PFNS and the high uncertainties of the experimental data in the wings of the PFNS [18].

The LAM PFNS ϕ^M and the first term of Cov^M satisfy the normalization conditions of Eqs. (1) and (2) contrary to the experimental ϕ^x and Cov^x . This normalization condition on the model covariances is broken by adding the second term on the right-hand side of Eq. (6), Cov^{MD} . Three different formulations of model defects Cov^{MD} are introduced in Section 3.2.2 and their influence on the evaluation and benchmarks is discussed in Section 4 and Appendix B.

2.4 Evaluation

2.4.1 Evaluation with Non-normalized Experimental and Model Data

The present evaluation is undertaken in PFNS space as the normalization condition of the model covariances could be easily relaxed by adding $\mathbf{Cov}^{MD}$ in Eq. (6). In addition, model deficiencies can be more easily addressed in PFNS space by adding an adequate $\mathbf{Cov}^{MD}$. This is contrary to the preceding work of [9, 10] where updated LAM parameters and covariances are obtained as evaluation results. A drawback of evaluating in PFNS space is that we do not obtain evaluated model parameters and thus cannot judge whether the evaluated parameters are reasonable. Some of the PFNS model parameters are highly non-linear therefore their Generalized Least Squares (GLS) evaluation may be affected by the implicit linearization. Such a potential problem does not affect the evaluation in PFNS space. On the other hand, there is no warranty of the existence of one LAM parameter set that reproduces the evaluated solution because a true average of experimental and model PFNS in a least-square sense (evaluation) is obtained.

The qualitative differences between experimental data and model results described in Section 2.1 are resolved by: (a) treating experimental data as shape data and rescaling them with respect to model values by Eq. (3), (b) converting LAM predicted PFNS values to bin-averaged PFNS by Eq. (5) and (c) breaking the normalization condition on the model covariances by considering $\mathbf{Cov}^{MD}(\phi_i^M, \phi_j^M)$ in Eq. (6). The model and experimental data and covariances can be now used on an equal basis in an evaluation.

The evaluated bin-averaged PFNS $\underline{\phi}^{post}$ and the associated covariance matrix $\mathbf{Cov}^{post}$ are obtained by a GLS algorithm

$$\begin{aligned}\underline{\phi}^{post} &= \underline{\phi}^M + \mathbf{Cov}^{post} \mathbf{S}^+ (\mathbf{Cov}^x)^{-1} (\underline{\phi}^x - \mathbf{S} \underline{\phi}^M), \\ \mathbf{Cov}^{post} &= \mathbf{Cov}^M - \mathbf{Cov}^M \mathbf{S}^+ (\mathbf{S} \mathbf{Cov}^M \mathbf{S}^+ + \mathbf{Cov}^x)^{-1} \mathbf{S} \mathbf{Cov}^M.\end{aligned}\quad (8)$$

The matrix $\mathbf{S}$ is the design matrix and $\mathbf{S}^+$ its transpose. The design matrix elements are computed by linear interpolation, see e.g. [19],

$$S_{ij} = \begin{cases} (\mathcal{E}_i^x - \mathcal{E}_{j-1}^M) / (\mathcal{E}_j^M - \mathcal{E}_{j-1}^M) & \mathcal{E}_{j-1}^M \leq \mathcal{E}_i^x < \mathcal{E}_j^M \\ (\mathcal{E}_i^x - \mathcal{E}_{j+1}^M) / (\mathcal{E}_j^M - \mathcal{E}_{j+1}^M) & \mathcal{E}_j^M \leq \mathcal{E}_i^x < \mathcal{E}_{j+1}^M \\ 0 & \text{otherwise} \end{cases}, \quad (9)$$

with an experimental lattice $\mathcal{E}_i^x$ and a model/evaluated one $\mathcal{E}_j^M$. The evaluated relative uncertainties associated with $\mathbf{Cov}^{post}$ are given by $\delta\phi_i^{post} = 100.0 \sqrt{\mathbf{Cov}(\phi_i^{post}, \phi_i^{post})} / \phi_i^{post}$.

The factor,

$$\chi^2 = \left[(\underline{\phi}^x - \mathbf{S} \underline{\phi}^M)^+ (\mathbf{S} \mathbf{Cov}^M \mathbf{S}^+ + \mathbf{Cov}^x)^{-1} (\underline{\phi}^x - \mathbf{S} \underline{\phi}^M) \right] / n, \quad (10)$$

provides a measure of the discrepancy of the model and experimental shape if the experimental data are properly scaled. The variable n is the number of experimental data points and yields the degrees of freedom of the χ^2 . A $\chi^2 > 1$ indicates discrepancies in the experimental data themselves and between experimental data and model values and points towards unknown systematic errors.

2.4.2 Normalization of Evaluated Results

The evaluated bin-averaged PFNS $\underline{\phi}^{post}$ and the evaluated covariance matrix $\mathbf{Cov}^{post}$ do not satisfy the ENDF-6 format requirements [13], Eqs. (1) and (2), if $\mathbf{Cov}^{MD}(\phi_i^M, \phi_j^M)$ is considered in $\mathbf{Cov}^M$, as discussed in Section II.D of [12]. In order to satisfy the ENDF-6 format requirements, the evaluated ϕ_i^{post} consisting of m data points is re-normalized by

$$\psi_i^{post} = \phi_i^{post} / G, \quad \text{with} \quad G = \sum_{i=1}^m \phi_i^{post} \Delta \mathcal{E}_i^M, \quad (11)$$

219 and the normalized covariance matrix is obtained by means of linear error propagation:

$$\mathbf{Cov}_{\psi}^{post} = \mathbf{T} \mathbf{Cov}_{\phi}^{post} \mathbf{T}^+. \quad (12)$$

220 The matrix $\mathbf{T}$ is a $m \times m$ sensitivity matrix with elements [12],

$$T_{ij} = \partial \psi_i^{post} / \partial \phi_j^{post} = \left(G \delta_{ij} - \phi_i^{post} \Delta \mathcal{E}_j \right) / G^2, \quad (13)$$

221 with the Kronecker delta δ_{ij} .

222 2.4.3 Calculation of Mean Energy

223 The mean energy $\langle \mathcal{E} \rangle$ of the normalized evaluated bin-averaged PFNS $\underline{\psi}^{post}$ is calculated by:

$$\langle \mathcal{E} \rangle = \sum_{i=1}^m \mathcal{E}_i \psi_i^{post} \Delta \mathcal{E}_i, \quad (14)$$

224 and the associated scalar variance $\text{var}(\mathcal{E})$ is given by means of linear error propagation by,

$$\text{var}(\mathcal{E}) = \underline{W}^+ \mathbf{Cov}_{\psi}^{post} \underline{W}, \quad \text{with} \quad W_i = \partial \langle \mathcal{E} \rangle / \partial \psi_i^{post} = \mathcal{E}_i \Delta \mathcal{E}_i. \quad (15)$$

225 2.5 Calculation of the Criticality

226 We use the newly evaluated PFNS and its covariance matrix for criticality calculations of the multipli-
 227 cation factor k_{eff} employing the MCNP6.1 code [20, 21]. The NJOY nuclear data processing code [22]
 228 is used to convert the evaluated data for the ^{239}Pu isotope from ENDF-6 format [13] to the continuous
 229 energy ACE format needed by MCNP. Then, k_{eff} is computed for a variety of applications using these
 230 new PFNS data and compared to the [k_{eff} using the ENDF/B-VII.1 evaluation](#). Other than statistical
 231 variations coming from the Monte Carlo simulation, the discrepancies can be directly attributed to the
 232 change in the PFNS of the neutron-induced fission of ^{239}Pu .

233 In the recent release of MCNP6.1 [23], a sensitivity/uncertainty method was implemented allowing
 234 the user to compute the sensitivity of k_{eff} to perturbations in the nuclear data. Using the linear algebra
 235 ‘sandwich’ rule,

$$\frac{\sigma_k^2}{k_{\text{eff}}^2} = \mathbf{S}_{\mathbf{k}} \left(\frac{\mathbf{Cov}_{\psi}^{post}}{\psi \psi^T} \right) \mathbf{S}_{\mathbf{k}}^T, \quad (16)$$

236 the relative variance on the left-hand-side is computed using the relative sensitivity coefficients, $\mathbf{S}_{\mathbf{k}} =$
 237 $(\psi/k_{\text{eff}})/(\text{d}k_{\text{eff}}/\text{d}\psi)$, computed by MCNP and the element-wise relative covariance matrix in parenthe-
 238 ses. The recently released version of NJOY does not include covariance matrix processing from ENDF-6
 239 to ACE formats, so this simple post-processing procedure is presently the only non-automated step
 240 necessary to determine the uncertainties in k_{eff} due to the nuclear data.

