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# The application of the multiple transient technique for the experimental determination of the relative abundances and decay constants of delayed neutrons of the IPEN/MB-01 reactor

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**Abstract**

An in-pile experiment for the determination of the relative abundances and decay constants of delayed neutrons has been successfully performed at the IPEN/MB-01 research reactor facility. The experimental data are of good quality and can be used to validate theoretical predictions of the delayed neutron group constants based on the current knowledge of the fission products yields and emission probabilities for known precursors. The theory/experiment comparison shows that the current release of ENDF/B-VI, namely release 8, shows severe discrepancies in both relative abundances and in its first decay constant. The version revised at LANL shows very good progress in both aspects. JENDL3.3 shows the best performance in the C/E comparison. One of the main achievements of the experiment was the consistency of the measured first decay constant to that of $^{87}$Br. Finally, it is also shown preliminary experimental results for an eight-group model.

## 1. Introduction

Delayed neutrons play an important role on the reactor physics field. Even though comprising less than 1% of the total neutrons from fission, they are very important in the safety area such as in the accident analysis and in the conversion of period into reactivity. In the event of the fission reaction chain there are many fission products (approximately 270) which can be considered potential emitters of delayed neutrons. However, an experimental characterization of all these emitters is very difficult due to their very low yield and/or low half-lives and also due to their very complex transmutation chain. However, it is possible to determine experimentally the aggregate behavior and generate a few-group model where the decay constants and abundances are mean values of various emitters with similar decay constant.

There are several experimental procedures to determine the delayed neutron parameters (Sprigss, 1993) and they are generally classified as "in-pile" and "out-of-pile" experiments. The so called "in-pile" experiments are performed in the multiplying system where there is no separation of the prompt and delayed neutrons. For the point reactor model this category of experiments are very important because they can provide valuable experimental information related to the effective delayed neutron parameters used in such a model. Such effective delayed neutron parameters are system-dependent.

To date, there are five techniques to perform in-pile experiments: the rod-drop method; the small negative-reactivity insertion; the reactivity oscillator; the multiple transient method (see Sprigss, 1993 for some details on the first three techniques); and the reactor-noise analysis in the frequency domain (Diniz and Dos Santos, 2006). However, it seems that only the two last techniques are still being used to obtain these important delayed neutron kinetic parameters. With the exception of a work by Vilim and Brock (1996), all published work involving the first three techniques are of the fifties, sixties and seventies.

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The available experimental support applied to the effective parameters of delayed neutrons to be used in the point reactor model is scarce and in many cases the utilization of the data of such experiments is not so straightforward and very well established. The main purpose of this work is an attempt to fulfill this need for a thermal reactor application. For such a purpose the in-pile experiment performed at the IPEN/MB-01 reactor for the determination of its relative abundances and decay constants of delayed neutrons will be presented and the adequacy of the nuclear data related to the delayed neutrons of several nuclear data libraries will be verified and discussed.

The experimental procedure employs the multiple transient technique developed by Sprigss (1993). The IPEN/MB-01 research reactor facility consists of a 28 × 26 array of $UO_2$ fuel rods, 4.3% enriched and clad by stainless steel (type 304) inside of a light water tank, with a maximum power level of 100 W. A complete description of the IPEN/MB-01 reactor can be found in Dos Santos et al. (1999) and Dos Santos et al. (2004). The theoretical analyses will consider the verification of the adequacy of the nuclear data of several libraries such as ENDF/B-VI.8 (BNL), ENDF/B-VI.8-LANL revision UEVAL,[1] and JENDL-3.3 (Shibata et al., 2002).

## 2. The Spriggs' method

The multiple transient technique developed by Spriggs is based on a least-squares-fitting algorithm that simultaneously fits a series of transients produced by small reactivity perturbations of arbitrary size. The function that is least-squares fitted is the analytic solution for a step change in reactivity as given by the point reactor model for an arbitrary number of delayed neutron groups. The Spriggs' method is based solely on the measurable quantities of relative power, time, and one of the roots of the Inhour equation. The number of delayed neutron groups that can be resolved is constrained only by the quality and range of the transient data used in the fit.

