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Test of models for $^{235}U(n_{th},f)$ charge distributions and their impacts on the covariance analysis

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Abstract. Fission yields are of major interest for the nuclear industry and the study of the fission process. Fission yields are determined by evaluation process based on the analysis of experimental datasets and completed by phenomenological models. The aim is to provide the best estimation of independent yields of $^{235}U(n_{th},f)$ reaction. In this paper, we will focus on the methodology of assessment of the nuclear charge distribution per mass. In the JEFF-3.1.1, the Wahl's parametrization of the Z_p-model is used to complete the experimental knowledge. Nevertheless, the lack of this parametrization consists in the absence of uncertainty information. Consequently, it is impossible to generate the components of independent yield correlation matrix from the Z_p-model. In this context, we propose to use the Z_p model with a new parametrization to assess the charge distributions. Two different calculations have been developed: the first one is based on a Wahl-like linear approximation of the parameters as a function of mass. The second approach consists to apply a "direct-Z_p model" without linear assumptions. This analysis is achieved using the Markov-Chain Monte-Carlo (MCMC) method to adjust the model parameters and then to deduce the covariance of charge distributions. Moreover, with our development, we have directly and consistently described the pre-neutron and post-neutron charge distributions.

1 Introduction

The exploitation of nuclear reactors dedicated to the production of electricity by chain reactions requires the control of a large number of physical quantities, throughout the fuel cycle. This control is the basis of the operation and safety of the installation and is partly based on nuclear data. In this sense, several research projects in different fields are underway, in particular work aimed to improving the quality of nuclear data. Among these data, there are the fission yields, which represent the production rates of fission products. These data are a major interest for the nuclear industry and the study of the fission process. They are used in the calculation and quantification of several observables, in particular for the fuel inventory [1], the burn-up calculation [2] or still for the reactor decay heat after its shutdown.

Historically, very important efforts have been made to understand the fission process in a coherent theoretical framework [3]. Despite these efforts, the current understanding of fission process is patchy and incomplete, and then a consistent description of the fission observables (pre-neutron yields, post-neutron yields, cumulative yields, neutron emission) is not available. In particular, this consistency between pre and post neutron yields is the base of the description of the fission yields, which are very important for the applications and the test of the fission models. It is clear that the data resulting from those models are not sufficiently accurate to satisfy the requirements of the nuclear industry. However, it is undeniable that the best estimation of fission yields is provided by current evaluation process based on the analysis of experimental data completed by phenomenological models. Thus, the aim is to provide the best estimation of isobaric, isotopic and isomeric yields.

These phenomenological models such as Wahl's model [4] and Madland-England's one [5] have shown great success. They have been used for the development of several libraries of evaluations such as the JEFF-3.1.1 [6], ENDF/B-VIII.0 [7] and JENDL/FPY-2011 [8] libraries. In those libraries, the charge distribution of independent yields are completed using the empirical Z_p-model proposed by A. C. Wahl based on linear parameterization known as Wahl's systematics [4]. It is important to note that the Z_p-model is initially used to describe the fractional yields of pre-emission neutron and can be translated to the post-neutron using the prompt neutron multiplicity (so-called saw-tooth). But in libraries, it was used directly to calculate post-neutron charge distribution. This represents a limitation in those evaluated libraries. In addition, a limitation of Wahl's systematics parameterization is mainly related to the lack of information on the uncertainties and the correlations of the model parameters. Consequently, it is impossible to generate the component of correlation matrix of charge distribution from the Z_p model. In this context, we propose to use the Z_p model with new parameterizations to complete the charge distributions. The aim is to test and to improve the accuracy of the methodology in order to generate the covariance matrix of independent yields.

2 Description of Z_p-model

The Z_p model consists in the modelling of the pre-neutron fractional yields with a Gaussian distribution of mean $Z_p(A^*)$ (representing the most probable charge) and the standard deviation $\sigma_Z(A^*)$. Two effects perturb the shape of the Gaussian distribution. The first is the charge polarization effect [4], which appears as a shift of $Z_p(A^*)$ from the charge Z_{UCD} based upon the assumption of the "unchanged charge density (UCD)" [4]. The UCD postulates that the charge-to-mass ratio of the fissioning nucleus, Z_F/A_F, is maintained for both fragments. The charge polarization effect is taken into account by adding a corrective term $\Delta Z(A^*) = Z_p(A^*) - Z_{UCD}$. The second is the pairing effect, which is related to the structure of the nucleus. To take into account this parity effect, A. C. Wahl propose two parameters, $F_Z(A^*)$ and $F_N(A^*)$ [4].

