A study of the construction of the correlation matrix of $^{241}$Pu($n_{th}$,f) isobaric fission yields
S. Julien-Laferriere, A. Chebboubi, G. Kessedjian, O. Serot

To cite this version:
S. Julien-Laferriere, A. Chebboubi, G. Kessedjian, O. Serot. A study of the construction of the correlation matrix of $^{241}$Pu($n_{th}$,f) isobaric fission yields. 4th International Workshop on Nuclear Data Covariances, Oct 2017, Aix-En-Provence, France. cea-02339327

HAL Id: cea-02339327
https://hal-cea.archives-ouvertes.fr/cea-02339327
Submitted on 14 Dec 2019
A study of the construction of the correlation matrix of $^{241}$Pu(n$_{th}$,f) isobaric fission yields

Sylvain Julien-Laferrière$^{1,2a}$, Abdelaziz Chebboubi$^{1b}$, Grégoire Kessedjian$^2$, and Olivier Serot$^1$

$^1$ CEA, DEN, DER, SPRC, F-13108 Saint-Paul-Lez-Durance, France
$^2$ LPSC, Université Grenoble-Alpes, CNRS/IN2P3, F-38026 Grenoble, France

Received: date / Revised version: date

Abstract. Two blind analyses for $^{241}$Pu(n$_{th}$,f) isobaric fission yields have been conducted, one analysis using a mix of a Monte-Carlo and an analytical method, the other one relying only on analytical calculations. The calculations have been derived from the same analysis path and experimental data, obtained on the LOHENGRIN mass spectrometer at the Institut Laue-Langevin. The comparison between the two analyses put into lights several biases and limits of each analysis and gives a comprehensive vision on the construction of the correlation matrix. It gives confidence in the analysis scheme used for the determination of the fission yields and their correlation matrix.

PACS. 25.85.Ec Neutron-induced fission – 29.85.Fj Data analysis – 07.75.+h Mass spectrometers

1 Introduction

Nuclear fission yields are key parameters for understanding the fission process [1] and to evaluate reactor physics quantities, such as decay heat [2]. Despite a sustained effort allocated to fission yields measurements and code development, the recent evaluated libraries (JEFF-3.3 [3], ENDF/B-VII.1 [4] ...) still present shortcomings, especially in the reduction of the uncertainties and the presence of reliable variance-covariance matrices. Yet these matrices are compulsory to use nuclear data, in particular when it comes to propagate uncertainties. A collaboration among the French Alternative Energies and Atomic Energy Commission (CEA), the Laboratory of Subatomic Physics and Cosmology (LPSC) and the Institut Laue-Langevin (ILL), started in 2009, aims at tackling these issues by providing precise measurements of fission yields with the related experimental covariance matrices for major actinides [5]. In particular for the $^{241}$Pu nuclide which is a major isotope in the context of multi-recycling and which is investigated in the present work.

Two measurements for thermal neutron induced fission of $^{231}$Pu have been carried out at the ILL in Grenoble (France), using the LOHENGIRN mass spectrometer [6,7] in May 2013 [8], labelled as Exp.1, and November 2015 [9], labelled as Exp.2. This instrument allows a selection of fission products regarding the $A/q$ and $E_k/q$ ratios, supplying an ion beam of of mass $A$, kinetic energy $E_k$ and of ionic charge $q$ with an excellent mass resolution ($\Delta A/A \simeq 1/400$). A double ionization chamber with a Frisch grid, similar to the one used in [10], is used to measure the count rate $N(A,t,E_k,q)$. The entire heavy peak region has been measured as well as a significant number of light masses.

The analysis scheme for the high yields region$^1$ (85 to 111 for the light masses and 130 to 151 for the heavy masses) has been subjected to two independent blind analyses [11], from the same raw data and analysis scheme.

The first one relies on a Monte-Carlo (MC) approach coupled with analytical calculations while the second one is based on a full analytical procedure. The main difference stands in the uncertainties propagation. In the analytical approach, for each step of the analysis, each parameter is supposed to have a Gaussian distribution. A classic approach is adopted, where functions are linearised by approximation to the first-order Taylor series expansion. On the contrary, in MC, no approximation is made when propagating uncertainties. The initial count rates are randomly generated from a Gaussian distribution with a large number of MC events. The mean value, standard deviation and covariances for each step are computed directly from the probability density functions.

