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Measurement of the delayed-neutron multiplicity and time constants in the thermal neutron induced fission of ^{235}U at ILL

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Abstract. Delayed-neutron (DN) data is essential in inherent reactor safety and reactor control since it is needed for the estimation of the reactivity. Nowadays, discrepancies among the data in various international databases (JEFF, ENDF, JENDL) are large and bring excessive conservatism in the safety margins. The ALDEN (Average Lifetime of DElayed Neutrons) experiment, built in a collaboration between CEA and CNRS, aimed at re-measuring the DN data associated with several fissioning systems (average delayed-neutron yield and kinetic parameters). The first experimental campaign consisted in the integral measurement of the DN activity after the irradiation of an ^{235}U target. It took place under the cold neutron flux of ILL (Institut Laue-Langevin) at the beginning of September 2018, in the PF1b experimental zone (doi:10.1016/j.nima.2006.03.020). The data analysis gave an average DN yield of 1.631E-02(2) DN/fiss and a mean precursors' half-life of 8.93(9) s. The results are consistent with the literature, but they are affected by one third of the uncertainty.

1 Introduction

Right after the fission, fission products are created together with the emission of 2 or 3 prompt neutrons. These neutron-rich fission products are naturally unstable and try to reach stability through a series of β^--decays. If, after the decay, the excitation energy of the daughter is larger than the separation energy of the last neutron, the daughter can reach stability through the emission of a neutron. This second decay is instantaneous so that the neutron is considered to be emitted by the father of the emitter, the precursor. The neutron is called delayed because it appears some time after the fission took place, and its delay only depends on the half-life of the precursor. It is common practice to sort the precursors into 6 or 8 groups, each characterized by a specific abundance a_i and decay constant λ_i. Figure 1 illustrates the delayed-neutron emission process for one of the main precursors, ^{137}I. Delayed neutrons are important in many sectors: nuclear structure studies, astrophysical applications, determination of nuclear content, etc. The main use of delayed-neutron data is, however, the determination of the reactivity for reactor physics applications. The reactivity is an essential quantity in reactor control because it is a measure of the neutron balance. One way to determine it is to apply the Nordheim equation (Eq. 1) after the measurement of the reactor period τ.

$$\rho = \frac{\Lambda}{\tau} + \beta_{eff} \sum_{i=1}^{8} \frac{a_i}{\lambda_i \tau + 1} \quad (1)$$

In Eq. 1, Λ is the prompt neutron generation time, β_{eff} the effective delayed-neutron fraction, a_i and λ_i the abundance and the decay constant of the delayed-neutron group i.

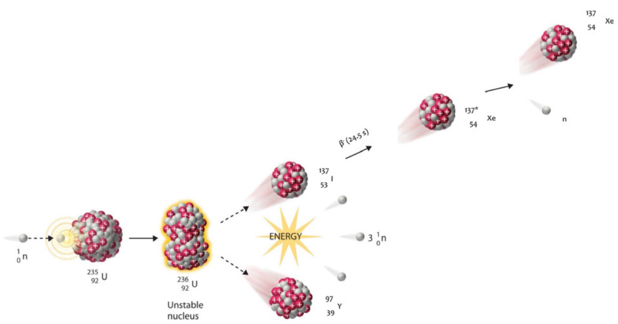

Figure 1: Illustration of the delayed-neutron emission from ^{137}I

To use the Nordheim equation, some parameters depending on delayed-neutron data are needed. The β_{eff} and the mean precursors' half-life ($\overline{T_{1/2}}$)

$$\overline{T_{1/2}} = \frac{\sum_{i=1}^{8} a_i T_{1/2,i}}{\sum_{i=1}^{8} a_i} \tag{2}$$

both depend on DN data and are known today with an uncertainty of 3% and 5%, respectively. The DN parameters are generally estimated through a microscopic calculation or through an integral experiment. In the framework of her Ph.D., Foligno [1] combined the two approaches with the aim of reducing the uncertainties associated with the data, and producing the respective correlation matrix. The experiment consisted in an integral measurement of the DN activity, which represents the global behavior of hundreds of precursors. Keepin performed the same experiment in the late '50 [2] and his results are still the reference for the European library JEFF-3.1.1 [3].