The pre-neutron emission charge distribution is therefore written:

$$P(Z|A^*) = \frac{F(A^*,Z)N(A^*)}{2}[erf(V) - erf(W)] \qquad (1)$$

With:

$$V = \frac{Z - Z_p(A^*) + 0.5}{\sigma_Z(A^*)\sqrt{2}}$$

$$W = \frac{Z - Z_p(A^*) - 0.5}{\sigma_Z(A^*)\sqrt{2}}$$

Where A^* represents the pre-neutron mass of the fission fragment, $F(A^*, Z)$ is the term to include the pairing effect and $N(A^*)$ is the normalization coefficient. To complete the description of the Z_p-model, A. C. Wahl proposed a parameterization to determine $\Delta Z(A^*)$, $\sigma_Z(A^*)$, $F_Z(A^*)$ and $F_N(A^*)$ known as Wahl's systematics.

In the following, we will test the compatibility between the values from Wahl's systematics and the experimental data. We will also introduce two approaches to propose new parameterizations for the Z_p model. Then, a comparison of different types of calculations will be presented and the results on the pre-neutron charge distribution and post-neutron charge distribution will be discussed.

3 Evaluation method

3.1 Pre and Post-neutron transition

The Z_p model is developed to determine the charge distribution of the fission fragments before prompt neutron emission. To determine the parameters of this model, we need to adjust those parameters on the available experimental data. However, these data corresponds to fission products after prompt neutron emission. It is therefore necessary to formulate a connection between the pre-neutron and post-neutron data. This connection is established through neutron emission probabilities. Indeed, we know that a given nucleus of mass A and charge Z is formed either by cold fission (zero neutron emission) or by several emissions of neutrons as indicated in the **Fig. 1**. We can therefore calculate the post-neutron fission yields from the pre-neutron fission yields using the formula:

$$N(A,Z) = \sum_{\nu=0}^{\infty} Y(A^*)P\left(Z\middle|A^*|\theta_{zp}\right)P(\nu|A^*) \qquad (2)$$

$$P(Z|A) = \frac{N(A,Z)}{\sum_Z N(A,Z)} \qquad (3)$$

With:

$$A^* = A + \nu$$

Where A* is the mass of fission fragment before neutron emission, A the mass of fission products, $P(\nu|A^*)$ represents the probability that a primary fission fragment of mass A^* emitting ν prompt neutrons, $Y(A^*)$ represents the mass yields, $P\left(Z\middle|A^*|\theta_{zp}\right)$ is the pre-neutron charge distribution, θ_{zp} is the parameters of Z_p model and P(Z|A) is the post-neutron charge distribution.

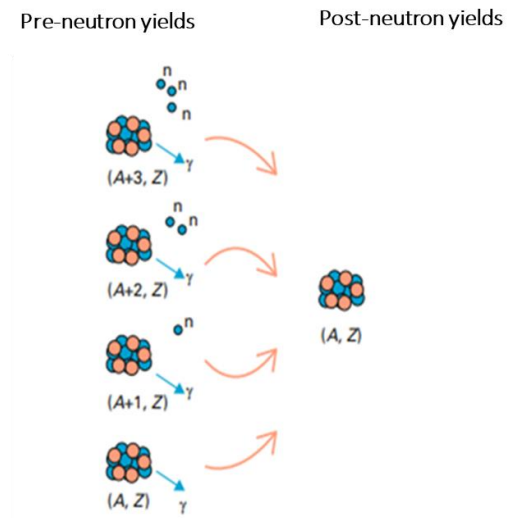

Fig. 1. Transition between pre-neutron data and post-neutron data [9].