### 2.1. Theoretical background of the Sprigss' method

The main assumption of the Spriggs' method is the validity of the point reactor model. In the special case of no reactivity feedback, initially critical system, the solution of the point kinetics model, following a step change of reactivity at $t = 0$, can be written as

$$\frac{n - n_\mathrm{b}}{n_0 - n_\mathrm{b}} = \sum_j \frac{\omega_\alpha \Lambda + \sum_i \frac{\omega_\alpha \beta_i}{\omega_\alpha + \lambda_i}}{\omega_j \left[\Lambda + \sum_i \frac{\beta_i \lambda_i}{(\omega_j + \lambda_i)^2}\right]} \mathrm{e}^{\omega_j t}, \qquad (1)$$

where $n$ = power level at time $t$; $n_0$ = initial power level; $n_\mathrm{b}$ = background power level associated with intrinsic/external neutron sources; $\omega_j$ = $j$th root of the Inhour equation; $\Lambda$ = prompt neutron generation time; $\lambda_i$ = decay constant of the $i$th precursor group; $\beta_i$ = delayed fraction of the $i$th precursor group; $\omega_\alpha$ is either the asymptotic inverse period or the prompt neutron-decay-period; a measured quantity in the experiment.

Eq. (1) states that the relative power is a function of two independent variables (time and one of measurable roots of the Inhour equation) and $2m + 1$ parameters: $\beta_1, \beta_2, \beta_3, \ldots, \beta_m, \lambda_1, \lambda_2, \lambda_3, \ldots, \lambda_m$, and $\Lambda$. Given a set of delayed neutron parameters, a neutron lifetime and one root of the Inhour equation, one can readily obtain the remainder roots ($\omega_j$). Eq. (1) is the fundamental equation for the Spriggs' method. Given a specific number of transients produced by step changes in reactivity, the $2m + 1$ parameters, interpreted as unknown parameters, can be found by simultaneously fitting the relative power as function of time into Eq. (1) for all transients. It must be noted that there is no need of previous knowledge of the reactivity for each transient. Reactivity is a calculated quantity based on the measured root of the Inhour equation and a given set of delayed neutron parameters.

The least-squares-fitting scheme is shown in Fig. 1 (Sprigss, 1993). Basically, the scheme starts with the input data:

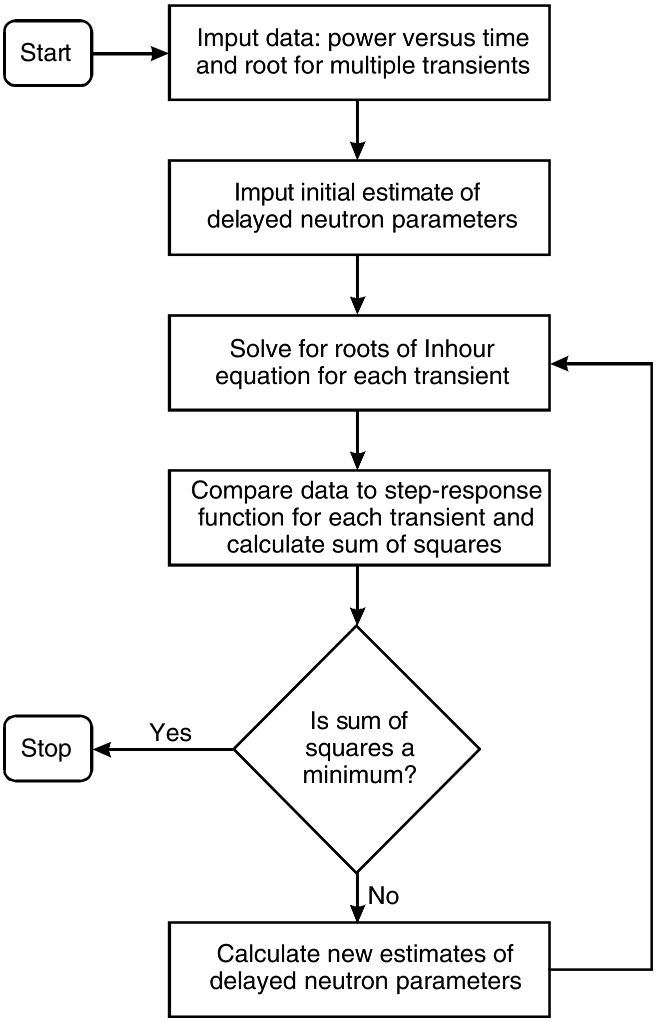

Fig. 1. Least-squares-fitting scheme applied in the multiple transient technique.

