235U capture cross-section adjustment in criticality benchmarks using ENDF/B-VII.1 evaluation

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ABSTRACT
This present study aims to evaluate the nuclear data uncertainty on the neutron multiplication factor ($k_{\text{eff}}$) in order to adjust the 235U capture multigroup cross sections and the covariance matrix of the ENDF/B-VII.1 evaluation. For this purpose, the generalized linear-least squares method is applied. The 44 multigroup cross sections and the covariance matrix are processed with NJOY99 nuclear software, while MCNP6.1 nuclear code is used for the $k_{\text{eff}}$ and sensitivity profile calculations. The obtained results show that the 235U capture cross sections require an adjustment of approximately −1% in 0–6 eV, −0.5% in 6 eV to 3 keV, 2% in 3 keV to 0.4 MeV, and −10% in 0.4–20 MeV.

1. Introduction
During these last years, the deterministic and Monte Carlo methods used in neutronic calculations have been improved. The main parameters of these calculations are the neutron cross sections, which are based on theories and nuclear experiments. Nevertheless, the values of these cross sections always still contain uncertainties and a necessary improvement. In order to reduce these uncertainties, several methods have been adopted: the generalized least squares method (Dupont, Ishikawa, Palmiotti, Salvatores, & de Saint-Jean, 2010), etc. The 235U capture cross section has a large difference from 30 keV to 1 MeV in the latest nuclear data libraries (Chadwick & Trkov, 2016; Iwamoto & McKnight, 2010; Mingrone, Vannini, Calviani, Ferrari, & Wallner, 2017), which leads to differences in the prediction of nuclear calculations for certain neutron parameters in nuclear reactors, such as $k_{\text{eff}}$. According to Iwamoto, this cross section requires a relative change of about 15% in 1 keV to 10 MeV energy range. In addition, other authors studied this subject (Leal, Derrien, Larson, & Wright, 1999; Leal, Mueller, Arbanas, Wiarda, & Derrien, 2008) and concluded that the relative standard deviation of $k_{\text{eff}}$ due to this cross section, is approximately 200 pcm for the HST001.001 benchmark and ENDF/B-VII.0.

Thus, it is desirable to adjust this cross section given that it’s possible to calculate its sensitivity and covariance matrix (Dupont et al., 2010).

In the first part of this paper, theories of sensitivity, uncertainty, and adjustment are presented. In the second part, the prior and posterior uncertainties on the effective multiplication factor, the adjusted capture cross section of 235U nucleus, and its covariance matrix in ENDF/B-VII.1 have been evaluated. For these, many benchmarks from IHECSBE (Briggs, 2004) have been taken with different geometries (spherical, cylindrical, etc.) and diverse fuel forms (mixed metallic, solution, solid). Also, a large neutron spectrum (thermal, epithermal, and fast) which covers all neutron energy groups was used. In order to obtain the adjusted cross section and its covariance matrix, the generalized linear-least squares method (GLLSM) (Dragt & Dekker, 1977; Lewins & Becker, 1982) was used, which was based on chi-square (chi²) distribution. To apply the GLLS method, sensitivity and multigroup covariance matrices have been calculated and prepared using MCNP6.1 (Pelowitz, 2013) and NJOY99 codes (MacFarlane & Muir, 1994), respectively. Also, 44 neutron energy groups was used (Bowman, 2000; Zhu, 2015).

2. Methodology and study approach
2.1. Sensitivity and uncertainty theory
The sensitivity coefficients are the ratio of the resulting relative change in multiplication factor to the relative change in a system parameter $x$ (cross section, fission nubar, etc.) over some small energy range $E + dE$. Assuming that the change in parameter $x$ is small enough such that $k_{\text{eff}}$ changes linearly with respect the parameter, the following relationship holds (Kiedrowski, 2013)

$$S_{k_{\text{eff}}, x} = \frac{x}{k_{\text{eff}}} \cdot \frac{\partial k_{\text{eff}}}{\partial x}$$

(1)