For this work the mass yields $Y(A^*)$ are given by S. Zeynalov et al. [10], the $P(\nu|A^*)$ used is provided by the FIFRELIN code [11] and $P(Z|A^*)$ is determined by Z_p model. In the following, we will propose two approaches to provide the parameterization, but we limit our analysis to the calculation of $\Delta Z(A^*)$, $\sigma_Z(A^*)$. For $F_Z(A^*)$ and $F_N(A^*)$ we will use the Wahl's systematics for this first study. The objective of our analysis is therefore to adjust $\Delta Z(A^*)$, $\sigma_Z(A^*)$ to reproduce the post-neutron data (JEFF-3.1.1 or experimental data) as shown in **Fig. 2**.

FIFRELIN [11] is a Monte-Carlo code that simulates the de-excitation process of fission fragments and provides fission observables. In particular, we obtain the probability that a primary fission fragment of mass A^* emits ν prompt neutrons, prompt neutron spectrum, prompt gamma spectrum, neutron

multiplicity etc. To determine these observables FIFRELIN needs a number of input data, such as the mass yields Y(A*), kinetic energy and spin distributions of the fission fragments. In the FIFRELIN calculations, we use the mass yields and kinetic energy distributions measured by S. Zeynalov et al. [10] to be consistent in our analysis.

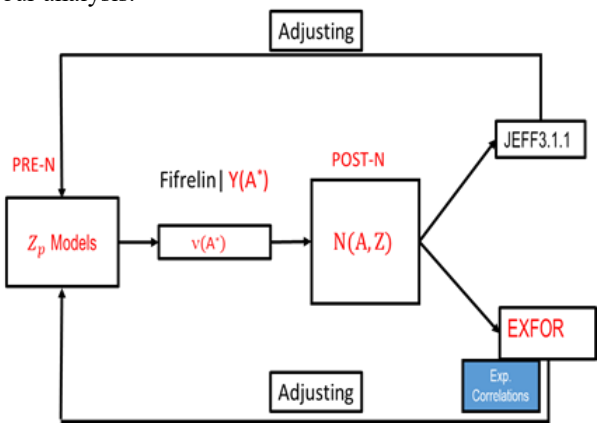

Fig. 2. The scheme for calculating and adjusting the Zp model parameters.

3.2 Adjustment of parameters

3.2.1 Linear parametrization (Wahl-like)

The principle of Wahl-like parameterization is to use the same assumptions as A.C. Wahl [4] i.e. that $\Delta Z(A^*)$ and $\sigma_Z(A^*)$ are linear in the pre-neutron mass region and then adjust the parameters of the lines to reproduce the post-neutron data. To determine $\Delta Z(A^*)$ for a mass region of [77-159] we need three parameters $\{\alpha_0, \alpha_1, \alpha_2\}$:

$$\begin{cases} \Delta Z(A^*) = \alpha_0 A^* + \alpha_1; \quad A^* \in [77-106] \\ \Delta Z(A^*) = \alpha_2(A^* - 106) + \Delta Z(106); \; [106-111] \end{cases} \tag{4}$$

By symmetry of the charge polarization, we determine the parameters in the complementary interval of [77-111] (i.e. [125-159]) and by continuity we determine the parameters in the interval of [111-125]. In a similar way we will use four parameters $\{\beta_0, \beta_1, \beta_2, \beta_3\}$ to determine $\sigma_Z(A^*)$:

$$\begin{cases} \sigma_Z(A^*) = \beta_0 A^* + \beta_1; \quad A^* \in [77,106] \\ \sigma_Z(A^*) = \beta_2; \; A^* \in [106,111] \\ \sigma_Z(A^*) = \beta_3; \; A^* \in [111,125] \end{cases} \tag{5}$$

By symmetry of $\sigma_Z(A^*)$, we calculate the parameters in the complementary interval of [77-111] (i.e. [125-159]).

To determine the best parameterization we will use an analysis based on a Classical Bayesian Monte-Carlo approach [12] :

$$P^{post}(\vec{\theta}|\vec{D}) = \frac{\mathcal{L}(\vec{D}|\vec{\theta})P(\vec{\theta})}{\int \mathcal{L}(\vec{D}|\vec{\theta})P(\vec{\theta})d\vec{\theta}} \tag{6}$$

$\vec{\theta} = \{\alpha_0, \alpha_1, \alpha_2, \beta_0, \beta_1, \beta_2, \beta_3\}$ is the parameter vector, $\vec{D}$ represents the data. The adjustment of the parameters is done with a classical Monte Carlo method by sampling the model parameters according to a prior distribution $P(\vec{\theta})$ (in our case a uniform distribution). The result is the posterior distribution of parameters given data used.