The goal of this work is to unveil the biases and limits of each method and see if they lead to similar results despite the hypothesis made. If not, these biases shall be understood and controlled. This gives confidence in the analysis path adopted while understanding the important steps of the analysis in particular in the construction of the covariance matrix.

$^1$ For the low yields regions (symmetric and very asymmetric masses), the analysis procedure is different due to a contamination phenomenon that will not be presented here.
At first, the analysis procedure from the raw \( N(A, t, E_k, q) \) data to the determination of the isobaric yields \( Y(A) \) will be presented in Sec. 2. Then the two methods will be compared in particular the experimental correlation matrices, and the important steps of this analysis will be underlined, in Sec. 3.

2 Analysis path

The analysis path followed by both MC and analytical approaches is described in this section.

The raw data obtained from the experiments, at time \( t \), are the count rates \( N(A, t, E_k, q) \) for a mass \( A \), at a kinetic energy \( E_k \) and at the ionic charge state \( q \). These count rates are considered as independent from each other since only the statistical uncertainty is accounted for. The definition of the total count rate for the mass \( A \) is:

\[
N(A) = \int_{E_k} \sum_q \frac{N(A, t, E_k, q)}{BU(t)} dE_k \tag{1}
\]

Where the Burn-Up, \( BU \), is the relative estimation over time, \( t \), of the amount of fissile material in the \( ^{241} \text{Pu} \) target used for the experiments (see for example Fig. 1). The time evolution of the \( BU \) is carefully estimated by regularly measuring the overall ionic charge and kinetic energy distribution of mass 136, since it corresponds experimentally to an optimal count rate and mass separation. It is a mandatory normalization step since the amount of fissile material is constantly decreasing over time due to the nuclear reactions consuming the \( ^{241} \text{Pu} \) and the loss of material because of the harsh environmental conditions of the target [12]. The \( BU \) also takes into account the ion beam fluctuations over time as well as the incident neutron flux variations. Additional details on the procedure used for the \( BU \) measurement can be found in [13]. The \( BU \) points are fitted in order to be extrapolated to the measurement times. No theoretical consideration is taken for the choice of the fit function due to the complexity of physical phenomena governing the target behaviour. For the Exp.2 for example, the \( BU \) behaviour is well reproduced by the sum of a polynomial of 1\(^{st}\) order and an exponential function, as seen in Fig. 1:

\[
BU(t) = a \cdot t + b + \exp(c \cdot t + d) \tag{2}
\]

Since the beam time is limited, the complete \((E_i, q)\) distribution for each mass cannot be measured. The experimental method, refined over time [14,8,15], is to transform the sum over the ionic charge states into a division by the probability density function of the ionic charge states \( P(q) \), and the integral over the kinetic energies into a summation over bins of a constant step. Eq. 1 becomes:

\[
N_q(A) = \sum_{E_k} \frac{N(A, t, E_k, q)}{P(q) \cdot BU(t)} \tag{3}
\]

One scan of the ionic charge states distribution at a constant kinetic energy is made for each mass and at least three scans of the kinetic energy distribution at different ionic charge states. Thanks to this, the ionic charge distribution \( P(q) \) can be estimated and at least three different values of the same \( N(A) \) are obtained: \( N_{q_1}(A), N_{q_2}(A) \) and \( N_{q_3}(A) \), one for each measured kinetic energy distribution. To properly expressed \( N_{q_i}(A) \), one should take into account the existing correlation between the kinetic energy and ionic charge distributions [8,15]. To simplify the comparison proposed in this work, this step has been by-passed.