The use of the first order in the perturbation theory gives the expression of the sensitivity coefficient of
$k_{\text{eff}}$ according to the following equation (Kiedrowski, 2013; Williams & Weisbin, 1978):

$$S_{k_{\text{eff}},x} = \frac{\langle \Psi^+, (\sum_x - S_x - k_{\text{eff}}^{-1} F_x) \Psi \rangle}{\langle \Psi^+, F \Psi \rangle}$$ (2)

where the brackets denote integration over all space (direction and energies), the term $\Psi$ is the neutron angular flux, and $\Psi^+$ its adjoint. $S_x$ is the macroscopic cross section of interest (zero if not a cross section), $S_x$ is the scattering integral for nuclear data $x$ (zero if not scattering), and $F_x$ is the fission integral for nuclear data $x$ (zero if not fission). At last, $F$ designates the integral fission operator for the entire system.

For each nuclear parameter $x$ and $y$, the sensitivity coefficients ($S_{k_{\text{eff}},x}$ and $S_{k_{\text{eff}},y}$) are evaluated by the Iterated Fission Probability method which is included in Monte Carlo N-Particle transport code MCNP6.1, whereas the covariance matrix (cov($x, y$)) between these two nuclear parameters $x$ and $y$ (cross sections, fission nubar, etc.) in the 44 energy groups is calculated via the NJOY99 software. Thus, the relative uncertainty on the multiplication factor due to the uncertainties of nuclear data can be assessed as follows (Kiedrowski, Wilson, & Brown, 2011; Salvatore et al., 2014):

$$\frac{\Delta k_{\text{eff}}}{k_{\text{eff}}} \leq \sqrt{S_{k_{\text{eff}},x} \cdot \text{cov}(x,y) \cdot S_{k_{\text{eff}},y}}$$ (3)
In the following this uncertainty is denoted by $\Delta k_{\text{eff}}$–nucl.

### 2.2. Adjustment theory

Quantifying the uncertainty of the $^{235}$U capture cross section in the preceding paragraph brings us to the second step of calculating where the least squares theory is applied to reduce these uncertainties. Figure 1 shows the main steps used in this study.

The following quantities are used in this work:

- $K_{E,i} (i = 1, N_x)$: Experimental value of $k_{\text{eff}}$ in benchmark $i$.
- $K_{C,i} (i = 1, N_x)$: ‘a priori’ calculated value of $k_{\text{eff}}$ in benchmark $i$.
- $K'_{E,i} (i = 1, N_x)$: ‘a posteriori’ calculated value of $k_{\text{eff}}$ in benchmark $i$.
- $\sigma_{ij}$: ‘a priori’ cross sections; $\sigma'_{ij}$: ‘a posteriori’ cross sections; $S_{ji}$: sensitivity

#### Table 1. Effective multiplication factor $k_{\text{eff}}$ of selected benchmarks and their statistical uncertainties (1o).