2 The principle of the ALDEN experiment

The main components of the ALDEN experiment are the fissile sample, the beam shutter, and the neutron detector. The first step of the experiment consisted in irradiating a fissile target to allow the build-up of DN precursors. During the irradiation, both prompt and delayed neutrons are emitted and measured by the LOENIE long counter. A beam shutter interrupts the neutron beam, fissions stops in the sample, and prompt neutrons disappear. Only DN precursors are left in the target and their decay can be easily registered by the detectors. A veto time Δt_0 needs to be set to take into account the fission tale, caused by slow neutrons emitted when the beam was open that hit the target after it was close and cause fission. Figure 2 shows a typical run of the ALDEN experiment, with 50 s of irradiation followed by more than 200 s of decay.

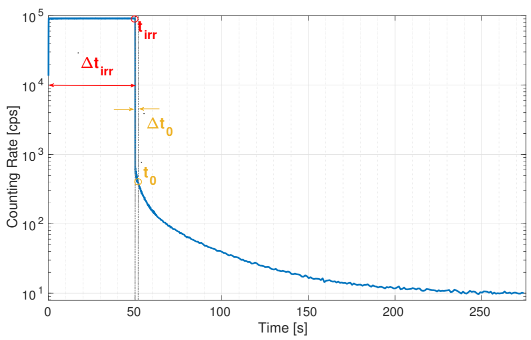

Figure 2: Typical irradiation run of the ALDEN experiment

The average counting rate during the irradiation phase is given by

$$\overline{n_i} = F(\nu_p \epsilon_p + \nu_d \epsilon_d) + b_i \tag{3}$$

while the delayed-neutron emission rate during the decay phase can be described as

$$n_d(t) = F\nu_d\epsilon_d \sum_{i=1}^{8} f_i a_i (1 - e^{-\lambda_i \Delta t_{irr}})(1 + e^{-\lambda_i \Delta t_m}) e^{-\lambda_i \Delta t_0} \cdot \cdot e^{-\lambda_i t} + b_d(t) \tag{4}$$

where F is the fission rate, ν_p and ν_d the average prompt and delayed-neutron yields, ϵ_p and ϵ_d LOENIE's efficiency to prompt and delayed-neutron spectrum, b_i and b_d the average background rate during irradiation and decay phase, f_i the factors $\epsilon_{d,i}/\epsilon_d$, a_i and λ_i the kinetic parameters, and Δt_{irr}, Δt_m and Δt_0 the irradiation, the measurement and the veto duration. Specific experimental campaigns have been designed to determine the efficiencies and the background rates. Deriving F from Eq. 3 and inserting it in Eq. 4, one obtains a formulation of the delayed-neutron activity which does not need the explicit fission rate value. The procedure to determine the ν_d consisted in extrapolating the counting rate at the end of the irradiation phase, after the disappearance of prompt neutrons. Only the beginning of the curve was needed for the fit, and the number of points taken into account has been chosen by minimizing the combination of systematic and statistical uncertainty. As far as the kinetic parameters are concerned, the abundances have been obtained through a fit of the whole decay curve.

2.1 The detector

The LOENIE long counter is made of two blocks of polyethylene with a central hole to host the target and 16 smaller holes to host the ^{3}He proportional counters. It is internally and externally shielded by flexibore to reduce the neutron background. LOENIE has been designed to have a flat efficiency as a function of the neutron energy, in the energy range of interest for delayed neutrons (100 keV - 1 MeV). This characteristic has been obtained through the arrangement of the detectors in three concentric rings at specific distances from the center. The optimization of the arrangement has been done by performing TRIPOLI4® [4] simulations. Figure 3 shows a picture of LOENIE and its 16 detectors.

Figure 3: LOENIE long counter with its 16 ^{3}He detectors

2.2 The target

The target is a miniaturized fission chamber designed and produced at the *Laboratory of Dosimetry, Sensors, and Instrumentation* of CEA Cadarache. It behaved as a target and a detector at the same time. The fissile material has been electroplated on a titanium support of 8 mm in diameter. The amount of fissile material has been determined in such a way not to exceed 10 kHz on each of the proportional counters during the irradiation phase, under the assumption of an equivalent 25 meV neutron flux of $4 \cdot 10^8$ cm^{-2}s^{-1}. In 2018, three fission chambers were produced: ^{235}U (210 μg), ^{239}Pu (140 μg), and a dummy chamber without fissile deposit for the background estimation. Figure 4 shows a picture of one of the chambers.