241 3 Input Quantities and Their Uncertainty Quantification

242 3.1 Experimental Covariance Estimation

243 In this evaluation, only experimental PFNS data [7, 24–30] of ^{239}Pu for incident neutron energies less
 244 than or equal to 500 keV, and which are well-enough documented to construct experimental covariance
 245 matrices, are taken into account. [The experimental data of \[7, 24, 25\] are shape data given either in](#)
 246 [arbitrary units in their EXFOR entry or normalized using the LAM. The neutron detectors of \[26\]](#)
 247 [were calibrated with the \$^{252}\text{Cf}\(\text{sf}\)\$ PFNS and in EXFOR ratio shape data with arbitrary normalization](#)
 248 [are provided. Only the measurements of \[27–30\] can be interpreted as absolute or absolute ratio data.](#)

We treat all experimental data as shape data because our experimental data base contains shape and absolute data, and due to the arguments given in Section 2.1.

The experimental data of [7] were originally published for an incident neutron energy $E_{\text{inc}} = 1.5$ MeV. They were transformed to $E_{\text{inc}} = 500$ keV by using the ratio of ^{239}Pu PFNS data $\psi^{\text{VII.1}}$ from ENDF/B-VII.1 at those two incident energies,

$$\phi_i^x(E_{\text{inc}} = 500\text{keV}) = \phi_i^x(E_{\text{inc}} = 1.5\text{MeV}) \frac{\psi_i^{\text{VII.1}}(E_{\text{inc}} = 500\text{keV})}{\psi_i^{\text{VII.1}}(E_{\text{inc}} = 1.5\text{MeV})}.$$

The data termed ‘Starostov’ throughout this paper are obtained by statistically combining four measurements ([27–29] and measurement cycle 2 of [30]), and their uncertainties, which are displayed in Fig. 1. These measurements were performed by the same group partly with the same fission counter in similar set-ups. These data and the ones of [26] were transformed to the outgoing neutron energy grid of the $^{252}\text{Cf}(\text{sf})$ PFNS of [31, 32]. The mentioned combination of NIIAR sets is done in the Bayesian sense to account for the strong correlations between these related experiment when transforming to the energy grid of [31, 32]. This treatment of [27–30] differs from that of [7, 24–26] in that the four sub-sets are rescaled with respect to each other before the combined data are rescaled to model values. The scaling factors of [27, 28, 30] are not impacted by this different treatment. The scaling factor of [29] is less than 2% smaller due to the differing shape of the model values at high outgoing neutron energies, which are not well-known, and leads to discrepancies between [29, 30] in their overlapping energy range. If the data of [29] are rescaled to model values, the evaluated PFNS changes by less than 0.7% and -0.5% between 100 keV and 10 MeV. The evaluated mean energy decreases by 5 keV (0.24%) to 2.075 MeV.

Figure 1: (Color online) The experimental data of [27–30] are shown.

The experimental uncertainties were estimated, as described in Section 2.2, by decomposing into partial uncertainties and estimating correlation matrices for the partial contributions. In Table I, the estimated partial uncertainties for the data of [24–30] are given. A preliminary version of this estimate can also be found in Table I. of [33]. A detailed description of the experimental covariance estimation for these experiments can be found in a LANL report [34]. The partial uncertainties for the data of [7] are taken directly from that reference, and they are not repeated here. The estimated total uncertainties are compared in Fig. 2 with model uncertainties. Clearly, the data sets of Starostov et al. [27–30] and Lestone et al. [7] have the lowest experimental uncertainties over most of the outgoing neutron energy range. The estimated experimental correlation matrices for [24–30] are given in Figs. 1 and 2 in [33] and the one of [7] is given in Fig. 3. The one standard deviation error bars obtained with the detailed experimental uncertainty quantification and information of [35] overlap for most of the experimental

data in Fig. 4 contrary to the previous evaluation [9]. In Fig. 2 of [9], the experimental one standard deviation error bars don't overlap for parts of [26,28] and [24,25]. The impact of using experimental data and covariances used for [9] compared to the present ones is discussed in Appendix A.

Table I: Ranges of relative uncertainties (in %) are given for the data of [24–30] for the main uncertainty sources.

Experiment	Knitter [24]	Staples [25]	Lajtai [26]	Starostov [27–30]
Counting Statistics	2.1–340	2.4–8.5	3.8–31.9	0.5–17.5
Time Resolution	0.01–68.5	0.03–29.9	0.1–36.1	0.06–79.6
TOF length	-	-	0.01–0.3	0.08–2.6
Detector Efficiency	2–7.9	7–10	0–7.4	0–3.0
Multiple Scattering/Background	0–20.6	0–16.7	3.8–12.4	0.6–15.0
Deconvolution	0–10	0–9.8	-	-
Reference	-	-	1.1–10.5	1.1–10.5

Figure 2: (Color online) The relative uncertainties estimated for the data of [7,24–30] are compared to relative uncertainties of $\mathbf{Cov}^M$ with $\mathbf{Cov}^{MD} = 0$.

In addition, correlations between uncertainties of the data of [24,25] which arise from common multiple scattering and deconvolution uncertainties are considered. Correlations between the uncertainties related to data of [26] and [27–30] arise from the covariances associated to the $^{252}\text{Cf}(\text{sf})$ PFNS data of [31,32]. These data were used to transform the ratio data of [26–30], giving ^{239}Pu data relative to $^{252}\text{Cf}(\text{sf})$, into ^{239}Pu PFNS data.

3.2 Model calculations

An extended Los Alamos model [11] is employed to calculate the model PFNS values $\underline{N}^M$ using a phenomenological approach. Changes compared to the LAM used in previous evaluations [9,10] were partly proposed in Ref. [9] and they will be highlighted in the text below. The model PFNS $N^M(E)$ at outgoing neutron energy E is a weighted superposition of PFNS for one average light and one heavy fragment, N_L and N_H respectively,

$$N^M(E) = (\bar{\nu}_L N_L(E) + \bar{\nu}_H N_H(E)) / (\bar{\nu}_L + \bar{\nu}_H). \quad (17)$$

They are weighted according to the average neutron multiplicities $\bar{\nu}_L$ and $\bar{\nu}_H$ of the light and heavy fragments [36]. In the original LAM as well as in [9,10], it was assumed that the light and heavy

Figure 3: (Color online) The correlation matrix and total relative uncertainties estimated for the data of Lestone et al. [7].

Figure 4: (Color online) The experimental data of [7, 24–30] as used for the evaluations are shown.

295 fragment PFNS are weighted equally, i.e. $\bar{\nu}_L = \bar{\nu}_H$. In the present implementation of the LAM,

$$\bar{\nu}_L = R_{\bar{\nu}} \bar{\nu}_H,$$

296 the weight of the light and heavy fragment can be different. In the laboratory frame, the light or heavy
 297 PFNS $N_x(E)$ with subscript $x = \{L, H\}$ are calculated by

$$N_x(E) = \frac{1}{2\sqrt{E_f}T_{m,x}^2(1+b/3)} \int_{(\sqrt{E}-\sqrt{E_f})^2}^{(\sqrt{E}+\sqrt{E_f})^2} \sigma_{c,x}(\varepsilon)\sqrt{\varepsilon} \left(1 + b\frac{(E-\varepsilon-E_f)^2}{4\varepsilon E_f}\right) I(\varepsilon)d\varepsilon, \quad (18)$$

298 with $I(\varepsilon)$ given by [37],

$$I(\varepsilon) = \begin{cases} \frac{1}{4s} \int_0^\alpha k_f(T)(s+1)^2 T \exp(-\varepsilon/T) dT, \\ \frac{s+1}{2(s-1)} \int_\alpha^{s\alpha} k_f(T) \left(-\frac{s+1}{2s} T + T_{m,x}\right) \exp(-\varepsilon/T) dT, \end{cases} \quad \alpha = \frac{2T_{m,x}}{s+1}, \quad (19)$$

299 and

$$k_f(T) = \left[\int_0^\infty \sigma_{c,x}(\varepsilon) \varepsilon \exp(-\varepsilon/T) d\varepsilon \right]^{-1}. \quad (20)$$

300 The variable E_f denotes the kinetic energy per nucleon in the fission fragment and $\sigma_{c,x}$ the inverse
 301 compound nucleus formation cross section. In previous evaluations [9, 10], the maximum temperature
 302 of the fission fragments $T_{m,x}$ was the same for the light and heavy fragments, $T_{m,L} = T_{m,H}$, while
 303 here the ratio $R_T = T_{m,L}/T_{m,H}$ of the temperatures can assume values different from unity. The
 304 maximum temperature in a fragment is given by $T_{m,x} = \sqrt{\langle E^* \rangle / \langle a_x \rangle}$, with $\langle a_x \rangle$, the average level
 305 density parameter. The variable $\langle E^* \rangle$ corresponds to the total average excitation energy, given by

$$\langle E^* \rangle = \langle E_r \rangle + E_{\text{inc}} + B_n - \langle TKE \rangle.$$

306 The average excitation energy depends on the average energy release $\langle E_r \rangle$, the energy of the incoming
 307 neutron E_{inc} , the neutron binding energy B_n and the average total kinetic energy $\langle TKE \rangle$ of the fission
 308 fragments.

