Summation calculation of delayed neutron yields for ^{235}U, ^{238}U and ^{239}Pu, based on various fission yield and neutron emission probability databases

Daniela Foligno[1,⋆], *Pierre* Leconte[1], *Olivier* Serot[1], *Olivier* Litaize[1], and *Abdelhazize* Chebboubi[1]

[1] *CEA Cadarache, 13115 Saint-Paul-lez-Durance*

Abstract. Summation calculations have been performed in order to compare the quality of several nuclear data libraries. The objective was to obtain the average delayed neutron yield, as well as the average delayed-neutron half-life for different fissioning systems (^{235}U, ^{238}U and ^{239}Pu) at different energies (thermal and fast) by using microscopic data. Each quantity is presented with a first evaluation of the uncertainty, computed under the assumption that the variables are all independent of each other.

1 Introduction to Summation calculation

The *Summation Method* consists on using microscopic data to compute a macroscopic quantity of interest by summing the contribution of each component (e.g. the contribution of each isotope).

- *Average delayed neutron yield ($\overline{\nu_d}$):* average number of delayed neutrons emitted per fission. It can be computed from the cumulative fission yields (CY) and the delayed-neutron-emission-probabilities ($\overline{P_{\rm n}}$[1]) from different libraries:

$$\overline{\nu_d} = \sum_i^N CY_i \cdot \overline{P_{n,i}} \tag{1}$$

  where i identifies a delayed neutron precursor and N the number of precursors. Notice that the use of the cumulative yields implies that the full decay chain has been considered and therefore Equation 1 is only valid for an *infinite* irradiation [2].

- *Precursor's importance (I_i):* the contribution of the precursor i to the $\overline{\nu_d}$.

$$I_i = \frac{CY_i \cdot \overline{P_{n,i}}}{\overline{\nu_d}} \tag{2}$$

⋆e-mail: daniela.foligno@cea.fr

[1] Defined as $\overline{P_{n,i}} = P_{n,i} + 2 \cdot P_{2n,i}$ since it takes into account both β_n^- and β_{2n}^- decays.

[2] Infinite with respect to the longest-lived precursor's half-life.

- *Average delayed-neutron precursors' half-life life ($< T_{1/2} >$):* average of the precursors' half-lives weighted on the precursor's importance. This parameter is typical of the fissioning system and it is an indication of the time dependence of the DN decay curves.

$$< T_{1/2} >= \frac{\sum_i^n \overline{P_{n,i}} \cdot CY_i \cdot T_{1/2,i}}{\sum_i^n \overline{P_{n,i}} \cdot CY_i} = \sum_i^n I_i \cdot T_{1/2,i} \qquad (3)$$

A calculated $< T_{1/2} >$ smaller than the experimental value would point out an underestimation of the long-lived precursors' contribution or an overestimation of the short-lived precursors' contribution in the summation process, and vice-versa. It is worth mentioning that the reactivity (ρ) is not a measurable quantity and it can only be estimated through a reactor period (T) measurement. The two quantities are linked to each other through the *simplified Inhour equation*, which contains the $< T_{1/2} >$. It is obvious that an incorrect value of the average precursors' half-life would lead to a wrong estimation of the reactivity, even in the case of accurate period measurements.

2 Results

The recommended values for the average delayed neutron yields are shown in Table 1, while the results of the calculations are shown in Tables 2 to 6. Notice that two error-values are shown for each quantity. One should be aware that in absence of measurements, nuclear data is computed through theoretical models. In this case, the values are *estimations*, and should be accompanied by a range of confidence. In reality, theoretically-derived data is presented in the libraries without any uncertainty value. The difference between the two presented errors is that the first one has been computed considering the theoretical values as completely reliable, and the second one by imposing the relative uncertainty on the data to be 100%. Table 7 reports the three most important precursors for all the considered fissioning systems, while Table 8 the average delayed neutron precursors' half-life, where the errors have been computed with the first of the two previously mentioned ways of dealing with theoretical models.

Table 1. $\overline{\nu_d}$ recommended values from the literature: Spriggs et al. [2] and WPEC-6 [3] respectively.