| Benchmark cases | $k_{\text{eff}}$ (ENDF/B.VII.1) | $k_{\text{eff}}$ (HEC580) |
|-----------------|---------------------------------|---------------------------|
| Hist01.002      | 0.99722 ± 0.00006               | 1.0021 ± 0.0072           |
| Hist01.004      | 0.99815 ± 0.00005               | 1.0008 ± 0.0053           |
| Hist01.007      | 0.99781 ± 0.00005               | 1.0008 ± 0.004            |
| Hist001.008     | 0.99797 ± 0.00005               | 0.9998 ± 0.0038           |
| Hist001.009     | 0.99412 ± 0.00006               | 1.0008 ± 0.0054           |
| Hist001.010     | 0.99241 ± 0.00005               | 0.9993 ± 0.0054           |
| Hist009.001     | 0.99695 ± 0.00005               | 0.999 ± 0.0043            |
| Hist009.001R    | 0.99745 ± 0.00005               | 1.000 ± 0.0057            |
| Hist009.002     | 0.99686 ± 0.00005               | 1.000 ± 0.0039            |
| Hist009.003     | 0.99556 ± 0.00005               | 1.000 ± 0.0036            |
| HST011.001     | 0.99859 ± 0.00004               | 1.0000 ± 0.0023           |
| HST011.002     | 0.99666 ± 0.00004               | 1.0000 ± 0.0023           |
| HST012.001     | 0.99723 ± 0.00003               | 0.9999 ± 0.0058           |
| HST013.001     | 0.99868 ± 0.00003               | 1.0012 ± 0.0026           |
| HST035.007     | 1.00467 ± 0.00005               | 1.000 ± 0.0035            |
| HMF003.001     | 0.99501 ± 0.00003               | 1.000 ± 0.005             |
| HMF003.002     | 0.99436 ± 0.00003               | 1.000 ± 0.005             |
| HMF003.004     | 0.99721 ± 0.00003               | 1.000 ± 0.005             |
| HMF003.005     | 1.00146 ± 0.00003               | 1.000 ± 0.003             |
| HMF003.008     | 1.00214 ± 0.00003               | 1.000 ± 0.003             |
| HMF003.009     | 1.00244 ± 0.00003               | 1.000 ± 0.005             |
| HMF003.010     | 1.00505 ± 0.00003               | 1.000 ± 0.005             |
| HMF014        | 0.99774 ± 0.00003               | 0.9989 ± 0.0017           |
| HMF021.002     | 0.9975 ± 0.00003                | 1.000 ± 0.0024            |
| HMF022.002     | 0.99746 ± 0.00003               | 1.000 ± 0.0021            |
| HMF026.011     | 1.00312 ± 0.00004               | 0.9982 ± 0.0042           |
| HMF028        | 1.00286 ± 0.00003               | 1.000 ± 0.0030            |

#### Table 2. Energy groups used in sensitivity and uncertainty analysis (Bowman, 2000; Zhu, 2015).

| Group number | Energy range (eV) | Group number | Energy range (eV) |
|--------------|-------------------|--------------|-------------------|
| 1            | 1.0000E-05–3.0000E-03 | 23           | 3.0000E-03–4.7500E+00 |
| 2            | 3.0000E-03–7.5000E-03 | 24           | 4.7500E-00–6.0000E+00 |
| 3            | 7.5000E-03–1.0000E-02 | 25           | 6.0000E-00–8.1000E+00 |
| 4            | 1.0000E-02–2.5300E-02 | 26           | 8.1000E-00–1.0000E+01 |
| 5            | 2.5300E-02–3.0000E-02 | 27           | 1.0000E-01–3.0000E+01 |
| 6            | 3.0000E-02–4.0000E-02 | 28           | 3.0000E-01–1.0000E+02 |
| 7            | 4.0000E-02–5.0000E-02 | 29           | 1.0000E-02–3.5000E+02 |
| 8            | 5.0000E-02–7.0000E-02 | 30           | 5.5000E-02–3.0000E+03 |
| 9            | 7.0000E-02–1.0000E-01 | 31           | 3.0000E-03–1.7000E+04 |
| 10           | 1.0000E-01–1.5000E-01 | 32           | 1.7000E-04–2.5000E+04 |
| 11           | 1.5000E-01–2.0000E-01 | 33           | 2.5000E-04–1.0000E+05 |
| 12           | 2.0000E-01–2.2500E-01 | 34           | 1.0000E-05–4.0000E+05 |
| 13           | 2.2500E-01–2.5000E-01 | 35           | 4.0000E-05–9.0000E+05 |
| 14           | 2.5000E-01–2.7500E-01 | 36           | 9.0000E-05–1.4000E+06 |
| 15           | 2.7500E-01–3.2500E-01 | 37           | 1.4000E-06–1.8500E+06 |
| 16           | 3.2500E-01–3.5000E-01 | 38           | 1.8500E-06–2.3540E+06 |
| 17           | 3.5000E-01–3.7500E-01 | 39           | 2.3540E-06–2.4790E+06 |
| 18           | 3.7500E-01–4.0000E-01 | 40           | 2.4790E-06–3.0000E+06 |
| 19           | 4.0000E-01–6.2500E-01 | 41           | 3.0000E-06–4.8000E+06 |
| 20           | 6.2500E-01–1.0000E+00 | 42           | 4.8000E-06–6.4340E+06 |
| 21           | 1.0000E+00–1.7700E+00 | 43           | 6.4340E-06–8.1873E+06 |
| 22           | 1.7700E+00–3.0000E+00 | 44           | 8.1873E+06–2.0000E+07 |
coefficients for $k_{\text{eff}}$ in benchmark $i$ with respect to cross section $j$; $C_{EC} = (C_E + C_C)$: $k_{\text{eff}}$ covariance matrix; $C_E$: $k_{\text{eff}}$ covariance matrix due to measurement; $C_C$: $k_{\text{eff}}$ covariance matrix due to calculation; $C_{\sigma}$: 'a priori' cross-section covariance matrix; $C_{\sigma}': 'a posteriori' cross-section covariance matrix; $\chi^2$: 'a priori' chi-square; $I$: unity matrix; $C_d = (C_{EC} + S C_{\sigma} S^T)$: total covariance matrix.

