

# New delayed neutron group constants and covariances for LWR core applications, combining summation calculations and integral experiments

P. Leconte<sup>a,\*</sup>, P. Archier<sup>a</sup>, C. De Saint Jean<sup>a</sup>, R. Diniz<sup>b</sup>, A. Dos Santos<sup>b</sup>, L. Fautrat<sup>a</sup>, D. Foligno<sup>a</sup>, B. Geslot<sup>c</sup>, E. Gilad<sup>d</sup>, P. Tamagno<sup>a</sup>, G. Truchet<sup>e</sup>, A. Zoia<sup>f</sup>

<sup>a</sup> CEA, DEN, DER/SPRC/LEPh Cadarache, F-13108 Saint Paul-Lez-Durance, France

<sup>b</sup> Instituto de Pesquisas Energéticas e Nucleares – IPEN/CNEN-SP, Av. Prof. Lineu Prestes 2242, Cidade Universitaria, São Paulo 05508-000, Brazil

<sup>c</sup> CEA, DEN, DER/SPEx/LPE Cadarache, F-13108 Saint Paul-Lez-Durance, France

<sup>d</sup> The Unit of Nuclear Engineering, Ben-Gurion University of the Negev, Beer-Sheva 84105, Israel

<sup>e</sup> CEA, DEN, DER/SPRC/LPN Cadarache, F-13108 Saint Paul-Lez-Durance, France

<sup>f</sup> DEN-Service d'études des réacteurs et de mathématiques appliquées (SERMA), CEA, Université Paris-Saclay, F-91191, Gif-sur-Yvette, France

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## 1. Introduction

The dynamic behavior of a nuclear reactor is commonly evaluated from the “inhour equation”, which is derived from the point kinetic equations, coupling the neutron density to the precursor concentrations as a function of time (Keepin, 1965). Its formulation, under the assumption of a negligible source contribution, relates the dynamic reactivity  $\rho$  (in pcm =  $10^{-5}$ ) to the asymptotic reactor period  $T = 1/\omega$  as follows:

$$\rho = \omega + \sum_{k=1}^{n_{\text{prec}}} \frac{\beta_k^{\text{eff}} \omega}{\omega + \lambda_k} \quad (1)$$

with  $\beta_k^{\text{eff}}$  the effective delayed neutron fraction for the  $k$ -th precursor group,  $\lambda_k$  the delayed neutron decay constant for the  $k$ -th precursor group,  $\Lambda$  the mean generation time. The concept of “effectiveness” in the delayed neutron group fractions is addressing the weighted average of data coming from various fissionable nuclides (indexed  $i$  below) and incident neutron energies. It can be expressed as follows:

$$\beta_k^{\text{eff}} = \frac{\sum_{\text{isotopes } i} \int_0^\infty \phi^+(E') \chi_{d,i,k}(E') dE' \int_0^\infty a_{k,i}(E) d_i(E) \Sigma_{f,i}(E) \phi(E) dE}{\sum_{\text{isotopes } i} \int_0^\infty \phi^+(E') \chi_{t,i}(E') dE' \int_{0t,i}^\infty (E) \Sigma_{f,i}(E) \phi(E) dE} \quad (2)$$

Here  $\phi$  and  $\phi^+$  are respectively the forward and adjoint flux,  $\Sigma_f$  the macroscopic fission cross section,  $a$  the delayed neutron relative abundance,  $\chi_d$  and  $\chi_t$  respectively the delayed and total (prompt + delayed) neutron spectrum,  $\chi_d$  and  $\chi_t$  respectively the number of delayed and total emitted neutrons per fission.

For an accurate prediction of the dynamic reactivity  $\rho$ , the choice of delayed neutron parameters ( $a_k, \lambda_k$ ) is crucial. One should keep in mind that reactivity is always inferred from experimental information, the reactor period, and input nuclear data as diverse as the delayed neutron yields, spectra and relative abundances, fission cross sections and total neutron multiplicities. When the dynamic reactivity is measured as a reactor physics parameter to be used for code validation, like for instance in sodium void reactivity coefficients performed in Zero Power Reactors (ZPR) (Tommasi et al., 2010) or in rod drop experiments (Geslot et al., 2007), it is important to evaluate properly the propagated uncertainty coming from the input nuclear data. In addition, there is a subtle issue when experiments from several institutes are analyzed using different sets of delayed neutron data. While sometimes corrections could be performed to have consistent reactivities, input delayed neutron data are generally not documented enough to correct the raw data.

As it was quoted by several papers on both evaluated data (Gremyachkin et al., 2015) and calculated dynamic reactivities (Santos and Diniz, 2014) (Zoia et al., 2016) (Zoia et al., 2017), cur-

\* Corresponding author.

E-mail addresses: [pierre.leconte@cea.fr](mailto:pierre.leconte@cea.fr) (P. Leconte), [pascal.archier@cea.fr](mailto:pascal.archier@cea.fr) (P. Archier), [cyrille.de-saint-jean@cea.fr](mailto:cyrille.de-saint-jean@cea.fr) (C. De Saint Jean), [rdiniz@ipen.br](mailto:rdiniz@ipen.br) (R. Diniz), [asantos@ipen.br](mailto:asantos@ipen.br) (A. Dos Santos), [leo.fautrat@cea.fr](mailto:leo.fautrat@cea.fr) (L. Fautrat), [daniela.foligno@cea.fr](mailto:daniela.foligno@cea.fr) (D. Foligno), [benoit.geslot@cea.fr](mailto:benoit.geslot@cea.fr) (B. Geslot), [gilade@bgu.ac.il](mailto:gilade@bgu.ac.il) (E. Gilad), [pierre.tamagno@cea.fr](mailto:pierre.tamagno@cea.fr) (P. Tamagno), [guillaume.truchet@cea.fr](mailto:guillaume.truchet@cea.fr) (G. Truchet), [andrea.zoia@cea.fr](mailto:andrea.zoia@cea.fr) (A. Zoia).

rent evaluated nuclear data libraries show strong differences, not only in the average number of delayed neutrons per fission  $\nu_d$ , but also in the description of the time dependence of their emission. These differences cannot be justified by delayed neutron data uncertainties, as covariance data are missing from most of evaluated nuclear data libraries. The challenge for future evaluated files is to provide new data with uncertainties and correlations, to be used in uncertainty analysis.

This paper proposes a contribution towards the evaluation of new delayed neutron data and associated covariances, based on a combination between microscopic and macroscopic data. The first part briefly recalls the evaluation work done during the last fifty years and the current status of nuclear data libraries. The second part describes the two-step approach that has been applied to derive delayed neutron group constants for  $^{235}\text{U}$  and  $^{238}\text{U}$ . It is based firstly on the application of the summation method, describing the number and time dependence of more than 400 known individual neutron precursors, and secondly on the measurement and fitting procedure of the amplitude and phase of the zero power transfer function, as measured in two independent reactor experiments. In the third part, our evaluated data is applied to compute dynamic reactivities and associated uncertainties in different Light Water Reactor (LWR) core applications, and will be compared to similar results based on the evaluated data from the main evaluated nuclear data libraries.

## 2. State of the art in the evaluation of delayed neutron group constants

There are two ways of evaluating the delayed neutron temporal group constants: one based on a macroscopic approach, by measuring the delayed neutron emission following burst or step irradiations, and another one based on a microscopic approach, by measuring the characteristics of individual neutron precursors and applying the summation method. Both approaches have advantages and disadvantages that we will detail.