309 The parameter s in $I(\varepsilon)$ introduced in [37] changes the original triangular distribution used in [9–11]
 310 to a more realistic fission fragment temperature distribution comparable to Terrell’s (see Fig. 6 in [38]).
 311 For $s = 1$, $R_T = 1$ and $R_{\bar{\nu}} = 1$, one obtains the same version of the LAM as in [10]. In [10], a
 312 phenomenological description of the anisotropy in the neutron evaporation [38] was already included
 313 in the model formulation. This anisotropy could be caused by large spins of the fission fragments,
 314 neutrons evaporated during the acceleration of the fragments or scission neutrons. The parameter b in
 315 Eq. (18) defines the strength of the anisotropy of evaporation. For $s = 1$, $R_T = 1$, $R_{\bar{\nu}} = 1$ and $b = 0$,
 316 one recovers the original LAM equations [11], also used in [9].

317 3.2.1 Model covariances due to uncertainties in the model parameters

318 The LAM PFNS were calculated for the fission fragment pair ^{100}Zr and ^{140}Xe at an incident energy
 319 of $E_{\text{inc}} = 500$ keV and with a binding energy $B_n = 6.534$ MeV using the CoH code [39] recently
 320 upgraded to include the LAM equations. The model mean values ϕ^M and model covariances $\mathbf{Cov}^M$
 321 were calculated by means of Eqs. (7) and (6) using the initial LAM parameter values $\underline{p}^0$ in Table II.
 322 The parameter values were sampled $K \approx 34,000$ times following a Gaussian probability distribution
 323 with one-sigma relative uncertainties as provided in Table II. In Table III, the mean value $\langle p \rangle$ of a
 324 specific model parameter p , relative uncertainty δp and the skewness γ_p are given. These are computed
 325 from the sampled parameter values p_k by,

$$\langle p \rangle = \frac{1}{K} \sum_{k=1}^K p_k, \quad \delta p = \frac{100}{\langle p \rangle K} \sqrt{\sum_{k=1}^K (p_k - \langle p \rangle)^2} \quad \text{and} \quad \gamma_p = \frac{100}{\langle p \rangle \delta p K} \sum_{k=1}^K (p_k - \langle p \rangle)^3.$$

326 The sampled parameter values do not all follow a Gaussian distribution, because of physical constraints
 327 applied to the parameter ranges of several parameters. Hence, their distribution has non-zero skewness
 328 and the initial parameter mean values $\underline{p}^0$ might be different from $\langle p \rangle$.

329 The mean values for the energy release $\langle E_r \rangle$, average total kinetic energy $\langle TKE \rangle$, average total
 330 gamma-ray emission energy $\langle E_\gamma^{\text{tot}} \rangle$ and neutron separation energy S_n were taken from [9], and they
 331 were chosen close to values presented in Ref. [40]. The initial relative uncertainties for $\langle E_r \rangle$ and $\langle TKE \rangle$
 332 have about the same size as in Refs. [9, 10]. The relative uncertainty for $\langle E_\gamma^{\text{tot}} \rangle$ was estimated based
 333 on the uncertainty of measured data by [41]. The parameters $\langle E_r \rangle$, $\langle TKE \rangle$, $\langle E_\gamma^{\text{tot}} \rangle$ and S_n are sampled
 334 under the following constraints

$$\langle E_r \rangle + E_{\text{inc}} + B_n - \langle TKE \rangle \geq \max(S_n, \langle E_\gamma^{\text{tot}} \rangle), \quad \langle E_r \rangle \leq 210 \text{ MeV}. \quad (21)$$

335 The first constraint guarantees that enough energy is available for a neutron or γ ray to be emit-
 336 ted. The second constraint is determined by using Monte-Carlo Hauser-Feshbach calculations [42] for

Table II: Initial model parameter values $\underline{p}^0$ of the LAM [11] and relative uncertainties $\delta \underline{p}^0$ used to calculate $\mathbf{Cov}^M$ and $\underline{\phi}^M$ of Eqs. (6) and (7).

Parameter	$\underline{p}^0$	$\delta \underline{p}^0$ (%)
$\langle E_r \rangle$	196.987 MeV	6
$\langle TKE \rangle$	177.6 MeV	3
$\langle E_\gamma^{\text{tot}} \rangle$	6.77 MeV	10
S_n	5.22 MeV	2
s	1.1	10
$R_{\bar{\nu}} = R_T$	1.14	20
b	0.1	50
$\langle a_L \rangle$	9.09091 MeV ⁻¹	10
$\langle a_H \rangle$	12.72727 MeV ⁻¹	10
m_{r_V}	1.0	4
m_{a_V}	1.0	4
m_V	1.0	4
m_{r_W}	1.0	5
m_{a_W}	1.0	6
m_W	1.0	10

Table III: Sampled model parameter mean values $\langle \underline{p} \rangle$, relative uncertainties $\delta \underline{p}$ and $\gamma_{\underline{p}}$ calculated from the sampled model parameters taking into account the constraints of Eqs. (21) and (22).

Parameter	$\langle \underline{p} \rangle$	$\delta \underline{p}$ (%)	$\gamma_{\underline{p}}$
$\langle E_r \rangle$	195.442 MeV	4.21	-0.297
$\langle TKE \rangle$	177.3 MeV	2.94	-0.02625
$\langle E_\gamma^{\text{tot}} \rangle$	6.76 MeV	10.02	5.012×10^{-4}
S_n	5.22 MeV	2.01	-1.1×10^{-4}
s	1.136	7.56	0.637
$R_{\bar{\nu}} = R_T$	1.14	19.97	-0.01882
b	0.103	45.89	0.223
$\langle a_L \rangle$	9.08925 MeV ⁻¹	10.06	-4.833×10^{-3}
$\langle a_H \rangle$	12.72801 MeV ⁻¹	10.02	1.990×10^{-3}
m_{r_V}	1.0	3.99	-0.01048
m_{a_V}	1.0	3.97	-0.01067
m_V	1.0	3.99	0.03397
m_{r_W}	1.0	5.02	0.03789
m_{a_W}	1.0	5.97	0.02247
m_W	1.0	10.01	-0.01722

²³⁹Pu(n_{therm},f). These calculations do not predict any energy release of more than 210 MeV in the fission process taking into account not only the mass-difference between the compound nucleus mass and those of possible fragmentation pairs but also the likelihood that a specific fragmentation occurs by means of sampling from primary fission fragment yields. The mean value of $\langle E_r \rangle$ calculated from the sampled parameter values is slightly shifted to lower values and its distribution has a negative skewness as shown in Fig. 5.

The initial values and relative uncertainty for the anisotropy parameter b are taken from [10]. For the temperature distribution parameter s , we chose a mean value and relative uncertainties to cover reasonable parameter values given information in [37] for ²⁵²Cf(sf). The constraints on b and s follow

Figure 5: The probability distribution functions of $\langle E_r \rangle$ and $\langle TKE \rangle$ sampled values under the constraint of Eq. (21) are shown. The vertical lines indicate the sampled mean value.

from [37],

$$s \geq 1 \quad \text{and} \quad b \geq 0. \quad (22)$$

The parameter R_T is used to redistribute the total excitation energy preferentially to the light fragment, as evidenced by the higher average number of neutrons coming from the light than from the heavy fragments, as calculated from [42]. Calculated values of $R_{\bar{\nu}}$ follow closely the values of R_T , and therefore we assumed $R_{\bar{\nu}} = R_T$. The initial mean values and relative uncertainty were derived from Fig. 16 in [42].

The initial values for the average level density parameters of the light and heavy fragments with mass number A , $\langle a_L \rangle$ and $\langle a_H \rangle$, are calculated from the approximation [11],

$$\langle a_x \rangle = A/11, \quad \text{for } x = \{L, H\}.$$

The initial relative uncertainties were taken from [9].

