

Systematic Quantification of Uncertainties for Evaluated Prompt Fission Neutron Spectra and Multiplicities

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Abstract. Uncertainties associated with evaluated average prompt fission neutron spectra and multiplicities are obtained for a suite of actinides in the Los Alamos model formalism. Systematics for the model input parameters are taken from the literature and used as prior values in a Bayesian updating procedure. Posterior systematics as well as associated posterior uncertainties are inferred. In addition, cross-isotope correlations are evaluated for the first time. The quantification of uncertainties associated with advanced Monte Carlo Hauser-Feshbach calculations of prompt fission neutron spectra is also discussed.

1 Introduction

Recent years have seen a surge in the interest of quantifying uncertainties associated with evaluated nuclear data. Thanks to ever increasing computing power, running one or a hundred nuclear reaction simulations for data evaluation purposes is easily achievable by most evaluators. Studying the sensitivity of the results to (modest) changes in model parameter values is therefore possible and provides a relatively straightforward access to evaluated uncertainties (see for example large-scale TALYS runs [1]).

Obviously, performing large-scale nuclear reaction simulations does not provide fully adequate uncertainties associated with evaluated data. Since an uncertainty is not a physical quantity, but is instead an information on how well we know a physical datum, quantifying this uncertainty requires to assess all sources of uncertainties originating in the evaluation process itself. In an ideal situation, all the information that entered into the evaluation should be reflected in the associated uncertainty.

Nuclear data evaluations are often the result of a combination of partial experimental measurements and theoretical model calculations, which carry with them some uncertainties. In addition, it has been a common practice to *adjust* the results to match integral or semi-integral benchmark better. While this *adjustment* is usually performed within evaluated error bars, it is never accounted for in the uncertainty quantification (UQ) process. The added information is therefore included to modify the central values, without touching the associated uncertainties and correlations. We will discuss this issue again in the conclusion.

In the present paper, we discuss our recent work on evaluating prompt fission neutron spectrum (PFNS) and multiplicity (PFNM) for a suite of uranium isotopes, and on quantifying their uncertainties consistently. This is done in the framework of the Los Alamos model developed by Madland and Nix [2]. Prior model input parameters are obtained from established systematics, and a Bayesian inference scheme is used to combine model calculations and

known experimental data. By using systematics as a function of isotope and incident neutron energy, correlations appear between isotopes as well as between energies.

The Bayesian inference scheme is presented in Section 2. Section 3 contains a brief introduction to the physics of prompt fission neutrons and gamma rays, as well as current theoretical models used to describe them. In particular, the Los Alamos (LA) model [2] used in the present work is discussed at some length. Recent work on advanced modeling of prompt fission neutrons and gamma rays is also briefly introduced, as it constitutes a natural extension of the present work. Some details on experimental efforts to measure PFNS and PFNM are discussed in Section 3.2. In the course of this work, a new evaluation code package was developed and is discussed briefly in Section 3.3. Numerical results for the suite of uranium isotopes studied here are presented in Section 4.3. Finally, we conclude by a discussion of the results and an outlook on the next steps to improve the current work for PFNS, and beyond.

2 Quantifying PFNS Uncertainties

Our approach to quantifying uncertainties on prompt fission neutron spectrum and multiplicity is very similar to the one we have used successfully for reaction cross sections [5]. It was described at some length in Ref. [6], but we are presenting it here briefly again for the sake of completeness.

A Bayesian approach is used to combine experimental and model parameter uncertainties. The equations for the Bayesian updating scheme used are as follows:

$$\begin{aligned} \mathbf{m}_1 &= \mathbf{m}_0 + \mathbf{P}\mathbf{C}^t\mathbf{V}^{-1}(\phi - f(\mathbf{m}_0)) \\ &= \mathbf{m}_0 + \mathbf{X}\mathbf{C}^t(\mathbf{C}\mathbf{X}\mathbf{C}^t + \mathbf{V})^{-1}(\phi - f(\mathbf{m}_0)), \end{aligned} \quad (1)$$

$$\begin{aligned} \mathbf{P} &= (\mathbf{X}^{-1} + \mathbf{C}^t\mathbf{V}^{-1}\mathbf{C})^{-1} \\ &= \mathbf{X} - \mathbf{X}\mathbf{C}^t(\mathbf{C}\mathbf{X}\mathbf{C}^t + \mathbf{V})^{-1}\mathbf{C}\mathbf{X}. \end{aligned} \quad (2)$$