The macroscopic approach considers the delayed neutron emission rate  $n(t)$  as an empirical law to be represented by a limited number of parameters. As  $n(t)$  is driven by the combination of hundreds of radioactive decays, each one of them being described by one or several exponential terms, it is convenient to apply the following empirical law<sup>1</sup>:

$$n(t) = F_d \sum_{k=1}^n a_k (1 - e^{-\lambda_k t_i}) e^{-\lambda_k t} \quad (3)$$

where  $F$  is the fission rate during the irradiation phase of duration  $t_i$  and  $\nu_d$  the delayed neutron yield,  $a_k$  the relative abundance of the  $k$ -th delayed neutron group,  $n$  is the number of groups in the model.

Following earlier works in the 1940 s and 1950 s where four to five groups were used to fit the experimental data, G.R. Keepin undertook a comprehensive work of delayed neutron measurements for thermal fission of  $^{233}\text{U}$ ,  $^{235}\text{U}$  and  $^{239}\text{Pu}$ , and fast fission of  $^{233}\text{U}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ , and  $^{232}\text{Th}$ . Several tests were done to evaluate the optimum number of parameters to fit the data. Six groups, i.e. 12 free parameters of relative abundances  $a_k$  and decay constants  $\lambda_k$ , were found to be the optimum for minimizing the error in the Least Square Fit (LSF) of the data.

Reference (Keepin, 1965) provides group constants and delayed neutron yields with associated errors, resulting from the fitting procedure. For a very long time, Keepin experiments were referred as a "reference" data sets for the isotopes previously mentioned, due to

a comprehensive consideration of experimental errors that could occur in other experiments, for instance the minimization of the transient time of the sample between the irradiation position and the counting position. The evaluated data for  $^{235}\text{U}$  and  $^{238}\text{U}$  relative abundance in JENDL-4.0 are adopted from the ones of Keepin. However, during the 1990 s, an in-pile reactivity experiment consisting of a series of super-prompt-critical bursts at the Arizona TRIGA reactor questioned the quality of Keepin data (Spriggs and Campbell, 2002). To investigate these discrepancies and to review the status of delayed neutron data, a workshop sponsored by the Nuclear Energy Agency working party on delayed neutron data (NEA WPEC/SG-6) was organized at IPPE, in April 1997. One of the conclusions of this workshop was the suggestion to increase the number of groups, in order to better describe the die away of long-lived neutron precursors and to simplify the kinetics equations by imposing the same set of decay constants for any fissionable isotope at any neutron energy. In 2002, following several tests done with 6, 7 and 8 groups, Spriggs and Campbell proposed a new 8-group delayed neutron model, with the first 3 decay constants set to the half-lives of the 3 longest-lived dominant precursors (i.e.  $^{87}\text{Br}$ ,  $^{137}\text{I}$  and  $^{88}\text{Br}$ ). Fixing the decay constants results in an invariant description of 8 differential equations, instead of multiplying the 6-group model by the number of fissionable isotopes occurring in the system. Unfortunately, as pointed out by Spriggs and Campbell, most of the original decay curve data for the 245 sets of parameters reported in the literature have never been published. To overcome this limitation, they proposed to apply the expansion method, as a temporary way to correct the deficiencies of the current 6-group model, before being able to acquire new data to be fitted by the 8-group model. This method consists of a LSF of a simulated time-dependent behavior of a theoretical system undergoing power transients for reactivities ranging from 0.01 to 0.95\$. The uncertainty on the 8-group model is transferred from the 12 decay constant and relative abundances uncertainties to the reactivity estimation and then lumped to the group relative abundances in the 8-group LSF. This method had the advantage to preserve the same uncertainty as in the 6-group model.

Among the available experimental datasets of relative abundance measurements for the thermal fission of  $^{235}\text{U}$  and fast fission of  $^{235}\text{U}$  and  $^{238}\text{U}$  (Spriggs and Campbell, 2002a, 2002b) and in spite of the previous comments regarding the reactivity experiments using Keepin data, Spriggs came to the conclusion that no better data were produced since, so the expansion from 6-group to 8-group was undertaken based on the single set of Keepin data. The resulting group constants are given in the appendixes of the SG-6 report and were adopted in the JEFF evaluated library since version 3.1. Concerning the relative abundance errors, even if they preserved the consistency with Keepin data, some questions arise. Indeed, these data are missing important information on the correlations between the 12 free parameters. These correlations cannot be recomputed a posteriori, because Keepin did not fit the 12 parameters simultaneously. Because this method gave higher calculated errors on  $a_k$  and  $\lambda_k$ , he proceeded in two steps by determining the four long-period groups from a long-irradiation, then the four shorter-period groups from a burst irradiation, and finally he applied a normalization of the two datasets (Keepin, 1965). This approach is obviously creating correlations that are difficult to evaluate, and also potential errors resulting from a different delayed neutron emission behavior between power step and pulse irradiations. Indeed, in the latter, as delayed neutron precursors are not in equilibrium with their parents, the kinetics of the delayed neutron emission at short-time is different and might not be represented similarly as for power-step irradiations (Foligno, 2017).

During the 1970s and 80s, with the much better understanding of the physics of delayed neutrons, a microscopic approach was

<sup>1</sup> Note that this formulation is not unique. Polynomial expansions could be applied as well but this would imply a larger number of free parameters for an equivalent chi-square.

introduced to overcome the limitation occurring in the measurement of shortest-lived delayed neutron groups and to extend the calculation of group constants to other fissionable isotopes. The method is usually referred to as the “summation method”, as the delayed neutron yield is summed from the contribution of all individual neutron precursors as follows:

$$v_d = \sum_{\text{precursor } k} P_{nk} Y_{nk} \quad (4)$$

Where  $P_{nk}$  is the neutron emission probability of precursor  $k$  and  $Y_{nk}$  its cumulative fission yield. For summation calculation of group constants, a more elaborate equation must be written, taking into account the kinetics of precursor decays and their build-up by  $\beta^-$  and/or  $(\beta, n)$  decay of their parent nuclides.

The work of Brady and England (1989) is a comprehensive application of the summation method to produce delayed neutron yields, group constants and delayed neutron spectra per group, covering 43 fissionable systems. One of the main advantages of this approach is that it correlates evaluated data for several fissionable isotopes, as they all share the same delayed neutron emission probabilities and only differ by the fission yield data. This approach is also expected to provide a much better description of delayed neutron emission as a function of incident neutron energy, considering the better understanding of the physics of the fission process. The Brady and England data were adopted in ENDF/B-VI nuclear data library and were not changed in latest versions (except in ENDF/B-VII.0 but next versions VII.1 and VIII.0 $\beta$ 4 came back to Brady and England data). Surprisingly, while reference (Brady and England, 1989) provides energy dependent data for thermal, fast and high-energy neutrons, the same set of data was imposed for energies from 0 to 30 MeV, based on the fast fission results. Although the work by Brady & England represents a strong effort to fill the gap for minor actinides (where few measurements exist), strong inconsistencies with the Keepin data for Uranium systems were pointed out by some authors (Spriggs, 1993; Williams, 1996). One should notice that Brady & England considered only 271 neutron precursors, while we have now experimental evidence for more than 400 delayed neutron precursors. In spite of this, the total delayed neutron yield is 13% higher than the Keepin value, so we should suspect some inconsistencies in the individual data. One may also question the way the delayed neutron emission rates were simulated with the CINDER-10 code, where only pulse-type irradiations were considered, while the experimental data by Keepin were acquired in both pulse-type and long irradiation cycles. The combination of them provide a much higher constraint in the fitting process between the short-lived precursors (emphasized by short irradiations) and the long-lived ones (emphasized by long-irradiations). As mentioned before, this approach does not lead to the same decay curves as for power-step irradiations, resulting in possible distortions in the evaluation of group constants. Finally, uncertainties were not provided, so that it is difficult to assess how inconsistent the group constants are with Keepin data.