If the three estimations of the same \( N(A) \) are compatible, the mean value \( \bar{N}(A) \) taking into account the experimental covariance matrix, \( C \in M_n(\mathbb{R}) \), is extracted by minimizing the generalized \( \chi^2 \) [16]:

\[
\bar{N}(A) = \left( \sum_{i,j} (C^{-1})_{i,j} \right)^{-1} \left( \sum_{i,j} (C^{-1})_{i,j} \cdot N_{q_i}(A) \right) \tag{4}
\]

With a reduced variance:

\[
\text{Var}(\bar{N}(A)) = \left( \sum_{i,j} (C^{-1})_{i,j} \right)^{-1} \tag{5}
\]

Where \( n \) is the number of energy scans for the mass \( A \). How \( C \) is obtained will be detailed in the Subsec. 3.1. The compatibility is checked through a generalized \( \chi^2 \) test taking into account the experimental covariances between the \( N(A) \)'s, such as the p-value is above 90% of confidence level:

\[
\chi^2(N(A)) = \sum_i \sum_j \left( N_{q_i}(A) - \bar{N}(A) \right) C_{i,j}^{-1} \left( N_{q_i}(A) - \bar{N}(A) \right) \tag{6}
\]

When this criterion is not met, an additional independent uncertainty, \( \delta \), is incrementally added to the diagonal of the covariance matrix \( C \) of the \( N(A) \)'s, see Eq. 7, until a satisfying \( \chi^2 \) is reached. This additional independent
Table 1. Set of masses measured in the two experiments and masses common to the two experiments.

| Exp.1 | Exp.2 | Common masses |
|-------|-------|---------------|
| 130 to 151 | 130, 133 to 142 |
| 85, 90, 92, 93, 94, 96, 98, 100 to 109, 111, 130, 133 | 146, 147, 149 |
| to 142, 146, 147, 149 | |

uncertainty is accounting for the $N(A)$’s dispersion due to the flawed control of the instrument.

$$C_{ii}(k) = C_{ii}(0) + k \cdot \delta$$

(7)

Where $k$ is the number of increments. This process is illustrated in Fig. 2.

As already specified in the introduction, two sets of experiments have been used in this analysis, the first one measured in May 2013, Exp.1, and the second one in November 2015, Exp.2. Since the experimental environment is different from one experience to the other (neutron flux, target etc …) the two sets are considered as independent even though both are obtained on the LOHENGRIN mass spectrometer. The next step is thus to combine these two experiments to obtain the merged vector $\overline{N}(A)_{\text{mgd}}$ (mgd stands for merged), by doing a relative normalization of one set to the other, relying on the value of the $\overline{N}(A)$ obtained for the 14 common masses, see Tab. 1.

If $X$ and $Y$ are respectively the $\overline{N}(A)$ vectors for the common masses of Exp.1 and Exp.2, then, to normalize Exp.1 with respect to Exp.2, one has to minimize the residual vector $\varepsilon$:

$$\varepsilon = Y - \kappa \cdot X$$

(8)

The best estimator of $\kappa$ is the normalization factor $k$ of Exp.1 with respect to Exp.2, obtained through the generalized least square method, in its matrix form [17]:

$$k = (X^T \Omega^{-1} X)^{-1} X^T \Omega^{-1} Y$$

(9)

Where $\Omega$ is the combined covariance matrix for Exp.1 and Exp.2, taken to be $\Omega = \Omega_X + \Omega_Y$. Eventually, the mean values of the normalized set of the two experiments are extracted for the common masses in a similar way to Eq. 4.

The last step is the absolute normalization to obtain the yields, $Y(A)$. Ideally, this normalization is done by taking $\sum_{i \in H} N(A_i)_{\text{mgd}} = 1$, $H$ being the masses of the heavy peak. This is true for pre-neutron yields when the contribution of ternary fission is not taken into account. In our experiments, post-neutron yields from the masses 121 to 159 are obtained. The sum of the mass yields for $A > 159$ represents 0.26 % in JEFF-3.3 of the heavy peak and 0.03 % for ENDF/B-VII.1. For the normalization step, a conservative uncertainty of 0.5% is taken, accounting for the absence of the very heavy masses, $A > 159$ and the approximation that the sum over the heavy masses is equal to 1 for post-neutron yields.

However, since this document is only focused on high yields, the completeness of the $Y(A)$ distribution for the heavy masses is not satisfyingly achieved. Nonetheless, in the scope of this document, the normalization will be done as previously explained, in order to discuss the impact of the absolute normalization.