For the $\vec{D}$ experimental set associated with a given experimental covariance C, the likelihood function can be chosen as a multivariate Gaussian:

$$\mathcal{L}(\vec{D}|\vec{\theta}) = e^{\frac{-1}{2}(\vec{D}-\vec{t})^T C^{-1}(\vec{D}-\vec{t})}$$

Here, $\vec{t}$ represents the model predictions, which are functions of the model parameters $\vec{\theta}$.

Then we calculate the average values of $\vec{\theta}$ and the associate covariance matrix according to the following equations:

$$\langle\vec{\theta}\rangle = \sum_{i=1}^{n} \vec{\theta}_i * P^{post}(\vec{\theta}_i|\vec{D}) \tag{7}$$

$$C(\langle\vec{\theta}\rangle_k; \langle\vec{\theta}\rangle_l) = \sum_{i=1}^{n} [\vec{\theta}_i^k - \langle\vec{\theta}\rangle_k][\vec{\theta}_i^l - \langle\vec{\theta}\rangle_l] * P^{post}(\vec{\theta}_i|\vec{D}) \tag{8}$$

3.2.2 Direct-Z_p parametrization

The direct-Z_p approach consists in adjusting the parameters of the model without using any assumptions. This allows the suppression of the linear constraints on mass regions in the adjustment of free parameters. On the other hand, the complexity of implementing this approach is due to the very high number of parameters that must be adjusted. For example, in the mass region [77-159] the number of masses is 83. Each mass is represented by two parameters, which makes a total of (83*2=166) parameters. Thanks to the symmetry between the light and the heavy fragments, the mass interval is limited to [77-118] (42 masses) and the parameter number is reduced to 84. It is therefore difficult to use a classical Monte Carlo approach. To overcome this complexity, we used an adjustment based on Markov Chain Monte Carlo (MCMC) method. An important question about MCMC algorithms is how quickly they will converge to their stationary distribution (posterior distribution). In our case, using a Metropolis-algorithm [13], we were quickly confronted by the difficulty of convergence of the algorithm. The solution of this problem comes with the use of Weighted Likelihood Bootstrap [14]. The principle of this approach is relatively simple; it consists in sampling the experimental data. For each sample, we calculate with a Simulated Annealing algorithm [15] the vector of parameters guaranteeing a maximum likelihood between the sample and the data calculated from the model. This allows us to have the vector of parameters and the value of the corresponding likelihood function as shown in **Fig. 3**. At the end of this procedure, we calculate the posteriori distribution of parameters. This allows us to calculate the mean vector of the parameters and the associated covariance matrix based on equations (7) and (8).

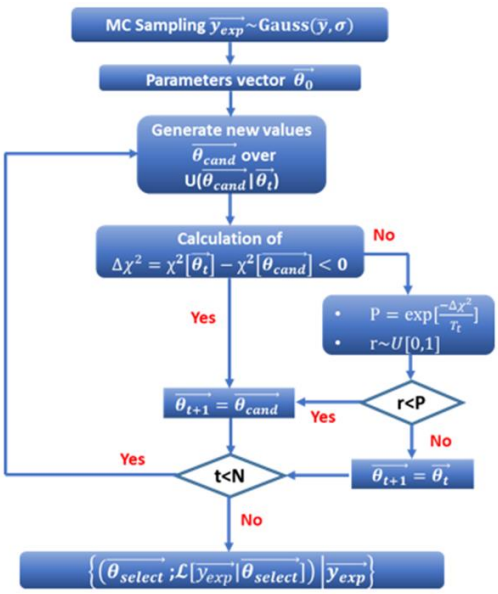

Fig. 3. Schematic diagram of the Bayesian Inference with the Bootstrap method and Simulated Annealing algorithm. The $\overrightarrow{y_{exp}}$ represents the sample of experimental data, $\overrightarrow{\theta_{cand}}$ represents the candidate parameter vector sampling according to a uniform distribution $U(\overrightarrow{\theta_{cand}}|\overrightarrow{\theta_t})$, $\overrightarrow{\theta_{select}}$ the selected parameter vector and N is the number of iterations.