We summarize in Table 1 the delayed neutron group constants for thermal fission of  $^{235}\text{U}$  and fast fission of  $^{238}\text{U}$  from the three following sets: Keepin 6-group data (=JENDL-4.0), Keepin 8-group expanded data (=JEFF-3.3) and Brady & England data (=ENDF/B-VIII.0 $\beta$ 4). The average half-life of delayed neutron emission  $\bar{T}_{1/2}$ , is a convenient way to appreciate the degree of agreement between two datasets:

$$\bar{T}_{1/2} = \ln(2) \sum_k \frac{a_k}{\lambda_k} \quad (5)$$

In the current table, one should notice that Brady & England data are 15% and 6% lower than the Keepin data for respectively  $^{235}\text{U}$  and  $^{238}\text{U}$ . This difference explains the 10% difference in the reactivity

estimation, as quoted in several papers (Santos and Diniz, 2014; Zoia et al., 2016; Zoia et al., 2017). The Keepin 8-group expanded data are leading to exactly the same average half-life  $\bar{T}_{1/2}$ , confirming the reliability of the Spriggs expansion method.

### 3. A two-step approach towards new evaluated relative abundances for U-235 and U-238

Our aim is to combine the best of the two approaches, using the microscopic approach with the most recent evaluated data of neutron precursors as a first guess, in addition to the macroscopic approach provided by reactor experiments. We will describe in more details each step in the next sub-sections.

#### 3.1. The microscopic approach: Derivation of group constants from summation calculations

Tests of combination between various evaluated cumulative fission yields (CFY) - JEFF-3, JENDL-4.0, ENDF/B-VII.0, GEFY-5.1 - and delayed neutron emission probabilities ( $P_n$ ) and half-lives ( $\lambda$ ) evaluations or compilations - Pfeiffer, Audi, JEFF-3.1.1/DD, ENDB/V-II.0, ENDB/V-II.1, ENSDF, RIPL-3, RIPL-3/2015 - were recently carried out at CEA, focusing on the three main isotopes  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{239}\text{Pu}$  (Foligno, 2017). Similar results were also presented by other authors, covering less combinations of evaluated data but on a wider range of fissionable isotopes, for instance in (Huynh and Jouanne, 2014; Dillmann et al., 2015). The consistent conclusions of these works are that the JEFF-3.1.1 library is the most reliable to predict the delayed neutron yields of  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{239}\text{Pu}$ . However, as shown in (Mathieu et al., 2012), JEFF-3.1.1/DD are missing many important delayed neutron precursors with neutron emission probabilities set to zero. On the contrary, the data taken from Pfeiffer, RIPL-3 and ENDF/B-VII.1 were much more complete, with more than 400 neutron precursors. Among these datasets, ENDF/B-VII.1 was preferred because most of evaluated data have uncertainty values on half-lives and neutron emission probabilities. For the few missing data, a conservative 100% uncertainty was assumed.

The combination of JEFF-3.1.1 fission yields with ENDF/B-VII.1 decay data leads to consistent results on the delayed neutron yields of  $^{235}\text{U}$  and  $^{238}\text{U}$  with experimental data, as recalled in Table 2.

Considering the satisfactory agreement obtained on the absolute delayed neutron yields, we will now consider the evaluation of delayed neutron group constants by the summation method. To this purpose, we wrote our own code to describe the delayed neutron emission rate of individual precursors. Bateman equations were numerically solved for the 400+ delayed neutron precursors with non-zero  $P_n$  in ENDF/B-VII.1 and non-zero CFY in JEFF-3.1.1. We recall that the balance equations describing the evolution in time of the atomic density of a precursor  $C$ , as well as of the ancestors  $B_i$ , during the irradiation phase, under the assumption of a constant fission rate  $F$ , are:

$$\begin{cases} \frac{dB_i(t)}{dt} = -\lambda_i B_i(t) + IY_{B_i} F \\ \frac{dC(t)}{dt} = -\lambda_C C(t) + IY_C F + \sum_{i=1}^K \lambda_{B_i \rightarrow C} B_i(t) \end{cases} \quad (6)$$

where  $\lambda_X$  is the decay constant of the isotope  $X$ ,  $\lambda_{B_i \rightarrow C}$  is the branching ratio, giving the probability for  $B_i$  to decay towards  $C$ ,  $IY_X$  is the independent fission yield of the  $X$  nuclide. The same equations are written with  $F = 0$  during the decay phase.

Irradiation times of 500 s, followed by 300 s of decay were simulated for  $^{235}\text{U}$  thermal fission and  $^{238}\text{U}$  fast fission (Fig. 1). A non-linear LSF of the total delayed neutron emission rate during

**Table 1**  
Delayed neutron relative abundances for <sup>235</sup>U thermal and <sup>238</sup>U fast fission.

Group	<sup>235</sup> U thermal						<sup>238</sup> U fast					
	Keepin 8-group expanded		Brady & England		Keepin 6-group		Keepin 8-group expanded		Brady & England		Keepin 6-group	
	$\lambda_k(s^{-1})$	$a_k$	$\lambda_k(s^{-1})$	$a_k$	$\lambda_k(s^{-1})$	$a_k$	$\lambda_k(s^{-1})$	$a_k$	$\lambda_k(s^{-1})$	$a_k$	$\lambda_k(s^{-1})$	$a_k$
1	0.0125	0.0328	0.0134	0.035	0.0124	0.033	0.0125	0.0084	0.0136	0.0139	0.0132	0.013
2	0.0283	0.154	0.0327	0.1807	0.0305	0.219	0.0283	0.104	0.0313	0.1128	0.0321	0.137
3	0.0425	0.0914	0.1208	0.1725	0.111	0.196	0.0425	0.0375	0.1233	0.1310	0.139	0.162
4	0.1330	0.197	0.3028	0.3868	0.301	0.395	0.1330	0.137	0.3237	0.3851	0.358	0.388
5	0.2925	0.331	0.8495	0.1586	1.14	0.115	0.2925	0.294	0.9060	0.2540	1.41	0.225
6	0.6665	0.0903	2.853	0.0664	3.01	0.042	0.6665	0.198	3.0487	0.1031	4.02	0.075
7	1.6348	0.0812					1.6348	0.128				
8	3.5546	0.0229					3.5546	0.0931				
$\bar{T}_{1/2}(s)$	9.03		7.66		9.03		5.32		4.98		5.32	

**Table 2**  
Delayed neutron yields  $v_d$  from summation calculations, compared with measured values.

Origin	<sup>235</sup> U thermal fission	<sup>238</sup> U fast fission
Keepin (Keepin, 1965)	(1.58 ± 0.05)%	(4.12 ± 0.17)%
Tuttle (1979)	(1.62 ± 0.05)%	(4.39 ± 0.10)%
Parish (1997)	(1.59 ± 0.04)%	-
Piksaikin (2002)	-	(4.61 ± 0.18)%
Our work: JEFF-3.1.1 (CFY) + ENDF/B-VII.1 ( $P_{nr}, \lambda$ )	(1.57 ± 0.08)%	(4.51 ± 0.25)%

are randomly sampled, according to normal laws. For the time being, all the input parameters are assumed to be independent, resulting in a conservative uncertainty on the simulated curves. For each set of sampled parameters, the LSF is performed, and the process is repeated  $m$  times (with typically greater than 1000m).