In [9, 10], the optical model potential (OMP) of Becchetti and Greenlees [43] was employed to calculate the inverse compound nucleus formation cross section $\sigma_{c,x}$ in Eq. (18). Another frequently used OMP is that of Koning and Delaroche [44]. If all parameters except the OMP remain the same, the resulting LAM PFNS using these two OMPs differ significantly, especially for the lowest energy neutrons as shown in Fig. 6. However, both OMPs assume that nuclei are spherical and their parameters were fitted to data near the valley of stability. Their use for fission fragments that are neutron-rich and deformed is therefore questionable.

Here, we use a modified version of the OMP of [44], where we rescaled the depth W , the radius r_W and the diffuseness a_W of the imaginary surface contribution of the potential depending on the incident neutron energy E_{inc} up to 2 MeV,

$$W = W m_W (0.8 + 0.2 E_{\text{inc}}/2.0), \quad (23)$$

$$r_W = r_W m_{r_W} (1.21 - 0.21 E_{\text{inc}}/2.0), \quad (24)$$

$$a_W = a_W m_{a_W} (0.9 + 0.1 E_{\text{inc}}/2.0). \quad (25)$$

The constants to parametrize the potential below 2 MeV were obtained by fitting the calculated strength functions S_0 and S_1 of $^{86,87,88}\text{Sr}$ and $^{90,91,92,93,94,96}\text{Zr}$ to their values in the Atlas of Neutron Resonances [45]. These isotopes were chosen as they are closest to the light fragment ^{100}Zr . At incident neutron energies above 2 MeV, the OMP of [44] was left unchanged. In CoH, the radius $r_{V/W}$, diffuseness $a_{V/W}$, the height V/W of the real volume (subscript V) as well as the imaginary surface contribution (subscript W) to the OMP can be changed by means of multiplicative parameters,

Figure 6: LAM PFNS are shown for the same input parameters except for the optical model potential in comparison to the ENDF/B-VII.1 evaluation [8].

$m_{rV}, m_{aV}, m_V, m_{rW}, m_{aW}, m_W$ respectively. These OMP parameters were varied within the parameter boundaries given in Table II. Their uncertainties were chosen higher than those proposed in Table IX of [46] for the OMP of [44] to account for the fact that the performance of the chosen OMP cannot be directly compared to ^{100}Zr data. In order to test the chosen OMP and its parameter boundaries, we compared with total and elastic experimental cross section data of ^{165}Ho which has a comparable deformation parameter.

Compared to the previous evaluation [9], we also include uncertainties for the LAM parameters $\langle E_\gamma^{\text{tot}} \rangle$, S_n , s , $R_{\bar{\nu}} = R_T$, m_{rV} , m_{aV} , m_V , m_{rW} , m_{aW} and m_W . In the previous evaluation, only uncertainties for $\langle TKE \rangle$, $\langle E_r \rangle$, an average level density parameter and B_n were included leading to strongly correlated model covariances, which were partly responsible for unreasonably small evaluated uncertainties [12]. The impact of using the model data and covariances of [9] compared to the present ones is discussed in Appendix A.

3.2.2 Model uncertainty covariances

We investigated three different formulations of the model defect covariance matrix Cov^{MD} introduced in Eq. (6):

Case A

$$\text{Cov}^{MD}(\phi_i^M, \phi_j^M) = \phi_i^M \phi_j^M (\Delta No)^2, \quad (26)$$

This covariance term introduces an uncertainty with standard deviation ΔNo in the scaling of the PFNS. The relative scaling uncertainty was taken to be $\delta No = \Delta No / No = 6\%$. This value of δNo was chosen such that model uncertainties at approximately 2 MeV are larger than the experimental uncertainties estimated for the data of [7, 24, 29, 30] shown in Fig. 2. The uncertainty δNo was added to break the normalization condition on the model covariances such that both experimental and model covariances are not-normalized. We compare evaluated uncertainties obtained by a statistical analysis of experimental data only to those obtained with model and experimental information in order to quantify the whether the latter evaluated uncertainties are reasonable. This comparison is only possible, if model and experimental covariances are either both not-normalized or both normalized, as normalizing covariances following Eq. (12) usually leads to an overall reduction of uncertainties [12]. This term has very little impact on the evaluated results, which is shown in Appendix B, as it drops out after normalizing the evaluated

PFNS and covariances [12]. Thus, this formulation of $\mathbf{Cov}^{MD}$ allows us to study the evaluated uncertainties obtained with experimental and LAM covariances only.

Case B

$$\mathbf{Cov}^{MD}(\phi_i^M, \phi_j^M) = \delta_{ij}(\Delta d_i)^2. \quad (27)$$

A diagonal covariance term with standard deviation Δd is added to the LAM covariances. We assume that the relative uncertainties associated with Δd shown in Fig. 7 are 10% up to 100 keV and decrease to 3 % at 600 keV. This term accounts for the assumption that the shape of the model is not perfect in an ad-hoc manner. It weakens too strong LAM correlations and thereby provides the flexibility for the evaluated shape and uncertainties to follow experimental data more closely. The chosen magnitude of the uncertainties below 100 keV reflects our belief in the possible accuracy of experimental data at this energy. The uncertainty Δd above 600 keV increases the prior uncertainties such that they are significantly larger than the experimental ones of [7, 27–30] up to 10 MeV. Thus evaluated PFNS and uncertainties are guided above 600 keV by experimental information. The choice of the diagonal covariance term is purely motivated by the aim to weaken model correlations, and it leads to a prior correlations matrix different from Case A, as is clearly visible in Figs. 8(a) and 8(b).

Case C

$$\mathbf{Cov}^{MD}(\phi_i^M, \phi_j^M) = N_i^{Ma} N_j^{Ma} \left[\bar{\nu}_s^2 \eta_{ij} \left(\frac{\varepsilon_i}{T^{Ma}} - \frac{3}{2} \right) \left(\frac{\varepsilon_j}{T^{Ma}} - \frac{3}{2} \right) (\delta T^{Ma})^2 + \text{var}(\bar{\nu}_s) \right]. \quad (28)$$

This covariance term is based on limited information about potential scission neutron contributions to the PFNS of [47–49]. According to Ref. [47] and references therein, the scission neutron spectrum could be roughly described by a Maxwellian spectrum, $N^{Ma}(\varepsilon) = 2\sqrt{\varepsilon} \exp(-\varepsilon/T^{Ma})/(\sqrt{\pi T^3})$, with Maxwellian temperatures T^{Ma} for ^{252}Cf ranging from about 0.3 MeV to one indistinguishable from the PFNS without scission neutron contributions. We estimate the Maxwellian temperature here to be $T^{Ma} = 0.9$ with relative uncertainty $\delta T^{Ma} = 50\%$ to cover possible ranges. The first term on the right-hand side of Eq. (28) corresponds to a covariance term estimated with a Maxwellian assumption and weakened by the arbitrary factor $\eta_{ij} = \exp\left\{-0.3[(\varepsilon_i - \varepsilon_j)/\max(\varepsilon_i, \varepsilon_j)]^2\right\}$. This arbitrary factor weakens the assumption that the scission neutron spectrum has a Maxwellian shape, as this assumption is questionable. The variable $\bar{\nu}_s$ denotes the average scission neutron contribution relative to the total prompt neutron multiplicity $\bar{\nu}$. We assume a $\bar{\nu}_s = 15\%$ and a standard deviation $\sqrt{\text{var}(\bar{\nu}_s)}$ of 50% such that it covers values put forward in [47–49]. It is very difficult to measure scission neutrons and hence their existence, spectrum and magnitude of $\bar{\nu}_s$ is still controversial and hence, such a covariance term can be questioned. In addition, this term accounts for only one possible defect and approximation made in the LAM while there are other possible defects (e.g., the use of only one fragmentation pair rather than including all possible fragmentations) not included in these considerations. The prior covariance matrix including $\mathbf{Cov}^{MD}$ of Case B differs from Case A (compare Figs. 8(a) and 8(c)). This formulation of $\mathbf{Cov}^{MD}$ leads to a substantial relative uncertainty contribution to the prior up to 3 MeV outgoing neutron energy which is shown in Fig. 7.

It should be stressed that the model defect terms put forward here as Cases B and C are speculative in character. They address in an ad-hoc manner that neither the LAM nor the experimental data on their own provide perfect information for an evaluation. Ideally, these issues should be resolved by using improved models as well as new high-precision experimental data once they are available and adequate to be used for an evaluation.