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[1] UEVAL home page, http://www.nea.fr/lists/ueval.

the relative power versus time and its corresponding measured root of the Inhour equation for each transient, and an initial estimate of the delayed neutron parameters. The next steps are iterative. For each iteration, the remaining roots of the Inhour equation are calculated and a basic non-linear least-square approach is employed. New estimates of the delayed neutron parameters are determined as well as the sum of squares of the step-response function for the whole set of the transients. The process is continued until the sum of squares of the deviations of the step-response function is minimized.

## 3. The experiment performed at the IPEN/MB-01 reactor

The standard configuration of the IPEN/MB-01 reactor core was employed for all the experimental work performed in this work. The perturbations to produce the transients were induced at the central position of the reactor core. An empty tube of SS-304 of the same diameter and thickness of the fuel rods but very high was employed for this purpose. A stopper was placed in the interior of this tube to accommodate a small sample of Ag–In–Cd (80% Ag, 15% In, and 5% Cd). The stopper was designed such that it could have its height adjusted so that the sample reactivity could be changed. The sample was placed very close to the central position of the active core in order to optimize its reactivity. The transients were produced by removing rapidly the sample from the active core.

According to Spriggs there are four key experimental conditions for the successful execution of the experiment: (a) a very fast sample removal system (removal time less than 20 ms), (b) a very fast acquisition system, (c) accurate determination of the initial point, i.e., the time beyond which the solution of the relative power is a sum of exponential functions and (d) accurate measurement of one root of the Inhour equation. The IPEN/MB-01 removal system consists of a high speed electric motor coupled with a beam catcher. The measured removal time is of the order of 6 ms ($10^{-3}$ s) which is more than adequate for the experimental purposes of this work. Therefore condition (a) is satisfactory attended.

The experiment has several features that must be contemplated by the acquisition system. Initially, when the sample is removed from the core there is a very fast change in the relative power due to the rapid decay of the largest (negative) root of the Inhour equation. After that the relative power change is less pronounced but still very important for the determination of the relative abundance and decay constants of delayed neutrons. Therefore, the experimental acquisition system has to be fast enough to describe all the physics details of the transient (conditions (b) and (c)). The experimental set up for the data collection for this work is composed of two compensated ionization chambers working in current mode and located in the reflector, approximately 11 cm away from the first fuel rods. The signals from these chambers are fed into the electrometers where there is a conversion from current to voltage (0–2 V). After that, the data are input into a data acquisition system which was set to a sample rate of 1 kHz which guarantees a very high acquisition rate. At every 1 ms one experimental data is collected. This last aspect is very important to describe the initial points of the transient mainly for the determination of the starting point ($t = 0$) of the transient. The starting point is the time beyond which the data can be described by a sum of exponential functions. The fourth condition is the measurement of one root of the Inhour equation. Also, here the experiment performed at the IPEN/MB-01 reactor addressed this aspect adequately by collecting the data in a long run to let the terms of all negative roots do decay to nearly zero.

The experiment basically can be described as follows: initially the reactor is criticalized at 1 W with the sample inside of the core. The automatic control system is turned off and the reactor is run in the manual mode. The IPEN/MB-01 reactor possesses a very accurate mechanism for the relative control rod positioning (Dos Santos et al., 1999). Hence, an experienced operator positions the control rod such that the reactivity is a few cents from the true critical condition. This aspect is very important for the experiment and will guarantee that in the initial condition the reactor is really pretty close to the criticality. Following the experimental procedure, the whole system is stabilized .The data collection system is then activated and the detector currents which are proportional to the reactor power are writing in a computer file for subsequent power normalization. These data are collected for at least 5 min before the removal of the sample from the core. This aspect will guarantee a proper power normalization before the transient starts. The sample is then removed from the reactor core and the transient starts. The data are acquired in a length of time long enough such that all the events necessary for the analyses are contemplated. Later on, the experimental data are normalized taking into consideration the power before the transient and the final result is written in a computer file. Fig. 2 shows an example of the experimental data collected in this work.

Fig. 2 shows clearly the fast change of the relative power density at the beginning of the transient as well as the region where the behavior becomes asymptotic which allows the determination of the first root of the Inhour equation ($\omega_\alpha$). A total of 52 transients ranging from approximately 40 to 110 pcm were produced in this work. They constitute the experimental data basis for the delayed neutron parameters determination.