The relative abundances and associated uncertainties + correlations were derived from the computation of the standard deviation and variance of the  $(a_{k,l})_{k=1..8,l=1..m}$  matrix.

3.2. The macroscopic approach: Assimilation of reactor experiments

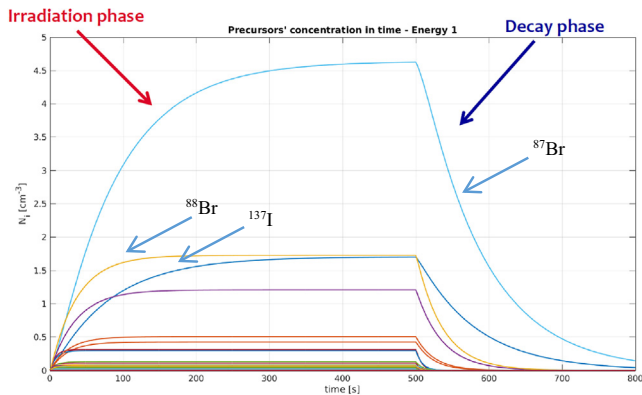
This first set of delayed neutron abundances, based on summation calculations, is now going to be constrained by two accurate reactor experiments related to the measurement of the Zero Power Transfer Function (ZPTF). We describe briefly the principles of each experiment before presenting how the data were analysed to derive a new set of delayed neutron group constants.

3.2.1. The IPEN/MB-01 neutron noise experiment

We consider the IPEN-LWR-RESR-001 benchmark, available from the International Reactor Physics Evaluation Project (IRPhEP). This experiment is described more in details in references (Santos and Diniz, 2014; International Handbook of Evaluated Reactor Physics Benchmark Experiments (IRPhEP), 2006).

IPEN/MB-01 is a zero power (<100 W) research reactor located in São Paulo, Brazil. The core configuration is a 28x26 lattice of 4.35% enriched uranium oxide fuel pins. The reactor is moderated with light water. The criticality is adjusted by the insertion of 2 groups of 12 AgInCd control rods. The square pitch of the core is chosen to be close to the optimum moderation ratio.

Measurements of the effective delayed neutron fraction and related group constants were carried out, using two kinds of neutron noise techniques. In a first step, the macroscopic neutron noise technique was applied to measure the neutron noise spectral distribution. Using two highly efficient fission chambers, located symmetrically in the water reflector of the core, two auto-power spectral densities (APSD) and one cross-power spectral density (CPSD) were recorded. Although this experimental technique is well-established for the measurement of  $\beta_{eff}$ , the innovation of the IPEN experiment was to acquire data during a very long time to resolve the low frequency range (<1 Hz), where the sensitivity to group constants is higher, while standard methods often consider the prompt neutron behavior above a few Hz. The IPEN team applied a non-linear LSF to evaluate the group constants, i.e.  $\lambda_k$  and  $\beta_k$ , in a 6-group model. We remind the theoretical formulation of



**Fig. 1.** Simulation of delayed neutron precursors build-up and decay in <sup>235</sup>U thermal fission.

the decay phase was performed with the MATLAB software. While in the Brady and England method the delayed neutron emission rates were fitted with 12 variable parameters in a 6-group model, we considered here fixed decay constants in a 8-group model, in agreement with that of Spriggs. To ensure the normalization of  $a_k$  parameters, there are two ways to proceed: we can fit 7 parameters and deduce the last one by  $1 - \sum_k a_k$ , or instead of fitting  $a_k$  we can substitute them by  $a_k = \mu_k / \sum_i \mu_i$  in Eq. (3) and fit the  $\mu_k$  parameters. The latter method was chosen because it was shown to be more robust to avoid potential negative values for the last parameter. Note that  $a_k$  standard deviations and correlations are evaluated a posteriori, based on the correlation matrix and standard deviations of  $\mu_k$ .

The uncertainty analysis of the relative abundances was done by using a Monte Carlo approach. All the unknowns in Eq. (5), i.e. independent fission yields, decay constants and branching ratios,



the CPSD, (the APSD being identical, with the addition of a constant term):

$$(\omega) = c \left| j\omega + \sum_k \frac{\beta_k^{\text{eff}} j\omega}{j\omega + \lambda_k} \right|^{-2} \quad (7)$$

The  $c$  constant includes several terms, including the Diven factor, the reactor power, the fission chamber currents and some other correction factors. The prompt neutron generation time  $\Lambda$  was fixed at 32  $\mu\text{sec}$  (Dos Santos et al., 2006), based on a previous experiment and the first decay constant associated to  $^{87}\text{Br}$  was fixed to the value  $1.2456\text{E-}02 \text{ s}^{-1}$ , also found in another experiment (Dos Santos et al., 2006). The remaining parameters  $(\lambda_k)_{k=2..6}$  and  $(\beta_k^{\text{eff}})_{k=1..6}$  as well as the  $c$  constant were let free in the LSF.

In a second step, the microscopic neutron noise technique was applied, by measuring the Rossi- $\alpha$  and Feynman- $\alpha$  neutron noise distributions in several sub-critical configurations. Using two detectors located in the core center and in the water reflector, and applying an enhanced two-region model, the ratio  $\alpha = -\beta^{\text{eff}}/\Lambda$  and  $\beta^{\text{eff}}$  were determined by fitting the theoretical formulations of the Rossi- $\alpha$  and Feynman- $\alpha$  distributions.

The interest of these macroscopic and microscopic neutron noise techniques is that no calculated corrections for the integral fission rate determination or for the Diven factor evaluation were necessary to derive the  $\alpha$  ratio, the  $\beta_{\text{eff}}$  and the group constants. It is currently the only available benchmark to allow the validation of the reactivity versus reactor period relationship, as detailed in reference (Leconte et al., 2016).

### 3.2.2. The MINERVE reactivity modulation experiment

During the past years, several kinetics parameters measurements were performed in the MAESTRO core configuration of the MINERVE zero power reactor, within the framework of several collaborations: between CEA and PSI using the neutron noise technique (Perret et al., 2017), and between CEA and Ben-Gurion University using the reactivity modulation technique (Gilad et al., 2015). We will consider the latter in this section.

MINERVE is a zero power (<100 W) pool-type reactor, located at CEA Cadarache. It is mainly used for nuclear data applications, based on the pile-oscillation technique. Aside from usual applications of sample oscillations, dedicated to the validation of reaction cross sections of various isotopes, materials or fuels, the measurement of  $\beta^{\text{eff}}$  using a reactivity modulation of the core was successfully tested in 2014. The experimental technique was inspired from previous works by Yedvab et al. (2006).

The reactivity perturbation was generated by the periodic insertion of a small water sample, using a dedicated mechanical piston. Under this reactivity excitation, the reactor undergoes a power modulation. Its amplitude corresponds to the ZPTF module and its phase shift in time is the ZPTF argument. In reference (Gilad et al., 2015), only the ZPTF module was considered, because the key goal of the experiment was to measure the  $\beta^{\text{eff}}$  parameter. Such experiments are very sensitive to power drifts that could be due for instance to temperature changes. Compared with neutron noise techniques, the acquisition of the ZPTF amplitude in the low frequency domain is much more time consuming, so the experimental data may not be of sufficient quality to constrain the fit of delayed neutron parameters, like in the IPEN experiment. However, the phase signal, which is obtained from the ZPTF argument, has the advantage to be insensitive to any reactor drift during the measurement. Its theoretical formulation reads:

$$\varphi(\omega) = -\text{atan} \left[ \left( \omega + \sum_k \frac{\beta_k^{\text{eff}} \omega_k}{\omega^2 + \lambda_k^2} \right) / \sum_k \frac{\beta_k^{\text{eff}} \omega^2}{\omega^2 + \lambda_k^2} \right] \quad (8)$$

The ZPTF phase was extracted from a Fourier transform of the periodic signal, recorded by a Boron ionization chamber and by a  $^{235}\text{U}$  fission chamber, both located in the core reflector.