Figure 7: The relative uncertainties associated with $\mathbf{Cov}^{MD}$ of Cases A, B and C are compared with those calculated by the LAM. The total prior uncertainties (associated with $\mathbf{Cov}^M$) are given by a combination of either Cases A, B or C with the LAM covariances following Eq. (6).

Figure 8: (Color online) The correlation matrices corresponding to $\mathbf{Cov}^M$ are shown, as obtained with different formulations of $\mathbf{Cov}^{MD}$.

4 Evaluated Results and Discussion

4.1 Evaluation Results in Comparison with Experimental Data and Uncertainties

The evaluations were performed according to Eq. (8) with experimental data and model calculations and associated covariances as presented in Section 3. The χ^2 value using $\mathbf{Cov}^{MD}$ of Eq. (26) is 0.62 and thus shows that experimental data and LAM values agree reasonably well within their estimated uncertainties.

In order to quantify if the evaluated uncertainties using the LAM covariances and $\mathbf{Cov}^{MD}$ of Eq. (26) are reasonable, given the experimental input, and to study the impact of the normalization condition on the evaluated results, the statistical analysis was undertaken in three steps:

1. In the first step, the evaluation was undertaken with experimental data and $\mathbf{Cov}^x$ of Section 3.1 and a very uncertain model covariance matrix, i.e., diagonal $\mathbf{Cov}^M$ with a relative uncertainty of 500% which is higher than any of the experimental uncertainties. This evaluation step is called ‘experiment only’.
2. In the second step, $\mathbf{Cov}^M$ including $\mathbf{Cov}^{MD}$ of Eq. (26) (Case A) and $\mathbf{Cov}^x$ of Section 3.1 were used. Thus the impact of the LAM covariances can be studied. The evaluated covariances

$\mathbf{Cov}_A^{post}$ and mean values ϕ_A^{post} do not satisfy the normalization conditions of Eqs. (1) and (2).

3. In the last step, the evaluated results are normalized according to Eqs. (11) and (12).

We expect the evaluated uncertainties to be of comparable size to statistically combined experimental uncertainties in an energy range where many well-known experimental data are available, which is from approximately 800 keV to 10 MeV. Otherwise, they should be guided by the model and model correlations. In Fig. 9, we compare the not-normalized evaluated PFNS uncertainties of Step 1 and 2 in the evaluation procedure, as experimental PFNS are intrinsically not-normalized quantities as described in Section 2.1. The normalization conditions on the PFNS and its covariances, Eqs. (1) and (2), lead to reduced uncertainties compared to not-normalized covariances (Fig. 9). This reduction of uncertainties is expected as the number of degrees of freedom of the covariance matrix and mean values are reduced by the normalization conditions of Eqs. (1) and (2) [12]. The evaluated uncertainties obtained in Step 2 are a factor of 1.6 lower at an outgoing neutron energy of 2 MeV than those of Step 1. The evaluated uncertainties at 2 MeV are distinctly higher than those obtained in Ref. [9], which were a factor 10 smaller than any experimental input uncertainty, but they are still slightly too small compared to an evaluation with experimental data only. In addition, the evaluated and normalized uncertainties in Fig. 10 are approximately 3.6% below 100 keV outgoing neutron energy. This magnitude is questionable given that experimental uncertainties are larger than 10% below 100 keV in Fig. 2. This reduction of uncertainties is clearly caused by the still strong correlations calculated using the LAM, despite the extensions made in Section 3.2. The LAM is indeed an effective but simplified model taking averages over many fragmentations of physically observable fission pairs, and thus the simplified parameter space restrains our evaluation too much, i.e., the model is too stiff. More sophisticated models such as [42, 50, 51] may eventually be used once their predictive power of the PFNS is suitably accurate for an evaluation. These models consider many fragmentation pairs to calculate the PFNS. Also, scission neutrons are not taken into account in the LAM, and this could lead to an under-prediction of the spectrum values at the lowest energies. The effect of a model parameter space which restrains uncertainty estimates too much has been studied in great detail in Ref. [52] for many different subject areas.

Figure 9: The evaluated relative uncertainties associated with $\phi_i^{\text{Experiment Only}}$ (step 1 of the statistical analysis), ϕ_A^{post} using $\mathbf{Cov}^M$ including $\mathbf{Cov}^{MD}$ of Eq. (26) (step 2, not-normalized) and ψ_A^{post} (step 3, normalized) are compared.

Our disbelief in the obtained evaluated uncertainties and the strong model correlations is quantified by adding $\mathbf{Cov}^{MD}$ of Cases B and C in Section 3.2.2 to the model covariances. The obtained evaluated and normalized relative uncertainties in Fig. 10 agree better with expectations based on experimental knowledge. The evaluated PFNS in Fig. 11 agree well with each other above 2 MeV and below 20

Figure 10: (Color online) Evaluated relative uncertainties using $\mathbf{Cov}^{MD}$ of Cases A, B and C and those rescaled a-posteriori of Case A are compared with each other. The evaluated uncertainties of Case B were smoothed. All associated evaluated covariances were normalized.

keV.³ All three evaluated PFNS follow closely the experimental data of Starostov et al. [27–30] and Lestone et al. [7] above 2 MeV outgoing neutron energy. Below 2 MeV, the evaluations are in good agreement with [27–30]. This good agreement is expected as the experimental data sets of [7, 27–30] have the lowest uncertainties of all experiments as shown in Fig. 2. Differences between the evaluated PFNS using $\mathbf{Cov}^{MD}$ of Cases A, B and C can be observed around 1.2 MeV, 400 keV and between 30 and 80 keV. The mean energy of the evaluated PFNS is 2.080 ± 0.015 MeV for Case A, 2.075 ± 0.016 MeV for Case B and 2.074 ± 0.017 MeV for Case C, which is a maximum change of less than 0.3%. However, the mean energies decreases by 1.4-1.7% compared to the ENDF/B-VII.1 value, 2.11 MeV. The evaluated covariances in Figs. 12(a), 12(b) and 12(c) differ visibly. They show features similar to their respective $\mathbf{Cov}^M$ although the prior correlations are softened in all three cases.

Figure 11: (Color online) The normalized evaluated PFNS ψ^{post} using $\mathbf{Cov}^{MD}$ of Cases A, B and C are compared to experimental data ϕ^x of [7, 27–30] and the ENDF/B-VII.1 results. These experimental data sets have the lowest uncertainties up to 10 MeV (Fig. 9), and they were rescaled to the evaluated results of Case A.

The estimation of $\mathbf{Cov}^{MD}$ of Cases B and C in Section 3.2.2 involved to some degree arbitrary assumptions, so a change in the evaluated PFNS compared to Case A can be questioned. These arbitrary assumptions of $\mathbf{Cov}^{MD}$ can be avoided by rescaling the evaluated uncertainties obtained

³The evaluated PFNS and relative uncertainties of Case B were smoothed a-posteriori using the Savitzky-Golay algorithm with third order polynomials.

Figure 12: (Color online) The correlation matrices corresponding to $\mathbf{Cov}_\psi^{post}$ are shown, as obtained with different formulations of $\mathbf{Cov}^{MD}$.

from Case A with a factor of 1.6 and adding an additional uncertainty term at low outgoing neutron energy following the procedure in [9]. This ad-hoc increase of evaluated uncertainties runs contrary to proper statistical procedures and the Bayesian approach where expert knowledge should be included in the evaluation process. However, one can study the impact of different $\mathbf{Cov}^{MD}$ on benchmarks including reasonable uncertainties given experimental knowledge.

4.2 Impact on Criticality and Reaction Rates

Our new evaluated PFNS for the $n(500 \text{ keV}) + {}^{239}\text{Pu}$ reaction and associated covariance matrices are tested in the simulation of critical assemblies. In [9], PFNS sensitivity studies were performed and in [53] forward uncertainty propagation sampling methods were used independently to compute the uncertainty of k_{eff} for the Jezebel Pu critical assembly. While both sensitivity/uncertainty approaches in [9] and [53] had significant differences, the results were in general agreement with the k_{eff} uncertainty between 0.16–0.25%. Here, we perform similar studies using the new MCNP6.1 [21] built-in sensitivity/uncertainty method.