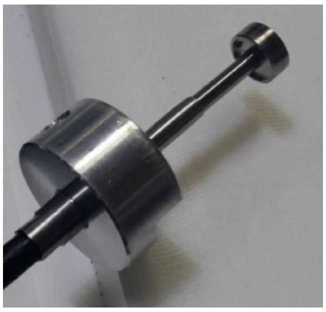

Figure 4: Miniaturized fission chamber

2.3 The fast-shutter

The standard beam shutter of the PF1b line was too slow for the needs of the ALDEN experiment and could not be remotely controlled. As a consequence, it has been necessary to design a new beam shutter, compact and light. The final design consisted of a rotative brushless motor with two screens of B_4C (boron carbide) and Cd (cadmium). Figure 5 shows a picture of the fast-shutter. The size, the thickness, and the materials have been chosen in such a way to reduce the neutron flux hitting the target by a factor of 10^8. Thanks to this system, the neutron beam could be turned off in less than 4.2 ms.

Figure 5: Fast-shutter

2.4 The experimental campaigns

The experimental campaign, performed in the framework of the ALDEN project, took place at ILL in September 2018 and consisted in the measurement of the delayed-neutron activity, the average background and the acquisition system loss of counts estimation. At the beginning of 2019, the long counter was sent to NPL (Nuclear Physics Laboratory, UK) for the estimation of its absolute efficiency at different energies using various neutron sources. In March, another campaign took place at the AMANDE accelerator (IRSN, France) for the relative efficiency estimation using several neutron fields resulting from proton interaction with different targets. Finally, a test with a pulser allowed the estimation of dead time and pile-up correction.

3 Data regression analysis

Each acquisition was accompanied by a binary file containing, for each event, the arrival time, the energy channel, and the detector in which it took place. The first step of the data regression analysis consisted in the conversion of the binary file into a histogram in time, the MCS (Multi-Channel Scaling). Notice that the MCS had to be computed for each proportional counter, in order to apply the respective loss-of-count correction due to the acquisition system's pile-up and dead-time. After that, all the MCS of the same run could be summed up to determine the global long-counter response. Finally, after scaling the different runs with respect to a reference point, they could be summed up and processed. Notice that an energy range of interest has been defined by analyzing the PHA (Pulse Height Amplitude), which allowed the discrimination between neutron and noise signals. Two cycles have been performed, both of them with 50 s of irradiation.

4 Results

The fit of the first 500 ms of decay curve with the CONRAD® code [5] resulted in a ν_d of 1.631E-02 ± 1.4%. All the parameters of the model have been marginalized, and their uncertainty is reflected in the 1.4%, of which 1% is due to the detector efficiency, 0.4% to the statistics, and 0.9% to a corrective factor (a_i, λ_i). The result of the experimental campaign is compatible with the WPEC-6 recommendations [6] (first line of Tab. 1), as well as with previous measurements (Fig. 6). The recommended values of ν_d come without any associated uncertainty, but the measurements from which they have been derived had an uncertainty of about 3%.
As far as the kinetic parameters are concerned, Tab. 1 reports both the abundances and the mean precursors' half-life resulting from the ALDEN experiment, together with the values recommended by the WPEC-6. Notice that the guessed values for the abundances have all been set to 0.125 +/- 1E+03 in order to be completely free parameters of the fit. In the end, they naturally converged towards the Foligno et al.'s set of abundances reported in Tab. 1. The uncertainties in the a_i of the long-lived groups have been strongly reduced, especially for a_1, which is particularly important after 100 s of decay. The global behavior of the system is determined by the mean precursors' half-life, which is well estimated and whose uncertainty has