	^{235}U$_{th}$	^{235}U$_{fast}$	^{238}U$_{fast}$	^{239}Pu$_{th}$	^{239}Pu$_{fast}$
[2]	1.62E-02(3.1%)	1.63E-02(1.8%)	4.65E-02(2.4%)	6.50E-03(6.2%)	6.51E-03(2.5%)
[3]	1.62E-02	1.66E-02	4.65E-02	6.50E-03	6.55E-03

Table 2. $\overline{\nu_d}$ calculation by Summation Method - ^{235}U$_{thermal}$

^{235}**U**		*CY*	
		thermal	
		JEFF-3.1.1	**ENDF/B-VII.0**
P$_n$	**ENDF/B-VII.1**	1.57E-02 (5.2-5.3%)	1.90E-02 (5.5-8.4%)
	JEFF-3.1.1	1.48E-02 (5.3-6.0%)	1.73E-02 (3.4-11.5%)
	Pfeiffer	1.62E-02 (5.5-5.6%)	1.84E-02 (5.7-7.5%)

Table 3. $\overline{\nu_d}$ calculation by Summation Method - $^{235}U_{fast}$

^{235}U		CY	
		fast	
		JEFF-3.1.1	**ENDF/B-VII.0**
P$_n$	**ENDF/B-VII.1**	1.72E-02 (5.0-5.0%)	1.76E-02 (6.6-6.8%)
	JEFF-3.1.1	1.70E-02 (5.2-5.6%)	1.66E-02 (5.5-6.5%)
	Pfeiffer	1.81E-02 (5.5-5.5%)	1.81E-02 (6.9-6.9%)

Table 4. $\overline{\nu_d}$ calculation by Summation Method - $^{238}U_{fast}$

^{238}U		CY	
		fast	
		JEFF-3.1.1	**ENDF/B-VII.0**
P$_n$	**ENDF/B-VII.1**	4.51E-02 (3.5-5.5%)	4.22E-02 (6.6-7.7%)
	JEFF-3.1.1	4.04E-02 (3.2-5.9%)	3.79E-02 (6.0-8.0%)
	Pfeiffer	4.53E-02 (4.4-4.5%)	4.29E-02 (7.1-7.1%)

Table 5. $\overline{\nu_d}$ calculation by Summation Method - $^{239}Pu_{thermal}$

^{238}U		CY	
		thermal	
		JEFF-3.1.1	**ENDF/B-VII.0**
P$_n$	**ENDF/B-VII.1**	6.03E-03 (6.5-6.6%)	7.18E-03 (3.2-3.5%)
	JEFF-3.1.1	6.05E-03 (7.1-7.5%)	7.02E-03 (4.0-8.0%)
	Pfeiffer	6.52E-03 (7.1-7.2%)	7.45E-03 (4.4-4.4%)

Table 6. $\overline{\nu_d}$ calculation by Summation Method - $^{239}Pu_{fast}$

^{238}U		CY	
		fast	
		JEFF-3.1.1	**ENDF/B-VII.0**
P$_n$	**ENDF/B-VII.1**	6.54E-03 (6.4-6.4%)	6.32E-03 (9.7-9.8%)
	JEFF-3.1.1	6.75E-03 (6.8-7.1%)	6.04E-03 (10.0-10.3%)
	Pfeiffer	7.21E-03 (7.0-7.1%)	6.56E-03 (9.6-9.6%)

3 Analysis

3.1 FY-Libraries:

- An overestimation of the ^{86}As's CY is present in the ENDF/B-VII.0 library, especially considering the fact that in JEFF-3.1.1 there is no difference between the independent and the cumulative yield for this isotope (see Table 9). An explication for this large discrepancy has been found in the paper from A. Sonzogni [5], who noticed an overestimation in his summation calculation of antineutrino spectra due to the excessive CY of ^{86}Ge (^{86}As's father). Looking at the previous versions of the library he found out that in 1993, when passing from ENDF/B-VI.1 to ENDF/B-VI.2, the ^{86}Ge has been erroneously given the CY of the ^{86}Se, while keeping the same IY. Substituting ENDF/B-VII.0's CY for ^{86}As with JEFF-3.1.1's one, the average DN yield for the thermal fission of ^{235}U undergoes a reduction of about 3%

Table 7. Precursors' Importance computed with CY from JEFF-3.1.1 and P_n from Pfeiffer

TOP 3 Precursors	^{235}U$_{th}$	^{235}U$_{fast}$	^{238}U$_{fast}$	^{239}Pu$_{th}$	^{239}Pu$_{fast}$
1st	^{137}I (15.5%)	^{137}I (12.3%)	^{137}I (8.7%)	^{137}I (24.7%)	^{137}I (17.1%)
2nd	^{89}Br (11.5%)	^{94}Rb (12.3%)	^{94}Rb (6.7%)	^{98m}Y (9.7%)	^{94}Rb (12.5%)
3rd	^{94}Rb (8.4%)	^{89}Br (12.0%)	^{90}Br (6.6%)	^{94}Rb (9.7%)	^{98m}Y (11.2%)