To this end, the evaluated PFNS obtained with $\mathbf{Cov}^{MD}$ of Case A (Eq. (26)) was extrapolated to cover an incident neutron energy range from 0 to 5 MeV by using the shape-change of the evaluated ${}^{239}\text{Pu}$ PFNS of [10] from 500 keV to the desired incident neutron energy. Between 5 MeV and 6 MeV incident neutron energy the PFNS is linearly interpolated between the new and ENDF/B-VII.1 data and above 6 MeV ENDF/B-VII.1 data are taken. The selected criticality benchmarks, which range from fast to thermal neutron spectra, are minimally impacted by the few neutrons above $E_{\text{inc}} = 5$ MeV which induce fission. The evaluated covariances obtained in this work were used to calculate uncertainties in k_{eff} according to the “Sandwich rule” (Eq. (16)). In comparison to the evaluation of Case A, the mean PFNS values and covariances of [9] in ENDF/B-VII.1 were used for the benchmark calculations. The k_{eff} for $i = \{\text{Case B, Case C}\}$ were calculated by:

$$k_{\text{eff}}^i = k_{\text{eff}}^{\text{VII.1}} \left[1 + \sum_j S_{k,j} \left(\psi_j^{post,i} / \psi_j^{\text{VII.1}} - 1 \right) \right],$$

where $k_{\text{eff}}^{\text{VII.1}}$ was calculated with the ENDF/B-VII.1 ${}^{239}\text{Pu}$ PFNS, $\psi_i^{\text{VII.1}}$. The respective uncertainties of k_{eff} were calculated following Eq. (16). For all the sensitivity/uncertainty calculations using the newly evaluated covariances and the ENDF/B-VII.1 covariances, the uncertainties were only propagated for incident neutron energies up to 5 MeV.

A set of criticality benchmarks used for the validation of MCNP was chosen in the present work to offer an idea of the impact of the new PFNS evaluation work. All these criticality safety benchmarks

can be found in the ICSBEP handbook [54], which are used widely for validation of transport codes like MCNP as well as validation of nuclear data. The calculated k_{eff} using evaluated PFNS of Cases A, B and C agree very well within their uncertainties despite the different assumptions made for Cov^{MD} in Section 3.2.2. The k_{eff} uncertainties of Cases B and C are also similar in size while those using the a-posteriori rescaled evaluated uncertainties of Case A are approximately a factor 1.4-1.5 larger for the benchmarks in Fig. 13. The k_{eff} uncertainties calculated using the evaluated covariances of Case A are slightly smaller than a factor of 1.6 compared to those calculated with a-posteriori rescaled uncertainties. This difference between calculated k_{eff} uncertainties shows that the uncertainty contribution added at low outgoing energies to the latter evaluated covariances has little impact on k_{eff} uncertainties (see also Fig. 6 of [1]). Hence, the choice of Cov^{MD} has little impact on k_{eff} values.

For the ^{239}Pu Jezebel (PMF1 in Fig. 13) critical assembly, the uncertainties were calculated to be 0.160% and 0.131% using the ENDF/B-VII.1 evaluation and the evaluation of Case A, respectively. In

Figure 13: (Color online) k_{eff} and the uncertainties due to the PFNS of the $n+^{239}\text{Pu}$ fission reaction for the ENDF/B-VII.1 evaluation and the present evaluation work. The Monte Carlo statistical uncertainties (~ 10 pcm for all cases) are not included on the plot.

Fig. 13, the results of the new PFNS evaluation show some notable trends. The k_{eff} is decreased for the fast spectrum systems (PMF1 – PMF8 on the left-hand-side), nearly unchanged for the intermediate spectrum systems (PMF11 and PCI1) and increased for the thermal spectrum systems (PST21 and MCT2). These changes are due to the shift toward a softer energy spectrum in the new evaluation of the PFNS. The uncertainties in Fig. 13 are smaller in the new evaluation.

The mean value of k_{eff} for many of the selected ICSBEP benchmarks are further away from unity, as would be desired to signify consistency between the evaluated differential and integral data, but many of the error bars in the present work overlap with those of the ENDF/B-VII.1 results. These error bars contain only uncertainties due to the PFNS covariances. If uncertainties of other evaluated data, e.g. ^{239}Pu total inelastic cross sections, are considered, the error bars would increase. Given that this evaluation of the mean value and uncertainty of the PFNS for the $n(500 \text{ keV})+^{239}\text{Pu}$ fission reaction includes model improvements and a new experimental data set, along with the older re-evaluated experimental data sets, other sources (i.e. cross sections, fission $\bar{\nu}$, angular distributions, etc.) of discrepancies with the ICSBEP benchmarks should be investigated.

In addition to criticality simulations, we have also calculated selected spectral indexes (i.e., the ratio of a specific reaction rate to that of $^{235}\text{U}(n,f)$) in the central region of the Jezebel critical assembly. Measured data are available for (i) the ^{169}Tm , ^{191}Ir and $^{238}\text{U}(n,\gamma)$ capture reactions; (ii) the ^{239}Pu , ^{237}Np and $^{238}\text{U}(n,f)$ fission reactions, and; (iii) the ^{238}U , ^{169}Tm and ^{191}Ir high energy ($n,2n$) threshold reactions. These data were recently re-evaluated and calculation-to-experiment (C/E) comparisons with ENDF/B-VII.1 nuclear data were reported in Refs. [5] and [8].

C/E values for calculations using ENDF/B-VII.1 nuclear data and for the same data except for

the new ^{239}Pu PFNS are provided in Table IV. The calculated average energy⁴($E_{50\%}$) for the reaction of interest is also given. The results are very similar for Cases A, B and C considering the MC uncertainties on the calculated values. Only for the higher threshold reactions, small differences are observed but the MC uncertainties are also higher for those reactions.

Table IV: Spectral Index C/E Values and their average energy ($E_{50\%}$) for selected reactions for the central region of the Jezebel (ICSBEP Pu-MET-FAST-001 Benchmark) Critical Assembly. The uncertainties quoted for “Case A” are also applicable for Cases B and C and. All uncertainties given are only MC uncertainties based on the number of neutron histories used to calculate the spectral indexes.

Reaction	$E_{50\%}$	C/E	C/E	C/E	C/E
	(MeV)	ENDF/B-VII.1	Case A	Case B	Case C
$^{169}\text{Tm}(n,\gamma)$	0.669	$1.168 \pm 0.06\%$	$1.192 \pm 0.2\%$	1.194	1.192
$^{191}\text{Ir}(n,\gamma)$	0.807	$0.950 \pm 0.06\%$	$0.966 \pm 0.2\%$	0.967	0.965
$^{238}\text{U}(n,\gamma)$	0.846	$0.966 \pm 0.06\%$	$0.980 \pm 0.2\%$	0.981	0.977
$^{239}\text{Pu}(n,f)$	1.877	$0.996 \pm 0.06\%$	$0.993 \pm 0.2\%$	0.993	0.994
$^{237}\text{Np}(n,f)$	2.346	$0.997 \pm 0.06\%$	$0.985 \pm 0.2\%$	0.984	0.985
$^{238}\text{U}(n,f)$	3.280	$0.983 \pm 0.06\%$	$0.961 \pm 0.2\%$	0.959	0.961
$^{238}\text{U}(n,2n)$	8.464	$1.253 \pm 0.28\%$	$1.143 \pm 0.9\%$	1.148	1.152
$^{169}\text{Tm}(n,2n)$	10.672	$1.177 \pm 0.6\%$	$1.030 \pm 2.0\%$	1.016	1.002
$^{191}\text{Ir}(n,2n)^5$	10.734	$1.137 \pm 0.6\%$	$0.995 \pm 2.0\%$	0.982	0.968

The average energy for these nine reactions cluster into two distinct energy intervals; with the six (n, γ) capture and (n,f) fission reaction average energies occurring below a few MeV while the three (n,2n) reaction average energies occur closer to 10 MeV. As such, these data probe very different portions of the PFNS. At low energies the C/E spectral index values obtained with the new PFNS are close to unity (the $^{169}\text{Tm}(n,\gamma)$ result being a clear outlier, but with the other five low energy C/E values near unity it seems likely that the basic $^{169}\text{Tm}(n,\gamma)$ cross section data are deficient). This is an expected result as at low energies the new ^{239}Pu PFNS is similar to the existing ENDF/B-VII.1 ^{239}Pu PFNS. This is also a desirable result as the C/E values at these lower average reaction energies are already close to unity. A clear improvement is seen in the C/E values for the high energy threshold (n,2n) reactions, as the ENDF/B-VII.1 based results vary from about 1.14 to 1.25, dropping to about 0.97 to 1.15 with the new ^{239}Pu PFNS. The $^{238}\text{U}(n,2n)$ using the new evaluations is improved compared to ENDF/B-VII.1, but still an outlier.