### Summary

The analysis path can be summarized in the following steps:

1. Sum over the kinetic energy distribution:

$$N(A,t,q_i) = \sum_{E_k} N(A,t,E_k,q_i)$$

2. Determine the $P(q_i)$ distribution and divide step (1) by $P(q_i)$:

$$N_{q_i}(A,t) = \sum_{E_k} \frac{N(A,t,E_k,q_i)}{P(q_i)}$$

3. Evaluate the $BU$ at $t$ and divide step (2) by $BU(t)$:

$$N_{q_i}(A) = \sum_{E_k} \frac{N(A,t,E_k,q_i)}{P(q_i) \cdot BU(t)}$$

4. Compute the mean value for each mass with eventual additional uncertainties: $\overline{N}(A)$

5. Normalize (relative normalization) and merge the two sets of experiments: $\overline{N}(A)_{\text{mgd}}$

6. Process the absolute normalization: $Y(A)$

### 3 Covariance matrices comparison

#### 3.1 Differences between Monte-Carlo and analytical methods

In the MC method, all count rates are sampled from a Poisson law. In order to assess $BU(t)$, the fitted $BU$ parameters are first decorrelated and then sampled from a
Gaussian law with a unit standard deviation. The decorrelation is obtained through the Eq. 10 [18]:

$$S = \Omega_R^{-1/2} R$$

(10)

With $\mathbf{R}$ the fitted parameters, $S$ the free parameters and $\Omega_R$ the covariance matrix of the fitted parameters. However, when the BU is computed from the sampling of the uncorrelated fit parameters, the probability density function obtained is no more a Gaussian distribution, see Fig. 3. In the analytic method, when propagating uncertainties each parameter is supposed to have a Gaussian distribution, if it is not the case the biases of this hypothesis should be controlled. This issue is under investigation. In order to still be able to compare the two analyses for the next steps, in the present work, the propagation of the BU uncertainties by the MC method is achieved analytically.

The covariance between $N(A)$’s, used in Eq. 4, can be decomposed between the covariances induced by the BU and the statistical covariances:

$$C = \text{Cov}(N_q(A_i), N_q(A_j)) = \text{Cov}_{\text{stat}} + \text{Cov}_{\text{BU}}$$

(11)

Only covariance at the step (2) is computed through MC as expressed by Eq. 12, with $m$ the MC event and $N_q(A_i,t)$ the arithmetic mean value of the $N_q^{m}(A_i,t)$’s. $\text{Cov}_{\text{stat}}$, the statistical part of the covariance matrix is then defined as follows:

$$\text{Cov}_{\text{stat}}(N_q(A_i,t), N_q(A_j,t')) = \sum_m (N_q^{m}(A_i,t) - N_q(A_i,t)) (N_q^{m}(A_j,t') - N_q(A_j,t'))$$

(12)

Where $t$ and $t'$ are respectively the times at which the mass $A_i$ at the ionic charge state $q_i$ and the mass $A_j$ at the ionic charge state $q_j$ have been measured. In the analytic case, Eq. 12 is written as Eq. 13:

$$\text{Cov}_{\text{stat}}(N_q(A_i,t), N_q(A_j,t')) = \frac{N_q(A_i,t) N_q(A_j,t')}{P_{q_{i}}(A_{i}) P_{q_{j}}(A_{j})} \times \text{Cov}(P_{q_{i}}(A_{i}), P_{q_{j}}(A_{j}))$$

(13)

Where $\text{Cov}(P_{q_{i}}(A_{i}), P_{q_{j}}(A_{j}))$ is the covariance coming from the ionic charge distribution. Therefore if $A_i \neq A_j$, this term is null. Otherwise by propagating in a classical way the covariance, it is written:

$$\text{Cov}(P_{q_{i}}(A_{i}), P_{q_{j}}(A_{j})) = \frac{P_{k_{i}}P_{k_{j}}}{I_{k_{i}}I_{k_{j}}}$$

$$\begin{align*}
& (I_{tot} - I_{k_{i}})(I_{tot} - I_{k_{j}}) \text{Cov}(I_{k_{i}}, I_{k_{j}}) \\
& - (I_{tot} - I_{k_{i}}) I_{k_{j}} \sum_{m \neq k} \text{Cov}(I_{k_{i}}, I_{m}) \\
& - (I_{tot} - I_{k_{j}}) I_{k_{i}} \sum_{n \neq k} \text{Cov}(I_{n}, I_{k_{j}}) \\
& + I_{k_{i}} I_{k_{j}} \sum_{m \neq n} \text{Cov}(I_{m}, I_{n})
\end{align*}$$

(14)

With $I_k$ the count rates of the charge $k$, $I_{tot} = \sum_k I_{k}$ the total count rate of the ionic charge distribution, and $P_{k} = \int_{t_{tot}}^{t_{tot}}$ the normalized count rate. One has to note, since each measurement is independent, that only the diagonal of the covariance matrix of the count rate is not null.