### 3.1. The experimental results

The least-squares-fitting approach considered in this work assumes six and eight groups of delayed neutrons. In all cases, the neutron generation time was fixed to 32 µs. This value was obtained experimentally in a previous work (Dos Santos et al., Physor 2004). The sensitivity of the relative abundances and the decay constants to variations of $\Lambda$ by a direct approach has been analysed and

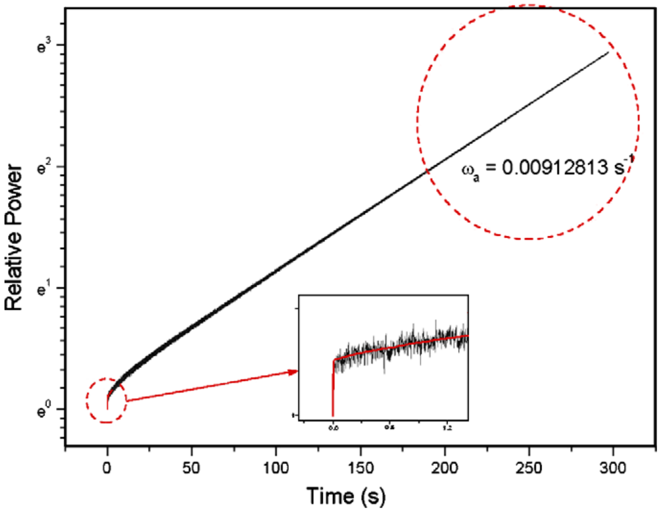

Fig. 2. Example of a transient for the effective delayed neutron parameters determination.

studied and it was found to be very low. In fact, the sensitivity of $\beta_i/\beta_{\text{eff}}$ can be neglected, while that of $\lambda_i$ presents in general a 1/10 ratio, i.e., a change of 5% on $\Lambda$ gives a maximum deviation on $\lambda_i$ of only 0.5%. These results make the assumption made here adequate for the purposes of this work.

Due to the complexity of the comparison between theory and experiment, this work will consider a simplified approach in the least-square fitting process. Each nuclear data library has its own set of decay constants which depends on the fissionable nuclide. The only exception here is the revised version of ENDF/B-VI performed at Los Alamos which possesses the same set of decay constants for $^{235}$U and $^{238}$U. The procedure adopted in this work to make feasible the comparison between theory and experiment will be to fix the decay constants of the major fissionable nuclide from the library under consideration and to fit the relative abundances. By major is meant the nuclide that most contribute to the total fission rate. For the specific case of the IPEN/MB-01 reactor $^{235}$U is by far the most important fissile nuclide since it contributes to about 97% of the total fission density. Therefore, the choice of the decay constants of $^{235}$U will not impose a very heavy restriction to the analyses of the problem and will make the comparison between theory and experiment feasible. The fitting of the decay constants was also performed in a six-group model but in this case it was considered an iterative approach. First, the relative abundances are fixed and the decay constants are fitted. Then, the process is inverted and the relative abundances are fitted. The process is repeated until convergence is achieved. The only fixed parameter was the decay constant of the sixth group, the group of smallest half-life.

Table 1 shows the decay constants arising from the iterative approach, the relative abundances for the decay constants of the ENDF/B.8 (LANL Revision), both in a six-group model, and the measured relative abundances for an eight-group model. In this last case, it was considered the decay constants from Spriggs et al. (2000). The 52 transients were simultaneously analyzed using the least-squares-fitting scheme previously described. The intention here is to show the degree of accuracy of the measured parameters as well as to show that the first decay constant from the in-pile experiment is consistent to that of $^{87}$Br. This aspect demonstrates the consistency and the accuracy of the experiment performed at the IPEN/MB-01 reactor. In a general way, Table 1 shows that the uncertainties increase as the decay constants of the delayed neutron group increase. This aspect is very predictable since the sensitivity of the relative power to the delayed neutron parameters decreases as the group half-live decreases. Also, Table 1 shows that the uncertainties for the six-group model is quite adequate to verify the applicability of any nuclear data library. In the eight-group model the uncertainty increases drastically due to the higher number of parameters to be resolved in the least-squares approach.