With these parameters, we can define the $\chi^2$ before adjustment (David & Jackson, 1988; Gibbs, 2011; Palmiotti, Salvatores, & Aliberti, 2015; Williams & Weisbin, 1978):

$$
\chi^2 = (\sigma' - \sigma)^T C_{\sigma}^{-1} (\sigma' - \sigma) + (K_E - K_C)^T C_{EC}^{-1} (K_E - K_C)
$$  (5)

The minimization of $\chi^2$ with respect to cross section gives the following two formulas:

$$
\sigma' - \sigma = C_{\sigma} S^T C_d^{-1} (K_E - K_C)
$$  (6)

and

$$
C_{\sigma}' = C_{\sigma} - C_{\sigma} S^T C_d^{-1} S C_{\sigma} = C_{\sigma} (I - S^T C_d^{-1} S C_{\sigma})
$$  (7)

where the above equations, respectively, give the adjusted cross section $\sigma'$ and the associated covariance matrix $C_{\sigma}'$.

In order to select the appropriate benchmarks to adjust the nuclear data and their covariance matrices, the reduced $\chi^2_N$ (individual $\chi^2$ divided by the total number of benchmarks) is calculated. This parameter must be included in the confidence interval of $\chi^2$ distribution for each selected benchmark, as shown below (Hirotsu, 2017; Lewins & Becker,
The posterior cross section and its covariance matrix found previously allow to calculate the posterior parameters (\( k_{\text{eff}} \) and nuclear data uncertainty) using Equations (9) and (10), respectively.

\[
K_0 = K_C \left[ 1 + S^T \frac{(\sigma' - \sigma)}{\sigma} \right]
\]

where \( \text{cov}'(x,y) \) denotes the adjusted covariance matrix.

### 2.3. Study approach

In this study, to perform the adjustment of the \( ^{235}\text{U} \) capture cross section, we selected several benchmarks: 12 fast, 15 thermal, and in Table 1, we presented these benchmarks, their standard deviations, and \( k_{\text{eff}} \). The \( k_{\text{eff}} \) calculations are performed using the Monte Carlo code MCNP6.1 and the ENDF/B-VII.1 evaluation where, in all simulations, a relatively high number of neutron histories (4000 active cycles of 100,000 neutrons) are used to neglect statistical uncertainties (3–6 pcm). In addition, the 44 energy groups used in these simulations are listed in Table 2.

Since the value of each physical quantity is given with uncertainty, our job is to reduce this uncertainty by applying the least squares method. For this, we need input parameters (anterior \( k_{\text{eff}} \) anterior covariance and sensitivity matrix, etc.), which allows us to calculate the adjusted output parameters (posterior \( k_{\text{eff}} \) posterior covariance matrix, and cross section) using Equations (6), (7), and (9).

The files used in this study are carefully processed and include the MCNP6.1 input files describing the benchmark simulation, the ACE (A Compact ENDF) nuclear data libraries, and the NJOY99 input files required for the processing of data covariance. The MCNP6.1 code uses this format (ACE) to calculate the multiplication factors using the KCODE card and sensitivity matrices using the KSEN card. As shown in Equations (3) and (4), the sensitivity matrices of the nuclear data are multiplied by their corresponding covariance matrices to calculate the uncertainty.