4 Results

4.1 Adjustment based on JEFF-3.1.1

The adjustment of the parameters was carried out on the JEFF-3.1.1 library for ^{235}U(n$_{th}$, f). The JEFF-3.1.1 data were obtained from the evaluation process. They represent a synthesis of the experimental data completed by Wahl's model. However, we know that at the maximum only three or four charges Z per mass are experimentally determined. Then, we assume that the isotopes (A,Z) with probability P(Z|A) (defined in equation (3)) lower than an absolute value of 0.02 comes from the model. Therefore, we eliminate every isotope whose P(Z|A)<0.02. This allows us to avoid as much as possible using data from the Wahl's model in the JEFF-3.1.1 [6].

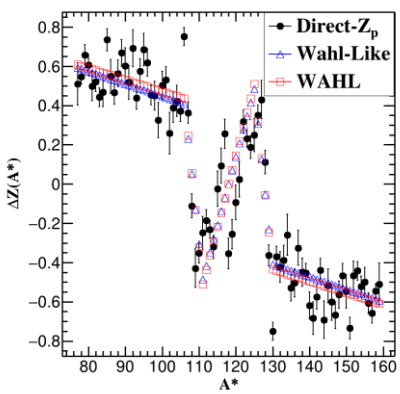

Fig. 4. Parameterization of the charge polarization based on Wahl's systematics (in red), Wahl-like approach with uncertainties (in blue) and direct-Z$_p$ approach (in black).

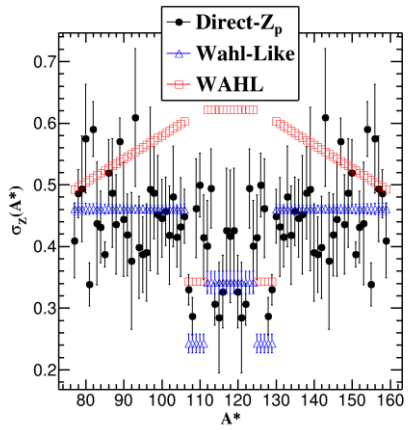

Fig. 5. Parameterization of σ$_Z$(A*) based on Wahl's systematics (in red), Wahl-like approach with uncertainties (in blue) and direct-Z$_p$ approach (in black).

Fig. 4 and **Fig. 5** are comparing the Zp model parameters, ΔZ(A*) and σ$_Z$(A*) as a function of fragment mass A* obtained from the Wahl-like and Direct-Zp methods compared with the original Wahl's parameterization. We note that the charge polarization is very similar to those proposed by A. C. Wahl as indicated (**Fig. 4**). Nevertheless, the σ$_Z$(A*) trends are very different between Wahl-like and Wahl (**Fig. 5**) due to the fact that we determine directly the pre-neutron parameters in our calculation. Indeed, considering the neutron emission per mass (so-called saw-tooth) the charge distribution per (post-neutron emission) mass A is adjusted by several pre-neutron charge distributions at different pre-neutron masses ($A_0^* = A + 0$, $A_1^* = A + 1$, $A_2^* = A + 2$, $A_3^* = A + 3$…). The difference on the σ$_Z$(A*) trend for both calculations comes from the difference with de Wahl's methodology which determine the Zp parameters on the post-neutron charge distribution and apply a correction to determine the pre-neutrons polarization and its width as a function of pre-neutron mass (A*).

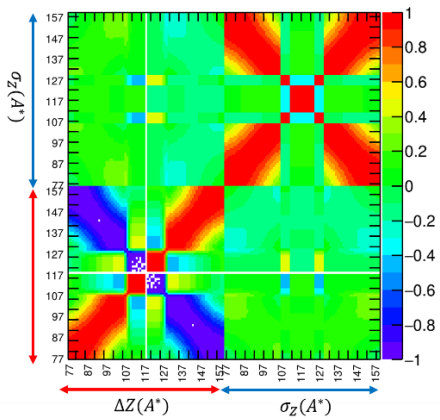

Fig. 6. The correlation matrix of the Wahl-like parameters.