The BU correlation is taken into account analytically in both analyses through:

$$\text{Cov}_{\text{BU}}(N_q(A_i), N_q(A_j)) = \frac{N_q(A_i)N_q(A_j)}{BU_{ik}BU_{jl}} \text{Cov}(BU_{ik}, BU_{jl})$$

(15)

With $\{r_{k}\}$ the BU parameters. As a consequence, for the analysis using MC, the following steps, (4) to (6), have to be computed analytically.

A final difference can be underlined. For independent parameters, the MC induces small correlations, even for pseudo-random numbers generated by different seeds. Since these correlations are low compared to the real experimental correlations, it is not an issue. It is worth noting that this MC numerical correlation artefact is dependent on the number of MC events. In this work, for $10^5$ events, the order of magnitude of this correlation artefact is $10^{-5}$.

3.2 Step (3): Impact of the BU on correlations

The BU is at this stage the only source of inter-masses correlations. To illustrate this, a mean experimental time $\overline{t}(A)$ has been constructed for each mass. $\overline{t}(A)$ is the mean time at which the mass $A$ is measured.

In Fig. 4, one can see that masses having close experimental time have high correlation and vice versa. For example, the masses 100 to 109 (black circle) have been measured very closely in time, the same goes for the masses 138 to 141 (green circle). The correlations in these groups
are very high, whereas inter groups correlations are close to 0.

The correlations obtained at this stage for the mix MC and analytic and the analytic methods are very similar, the highest absolute difference observed in the correlation matrix is $\Delta \text{Corr} = 0.0032$ which represents a relative difference of $\frac{\Delta \text{Corr}}{\text{Corr}} = 1.3\%$, see Fig. 5.

### 3.3 Step (4): Impact of the additional dispersion uncertainties

As it has just been explained, the correlation matrix is at this stage governed by the $BU$. The additional dispersion uncertainties introduced after Eq. 6, as independent uncertainties, wash away the initial structures. The weight of the common uncertainties coming from the $BU$ is reduced.

The impact of these additional uncertainties are shown in Fig. 6 and Fig. 7.

### 3.4 Step (5): Impact of the relative normalization

The definition of the relative normalization factor $k$, in Eq. 9, is dependent on which experiment is the reference. Both the out-coming $\overline{N}(A)_{\text{mgd}}$ and the associated correlation matrix will be affected, as it can be observed from Fig. 8 and 9. Indeed, if Exp.1 is normalized with respect to Exp.2, the variance of $\mathbf{Z} = k \mathbf{X}$, the normalized vector
The relative difference on $\mathcal{N}(A)_{\text{mgd}}$ in black on the left axis, and on the uncertainty $\sigma_{\text{mgd}}$ in blue on the right axis, when the reference is Exp.1 or Exp.2. For the sake of the comparison, both sets of $\mathcal{N}(A)_{\text{mgd}}$ have been normalized to 1.

Fig. 8. The relative difference on $\mathcal{N}(A)_{\text{mgd}}$ in black on the left axis, and on the uncertainty $\sigma_{\text{mgd}}$ in blue on the right axis, when the reference is Exp.1 or Exp.2. For the sake of the comparison, both sets of $\mathcal{N}(A)_{\text{mgd}}$ have been normalized to 1.

Fig. 9. The correlation matrix after the relative normalization when Exp.1 is the reference, on the left, and when Exp.2 is the reference, on the right. X and Y labels are identical to Fig. 12 (top).}

3.5 Step (6): Impact of the absolute normalization

The impact on the $Y(A)$ correlation matrix of the absolute normalization is illustrated in Fig 11. In Fig. 11, a diagonal correlation matrix is displayed (top left), while the analytic $^{241}\text{Pu}(n_{th},f)$ experimental $\mathcal{N}(A)_{\text{mgd}}$ correlation matrix is shown (bottom left). On the right, the correlation matrices after the absolute normalization depending on which is the input correlation matrix. Thus, the effect of the absolute normalization step alone is shown on the top right of Fig. 11. The $Y(A)$ correlation matrix (bottom right), is the combination of the effect of the absolute normalization alone and the experimental $\mathcal{N}(A)_{\text{mgd}}$ correlation matrix (bottom left).