Table 1
Effective delayed neutron parameters of the IPEN/MB-01 reactor

| Group | Decay constant (s$^{-1}$) | $\beta_i/\beta_{\text{eff}}$ | $\beta_i/\beta_{\text{eff}}$ |
|---|---|---|---|
| 1 | 1.2456E−02 (0.25%) | 0.0510 (2.95%) | 0.0462 (3.76%) |
| 2 | 3.2738E−02 (0.34%) | 0.1908 (2.97%) | 0.0940 (4.56%) |
| 3 | 1.2084E−01 (0.58%) | 0.2061 (2.97%) | 0.1307 (4.61%) |
| 4 | 3.2814E−01 (0.61%) | 0.3305 (3.05%) | 0.1642 (4.22%) |
| 5 | 1.7971E+00 (4.45%) | 0.1537 (4.21%) | 0.3430 (3.83%) |
| 6 | 3.87 (fixed) | 0.0679 (5.00%) | 0.092 (15.43%) |
| 7 | | | 0.1030 (11.32%) |
| 8 | | | 0.0275 (22.95%) |

With regard the first decay constant, it should be stressed here that the ENDF/B-IV.8 library overestimates the value of this parameter. The comparison with the values presented by its revised version at LANL, the JENDL 3.3 and the results of this work, shows a discrepancy of about 7%. This fact can lead to very severe restrictions in reactivity calculations, as shown by Sprigss (1993) and Diniz and Dos Santos (2006).

## 4. The theory/experiment comparison

Following a standard approach, it can be shown (Bell and Glasstone, 1970) that the effective delayed neutron fraction for the delayed neutron group $j$ ($\beta_{\text{ef}_j}$) can be found as

$$\beta_{\text{ef}_j} = \frac{1}{F}\int \cdots \int \chi_{d_j}(E)\beta_j \nu \Sigma_f(r,E')\phi(r,\Omega',E')\phi^*(r,\Omega,E) \times \mathrm{d}r\,\mathrm{d}\Omega'\,\mathrm{d}E'\,\mathrm{d}\Omega\,\mathrm{d}E, \tag{2}$$

where the symbols follow the same meaning as in Bell and Glasstone (1970).

The relative effective abundances of delayed neutrons in group $j$ is obtained as $\beta_{\text{ef}_j}/\beta_{\text{eff}}$, where $\beta_{\text{eff}}$ is the sum of $\beta_{\text{ef}_j}$ for all groups $j$. This is the quantity to be compared to the experimental values. Compared to another reactor physics responses such as $k_{\text{eff}}$ and reaction rates that can employ a very powerful and nearly exact capability of MCNP-4C

(Briesmeister, 1993), the effective delayed neutron parameters do not have a proven and well stabilized calculational methodology that can reduce the uncertainty in the reactor modeling and in the solution of the neutron transport equation to a value smaller than that inherent in the nuclear data libraries used in the analyses. Consequently, in the theory/experiment comparisons the discrepancies of the calculational quantities will not be due solely to the nuclear data library used in the process but it will carry also some part due to the methodology itself. The approach used here will be twofold. Initially, the commonly methodology used at IPEN for the reactor analysis in general based in the coupled systems HAMMER-TECHNION (Barhen et al., 1978) for the cross section generation and weighting and CITATION (Fowler et al., 1971) for neutron diffusion in the reactor will be employed for the theoretical analysis of the experiment. In an attempt to make a methodology based on transport theory, this work will also employ the coupled NJOY/AMPX-II/TORT (Rhoades and Simpson, 1991) systems for the determination of the effective delayed neutron parameters. The advantage of TORT (Dos Santos et al., Physor 2002) approach, as already said, is the solution of the neutron transport equation (forward and adjoint) as well as the flexibility to use several neutron groups. The nuclear data libraries used in this work are the ENDF/B-VI.8, ENDF/B-VI (LANL review) and JENDL3.3.

A characteristic of all nuclear data libraries used in this work is that all have the same delayed neutron spectrum which is originally from ENDF/B-VI.0. The older six-group spectra are used with the new six-group parameters, regardless of the inconsistency of the older and newer group decay constants. Only ENDF/B-VI.8 has a consistent set of decay constants. This aspect, which is beyond of the scope of this work, may impose some sort of systematic bias in the comparison of theory and experiment.

