Delayed neutron yields and spectra from photofission of actinides with bremsstrahlung photons below 20 MeV.

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Abstract. Experimental results for the photofission of $^{238}$U with an endpoint Bremsstrahlung emission of 15 MeV are presented. Absolute yield and time characteristics of the delayed neutrons are extracted. In parallel, calculations for fission fragment distributions and corresponding delayed neutron parameters are given and compared to data.

1. Introduction
The renewed interest in photonuclear processes is motivated by applications as radioactive ion beam (RIB) production, intense photo-neutron sources, radioprotection and decommissioning of electron accelerators, etc. Today, a particular attention is paid to the non-destructive characterization of waste barrels [1] and detection of nuclear materials [2], both based on photofission process and the associated delayed neutron (DN) emission. Unfortunately, available DN data are scarce, incomplete and in some cases even contradictory. Therefore, a new experimental program has been undertaken in order to provide the good quality photofission DN data being the essential information for the above applications including the evaluations and data libraries in the energy range of the endpoint Bremsstrahlung photons from 6 MeV to 19 MeV, i.e. covering the entire Giant Dipole Resonance (GDR) region [3]. Absolute yields and time spectra of DNs for high priority nuclei as uranium and plutonium isotopes including some minor actinides (Am and Np) will be investigated. In this paper, after a brief description of the DN parameterization in Section 2, experimental results for $^{238}$U are presented in Section 3. In parallel to the experimental program, systematic calculations are performed with the fission-evaporation code ABLA+PROFI [4-5] in order to reproduce experimental mass distributions of fission fragments and DN data. In this paper, experimental results and calculations are compared for the $^{238}$U nucleus.

2. Delayed neutrons
Absolute yields and time characteristics of delayed neutron emission for neutron-induced fission have been extensively studied for the last fifty years. This neutron emission follows the beta decay of some fission fragments which are called precursors. The precursors are usually lumped in 6 groups according to their half-lives [6]. In the case of photofission the same classification has been adopted here. The time dependence of the delayed neutron yield is then expressed by $Y_{DN}(t) = \Sigma \lambda \exp(-\lambda t)$
where \( i \) goes from 1 to 6. This holds for infinite irradiation time for which all precursors are at equilibrium. The left panel of figure 1 shows the relative contribution of the \(^{238}\text{U} \) DN yield of each group according to the time. One can note that over 100 s, groups 1 (\( T_{1/2} = \ln 2/\lambda \approx 55 \) s) and 2 (\( T_{1/2} \approx 20 \) s) are predominant. On the other hand, for finite irradiation time (e.g. 1 ms), the expression for \( Y_{\text{DN}}(t) \) becomes \( \sum a_i \exp(-\lambda_i t) (1-\exp(-\lambda_i T)) \) where \( T \) is the irradiation time. In this case, one remarks in right panel of figure 1 the relative enhancement for the groups 5 (~0.5 s) and 6 (~0.2 s) with respect to the other groups. Therefore, the choice of irradiation time (\( T_{\text{irr}} \)) and counting time (\( T_{\text{count}} \)) is an important parameter for the determination of the delayed neutron characteristics. To obtain a good accuracy for all groups, it is then necessary to perform measurements with different combinations for \( T_{\text{irr}} \) and \( T_{\text{count}} \).

![Figure 1](image-url)

**Figure 1.** Relative DN yield contributions with respect to the total DN yield of \(^{238}\text{U} \) as a function of the decay time for the six groups; for infinite irradiation on the left panel and 1 ms of irradiation on the right panel. 6 groups are numbered in both panels.

### 3. Experiment

The DN measurements were made with typical \(^3\text{He} \) counter (4 bars), 30 cm long and with a diameter of 2.5 cm. The \(^3\text{He} \) tube was placed in the center of a \( \text{CH}_2 \) box of 37x20x10 cm\(^3\), which was surrounded by a Cd foil (1 mm) to reject low energy scattered neutrons. The intrinsic efficiency of the detector (\(^3\text{He}+\text{CH}_2+\text{Cd} \)) has been measured with the use of \(^{252}\text{Cf} \) and AmBe sources, and mono-energetic neutron beams of different energies. The VDG 4MV at Bruyères-le-Châtel (CEA/DIF) has provided proton beams. The \( \text{p}+\text{T} \) and \( \text{p}+\text{Li} \) reactions have been used in order to obtain neutron beams in the range from 100 keV to 2 MeV. During these calibration experiments it has been shown that the intrinsic efficiency of the detector is 5% and remains constant within the delayed neutron energy range (0.1 to 1 MeV). The efficiency calculated with Monte Carlo simulations using MCNPX is in agreement with this value.