### 3.2.3. Assimilation of the two experiments

The detailed analysis of both experiments revealed several possible improvements. First, fitting the ZPTF module and argument by a more constrained model, with fixed decay constants and variable abundances, to be consistent with the Spriggs model, would allow a more straightforward comparison with the 8-group expanded values from Keepin 6 group data. Second, fitting the relative abundances instead of the macroscopic parameters  $\beta_k^{\text{eff}}$  would ensure a more direct inference on the input nuclear data and related uncertainties, to be used for the calculation of other kinds of core configurations. Finally, as our goal is to produce covariance data to be used in uncertainty analysis for the computation of the dynamic reactivity, it is important to evaluate the correlation matrix between the fitted parameters.

In agreement with the IPEN Institute, (which kindly provided the raw data of the spectral densities acquired in the IPEN/MB01 experiment), and the Ben Gurion University (which shared the data and processing tools for the Fourier transform of the period signal recorded in the MINERVE experiment), we have tested a new method for analyzing the ZPTF, to infer delayed neutron group constants for  $^{235}\text{U}$  and  $^{238}\text{U}$ , which we are going to detail hereafter.

It is common to express  $\beta_k^{\text{eff}}$  as the product of the total effective delayed neutron fraction  $\beta^{\text{eff}}$  and the effective relative abundance of the  $k$ -th precursor group  $a_k^{\text{eff}}$ . Considering that the relative abundances  $a_{ki}(E)$  for isotope  $i$ , in precursor group  $k$  are usually described in a coarse energy mesh of three groups for thermal (25 meV), fast (400 keV) and high energy (14 MeV) neutrons, we can rewrite Equation (2) with the following condensed formulation:

$$\beta_k^{\text{eff}} = \beta^{\text{eff}} a_k^{\text{eff}} = \beta^{\text{eff}} \sum_{g=1}^3 \sum_{\text{isotopes } i} P_{kig} a_{kig} \quad (9)$$

Where  $P_{kig}$  is the relative production weight of the  $k$ -th group of delayed neutrons from the fission of isotope  $i$  induced by neutrons in energy group  $g$ :

$$P_{kig} = \frac{\int_{E_g} \phi^+(E') \chi_{d,k,i}(E') dE' \int_{E_g} v_{d,i}(E) \Sigma_{f,i}(E) \phi(E) dE}{\sum_{g=1}^3 \sum_{\text{isotopes } i} \int_{E_g} \phi^+(E') \chi_{d,i}(E') dE' \int_{E_g} v_{d,i}(E) \Sigma_{f,i}(E) \phi(E) dE} \quad (10)$$

This coefficient can be decomposed into a precursor-dependent term, and a second term not depending on precursors:

$$\begin{aligned} P_{kig} &= \frac{\int_{E_g} \phi^+(E') \chi_{d,i,k}(E') dE'}{\int_{E_g} \phi^+(E') \chi_{d,i}(E') dE'} \\ &\times \frac{\int_{E_g} \phi^+(E') \chi_{d,k}(E') dE' \int_{E_g} v_{d,i}(E) \Sigma_{f,i}(E) \phi(E) dE}{\sum_{g=1}^3 \sum_{\text{isotopes } i} \int_{E_g} \phi^+(E') \chi_{d,i}(E') dE' \int_{E_g} v_{d,i}(E) \Sigma_{f,i}(E) \phi(E) dE} \\ &= c_{kig} p_{ig} \end{aligned} \quad (11)$$

In this formulation, the first term, denoted  $c_{kig}$ , represents the deviation of the adjoint-weighted delayed neutron spectrum of precursor group  $k$  from the adjoint-weighted aggregate delayed neutron spectrum. The second term, denoted  $p_{ig}$ , is almost independent of the aggregate delayed neutron spectrum, because the mean energy does not significantly change from one isotope to another.

After minor manipulation of Eqs. (7) and (8), and using the decomposition of  $\beta_k^{\text{eff}}$  into a linear combination of  $a_{kig}$  terms where

we considered only the thermal fission of <sup>235</sup>U and fast fission of <sup>238</sup>U (99.8% of the total delayed neutron emission), we end up with the following functions to be fitted:

$$\phi(\omega) = \kappa \left| \frac{j\omega}{\alpha} + \sum_{k=1}^8 \frac{(pc_{k,U5th}a_{k,U5th} + (1-p)c_{k,U8f}a_{k,U8f})j\omega}{j\omega + \lambda_k} \right|^{-2} \quad (12)$$

$$\varphi(\omega) = -atan \left[ \frac{\frac{1}{\alpha} + \sum_{k=1}^8 \frac{(pc_{k,U5th}a_{k,U5th} + (1-p)c_{k,U8f}a_{k,U8f})}{\omega^2 + \lambda_k^2}}{\sum_{k=1}^8 \frac{(pc_{k,U5th}a_{k,U5th} + (1-p)c_{k,U8f}a_{k,U8f})\omega}{\omega^2 + \lambda_k^2}} \right] \quad (13)$$

Where  $\kappa$  is a constant term,  $\alpha = -\beta_k^{eff} / \Lambda$ ,  $p$  is the rate of delayed neutron emitted from thermal fission of <sup>235</sup>U and  $c_{k,i}$  is consistent with the definition in Eq. (10).

The  $p$  and  $c_{k,i}$  coefficients were evaluated for each experiment thanks to TRIPOLI4/JEFF-3.1.1 calculations. TRIPOLI-4<sup>®</sup> is a three-dimensional and continuous-energy Monte-Carlo particle transport code developed by CEA, devoted to shielding, reactor physics, criticality safety and nuclear instrumentation (Brun et al., 2015). In TRIPOLI4, the quantities  $\beta_k^{eff}$  can be decomposed into isotopic and group contributions (Zoia et al., 2016; Zoia et al., 2017). The parameter  $p$  is close to 0.9 for both experiments and the maximum deviation from unit for the  $c_{k,i}$  coefficients is 3%. The results were also cross-checked with APOLLO2.8 calculations (Sanchez et al., 2010). A 0.3% (1 $\sigma$ ) uncertainty was considered on  $p$ , driven by the a conservative 3% uncertainty on the average delayed neutron yield  $\nu_d$  in the thermal fission of <sup>235</sup>U. A 0.3% (1 $\sigma$ ) uncertainty was taken for each  $c_{k,i}$  coefficient (=10% of the maximum deviation from unity). The  $\alpha$  ratios were taken from (Santos and Diniz, 2014) and (Perret et al., 2017).