Table 2 compares the effective and the effective relative delayed neutron abundances by both methodologies for the ENDF/B-VI (LANL review) case. TORT was run with 4 and 16 groups and with quadrature order 2 and 16. As

Table 2
Calculated effective parameters for ENDF/B-VI (LANL review) library

| Effective parameters | CITATION | TORT | | |
|---|---|---|---|---|
| | 4 groups | 4 groups – $S_2$ | 4 groups – $S_{16}$ | 16 groups – $S_{16}$ |
| $\beta_{\text{eff}}$ | $7.80130 \times 10^{-3}$ | $7.79916 \times 10^{-3}$ | $7.73725 \times 10^{-3}$ | $7.92376 \times 10^{-3}$ |
| $\beta_1/\beta_{\text{eff}}$ | $4.02682 \times 10^{-2}$ | $3.53992 \times 10^{-2}$ | $3.53746 \times 10^{-2}$ | $3.55385 \times 10^{-2}$ |
| $\beta_2/\beta_{\text{eff}}$ | $1.83515 \times 10^{-1}$ | $1.84562 \times 10^{-1}$ | $1.84516 \times 10^{-1}$ | $1.84774 \times 10^{-1}$ |
| $\beta_3/\beta_{\text{eff}}$ | $1.60349 \times 10^{-1}$ | $1.61397 \times 10^{-1}$ | $1.61234 \times 10^{-1}$ | $1.60446 \times 10^{-1}$ |
| $\beta_4/\beta_{\text{eff}}$ | $4.01761 \times 10^{-1}$ | $4.03431 \times 10^{-1}$ | $4.03651 \times 10^{-1}$ | $4.03917 \times 10^{-1}$ |
| $\beta_5/\beta_{\text{eff}}$ | $1.52425 \times 10^{-1}$ | $1.53233 \times 10^{-1}$ | $1.53234 \times 10^{-1}$ | $1.53282 \times 10^{-1}$ |
| $\beta_6/\beta_{\text{eff}}$ | $6.16881 \times 10^{-2}$ | $6.19781 \times 10^{-2}$ | $6.19901 \times 10^{-2}$ | $6.20434 \times 10^{-2}$ |

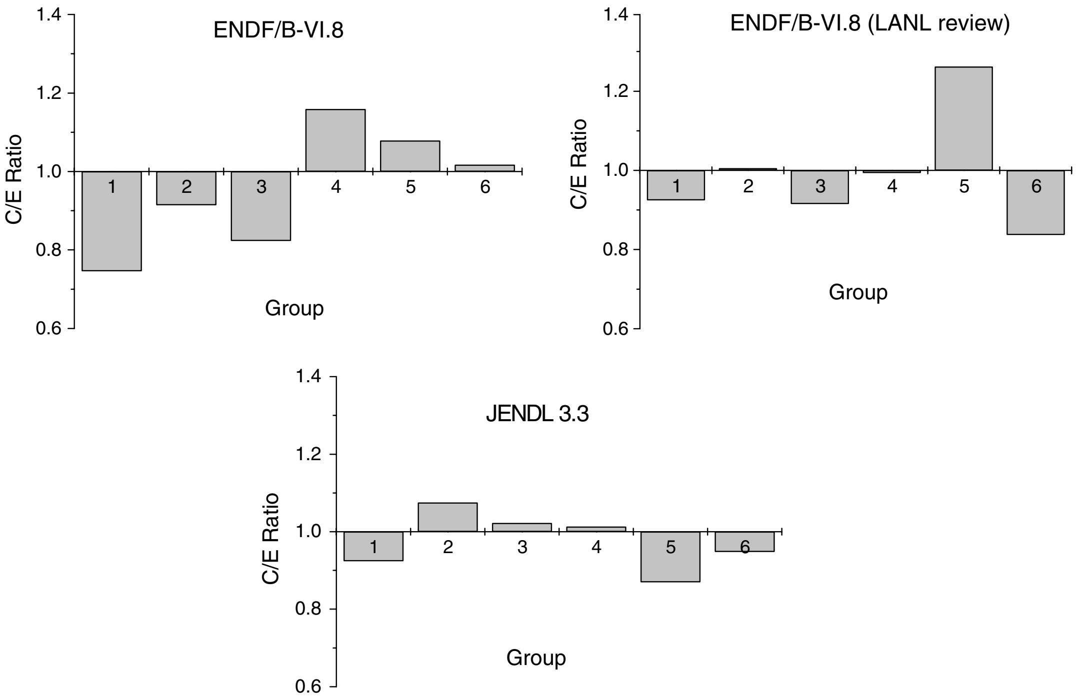

Fig. 3. Comparison theory/experiment for the relative abundances.