In those equations, \mathbf{m}_0 represents the vector of *prior* model parameters, and \mathbf{m}_1 is the vector of *posterior* model parameters. The quantity $f(\mathbf{m}_0)$ is the calculated spectrum using

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prior parameter values. ϕ is the experimental spectrum. \mathbf{X} and \mathbf{P} are the *prior* and *posterior* model parameter covariance matrices, respectively. Finally, \mathbf{V} represents the experimental covariance matrix, and \mathbf{C} the sensitivity matrix whose elements are given by

$$C_{ij} = \frac{\partial N(E_i)}{\partial p_j} \times \frac{p_j}{N(E_i)} \quad (3)$$

at the energy E_i for the parameter p_j . The evaluated covariance matrix \mathbf{F} for the prompt neutron spectrum is finally obtained as

$$\mathbf{F} = \mathbf{CPC}^t. \quad (4)$$

3 Prompt Fission Neutrons

In the present work, we refer to *prompt fission neutrons and gamma rays* as those emitted from the fission fragments formed right after the break up of a heavy nucleus. Those prompt particles are emitted within about 10^{-18} to 10^{-7} sec after scission [7], until fission *products* are formed in their ground-state or long-lived isomeric states. Those products may eventually decay by weak interaction, and lead to the production of additional β -*delayed* neutrons and gamma rays.

Most modern evaluated nuclear data libraries contain information on the average prompt fission neutron spectrum (PFNS) and average prompt fission neutron multiplicity (PFNM) as a function of the incident neutron energy. Those quantities are commonly evaluated using the so-called Madland-Nix or Los Alamos (LA) model [2], or a simpler representation such as a Watt or Maxwellian spectrum, or a combination thereof.

More recently, Monte Carlo studies following the de-excitation of the primary fission fragments by successive emissions of neutrons and gamma rays have been developed successfully (for example, see Ref. [8]) and will be used in upcoming revisions of evaluated libraries.

3.1 Theory

3.1.1 Los Alamos Model

Prompt fission neutron spectra in the ENDF/B-VII.0 library were evaluated using the Los Alamos model [2]. Because the uncertainty quantification methodology follows the one for the evaluation of the PFNS in the first place, we recall here the major results of the Los Alamos model and identify the model parameters that enter in the following sensitivity calculations. We refer the interested reader to Ref. [2] for more details.

The neutron energy spectrum $N(E)$, in the laboratory frame, for a fission fragment moving with a kinetic energy per nucleon E_f reads

$$N(E) = \frac{1}{2\sqrt{E_f T_m^2}} \int_{(\sqrt{E}-\sqrt{E_f})^2}^{(\sqrt{E}+\sqrt{E_f})^2} d\epsilon \sigma_c(\epsilon) \sqrt{\epsilon} \times \int_0^{T_m} dT k(T) T \exp(-\epsilon/T), \quad (5)$$

where ϵ is the center-of-mass neutron energy, $k(T)$ is the temperature-dependent normalization constant given by

$$k(T)^{-1} = \int_0^\infty d\epsilon' \sigma_c(\epsilon') \epsilon' \exp(-\epsilon'/T). \quad (6)$$

$\sigma_c(\epsilon')$ is the energy-dependent cross section for the inverse process of compound nucleus formation, and T_m is the maximum value of the triangular distribution of temperatures used to describe the excitation energy distribution in the fragments. Considering the most probable fragmentation only, the average neutron energy spectrum in the laboratory frame is given by a simple average over the spectra for the light and heavy fragments. Following the original formulation of the LA model, an equal number of neutrons emitted from the light and heavy fragments is assumed.

At higher incident energies, these formulas can be generalized to take into account the onset of multi-chance fission, i.e. the probability that one or more neutrons are emitted before the residual compound nucleus fissions. However, in the present work, we are only concerned with low-energy (0.5 MeV) fission for which only the first-chance fission component is non-zero.

3.1.2 Monte Carlo Hauser-Feshbach

In the Monte Carlo Hauser-Feshbach (MCHF) approach, as described in [3,4], the de-excitation of the primary fission fragments is followed exactly for each neutron and gamma-ray emission using the Hauser-Feshbach formalism and emission probabilities obtained from optical model calculations for the neutrons and gamma-ray strength functions for the gamma rays. Such simulations require to know the fission fragment yields as a function of mass and charge of the fission fragment. Also, the initial excitation energy, spin and parity distributions in each decaying fragment has to be known. Once those initial conditions are fixed, Hauser-Feshbach calculations are performed at each stage of the decay, and characteristics of the emitted particles are recorded and analyzed.