The fitting of the ZPTF module and argument with respectively Eqs. (12) and (13) was undertaken in the CONRAD code (Archier et al., 2014), under the framework of the Bayesian inference. The fitting procedure follows a standard Generalized Least Square (GLS) method, to minimize the sum of the two chi-squares associated to each experiment, namely the following function  $\vartheta(\vec{p})$ :

$$\vartheta(\vec{p}) = \sum_{x=1}^n \left( \vec{C}_x - \vec{E}_x \right) M_{E_x}^{-1} \left( \vec{C}_x - \vec{E}_x \right)^t + \left( \vec{p}^f - \vec{p}^i \right) M_{\vec{p}^i}^{-1} \left( \vec{p}^f - \vec{p}^i \right)^t \quad (14)$$

where  $\vec{C}_x$  and  $\vec{E}_x$  are the calculated and measured values of experiment no.  $x$ ,  $M_{E_x}$  is the covariance matrix of the experiment no.  $x$ ,  $\vec{p}^i$  and  $\vec{p}^f$  are the prior and posterior estimations of parameters  $\vec{p}$ ,  $M_{\vec{p}^i}^{-1}$  is the covariance matrix of the prior values of parameters  $\vec{p}^i$ .

The normalization constant  $C$  was first fitted alone, with the relative abundances  $a_{k,i}$  fixed to the prior values from summation calculations. Parameters  $\alpha$ ,  $p$  and  $c_{k,i}$  were kept constant as well. The uncertainties on the fixed parameters were propagated on  $C$  using the Monte-Carlo marginalization technique. The posterior fitted constant  $C'$  was then fixed and imposed in the fitting of  $a_{k,i}$  alone. The uncertainty on the parameters  $\alpha$ ,  $p$ ,  $c_{k,i}$  and on the normalization  $C$  were also propagated with a Monte-Carlo marginalization method. We present in Table 3a and Table 3b the results of the relative abundances, standard deviations and correlations that we obtained. Figs. 2a, 2b, 3a, 3b illustrate the agreement of the prior and posterior fitted parameters, on the ZPTF module and phase, compared with equivalent results produced with the relative abundances from Brady & England (=ENDF/B-VIII.0 $\beta$ 4), Keepin 6-group (=JENDL-4.0) and Keepin 8-group expanded (JEFF-3.3), assuming the same  $p$  and  $c_{k,i}$  parameters, in order to emphasize the differences due to the group constants alone.

**Table 3a**  
Relative abundances and uncertainties (matrix groups 1–8: <sup>235</sup>U thermal fission; 9–16: <sup>238</sup>U fast fission).

Group $k$	$\lambda_k$	$a_{k,U5th}$	$a_{k,U8f}$
1	0.0125	0.0369 ± 0.0061	0.0085 ± 0.0015
2	0.0283	0.1357 ± 0.0247	0.0941 ± 0.0119
3	0.0425	0.0961 ± 0.0245	0.0340 ± 0.0065
4	0.1330	0.1813 ± 0.0122	0.1328 ± 0.0145
5	0.2925	0.3622 ± 0.0127	0.2674 ± 0.0251
6	0.6665	0.0812 ± 0.0105	0.2273 ± 0.0168
7	1.6348	0.0781 ± 0.0061	0.1422 ± 0.0170
8	3.5546	0.0286 ± 0.0026	0.0937 ± 0.0104
$\bar{T}_{1/2}(s)$		8.87 ± 0.15	4.97 ± 0.28

These plots clearly confirm the disagreement of Brady & England group parameters in both experiments, while Keepin 6-group and 8-group expanded data show relatively good agreement. One should notice that the average delayed neutron half-life for <sup>235</sup>U and <sup>238</sup>U, presented in Table 3, are in 1 $\sigma$  agreement with the value derived from Keepin 6-group data.

#### 4. Application of the new set of relative abundances to infer dynamic reactivities and uncertainties

We will now test the set of group constants that was obtained in the previous section to calculate the kinetics parameters  $\beta_k^{eff}$  in various LWR core benchmark situations. For this purpose, we have used the APOLLO2.8/REL2005 (Leconte et al., 2016) calculation scheme to compute the  $p$  and  $c_{k,i}$  coefficients, as defined in Eq. (1), using pin-cell type geometries with an imposed buckling to be critical. <sup>235</sup>U enrichments  $e\%$  were modified from 2 to 20%. The resulting  $p$  coefficient is ranging from 0.80 to 0.92 and the  $c_{k,i}$  coefficients remain in [-3%; +6%] around unit. The maximum deviation of  $c_{k,i}$  from unit is obtained for group 1, due to its much lower mean energy of delayed neutron spectrum (211 keV). The coefficients are slightly dependent on the fuel enrichment, due to a different adjoint-weighting of the delayed neutron group spectra.

The inhour equation is computed as presented in Eq. (1), using the linear decomposition of  $\beta_k^{eff}$  into  $a_{k,U5th}$  and  $a_{k,U8f}$  of Eq. (9). For this application, we have assumed  $\Lambda = 32\mu\text{sec}$ , as in the IPEN experiment. As the objective of this work is to evaluate the impact of delayed neutron group constant covariances, we are not considering here the uncertainty contributions from the <sup>235</sup>U and <sup>238</sup>U yields, nor from the prompt neutron generation time. We present in Fig. 4 the dynamic reactivity uncertainty obtained from our evaluation of relative abundances as compared with Keepin 8-group expanded and Keepin 6-group data, and the comparison for various situations of fuel enrichments. To interpret the differences between the different datasets, it is also instructive to compute the sensitivity coefficients of the group decay constants and relative abundances (see Fig. 4).

We observe that the fuel enrichment has a minor impact on the evaluated uncertainty. Compared with other datasets, our evaluated abundance uncertainties are leading to a relatively constant 2.5% uncertainty on the positive reactivity range and a slowly increasing uncertainty from 2.5 to 4.5% in the negative reactivity range where it is dominated by the uncertainty on groups 1 and 2, as shown in Fig. 4. We verify, as expected that Keepin 6-group data lead to the same uncertainty of 3–4% on the dynamic reactivity as using Keepin 8-group expanded data in the positive reactivity range. However, in the negative reactivity range, a strong disagreement occurs between the 8-group expanded and the 6-group data. This is due to the difference of uncertainty on the common first group associated to <sup>87</sup>Br, where the Keepin 6-group dataset includes a 2.3% uncertainty in the decay constant  $\lambda_1$  while

**Table 3b**

Correlation matrix for relative abundances (matrix groups 1–8:  $^{235}\text{U}$  thermal fission; 9–16:  $^{238}\text{U}$  fast fission).

1.000	-0.756	0.551	-0.046	-0.159	0.285	-0.261	0.086	0.023	-0.028	0.046	-0.014	0.046	-0.013	-0.008	-0.056
-0.756	1.000	-0.951	0.376	0.066	-0.383	0.336	-0.066	-0.023	0.035	-0.043	0.018	-0.034	0.013	-0.023	0.063
0.551	-0.951	1.000	-0.564	0.070	0.318	-0.324	0.061	0.019	-0.062	0.035	-0.021	0.038	-0.005	0.029	-0.056
-0.046	0.376	-0.564	1.000	-0.772	0.230	0.050	-0.074	0.000	0.009	0.003	-0.044	0.021	0.029	-0.039	0.015
-0.159	0.066	0.070	-0.772	1.000	-0.714	0.285	0.019	-0.008	0.038	-0.033	0.042	-0.082	0.007	0.061	0.009
0.285	-0.383	0.318	0.230	-0.714	1.000	-0.737	0.089	0.000	0.003	-0.027	0.031	-0.061	-0.048	0.076	0.068
-0.261	0.336	-0.324	0.050	0.285	-0.737	1.000	-0.530	0.024	0.036	0.076	-0.015	0.136	-0.016	-0.174	-0.088
0.086	-0.066	0.061	-0.074	0.019	0.089	-0.530	1.000	0.027	-0.004	0.056	-0.022	0.083	0.015	-0.059	-0.128
0.023	-0.023	0.019	0.000	-0.008	0.000	0.024	0.027	1.000	-0.026	0.121	-0.104	0.098	-0.064	-0.071	-0.051
-0.028	0.035	-0.062	0.009	0.038	0.003	0.036	-0.004	-0.026	1.000	-0.275	0.153	-0.381	-0.176	-0.009	0.035
0.046	-0.043	0.035	0.003	-0.033	-0.027	0.076	0.056	0.121	-0.275	1.000	-0.385	0.401	-0.127	-0.217	-0.197
-0.014	0.018	-0.021	-0.044	0.042	0.031	-0.015	-0.022	-0.104	0.153	-0.385	1.000	-0.561	-0.119	0.078	0.104
0.046	-0.034	0.038	0.021	-0.082	-0.061	0.136	0.083	0.098	-0.381	0.401	-0.561	1.000	-0.340	-0.421	-0.218
-0.013	0.013	-0.005	0.029	0.007	-0.048	-0.016	0.015	-0.064	-0.176	-0.127	-0.119	-0.340	1.000	-0.181	-0.049
-0.008	-0.023	0.029	-0.039	0.061	0.076	-0.174	-0.059	-0.071	-0.009	-0.217	0.078	-0.421	-0.181	1.000	-0.276
-0.056	0.063	-0.056	0.015	0.009	0.068	-0.088	-0.128	-0.051	0.035	-0.197	0.104	-0.218	-0.049	-0.276	1.000