shown in Table 2, the effect of the number of groups and the $S_N$ order on the relative abundances is quite small. There is an important effect on $\beta_{\mathrm{eff}}$ as the number of groups increases. Comparing the CITATION values to those of TORT, there is only a significant difference in group 1. The other groups show only a slight difference.

The comparison of the calculated relative abundances to the experimental values is shown in Fig. 3. The calculated values are from CITATION. Note now that the decay constants for each library were kept fixed during the least-squares procedure.

Fig. 3 shows that there is a systematic underprediction of the calculated relative abundance for the first group. ENDF/B-VI.8 shows the worst performance with deviations as high as 26% for the first precursor group. On the other hand, in a global sense, the LANL revision of ENDF/B-VI shows a relatively good progress and the agreement to the experimental values can be considered reasonable, with exception of the fifth group which shows high deviation. The best performance is due to JENDL3.3 which shows a really remarkable comparison.

Finally, it should be stressed that the theory/experiment comparison of the delayed neutron parameters performed in this work is not equal to the several others comparisons performed in the reactor physics area. The analyses are valid only for the nuclear data libraries considered here. The analyses of a new library require the whole process shown in this work to be repeated. This imposes some difficulties to establish experimental benchmarks to validate effective relative abundances and decay constants of delayed neutrons simply because the whole set of experimental data (the 52 transient data in the case under consideration) plus the computer programs used to fit the experimental data have to be made available to the nuclear community. This is an aspect to be kept in mind and addressed in future evaluations of benchmark problems related specifically to the kind of the delayed neutron nuclear data addressed in this work.

## 5. Conclusions

The experiment for the determination of the relative abundances and decay constants of delayed neutrons of the IPEN/MB-01 reactor has been successfully performed. The sample removal system was designed and tested satisfactorily, and the measured removal time was around 6 ms which is more than adequate for the purposes of the experiment. The experimental data basis consisted of 52 transients which were simultaneously fitted to extract the relative abundances and decay constants. The final fitted results are of good quality and can be very helpful to validate theoretical predictions of the delayed neutron constants based on the current knowledge of fission yields and emission probabilities for known precursors. Furthermore, the first decay constant obtained experimentally in a six-group model, shows total consistency to that of $^{87}$B. The comparison between theory and experiment performed in this work shows that the current release of ENDF/B-VI, namely release 8, has the worst performance. Not only its relative abundances of delayed neutron show severe discrepancies but also its first decay constant is overpredicted which imposes severe restriction on the determination of the reactivity. The revised version of ENDF/B-VI performed at LANL shows very good progress and also its first decay constant is very close to the one determined in this work. The best performance is obtained by JENDL3.3 which shows excellent agreement in all types of comparison performed in this work.

The utilization of the same delayed neutron spectrum in all nuclear data files used in this work is a point that should be addressed in future evaluations of ENDF/B and JENDL files. The inconsistency of the older (ENDF/B-VI.0) and newer group decay constants in the case of JENDL3.3 and in the ENDF/B-VI revision performed at Los Alamos may introduce some sort of systematic bias. Whether or not the utilization of the same delayed neutron spectrum represents an important bearing remains an unknown question.

It is important to mention here that the multiple transient technique presents some difficulties to resolve the short-lived precursor groups, namely groups 5 and 6. Firstly, this occurs because the high fluctuation of the data at the beginning of the transient due to the low power level of the reactor when the experiment begins (only 1 W) and therefore, the low currents supplied by the ionization chambers. Also, the number of data points in this region seems not to be enough to give good results through the least-squares approach.

To overcome the first restriction, the increase of the maximum power of the IPEN/MB-01 reactor is already in course. In this way, the power level at the beginning of the experiment will be high enough so that the currents of the ionization chambers present less fluctuation. In the case of the second problem, a higher data acquisition rate than 1 kHz, say 10 kHz, is under consideration for further experiments.

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