In order to validate the experimental method, a delayed neutron measurement for the neutron-induced fission of \(^{238}\text{U} \) has been done. With an incident proton beam of 6 \( \mu \text{A} \) (VDG, CEA/DIF), nearly \( \approx 4.6e+6 \) neutrons/s of 2 MeV were impinging on the metallic \(^{238}\text{U} \) target of 405 g (cylinder, \( L=3 \) cm, \( \phi=3 \) cm; 0.3% of \(^{235}\text{U} \)) placed at 20° with respect to the proton beam. The detector was placed 10 cm behind the uranium target, giving a global DN efficiency of the detection system of 1.08%. The logical output of the preamplifier was sent to the multi-channel scaler. The irradiation and counting times have been set to 125 s and a measurement of 53 cycles has been performed. Using the number of fissions calculated with MCNPX, the total delayed neutron yield for 100 fissions has been evaluated to \( 4.56 \pm 0.62 \). This value is close to the ENDF reference value (\( \nu_d = 4.4 \)). The poor statistics accumulated during the experiment did not allow an extraction of the full time characteristics. Nevertheless, some ratios of different group contributions have been calculated (Table 1). The agreement between our
results and the ENDF values validates the measurement procedure for applying it to the photofission experiment.

**Table 1.** DN yield ratios in the case of 6-group model for n (2 MeV)-induced fission of $^{238}$U.

| Ratio  | ENDF-B VI | This work  |
|--------|-----------|------------|
| a2/a1  | 12.85     | 12.1 ± 8.7 |
| a4/a1  | 37.12     | 37 ± 25    |
| a3/a2  | 1.21      | 0.84 ± 0.47|
| a4/a2  | 2.89      | 3.0 ± 1.2  |
| a4/a3  | 2.39      | 3.6 ± 2.0  |

The experimental setup for the photofission DN measurements is shown on figure 2. The ELSA accelerator (CEA/DIF, Bruyères-le-Châtel) has provided a 15 MeV pulsed electron beam with an intensity of 1μA. The thickness of the tantalum converter was 1.2 mm providing a gamma flux of 6.48e+8 photons/μA·cm² (MCNPX calculations) on the uranium target placed at 1.50 m at 0 degree. Residual electrons were stopped in the 6cm aluminum attenuator (see figure 2). The photon beam angular aperture was limited to 5 degrees by the lead collimator. The detector was located at 17 cm of the uranium target and at 90 degrees with respect of the beam axis. With such a configuration, the global DN efficiency has been evaluated to be 0.62%.

![Figure 2. The experimental setup for DN measurements from photofission.](image)

Three combinations of irradiation and counting (decay) campaigns have been chosen: 140μs-30s, 5s-100s and 300s-300s. In figure 3 the time spectrum for the third one (300s-300s) is shown (symbols). The background measurement was obtained separately without uranium target giving ~1.4 counts/s. The number of fission has been calculated with MCNPX and the electron current value. For the long irradiation as 300 s, the absolute DN yield is proportional to the measured decay curve value at t = 0 and the corrections needed are negligible. The value of 3.05 ± 0.20 has been found for the absolute DN yield in this case. This is in agreement with previous value of 3.1 ± 0.4 obtained by Nikotin et al. [7] in 1965.
This long irradiation has also allowed the extraction of the parameters for the two first groups. Indeed, considering that only $^87\text{Br}$ contributes to the first group, we constrain its half-life to 55.6s. The a1 value has been extracted by fitting the end of the time spectrum. Then, with an iterative procedure and switching to the smaller time values, the parameters for the 2nd group have also been extracted. Finally other T$_{ir}$ and T$_{count}$ combinations allowed the determination of other group parameters. The grey curve in figure 3 is the sum of the six exponentials resulting from the global parameters fit. It reproduces the experimental curve remarkably well. The numerical results are presented in Table 2 and compared to Nikotin’s parameters. While the half-lives are close, some discrepancies are observed for the group contributions. The biggest difference is found for the 6$^{th}$ group where our result is by a factor of 2 smaller. In addition, the accuracy of our measurement is better. Kuhl et al [8] have also observed this discrepancy for $^{238}\text{U}$ but at 8 and 10 MeV. Their data, obtained with different irradiation times (250 ms, 12s, 300s) are in agreement with our work for the 6$^{th}$ group. Indeed, the 6$^{th}$ group is difficult to extract with the long irradiation time what was probably the case during the experiment by Nikotin et al..