Moreover, it is important to note that Wahl-like approach generates low uncertainties and extremely high correlations of the parameters. This is the result of the linearity assumption, which constrains the parameters and induces strong correlations (**Fig. 6**) and low uncertainties. The propagation of uncertainties and correlations of the parameters allows us to generate the

correlation matrix of the pre-neutron charge distribution based on the equation (1) and illustrated in **Fig. 7** (left).

The calculation of the correlation matrix of the charge distribution allows us to generate the correlation matrix of the post-neutron charge distribution based on the equations (2-3) and presented **Fig. 8** (left). Note that the step of self-normalization and propagation of uncertainties of conservation laws to generate the absolute post-neutron charge distribution and the associated covariance matrix is not applied here. Then, **Fig. 8** (left) illustrates only the correlation component from Zp model used. For more details, see [16] and [17].

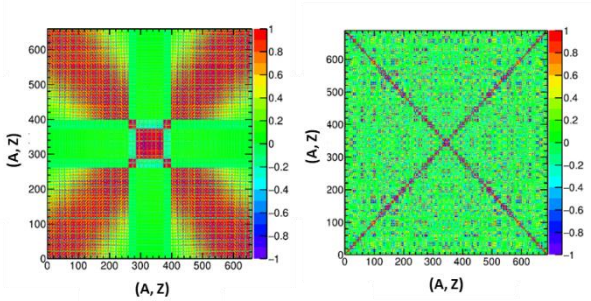

Fig. 7. The correlation matrices of the pre-neutron charge distribution based on Wahl-like approach (left) and direct-Z_p approach (right). The axes represent the isotope indexes.

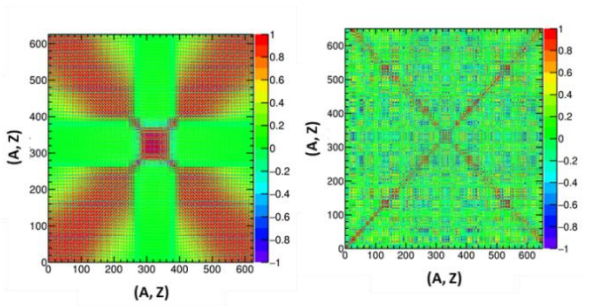

Fig. 8. The correlation matrices of post-neutron fission yields based on Wahl-like approach (left) and direct-Zp approach (right). The axes represent the isotope indexes.

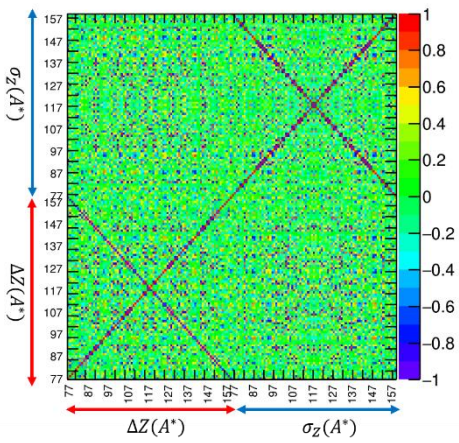

Fig. 9. The correlation matrix of direct-Zp parameters as a function of masses.

Similarly to the previous approach, we adjust the direct-Z_p parameterization to reproduce the major isotopes per mass of JEFF-3.1.1 data. The free adjustment reduces the correlation values between the parameters (**Fig. 9**) compared to Wahl-like parametrizations (**Fig. 6**), but it increases the uncertainties of these parameters as shown in **Fig. 4** and **Fig. 5**.

Based on the equation (1), we propagate the parameter uncertainties to generate the correlation of the pre-neutron charge distributions **Fig. 7** (right) and those of the post-neutron charge distribution **Fig. 8** (right) using the equation (2-3). The comparison of these distributions between calculations from Wahl's systematics and direct-Z_p approach shows that the Direct Z_p parameterization proposed better reproduces the data from JEFF-3.1.1 as illustrated in **Fig. 10**.