A fundamental advantage of MCHF calculations compared to the LA model is that various *exclusive* data can be assessed, as opposed to only two average quantities, i.e., average PFNS and PFNM. Modern experiments can explore this much richer phase-space and shed some light on the very interesting physics near the scission point.

MCHF calculations to study fission fragment decay implies to use a large number of data and model parameters for a large number of nuclei. Those parameters are the same as those that appear in standard Hauser-Feshbach calculations, and can be probed in various ways. One of them is the so-called Total Monte Carlo (TMC) approach as described in Ref. [1]. Besides those parameters, uncertainties in the determination of the primary fission fragment yields have also to be taken into account.

3.2 Experiments

Recently, significant experimental efforts have been initiated to improve our knowledge of prompt fission neutrons and in particular to obtain an accurate determination of

their average spectrum [9–12]. These renewed efforts come with the realization that uncertainties in evaluated spectra can lead to significant uncertainties in integral quantities that are relevant to many nuclear applications.

In the present work, experimental covariance matrices were estimated in a very simplified fashion, and no effort has been made to get a more realistic representation of the systematic uncertainties. In particular, known cross-correlations between measurements and isotopes have not been taken into account. This is a clear starting point for improving the preliminary results presented here.

3.3 Evaluation

In the course of this work, we have developed an evaluation package for analyzing and predicting PFNS and PFNM. Written in modern Fortran, it is made up of various modules that can be used to perform LA model calculations and parameter sensitivity studies, read experimental data and uncertainties, perform Bayesian statistics, create probability distribution functions of model parameters, generate covariance matrices, and read and write ENDF-formatted prompt fission neutron spectrum, multiplicity and covariance matrices. It also contains a module for producing high-quality figures easily.

It represents a powerful tool to perform any serious study related to the evaluation of prompt fission neutrons. Plans to expand its use to MCHF calculations are in development.

4 Study of a Suite of Uranium Isotopes

In the following, numerical results are presented for a suite of uranium isotopes, for which the LA model was used.

4.1 Model Parameter Systematics

In order to study a suite of uranium isotopes, we have used the model parameter systematics of Tudora [13] as *prior* values in the Bayesian analysis. At low incident neutron energies, i.e., below the threshold for second-chance fission, the most important input parameters entering in LA model calculations are the average Total Kinetic Energy $\langle TKE \rangle$, the average energy release $\langle E_r \rangle$, the average level density parameter $\langle a \rangle$, and the average neutron separation energy $\langle S_n \rangle$. In theory, $\langle E_r \rangle$ can be obtained from calculating the Q -value of the fission reaction for each fission fragment pair appearing in the observed mass and charge yields, and $\langle S_n \rangle$ can also be derived approximately from mass difference calculations over the entire chain of prompt neutron decay. However, the large number of unknowns leaves room for an adjustment on those parameters, and can be considered free within known limits.

By studying a large set of isotopes, Tudora has inferred [13] fitted formulae for those parameters at thermal neutron energy. They are of the following form:

$$\begin{aligned} \langle TKE \rangle_{th} &= \alpha_1 + \alpha_2 Z^2 / A^{1/3}, \\ \langle E_r \rangle_{th} &= \alpha_3 + \alpha_4 x + \alpha_5 x^2, \\ \langle S_n \rangle_{th} &= \alpha_6 + \alpha_7 x + \alpha_8 x^2, \\ \langle a \rangle &= A / \alpha_9, \end{aligned} \quad (7)$$

where x is the fissility parameter given by

$$x = \frac{Z^2}{A} \left[50.883 \left(1 - 1.7826 \frac{(N-Z)^2}{A^2} \right) \right]. \quad (8)$$

Numerical values for the $\{\alpha_i\}$ parameters can be found in [13] for suites of isotopes, and will not be repeated here.

Tudora also includes a slight linear dependence of the average Total Kinetic Energy $\langle TKE \rangle$ as a function of the incident neutron energy. At the lowest incident energies, it results in a very small change only. In this work, we do not include any energy dependence of the LA model parameters.

By starting from model parameter systematics, we are shifting the problem from the suite of parameters $\{\langle TKE^i \rangle, \langle E_r^i \rangle, \langle S_n^i \rangle, \langle a^i \rangle\}_{i=1..N}$, where N is the number of isotopes considered, to a set of nine parameters $\{\alpha_1, \dots, \alpha_9\}$ that determine the LA model parameter values for all isotopes at once. It allows a consistent evaluation of PFNS for a suite of isotopes, even for those with no existing measurement, thereby building correlations along the chain.