5 Summary and Outlook

Evaluations of the ^{239}Pu prompt fission neutron spectrum (PFNS) induced by neutrons of 500 keV and associated covariances were presented. In this work, we used the experimental data sets of [7, 24–30] as well as Los Alamos model (LAM) calculations [11]. We followed a consistent evaluation procedure while significantly improving the analysis of the experimental data and their uncertainties, extended the LAM following known physical constraints and experimental observations [9, 37, 38], included uncertainties for more model parameters and studied different approaches to estimate model defects.

The evaluation procedure takes into account the qualitative difference between normalized LAM predictions and bin-averaged experimental shape data and scales experimental data with respect to model values. This specific procedure is used to avoid biases in the evaluated results as shown in recent studies [12, 15].

⁴The average reaction energy given in Table IV is for ENDF/B-VII.1 nuclear data; use of the new ^{239}Pu PFNS generally decreases these energies by about 10 keV.

⁵For the $^{191}\text{Ir}(n,2n)$ reaction, the calculated daughter product is $^{190}(g+m1+m2+0.084*m3)\text{Ir}$.

The present work also addressed two important issues raised in previous works [9, 10] and blamed for very low evaluated uncertainties encountered there: We performed more realistic assessments of the experimental uncertainties. We extended the LAM, included uncertainties of more model parameters but we also included different model defect covariances in the prior to avoid very strong correlations in the final covariance matrix, which also led to very small uncertainties as shown in [12]. The results are evaluated PFNS in better agreement with available experimental data, and more reasonable evaluated uncertainties compared to experimental uncertainty information.

The k_{eff} values and C/E values for spectral indexes calculated with the different model defect uncertainties agree well within their uncertainties despite the different assumptions made. We note that the calculated multiplication factor k_{eff} in selected criticality benchmarks is in slightly poorer agreement with experimental data with the new evaluation than with the ENDF/B-VII.1 results. The C/E values for spectral indexes measured in the central region of the Jezebel critical assembly for the ^{169}Tm , ^{191}Ir and ^{238}U (n, γ) reaction and for the ^{239}Pu , ^{237}Np and ^{238}U (n,f) reaction are close to unity for both the newly evaluated PFNS and the original ENDF/B-VII.1 PFNS. The C/E values for high-energy threshold ^{238}U , ^{169}Tm , ^{191}Ir (n,2n) reactions are improved compared to calculations using ENDF/B-VII.1. With a variety of improvements to the measured data and theoretical models used throughout the evaluation process, these new evaluations of the ^{239}Pu PFNS need to be tested in other applications of interest, possibly including other reactor physics applications.

In summary, the evaluated PFNS and relative uncertainties obtained above are consistent with experimental input data and lead to reasonable benchmark results. Consequently, this work can and will be used as a starting point for an evaluation of the ^{239}Pu PFNS for incident neutron energies from thermal to 30 MeV which is of high interest for the CIELO [5] project.

This evaluation could be improved by including new and precise experimental PFNS data but also by improved model information. During the final phase of this paper, the experimental PFNS of [55] were recovered and it will be investigated whether they are suitable for an evaluation. Despite the extensions made in this work to the LAM compared to previous evaluations [9, 10], one should be aware that the extended LAM parameter space restrains the evaluated uncertainties too much. The proposed ad-hoc fixes lead to similar evaluated results and have little impact on the benchmarks. However, the ultimate goal is to use a more sophisticated model such as [42]. This model takes into account the many fragmentations occurring in the fission process, uses auxiliary experimental data (e.g., $\langle TKE \rangle$, fission yields) to constrain its model parameters and also predicts more observables such as e.g., the prompt fission gamma spectrum and the neutron multiplicity. Also, scission and pre-fission neutrons are not taken into account in the LAM. As the existence and magnitude of scission neutrons remains controversial, more direct experimental evidence is needed.

A Impact of Improved Uncertainty Quantification for Model and Experimental Data

The impact of the improved experimental uncertainty quantification is studied by a statistical analysis (E1) using the experimental data and covariances used in [9], and by a statistical analysis (E2) using the same experimental data sets and the experimental uncertainty quantification of Section 3.1. The model information of Section 3.2 is used for both evaluations. A similar study using preliminary data is presented in Ref. [56]. In Ref. [9], experimental data of [24–26, 28] were used. Experimental correlations were estimated in a simplified manner assuming a correlation of 0.5 between uncertainties of the same experiment and 0.0 between different experiments. If we use these experimental covariances, the evaluated mean values show a notable difference compared with using the same experimental data sets with the experimental covariances of Section 3.1 as shown in Fig. 14(a). The evaluated relative uncertainties shown in Fig. 14(b) are underestimated substantially by 15–65% if we use experimental covariances estimated for [9].

Figure 14: The ratio of evaluated PFNS and relative uncertainties using the model values and covariances of [9] and the current evaluation using $\mathbf{Cov}^{MD}$ of Case A as well as the ratio of evaluated PFNS using the experimental data and covariances of [9] and the current uncertainty quantification for the same data sets are shown.

The impact of the improved model uncertainty quantification is studied by a statistical analysis (E3) using model results and covariances of [9] in comparison to the current evaluation (E4) using $\mathbf{Cov}^{MD}$ of Eq. (26) (Case A). The experimental data and covariances of 3.1 are used for both evaluations. In [9], model covariances were estimated using non-zero uncertainties for only $\langle E_r \rangle$, $\langle TKE \rangle$, the average level density parameter and B_n (Table 1 in [9]). In the present work, we extended the LAM and used uncertainties for many more LAM parameters (Table II in Section 3.2.1). The resulting model relative uncertainties are of comparable size but the model covariances are less strongly correlated than those of [9]. The evaluated PFNS assume lower values below 500 keV in Fig. 14(a) using the previous model information and cannot describe Starostov et al. data equally well. This tendency is also observed in ENDF/B-VII.1 data in Fig. 11, which does not appear in the evaluated data of [10]. In [10] and the current work, an anisotropy in the emission of neutrons in the center-of-mass frame was introduced in the LAM which leads to higher PFNS at lower outgoing neutron energies. The evaluated relative uncertainties using the model information of [9] are substantially reduced by 5–70% compared to using the one of Section 3.2. This study is in agreement with the finding in Ref. [12] that the low evaluated uncertainties obtained in [9] were partly caused by the strong model correlations in combination with the normalization conditions.

B Impact of δNo on the Evaluation

Here, we show the impact of the magnitude of δNo on the evaluated results. A factor of $\delta No = 0\%$ corresponds to a model covariance matrix which still satisfies the normalization condition of Eq. (2). The corresponding evaluated PFNS and relative uncertainties are the most strongly affected ones compared to non-zero values of δNo as shown in Figs. 15(a) and 15(b). However, the impact of δNo is small.

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(a) PFNS

(b) Relative Uncertainties

Figure 15: The ratios of evaluated PFNS and relative uncertainties using $\delta No = \{0, 3, 10\}\%$ versus the final evaluation ($\delta No = 6\%$) are shown.