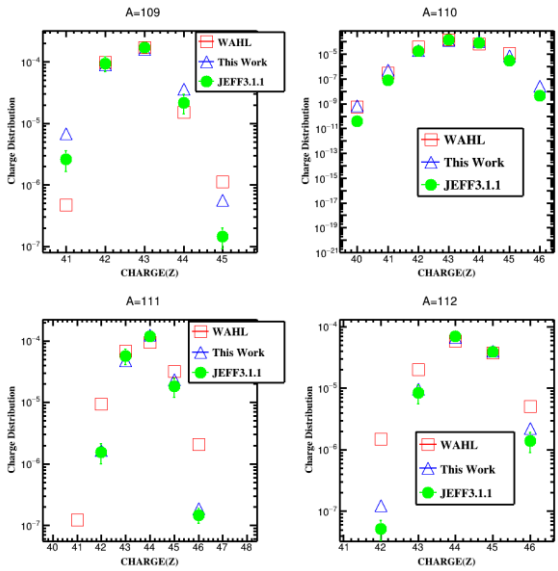

Fig. 10. The comparison of the charge distribution calculated using the Wahl's systematic (in red) and direct-Zp parameterization (in blue) with the JEFF-3.1.1 evaluation (in green) for the masses 109, 110, 111, 112.

A complementary comparison of the evaluations consists in the calculation of the empirical mean charges and the standard deviations of the post-neutron charge distributions according to the following equations:

$$\overline{Z(A)} = \sum_Z Z * \frac{N(A,Z)}{\sum_Z N(A,Z)}$$

$$\Delta Z(A) = \overline{Z(A)} - Z_{UCD}$$

$$\sigma_Z(A) = \sum_Z (Z - \overline{Z(A)})^2 * \frac{N(A,Z)}{\sum_Z N(A,Z)}$$

We compare the empirical mean values and standard deviations for the post-neutron charge distributions coming from the direct-Z_p approach. **Fig. 11** and **Fig. 12** show the evolution of the empirical mean values and the standard deviations as a function of the fission product mass obtained from the Direct-Z_p approach compared with JEFF-3.1.1. We observe a good agreement between both calculations.

Another check of consistency of the analysis is given by the generalized χ_g^2 statistical test. We obtain that the

direct-Z_p approach generates a consistent evaluation (P-value > 0.003, for 99.7% confidence level) of data unlike the Wahl's systematic and Wahl-like data as indicated in **Table 1**.

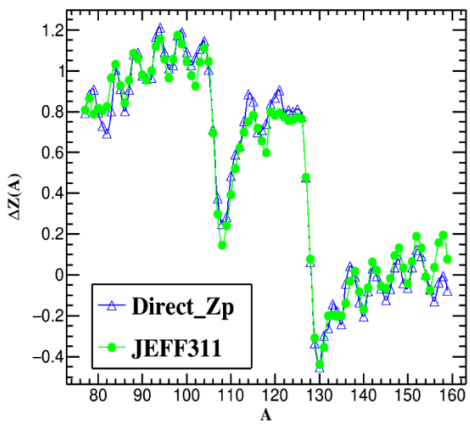

Fig. 11. Empirical calculation of post-neutron charge polarization from the JEFF-3.1.1 data (green) compared to the same calculation from charge distribution using the direct-Zp model (blue).

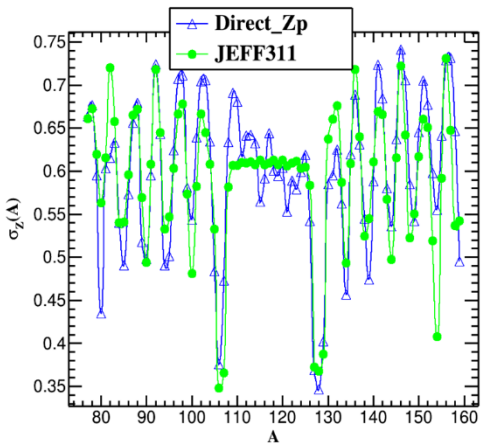

Fig. 12. Empirical standard deviation of post-neutron charge distributions calculated from the JEFF-3.1.1 library (green) compared to those from the direct-Zp approach (blue).

Table 1. Statistical test of charge distribution according to the different Zp parametrizations.