Table 8. Average delayed-neutron half-life

	Summation		**6-groups**	**8-groups**	**Recommended**
	JEFF-ENDF	**JEFF-Pfeiffer**	**ENDF/B-VII.1**	**JEFF-3.1.1**	[2]
^{235}U$_{th}$	9.42 ± 0.40	9.01 ± 0.44	7.67	9.02 ± 0.27	9.02 ± 0.34
^{235}U$_{fast}$	8.69 ± 0.27	8.20 ± 0.40	7.67	9.11 ± 0.09	9.03 ± 0.08
^{238}U$_{fast}$	5.03 ± 0.15	4.93 ± 0.22	3.05	5.32 ± 0.11	5.32 ± 0.14
^{239}Pu$_{th}$	11.22 ± 0.70	10.34 ± 0.75	9.21	10.69 ± 0.85	10.69 ± 1.11
^{239}Pu$_{fast}$	9.86 ± 0.65	8.97 ± 0.68	9.21	10.35 ± 1.09	10.09 ± 1.26

- Precursors' CYs increase with energy (0.025 eV → 400 keV) in the JEFF-3.1.1 library much more than in the ENDF/B-VII.0 library (see Fig. 1), leading to an increase of the $\overline{\nu_d}$, which is supposed to be energy-insensitive until the MeV scale. In the same figure, one could also notice the large decrease in the ^{86}As CY with energy according to the ENDF/B-VII.0 library. However, the apparent reduction is only due to the wrong value at thermal energy, as mentioned before
- A sensitivity analysis showed that most of the uncertainty on the delayed neutron yield comes from the uncertainty on the fission yields. Furthermore, it has to be stressed that FY-data comes from measurements and it is always accompanied by uncertainties.

3.2 P_n-Libraries:

- JEFF-3.1.1 P_n-library always gives an underestimated $\overline{\nu_d}$ because several precursors' emission probabilities are missing
- ^{98m}Y's P_n underwent an order of magnitude change from ENDF/B-VII.0 to ENDF/B-VII.1, leading to a huge difference in the ^{239}Pu's importance list. Indeed, this precursor contributes to 10% of the $\overline{\nu_d}$ according to ENDF/B-VII.0 and to 1% according to ENDF/B-VII.1 because, in the most recent version of the library, the metastable state has been given the same P_n as the ground state (see Table 10). This is likely to be a typing mistake, since JEFF-3.1.1 and ENDF/B-VII.0 agree on the ^{98m}Y's P_n value

Table 9. Cumulative vs Independent Yield Ratio for some precursors

^{235}U$_{th}$	CY_i/IY_i	
Precursors	**ENDF/B-VII.0**	**JEFF-3.1.1**
^{86}As	27.8	1.0
^{85}As	1.8	1.0
^{96}Rb	1.2	1.5
^{137}Te	1.2	1.0

Table 10. Neutron emission probabilities - Transcription Error (ENDF/B-VII.0 → ENDF/B-VII.1)

Precursors	**ENDF/B-VII.0**	**ENDF/B-VII.1**
^{98}Y	3.31E-03	3.31E-03
^{98m}Y	3.20E-02	3.31E-03