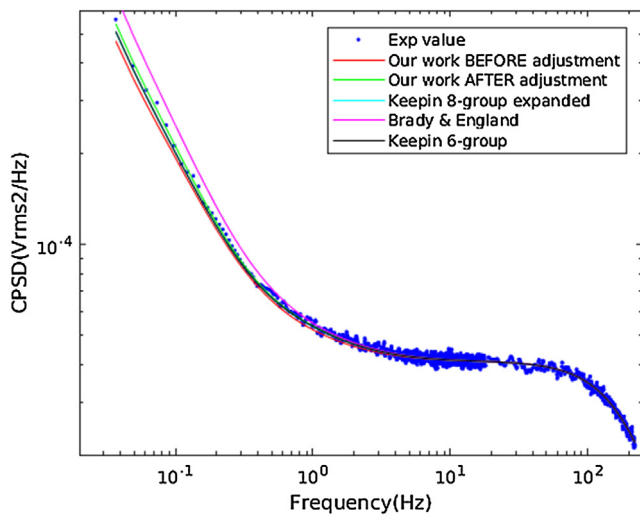


Fig. 2a. ZPTF module (IPEN).

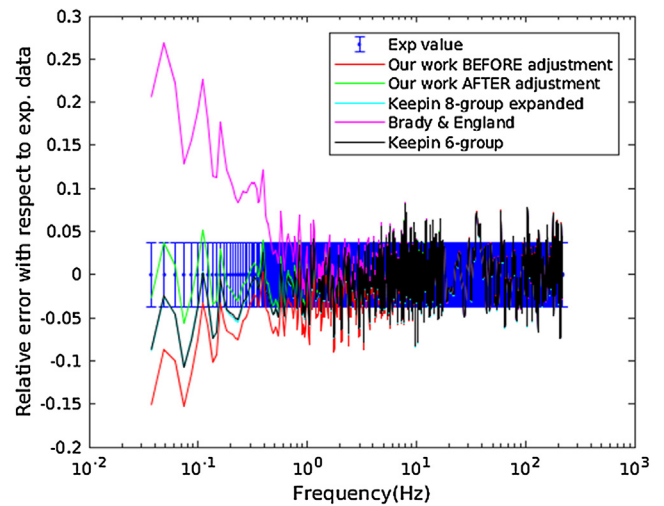


Fig. 3a. Error between theoretical and measured ZPTF module (IPEN).

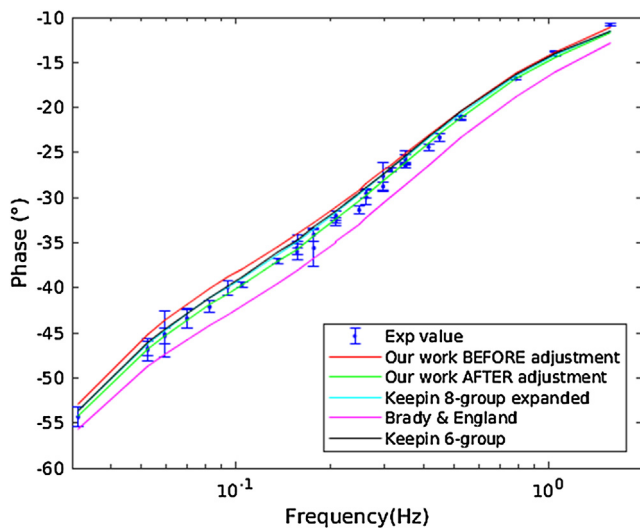


Fig. 2b. ZPTF phase (MINERVE).

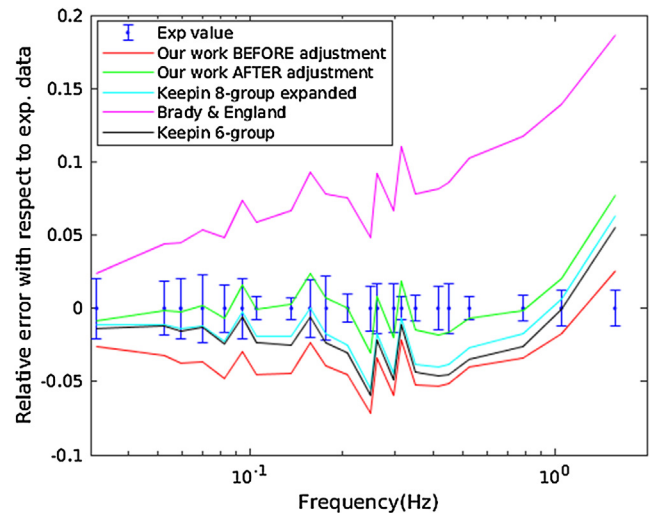


Fig. 3b. Error between theoretical and measured ZPTF phase (MINERVE).

Keepin expanded 8-group consider fixed decay constants without uncertainty, with the addition of a very strong sensitivity coefficient, as seen in Fig. 5. In the Keepin 8-group expanded data, the Spriggs method was applied to keep the consistency of reactivities

and uncertainties in the positive but not in the negative reactivity range. As a consequence, the uncertainty on  $\lambda_1$  was improperly transferred on the first group relative abundance, while its sensitivity in the positive reactivity range is low. Therefore such approach misses an important degree of freedom occurring in