### 3. Results and discussion

#### 3.1. Covariance of the \( ^{235}\text{U} \) capture multigroup cross section

In this step, the NJOY99 system was used to generate the \( ^{235}\text{U} \) capture multigroup cross section and
its covariance matrix of the ENDF/B-VII.1 evaluation. For this, the input file NJOY contains several modules, the main ones are GROUPR and ERROR for the multigroup cross sections (MF = 3) and the covariances (MF = 33), respectively. The results of the multi-capture group cross sections are shown in Figure 2 and the covariance generated with the VIEWR module of the NJOY system is shown in Figure 3.

Figure 3 shows the percent standard deviation of the $^{235}\text{U}$ capture cross-section uncertainty. These results can be used in turn to judge the adequacy of ENDF/B-VII.1 data. Covariance data improve the evaluation of cross sections using integral measurements (MacFarlane & Muir, 1994).

### 3.2. $k_{\text{eff}}$ and sensitivity

The sensitivities and multiplication factors ($k_{\text{eff}}$) are calculated for the selected benchmarks using the KSEN card of the MCNP6.1 code (Pelovitz, 2013). The sensitivity simulations of four benchmarks are shown in Figure 4.

The results obtained indicate that the $k_{\text{eff}}$ sensitivities caused by the $^{235}\text{U}$ capture cross section are remarkable for all energy groups greater than 10 and that large uncertainty is expected for fast benchmarks because they have a high sensitivity.

### 3.3. Uncertainty of the $^{235}\text{U}$ capture multigroup cross section

The uncertainty of nuclear data on the $k_{\text{eff}}$ caused by the capture multigroup cross section of the $^{235}\text{U}$ is calculated using Equations (3) and (4). Table 3 summarizes the values found.

From the above table, high uncertainty values are observed for fast benchmarks and mean values for the other benchmarks. This demonstrated that it is necessary to adjust the nuclear data of the $^{235}\text{U}$ capture cross section to reduce these uncertainties.

### 3.4. The relative correction of the $^{235}\text{U}$ capture cross section

Figure 5 represents the relative correction of the $^{235}\text{U}$ capture cross section using ENDF/B-VII.1 evaluation in 44 neutron energy groups.

### Table 3. Nuclear data uncertainty (in pcm) on $k_{\text{eff}}$ for the selected benchmarks.

| Benchmark cases | $\Delta k_{\text{eff-nucl ENDF/B-VII.1}}$ |
|-----------------|----------------------------------------|
| Hst001.002      | 72.249950                              |
| Hst001.004      | 70.865330                              |
| Hst001.007      | 32.842850                              |
| Hst001.008      | 68.380810                              |
| Hst001.009      | 72.183650                              |
| Hst001.010      | 34.785670                              |
| Hst009.001      | 242.456000                             |
| Hst009.001R     | 242.423800                             |
| Hst009.002      | 193.746700                             |
| Hst009.003      | 131.152100                             |
| HST011.001      | 27.772630                              |
| HST011.002      | 27.327540                              |
| HST012.001      | 14.374460                              |
| HST013.001      | 13.730140                              |
| HST035.007      | 69.771800                              |
| HMF003.001      | 1001.716000                            |
| HMF003.002      | 1038.204000                            |
| HMF003.003      | 1087.869000                            |
| HMF003.004      | 1111.644000                            |
| HMF003.005      | 1183.479000                            |
| HMF003.006      | 1299.690000                            |
| HMF003.010      | 1386.870000                            |
| HMF014          | 1018.521000                            |
| HMF021.002      | 963.558800                             |
| HMF022.002      | 928.469100                             |
| HMF026.011      | 793.130900                             |
| HMF028          | 1112.792000                            |

Figure 5. The relative correction of the $^{235}\text{U}$ capture cross section in 44 energy groups.
Table 4. Estimation of the relative correction (%) of $^{235}\text{U}$ capture cross section in 44 energy groups.

| Energy groups | 1–24 | 24–30 | 30–34 | 33–44 |
|---------------|------|-------|-------|-------|
| Cross section | 0–6 eV | 6 eV–3 keV | 3 keV–0.4 MeV | >0.4 MeV |
| $^{235}\text{U}$ (ENDF/B-VII.1) | −1% | −0.5% | 2% | −10% |

Figure 6. Ratio of posterior/prior covariance matrix of the $^{235}\text{U}$ capture cross section in the evaluation ENDF/B-VII.1.