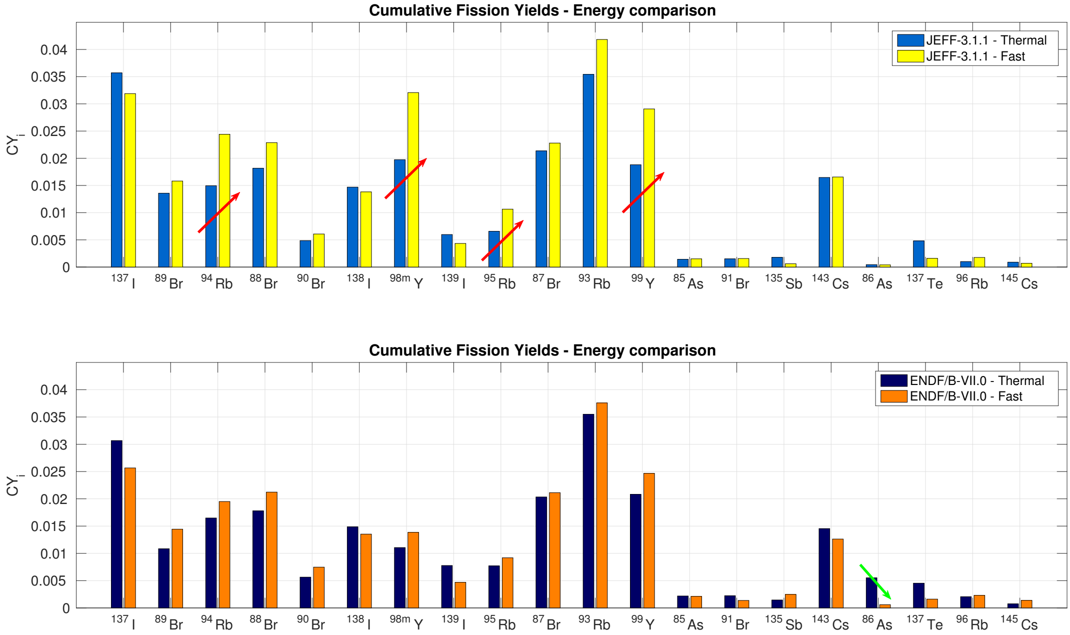

Figure 1. Precursors' Cumulative Yields - Thermal vs Fast Fission - JEFF-3.1.1 vs ENDF/B-VII.0

4 Conclusions

1. About 90% of delayed neutrons are emitted by odd-Z fission products [6], which at thermal incident neutron energies are less likely to be produced than even-Z isotopes. At higher excitation energies the proton pairing energy loses its importance and the even-odd effect weakens. A small increase of the DN yield with energy is therefore expected up to about 4 MeV, where the second chance fission occurs. Another drop in the $\overline{\nu_d}$ is expected around 12 MeV due to the third-chance fission. This phenomenon seems to be compensated by the fact that at higher incident neutron energies the fission becomes more and more symmetric, leading to a reduction in the $\overline{\nu_d}$. Since the P_n do not change with energy, the ony way to study this effect is to analyse the CYs

2. The best results are obtained when coupling CY data from JEFF-3.1.1 and P_n data from ENDF/B-VII.1, but the energy-dependence of JEFF-3.1.1 fission yields has to be improved. A new set of fast CYs has been obtained by using JEFF-3.1.1 thermal CYs and GEF energy model, but further studies are needed

3. The eight-groups DN set from JEFF-3.1.1, unlike the six-groups set from ENDF/B-VII.1, gives good estimations of the average DN precursors' half-lives, always in the range of the uncertainty with respect to the recommended values

4. Further work needs to be done on uncertainties, aiming at both reducing the uncertainties on fission yields, and estimating the uncertainty on the P_n derived by theoretical models when experimental data was missing.

References

[1] L. Mathieu, O. Serot, T. Materna, A. Bail, U. Köster, H. Faust, O. Litaize, E. Dupont, C. Jouanne, A. Letourneau, S. Panebianco, *New neutron long-counter for delayed neutron investigations with the LOHENGRIN fission fragment separator*, JINST 7, P08029 (2012). http://iopscience.iop.org/article/10.1088/1748-0221/7/08/P08029.

[2] G.D. Spriggs, J.M. Campbell, V.M. Piksaikin, *An 8-group delayed neutron model based on a consistent set of half-lives*, Prog. Nucl. Energy 41, 223 (2002).

[3] G. Rudstam, Ph. Finck, A. Filip, A. D'angelo, R.D. McKnight, *Delayed neutron data for the major actinides*, NEA/WPEC-6 (2002). https://www.oecd-nea.org/science/wpec/volume6/volume6.pdf.

[4] V. Zammit-Averlant, *Validation Intégrale des Estimations du Parametre Béta Effectif pour les Réacteurs MOX et Incinérateurs*, Ph.D. thesis, Université Aix-Marseille I (1998).

[5] A.A. Sonzogni, E.A. McCutchan, T.D. Johnson, P. Dimitriou, *Effects of Fission Yield Data in the Calculation of Antineutrino Spectra for* ^{235}*U(n,fission) at Thermal and Fast Neutron Energies*, Phys. Rev. Lett. 116, 132502 (2016).

[6] M.C. Brady, *Evaluation and Application of Delayed Neutron Precursor Data*, LA–11534-T United States (1989)