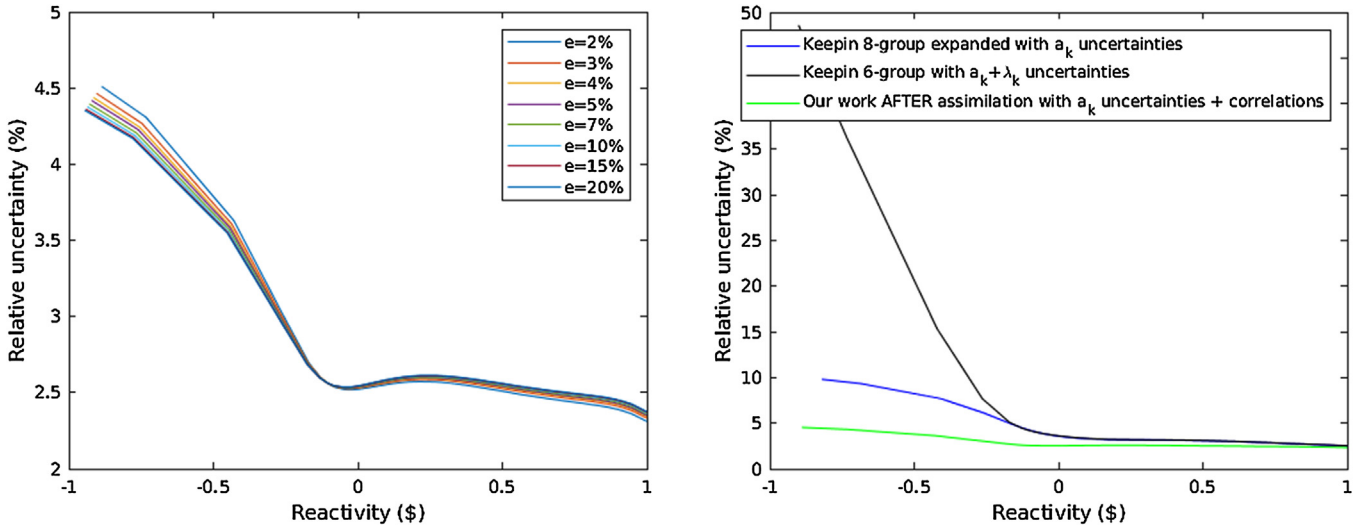


Fig. 4. Uncertainty on the dynamic reactivity from our evaluated relative abundances, for various fuel enrichments (left side) and compared to various libraries (right side, fuel enrichment  $e = 4\%$ ).

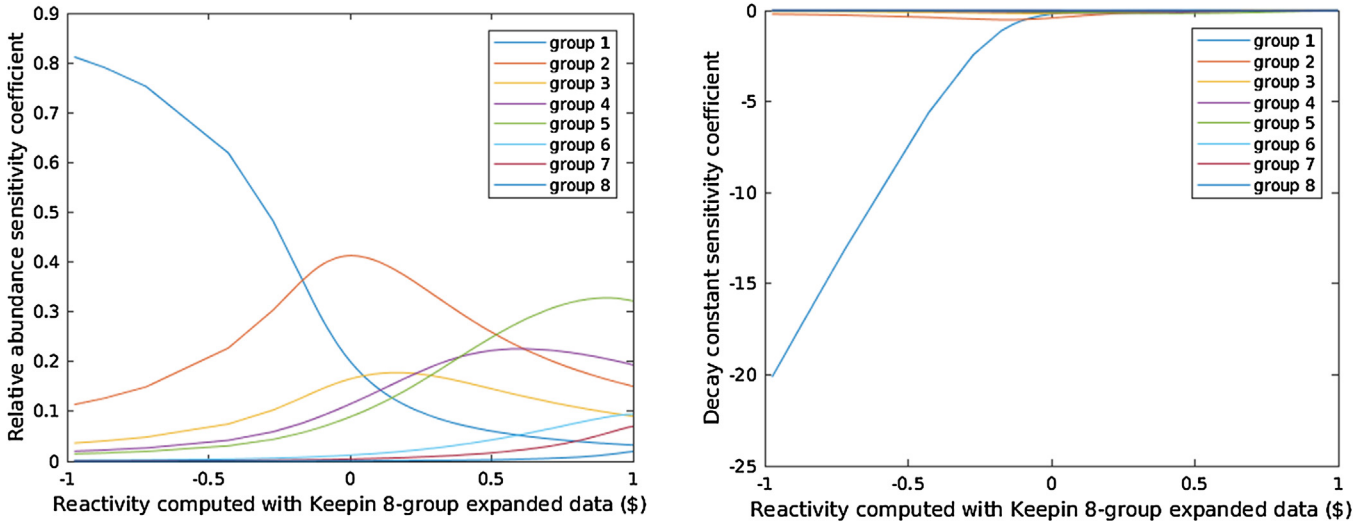


Fig. 5. Dynamic reactivity sensitivity coefficients due to  $a_k$  (left side) and  $\lambda_k$  (right side).

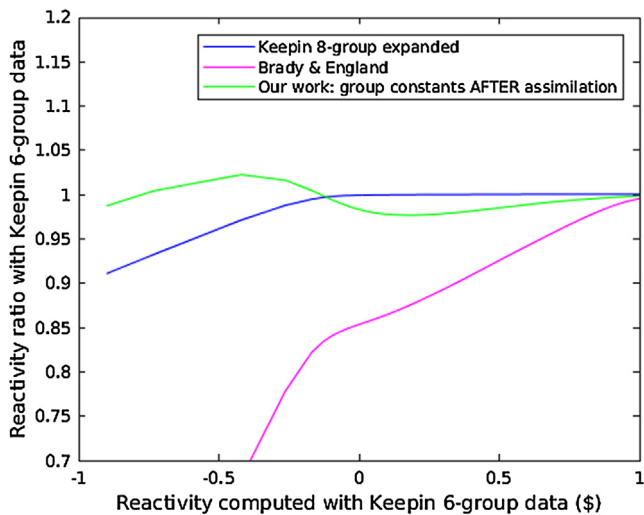


Fig. 6. Dynamic reactivity estimations from various datasets.

the initial fit of Keepin that does not exist in the Spriggs model, so we may conclude that the 8-group expanded data are not very reliable in the negative reactivity range. This is confirmed also when comparing the reactivity estimations from the various datasets, as plotted in Fig. 6. The Keepin 8-group expanded data is in <0.5% agreement with Keepin 6-group data for  $\rho$  greater than 0, while it tends to almost 10% difference for  $\rho$  close to  $-1$ \$. Our evaluation is performing well with respect to Keepin 6-group data, with a maximum difference of  $-2\%$  to  $+2\%$  over the whole reactivity range.

**5. Conclusions**

With the combination of summation calculation of relative abundances, based on a more recent re-evaluation of microscopic data of individual precursors than the work of Brady & England, with two independent integral experiments for the measurement of the ZPTF, we have produced new evaluations of delayed neutron relative abundances for the thermal fission of  $^{235}\text{U}$  and fast fission of  $^{238}\text{U}$ . We have estimated the uncertainties and correlations



between group parameters, as well as between the two isotopes, within the framework of the Bayesian inference, with the inclusion of normalization and correction factor uncertainties through a Monte-Carlo marginalization technique. The resulting dynamic reactivity is shown to be in better agreement than 2% with the one computed from the reference Keepin 6-group data, while Keepin 8-group expanded data are not consistent with the original data in the negative reactivity range. Our recommended uncertainty on the reactivity estimation is 2.5% ( $1\sigma$ ) in  $0 < \rho < 1$  [\$] and 2.5–4.5% ( $1\sigma$ ) in  $-1 < \rho < 0$  [\$], for U fuel enrichments covering the range from 2 to 20%.

This work is the first step of an effort towards a more reliable evaluation of delayed neutron data and associated covariances. Future work will include on one side more up-to-date data of fission yields (JEFF-3.3) and radioactive decay data recommended by the IAEA/CRP group on delayed neutron data, and new reactor experiments of neutron noise spectral distributions, currently being measured in the EOLE and MINERVE facilities. For a significant reduction of group parameter uncertainties, we are also considering new delayed neutron activity measurements, to be performed in the near future on the three main isotopes  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{239}\text{Pu}$ , so that a rigorous uncertainty analysis will be possible to overcome the limitations of the Spriggs method. Comparison of dynamic reactivity estimations from reactor period and reactivity equivalence method will be also useful to validate the proposed datasets of delayed neutron abundances.

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## Further reading

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