Figure 7. Prior, posterior, and experimental $k_{\text{eff}}$ with one standard deviation in the evaluation ENDF/B-VII.1.

Figure 8. Prior, posterior, and experimental $k_{\text{eff}}$ with two standard deviation in the evaluation ENDF/B-VII.1.
The relative corrections obtained for the $^{235}$U capture cross section in 44 neutron energy groups are summarized in Table 4 for the evaluation ENDF/B-VII.1.

### 3.5. Adjusted multigroup covariance matrix of the $^{235}$U capture cross section

Using the previous results and Equation (7), we calculated the posterior covariance matrix of the $^{235}$U capture cross section. Its ratio to that of the one prior is shown in Figure 6.

Figure 6 shows that the ratio of the posterior and prior covariance matrix of the $^{235}$U capture cross section is less than the unit in the energy groups 22 to 29 (1.77–550 eV). This indicates that the covariance improved. The studied ratio is null in the energy groups 29 to 44, for this reason, it is not presented in this figure.

### 3.6. Comparison between prior posterior and experimental $k_{\text{eff}}$

To validate the accuracy of the adjustment process of the $^{235}$U capture cross section in the ENDF/B-VII.1 evaluation, the $k_{\text{eff}}$ is calculated with the adjusted nuclear data using Equation (9). The following figures show the anterior, posterior, and experimental $k_{\text{eff}}$, where the experimental $k_{\text{eff}}$ is plotted with one and two standard deviations, respectively, in the two figures below.

Figures 7 and 8 show that in some thermal and fast benchmarks, the calculated posterior $k_{\text{eff}}$ is improved and there is an improvement in the adjusted values. But in the other benchmarks, the calculated anterior and posterior $k_{\text{eff}}$ stay the same. In addition, Figure 8 show that the anterior and posterior $k_{\text{eff}}$ are all included in the experimental confidence interval.

To investigate the preceding results, prior and posterior ratios of the calculated $k_{\text{eff}}$ to the experimental are calculated and presented in Figure 9.

The figure above clearly indicates that the adjusted $k_{\text{eff}}$ is close to the experimental one in almost studied benchmarks.

### 3.7. Comparison between prior and posterior nuclear uncertainty on $k_{\text{eff}}$

The last step of this work is to test the effect of the adjusted covariance matrix on the calculated nuclear uncertainties using Equation (10). Table 5 presents these uncertainties as well as the previous ones.

The results presented in Table 5 show a significant improvement in nuclear data uncertainty caused by the capture cross section of uranium 235. Posterior uncertainties on $k_{\text{eff}}$ were reduced by about 40% in thermal benchmarks and about 90% in fast benchmarks in the ENDF/B-VII.1 evaluation.
4. Conclusion

In this paper, nuclear data uncertainty on $k_{eff}$ was studied to adjust the $^{235}$U capture cross section taken from ENDF/B-VII.1, with the MCNP6.1 Monte Carlo code, NJOY99 nuclear data processing system, and the GLLSM processed by a FORTRAN 95 program. Several critical benchmarks showed that the $^{235}$U capture cross section and its covariance matrix require an adjustment in the studied evaluation. Also, the obtained results have proved amelioration of the calculated $k_{eff}$ and its nuclear uncertainty. The proposed adjustment in the $^{235}$U capture cross-section and its covariance matrix of the ENDF/B-VII.1 evaluation reduced the difference between the experimental $k_{eff}$ and the $k_{eff}$ calculated in several benchmarks. In addition, a significant decrease in nuclear uncertainties on the $k_{eff}$ was observed after adjustment. In general, all the results obtained in this paper are satisfactory, but to further increase the criticality safety margin, the difference between experimental and calculated $k_{eff}$ must be further reduced by improving the cross section studied and its covariance matrix.

Disclosure statement

No potential conflict of interest was reported by the authors.

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