### Table 2. Half-lives (s) and relative contributions (%/fiss) for $^{238}\text{U}$ at 15 MeV.

| Group | T$_{1/2}$ [Nikotin] | T$_{1/2}$ [this work] | a$_{1}$ (%) [Nikotin] | a$_{1}$ (%) [this work] |
|-------|---------------------|-----------------------|--------------------|-----------------------|
| 1     | 56.2 ± 0.8          | 55.6                  | 1.98 ± 0.08        | 1.7 ± 0.2             |
| 2     | 21.3 ± 0.3          | 21.88 ± 0.66          | 15.7 ± 0.5         | 16.5 ± 0.5            |
| 3     | 5.50 ± 0.20         | 5.01 ± 0.49           | 17.5 ± 0.7         | 18.3 ± 0.7            |
| 4     | 2.15 ± 0.10         | 2.07 ± 0.14           | 31.1 ± 0.8         | 37.3 ± 0.7            |
| 5     | 0.70 ± 0.06         | 0.584 ± 0.051         | 17.7 ± 0.9         | 18.0 ± 0.4            |
| 6     | 0.19 ± 0.02         | 0.174 ± 0.019         | 16.1 +2/-5         | 8.5 ± 0.8             |

### 4. Calculations

In order to obtain the delayed neutron table for actinides, a complete scheme of calculation procedure has been built. The experimental setup geometry has been implemented in MCNPX and the bremsstrahlung spectrum on the target has been calculated. Next, based on the GDR model, the absorption cross section was calculated using the sum of two lorentzians and the parameters of the RIPL2 table [9]. Result of the parameterization is close to the IAEA evaluation [10] for $^{238}\text{U}$ between 5 and 20 MeV. Then, the evaporation-step was performed with ABLA code [4], developed at GSI, which includes a fission fragment distribution modeling (PROFI [5]). Results for $^{235}\text{U}$ and $^{239}\text{U}$ at 15 and 25 MeV are shown in figure 4. Calculations (histograms) reproduce the experimental mass distributions (symbols) rather well with same input parameters. The position of the peaks and the
widths are similar while the relative heights are slightly different. For $^{237}$Np and $^{239}$Pu (figure 5, left and right panel) the code gives also good results. For these nuclei, only parameters governing the potential well depth of the two asymmetric paths had to be adjusted in order to reach this quality. Finally, fission fragment cumulative yields have been calculated with the evolution code CINDER'90 [14] using the independent yields given by ABLA. Delayed neutron precursors have then been identified and their contributions lumped into 6 groups according to their half-lives. The result is presented in Table 4 and compared to our experimental results. First, note that the calculated absolute yield ($\nu_d$) for hundred fissions is rather close to the experimental one. One also remarks that the half-lives are in agreement. However, discrepancies for the individual group yields are rather significant. The difference is probably due to the neutron rich part of the fission fragment distributions. Even if ABLA seems to reproduce mass distribution data rather well, it enhances the need of comparison for isotopic yields. Indeed, it is difficult to check the validity of ABLA for this observable since very little data is available. A way to bypass this difficulty could be an analysis of DN time spectrum in 8 or even 12 groups [15]. With this kind of analysis, some isotopic yields could be extracted and then directly compared to model calculations.

**Figure 4**. Mass distributions from photofission of $^{235}$U and $^{238}$U at 15 and 25 MeV. Data [11] (squares) are compared to calculations (histograms).

**Figure 5**. Mass distributions from photofission of $^{237}$Np [12] (left panel) and $^{239}$Pu [13] (right panel) at 14 and 28 MeV respectively. Data (squares) are compared to calculations (histograms).

| Group | $T_{1/2}$ (s) | DN Yield (%) |
|-------|---------------|--------------|
|       | Data | Calc. | Data | Calc. |

**Table 4**. Six-group DN parameters for photofission of $^{238}$U at 15 MeV.
5. Conclusion

An experimental campaign for actinide DN measurements from photofission below 25 MeV has been undertaken. For $^{238}$U at 15 MeV, our experimental results have improved the accuracy obtained previously by other teams and permitted to clarify the situation for the 6th group. DN measurements will be pursued. The analysis of the recently performed $^{232}$Th photofission DN measurement at 15 MeV endpoint Bremsstrahlung energy is in progress. For other actinides ($^{235}$U, $^{239}$Pu, $^{237}$Np, $^{241}$Am), the radioactivity is such that only small samples could be irradiated. Due to this limitation, the experimental setup will have to be improved.

The experimental effort is accompanied by a theoretical work and evaluations. Model calculations have to be improved in order to predict mass distributions of actinides. The lack of data for photofission is the major problem. Nevertheless, the DN parameters for $^{238}$U at 15 MeV are well reproduced. Furthermore, the relatively good reproduction of the existing data compared to ABLA predictions makes possible the photon activation file (PAF) elaboration [16]. The release of the PAF in the near future will include fission fragment distributions and delayed neutron tables.

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