The $Y(A)$ correlation matrix structure is marked by the normalization procedure that creates a correlation background.

4 Conclusion and Perspectives

The experimental $Y(A)$ obtained in this work are presented in Fig. 12 and compared to JEFF-3.3. The precision achieved in this experimental work is much better than the JEFF-3.3 evaluation and significant discrepancies are observed, in particular in the light masses.

An effect not taken into account in this work is the correlation between the kinetic energy and ionic charge distributions. Since the ionic charge scan is made at a specific kinetic energy, $E_k$, this correlation has to be taken into account in order to properly estimate $P(q_i)$ in Eq. 3 [20]. This is expected to significantly reduce the need of the additional dispersion uncertainties from Subsec. 3.3.

This work showed that both analyses give identical results for the estimation of the correlations matrix when the MC bias highlighted during the $BU$ sampling step is by-passed. The $BU$ sampling bias is under investigation in order to compare the MC and analytic methods on the full analysis scheme and not only on steps (1) to (3). For every steps of the analysis scheme, the correlation matrices obtained by the two different analyses have been compared and no larger differences than the one observed at step (3)
are seen, giving a strong confidence in our method. Mean values and uncertainties are also identical, validating both analyses. The structure of the correlation matrix is well understood, the importance of several steps in the construction of the correlation matrix have been emphasized: the BU creates strong positive correlations while the additional uncertainties due to the limits of the experimental method flatten the correlation matrix. Finally the absolute normalization creates a correlation background.

The analyses have been compared without the correlation between the kinetic energy and ionic charge distributions. In order to finalize this work, a supplementary step where it is taken into account will be included to the analysis scheme. In addition, a re-analysis of the Exp.1 with updated tool and the construction of a reliable observable to rank the relative normalization possibilities are on going.

This work was supported by CEA, IN2P3 and "le défi NEEDS". The authors are grateful for the support of the ILL and all the staff involved from CEA Cadarache and LPSC.

References

1. H.G. Börner and F. Gönnenwein, in The Neutron (World Scientific Publishing, Singapore, 2012)
2. N. Terranova et al., Ann. of Nucl. Energy 109, 469-489 (2017)
3. A. Plompen et al., Eur. Phys. J. A, (to be submitted)
4. M.B. Chadwick et al., Nucl. Data Sheets 112, 2887-2996 (2011)
5. O. Serot et al., Nucl. Data Sheets 119, 320-323 (2014)
6. P. Arbruster et al., Nucl. Instrum. Methods 139, 213-222 (1976)
7. H.R. Faust et al., ILL Internal Scientific Report 81FA45S (1981)
8. F. Martin, Ph.D. thesis, Université de Grenoble, 2013
9. S. Julien-Laferriere et al., Eur. Phys. J. Web of Conf. 169, 00008 (2018)
10. H.R. Faust, Z. Bao, Nucl. Phys. A 736, 55-76 (2004)
11. J. G. Heinrich, Report 6576, University of Pennsylvania (2003)
12. U. Köster et al., Nucl. Instrum. Methods A 613 (3), 363-370 (2010)
13. Y.K. Gupta et al., Phys. Rev. C 96 (2017)
14. A. Bail, Ph.D. thesis, Université Bordeaux I, 2009
15. A. Chebboubi, Ph.D. thesis, Université de Grenoble-Alpes, 2015
16. M. Schmelling, Phys. Scripta 51, 676-679 (1995)
17. A. C. Aitken, P. Roy. Soc. Edin. A 55, 42-48 (1935)
18. A. Kessey, A. Lewin and K. Strimmer, Am. Stat. (2018), DOI: 10.1080/00031305.2016.1277159
19. B. Voirin et al., Eur. Phys. J. N 4, (2018)
20. F. Martin et al., Nucl. Data Sheets 119, 328-330 (2014)