Approach	Wahl	Wahl-like	Direct Z_p
χ^2	1024	457.7	150.6
DOF	252	245	168
P-VALUE	10^{-93}	$9*10^{-15}$	0.83

4.2 Adjustment based on Lang's data

In this section, we will use the Direct-Z_p approach in order to compare the results of the adjustment of the parameters based on JEFF-3.1.1 [6] and on experimental data of W. Lang et al. [18]. Unlike the JEFF-3.1.1 data which are derived from the evaluation process. Lang's set are derived directly from the experimental process and have much smaller uncertainties, it covers a mass region between [80,110]. The **Fig. 13** and **Fig. 14** are comparing the results of adjusting the Z_p model parameters ($\Delta Z(A^*)$ and $\sigma_Z(A^*)$) using two different sources of data: JEFF-3.1.1 library and Lang's data. We observe many deviations, which show that a combination of all available experimental data is requested to assess the uncertainties on experimental knowledge. Moreover, the Lang's dataset remains limited and incomplete in the mass range of interest. In the near future, we plan to build a much larger experimental database containing an experiment data synthesis in order to use our approach on this database. This will allow us to improve the quality of the Z_p model parameters.

Nevertheless, we observe an important decrease of uncertainties of parameters with only one dataset. Thus, it will be interesting to introduce a similar analysis than mass yields (see [16] and [17]) of all available datasets to extract the experimental average dataset with realistic experimental standard deviations and correlation matrix.

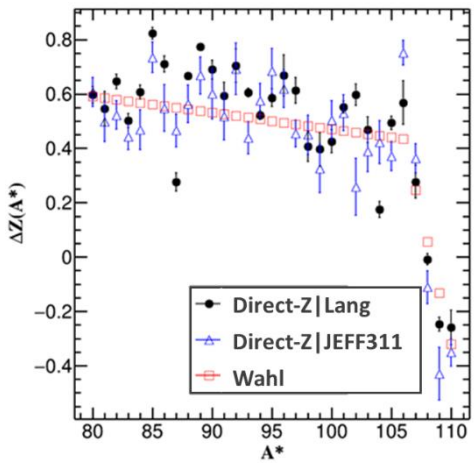

Fig. 13. The evolution of the pre-neutron emission charge polarization, in black the data determined via the adjustment based on Lang data, in blue the data determined via the adjustment based on JEFF-3.1.1 data and in red the data of Wahl's systematics parameterization.

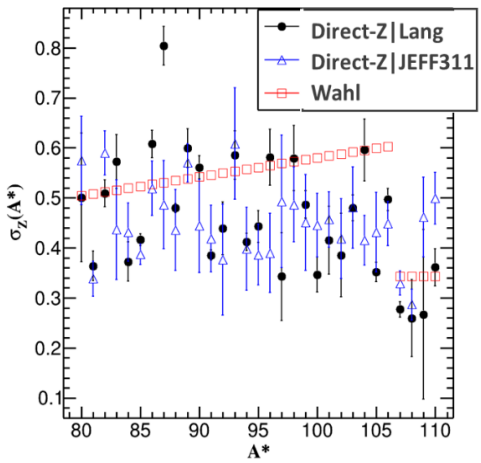

Fig. 14. The evolution of the pre-neutron emission $\sigma_Z(A^*)$, in black the data determined via the adjustment based on Lang results, in blue the data determined via the adjustment based on JEFF-3.1.1 library and in red the data of Wahl's systematics parameterization.

5 Conclusion

We have proposed two new approaches in order to determine Z_p model parameterization with their associated covariance matrix for $^{235}U(n_{th}, f)$ reaction. With these parameterizations, we propagated the parameter uncertainties to generate two covariance matrices of charge distribution associated to the JEFF library. These matrices will be used to fill the lack of charge distribution covariance information for the mixed analysis proposed for the fission yield JEFF-4 library [17]. For the direct-Z_p approach, an adjustment based on Lang's data was performed and the comparison with the adjustment based on JEFF-3.1.1 was made. This approach, starting from the pre-neutron distribution, allows the determination of Zp parameters the overall mass range using the charge conservation law. Thus, even if there is some lacks in the experimental data, the completeness of fission yield assessment is almost achievable for the $^{235}U(n_{th},f)$. With this method, we go back to the origin of the Z_p model but directly applied in the pre-neutron mass region. It will be interesting to apply this approach on the $^{252}Cf(sf)$ which is a standard reaction for fission yields.

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