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References

- [1] I. Kodeli et al., Nucl. Inst. Meth. A 610 (2009) 540.
- [2] W.S. Yang et al., Nucl. Data Sheets 109 (2008) 2852.
- [3] R. Capote et al., Prompt Fission Neutron Spectra of Major Actinides, INDC(NDS)-0541, International Atomic Energy Agency (2009).
- [4] R. Capote et al., Prompt Fission Neutron Spectra of Major Actinides, INDC(NDS)-0571, International Atomic Energy Agency (2010).
- [5] M.B. Chadwick et al., Nucl. Data Sheets 118 (2014) 1.
- [6] A. Chatillon et al., Phys. Rev. C 89 (2014) 014611.
- [7] J.P. Lestone and E.F. Shores, Uranium and Plutonium Prompt-fission-neutron Spectra (PFNS) from the Analysis of NTS NUEX Data and the Corresponding Uncertainty Budget, LA-UR-14-24087, Los Alamos National Laboratory (2014).
- [8] M.B. Chadwick et al., Nucl. Data Sheets 112 (2011) 2887.
- [9] P. Talou et al., Nucl. Sci. Eng. 166 (2010) 254.
- [10] M.E. Rising et al., Nucl. Sci. Eng. 175 (2013) 81.
- [11] D.G. Madland and J.R. Nix, Nucl. Sci. Eng. 81 (1982) 213.
- [12] D. Neudecker, R. Capote, D.L. Smith, T. Burr and P. Talou, Nucl. Sci. Eng. 179 (2015) 381.
- [13] ENDF-6 Formats Manual, BNL-90365-2009 Rev.2, Brookhaven National Laboratory (2012); edited by A. Trkov, M. Herman and D.A. Brown.
- [14] D.L. Smith, D. Neudecker and R. Capote, Prompt Fission Neutron Spectrum Evaluation Techniques, [INDC\(NDS\)-0678](#), [International Atomic Energy Agency \(2015\)](#).
- [15] A. Trkov, R. Capote and V. Pronyaev, Current Issues on Nuclear Data Evaluation Methodology: ^{235}U Prompt Fission Neutron Spectra and Multiplicity for Thermal Neutrons, Nucl. Data Sheets [123 \(2015\) 8](#).
- [16] D.L. Smith and N. Otuka, Nucl. Data Sheets 113 (2012) 3006.
- [17] J.W. Boldeman, Review of Measurements of the Prompt Fission Neutron Spectrum from the Spontaneous Fission of Cf-252, IAEA-TECDOC-410, 125-143 (1987).
- [18] G. Zerovnik et al., Random Sampling of Correlated Parameters - a Consistent Solution for Unfavourable Conditions, Nucl. Data Sheets [123 \(2015\) 185](#).
- [19] T. Kawano et al., J. Nucl. Sci. Techn. 37 (2000) 327.
- [20] J.T. Goorley, MCNP6.1.1-Beta Release Notes, Los Alamos National Laboratory, LA-UR-14-24680 (2014).
- [21] MCNP6 Development Team, Initial MCNP6 Release Overview MCNP6 Version 1.0, Los Alamos National Laboratory, LA-UR-13-22934 (2013).
- [22] A.C. Kahler III and R. Macfarlane, The NJOY Nuclear Data Processing System, Version 2012, LA-UR-12-27079 (2012).

- [23] B.C. Kiedrowski, MCNP6.1 k-Eigenvalue Sensitivity Capability: A User's Guide, Los Alamos National Laboratory, LA-UR-13-22251 (2013).
- [24] H.-H. Knitter, Atomkernenergie 26 (1975) 76, EXFOR-No. 20576.001.
- [25] P. Staples et al., Nucl. Phys. A 591 (1995) 41, EXFOR-No. 13982.001.
- [26] A. Lajtai et al., Energy Spectrum Measurements of Neutrons for Energies 30 keV - 4 MeV from thermal fission of main fuel elements, Proceedings of the Conf. on Nuclear Data for Basic and Applied Sciences, Santa Fe, USA, 1985, Vol. 1, p. 613 (1985), EXFOR-No. 30704.001.
- [27] V.N. Nefedov, B.I. Starostov and A.A. Boytsov, Precision measurements of ^{252}Cf , ^{233}U , ^{235}U and ^{239}Pu prompt fission neutron spectra (PFNS) in the energy range 0.04 - 5 MeV, INDC(CCP)-0457, International Atomic Energy Agency (2014), translation into English from: Proceedings of the All-Union Conf. on Neutron Physics, Kiev, USSR, 1983 2, p. 285 (1983), EXFOR-No. 40871.009.
- [28] A.A. Boytsov, A.F. Semenov and B.I. Starostov, Relative measurements of $^{233}\text{U}+\text{n}_{\text{th}}$, $^{235}\text{U}+\text{n}_{\text{th}}$ and $^{239}\text{Pu}+\text{n}_{\text{th}}$ prompt fission neutron spectra (PFNS) in the energy range 0.01 - 5 MeV, INDC(CCP)-0459, International Atomic Energy Agency (2014), translation into English from: Proceedings of the All-Union Conf. on Neutron Physics, Kiev, USSR, 1983, Vol. 2, p. 294 (1983), EXFOR-No. 40873.006.
- [29] B.I. Starostov, V.N. Nefedov and A.A. Boytsov, Precision measurements of ^{252}Cf , $^{233}\text{U}+\text{n}_{\text{th}}$, $^{235}\text{U}+\text{n}_{\text{th}}$ and $^{239}\text{Pu}+\text{n}_{\text{th}}$ prompt fission neutron spectra (PFNS) in the energy range 2 - 11 MeV, INDC(CCP)-0458, International Atomic Energy Agency (2014), translation into English from: Proceedings of the All-Union Conf. on Neutron Physics, Kiev, 2-6 Oct. 1983, 2, p. 290 (1983), EXFOR-No. 40872.003.
- [30] B.I. Starostov, V.N. Nefedov and A.A. Boytsov, Prompt neutrons spectra from the thermal neutron fission of ^{233}U , ^{235}U , ^{239}Pu and spontaneous fission of ^{252}Cf in the secondary neutron energy range 0.01 - 12 MeV, INDC(CCP)-293/L, INDC Report, p. 19 (1989), translation into English from: Nuclear Constants, 3, (1985), EXFOR-No. 40930.001.
- [31] W. Mannhart, IAEA Report IAEA-TECDOC-410, Evaluation of the Cf-252 Fission Neutron Spectrum between 0 MeV and 20 MeV, p. 158-171 (1987).
- [32] W. Mannhart, IAEA Report INDC(NDS)-220/L, Status of the Cf-252 Fission-Neutron Spectrum Evaluation with Regard to Recent Experiments, p. 305-336 (1989).
- [33] D. Neudecker et al., Preliminary Evaluation and Uncertainty Quantification of the Prompt Fission Neutron Spectrum of ^{239}Pu , Nucl. Data Sheets [123 \(2015\) 146](#).
- [34] D. Neudecker et al., Compilation of ^{239}Pu PFNS experimental covariance matrices, LA-UR-13-24743, Los Alamos National Laboratory (2013).
- [35] T.N. Taddeucci et al., Multiple-scattering Corrections to Measurements of the Prompt Fission Neutron Spectrum, Nucl. Data Sheets [123 \(2015\) 135](#).
- [36] T. Ohsawa, T. Horiguchi and M. Mitsuhashi, Nucl. Phys. A 665 (2000) 3.
- [37] F.-J. Hambsch et al., Ann. Nucl. Eng. 32 (2005) 1032.
- [38] J. Terrell, Phys. Rev. 113 (1959) 527.
- [39] T. Kawano, P. Talou, M.B. Chadwick, and T. Watanabe, J. Nucl. Sci. Technol. 47 (2010) 462.

- 729 [40] D.G. Madland, Nucl. Phys. A 772 (2006) 113.
- 730 [41] J. Frehaut, A. Bertin and R. Bois, Mesure de $\bar{\nu}_p$ et $\bar{E}_\gamma$ Pour la Fission de ^{232}Th , ^{235}U et ^{237}Np
731 Induite par des Neutrons D'Energie Comprise Entre 1 e 15 MeV, Proc. of the Con. on Nucl. Data
732 for Sci. and Technology, 78 (1983).
- 733 [42] B. Becker et al., Phys. Rev. C 87 (2013) 014617.
- 734 [43] F.D. Becchetti and G.W. Greenlees, Phys. Rev. 182 (1969) 1190.
- 735 [44] A.J. Koning and J.P. Delaroche, Nucl. Phys. A 713 (2003) 231.
- 736 [45] S.F. Mughabghab, "Atlas of Neutron Resonances: Resonance Parameters and Neutron Cross
737 Sections, Z = 1-100," (Elsevier, Amsterdam 2006).
- 738 [46] R. Capote et al., Nucl. Data Sheets 110 (2009) 3107.
- 739 [47] C. Wagemans, "The Nuclear Fission Process," (CRC Press, 1991).
- 740 [48] G.A. Petrov et al., Phys. of Atomic Nuclei 71 (2008) 1137.
- 741 [49] N. Kornilov, F.-J. Hambsch and A.S. Vorobyev, Nucl. Phys. A 789 (2007) 55.
- 742 [50] R. Vogt et al., Phys. Rev. C 80 (2009) 044611.
- 743 [51] O. Litaize and O. Serot, Phys. Rev. C 82 (2010) 054616.
- 744 [52] D.E. Vaughan and D.L. Preston, Physical Uncertainty Bounds, Los Alamos National Laboratory,
745 LA-UR-14-20411 (2014).
- 746 [53] M.E. Rising, A.K. Prinja and P. Talou, Nucl. Sci. Eng. 175 (2013) 188.
- 747 [54] International Handbook of Evaluated Criticality Safety Benchmark Experiments, OECD Nuclear
748 Energy Agency report NEA/NSC/DOC(95)03, September 2013 Edition.
- 749 [55] A. Smith et al., Nucl. Sci. Eng. 76 (1980) 357.
- 750 [56] D. Neudecker, P. Talou and T. Kawano, Transactions of the American Nuclear Society 111 (2014)
751 1415.