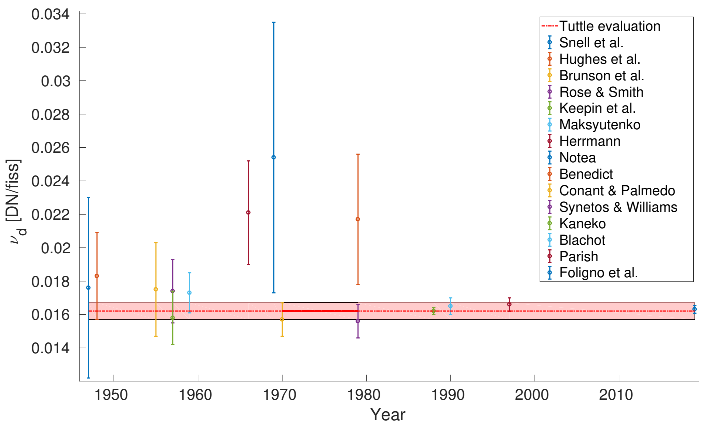

Figure 6: Experimentally measured ν_d for ^{235}U

been reduced from 3 to less than 1% when considering the correlation matrix (Fig. 7) in the uncertainty propagation procedure. Given the huge impact they have, it is worth spending a few words on correlations. A negative correlation is the relationship between two quantities that move in opposite directions. Let's take the portion of decay curve corresponding to a half-life of about 3 seconds. Due to the group assumption, this part of curve is the sum of the fifth ($T_{1/2,5} = 2.37s$) and the fourth ($T_{1/2,4} = 5.21s$) exponentials, and if a_5 increases, then a_4 must decrease to preserve the sum. This is why the abundances are anti-correlated with their adjacent groups. From a mathematical point of view, it is worth stressing the huge impact of correlations in the uncertainty quantification (see Eq. 5). If the correlation matrix is not used, it must be replaced by an identity matrix. On the other hand, using the correlation matrix, which in this case is mostly anti-correlated ($corr(a_i, a_j) < 0$), most of the terms in the sum become negative and balance the positive ones. The global effect is a strong reduction in the total uncertainty.

$$\sigma_{\overline{T_{1/2}}} = \sqrt{\sum_{i=1}^{8}\sum_{j=1}^{8}\frac{\partial\overline{T_{1/2}}}{\partial a_i}\frac{\partial\overline{T_{1/2}}}{\partial a_j}corr(a_i,a_j)\sigma_{a_i}\sigma_{a_j}} \quad (5)$$

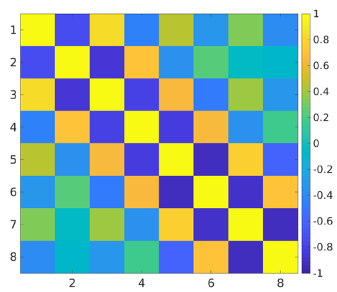

Figure 7: Correlation matrix associated with Foligno's abundances

5 Conclusions

In conclusion, the experiment resulted to be successful. The results of the first experimental campaign are consistent with the literature. The uncertainties of both ν_d and $\overline{T_{1/2}}$ have been reduced to at least half of the recommended values. In perspectives, a new experimental campaign should be launched to improve the estimation of the short-lived groups' abundances, with adapted cycles of irradiation and decay phase. In addition to that, after the thermal fission of ^{235}U, other fissioning systems will be studied in the framework of the ALDEN project, starting with ^{239}Pu, ^{241}Pu, and ^{233}U.

Table 1: Results of the ALDEN experiment. *Foligno et al.* are the results of the fit while *WPEC-6* are the values recommended by the WPEC-SG6

Quantity of interest	Foligno et al.	WPEC-6 [6]
ν_d [DN/fiss]	1.631E-02 (1.4%)	1.62E-02 (-)
a_1	3.64E-02 (2.7%)	3.28E-02 (12.8%)
a_2	1.31E-01 (3.3%)	1.54E-01 (4.4%)
a_3	1.15E-01 (4.9%)	9.14E-02 (9.8%)
a_4	1.66E-01 (4.8%)	1.97E-01 (11.7%)
a_5	3.55E-01 (5.2%)	3.31E-02 (2.0%)
a_6	6.92E-02 (40.8%)	9.03E-02 (5.0%)
a_7	8.15E-02 (49.3%)	8.12E-02 (2.0%)
a_8	4.58E-02 (66.3%)	2.29E-02 (41.5%)
$\overline{T_{1/2}}$ [s]	8.93 (5.72%)	9.02 (3.0%)
$\overline{T_{1/2}}$ with corr [s]	8.93 (0.98%)	9.02 (3.0%)

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