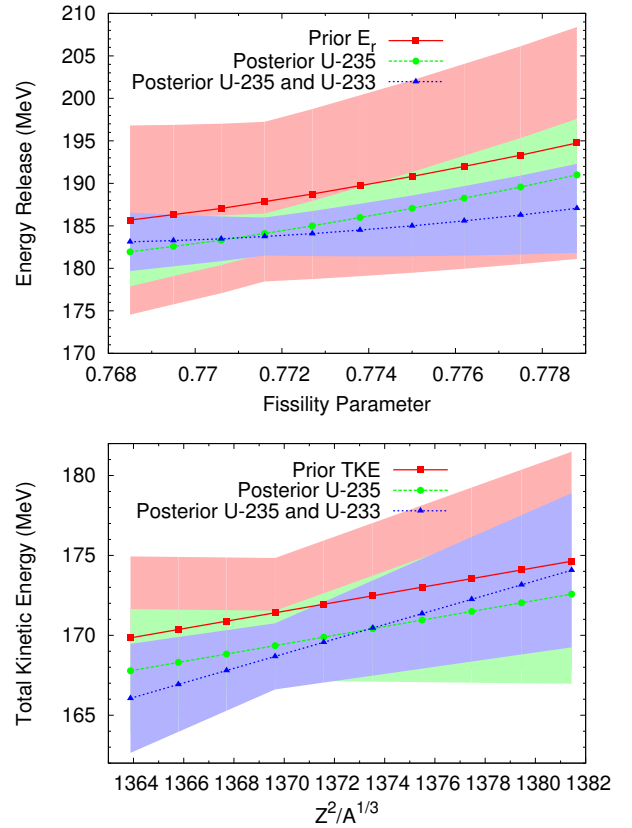


Fig. 1. Values of the model input parameters $\langle E_r \rangle$ and $\langle TKE \rangle$ appearing in the Los Alamos model. Prior values were taken from Tudora [13] and prior uncertainties were grossly estimated based on previous work [6]. Posterior values were obtained using ^{235}U experimental data first, then adding other data on ^{233}U thermal PFNS.

4.2 Experimental Data

Experimental data for the suite of uranium isotopes were collected and a selection of those is summarized in Table 1.

In particular, as expected the coefficients α_1 and α_3 , as well as α_4 , are strongly correlated to keep the effective temperature in the Los Alamos model equations within a reasonable range of values. A strong anti-correlation is also noted between α_3 and α_9 , i.e., between the average energy release and the average level density parameter. Those correlations had already been noted in [6].

5 Conclusion and Outlook

The results presented here represent a natural extension of our previous work on the quantification of uncertainties for the evaluated PFNS on $n(0.5 \text{ MeV})+^{239}\text{Pu}$ [6]. By studying more than one isotope and one incident neutron energy at a time, we are able to update evaluated PFNS even for those isotopes that have no associated measured data. It provides a more natural and encompassing framework not only for the quantification of uncertainties but also for the evaluations of prompt fission neutron spectra in general.

Several limitations of this preliminary work should be emphasized. First and foremost, the experimental covariance matrices used in the present study are rudimentary in their derivation. In addition no cross-correlations between data sets were considered, where they obviously exist. For instance, two Lajtai measurements [15] for ^{233}U and ^{235}U were performed in the very same experiment, and significant correlations in the determination of the experimental PFNS are expected.

Another limitation is the use of low-energy spectrum data only, while more data exist at higher incident neutron energies and even more data on the average neutron multiplicity $\bar{\nu}(E_{inc})$. Such data could easily be included in a further analysis using the exact same tools.

Finally, the Los Alamos model to represent a prompt fission neutron spectrum is based on several hypotheses and approximations that could now be lifted. In particular, Monte Carlo Hauser-Feshbach (MCHF) techniques are being developed [3,4,8] to study the de-excitation stage of the primary fission fragments in a much more detailed way. Such techniques can compute not only the average spectrum and multiplicity, as are obtained in the LA model, but also a wide array of physical quantities inaccessible with a simpler approach: $P(\nu)$, $\bar{\nu}(A,Z,TKE)$, exclusive spectra, n-n energy and angular correlations, etc. Experimental data exist for some of them, which could further constrain these more sophisticated calculations. Monte Carlo techniques such as described in [1] could be used to probe the large model parameter phase space, and infer realistic spectrum uncertainties.

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