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# Evaluation of delayed neutron yields and time spectra from photofission of $^{238}\text{U}$ induced by *Bremsstrahlung* photons below 9 MeV

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**Abstract.** Few data exist regarding the yields and time spectra of delayed neutrons from the photofission reaction on actinides. In this paper, we report the characteristics of both six-group and eight-group delayed neutron time models for the photofission of  $^{238}\text{U}$  induced by a *Bremsstrahlung* photon spectrum with an end-point energy at 9 MeV. This study was conducted using a 9 MeV linear electron accelerator housed at the SAPHIR platform located at CEA Paris-Saclay, France. A reference sample of depleted uranium was positioned inside a neutron detection block, previously designed and optimized by Monte Carlo simulation, made of high-density polyethylene covered by cadmium and including ten one meter-long  $^3\text{He}$  proportional counters. Based on the consistent set of half-lives proposed by Spriggs *et al.* as early as 1999 for neutron-induced fission, our description in eight delayed neutron groups for  $^{238}\text{U}$  at 9 MeV is the first published for photofission. These new results bring valuable information to the scarce data available in the literature and represent useful inputs for the development of photofission-based nuclear measurement systems as they meet a significant interest in the fields of nuclear data, radioactive waste package characterization and for security-related applications.

## 1 Introduction

The reaction in which the fission of an atomic nucleus is induced by a high-energy photon is known as photofission [1, 2]. Both *fissile* actinides ( $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , etc.) and *fertile* actinides ( $^{238}\text{U}$ ,  $^{240}\text{Pu}$ , etc.) can undergo photofission. The energy threshold of this reaction is around 6 MeV for all actinides. The applications of photofission range from security at borders to radioactive waste package characterization. To implement this technique, the high-energy photon beam is usually produced by *Bremsstrahlung* in the conversion target of a linear electron accelerator. Moreover, a strong interest has emerged in the recent years for the use of electron accelerators operated below 10 MeV in order to develop photofission mobile systems [3, 4]. Both the design of these measurement stations and the evaluation of their performance require Monte Carlo simulation studies and post-processing calculations, which justifies the need for the most reliable nuclear data concerning the photofission reaction in the energy range of interest.

A six-group multi-exponential theoretical description was originally provided by Keepin *et al.* in 1957 for delayed neutrons from thermal or fast neutron-induced fission of several isotopes [5]. A few years later, this model was applied to delayed neutrons from photofission. In 1965, Nikotin *et al.* published data for the photofission of  $^{232}\text{Th}$ ,  $^{235}\text{U}$ ,  $^{238}\text{U}$  and  $^{239}\text{Pu}$  induced by a *Bremsstrahlung* photon spectrum with an end-point

energy at 15 MeV [6]. In 1970, Kull *et al.* presented data for the photofission of  $^{235}\text{U}$  and  $^{238}\text{U}$  at both 8 and 10 MeV end-point energies [7]. In 2006 and 2007, Doré *et al.* reported data for the photofission of  $^{235}\text{U}$  and  $^{238}\text{U}$  at various end-point energies below 20 MeV [8, 9]. In 2009, Macary *et al.* provided data for the photofission of  $^{235}\text{U}$  and  $^{237}\text{Np}$  for a couple of end-point energies below 20 MeV [10].

From a theoretical point of view, each delayed neutron group  $i$  is characterized by a half-life time  $(t_{1/2})_i$  or an equivalent decay constant  $\lambda_i$  and a relative yield  $a_i$ . Following an irradiation of duration  $T_{\text{irr}}$ , the delayed neutron signal decays over time and can be described by Eq. 1 considering a number of groups noted  $n$ .

$$Y_{\text{DN}}(t) = \sum_{i=1}^n a_i e^{-\lambda_i t} (1 - e^{-\lambda_i T_{\text{irr}}}) \quad (1)$$

In this paper, we evaluate the delayed neutron yields and time spectra for the photofission of  $^{238}\text{U}$  induced by *Bremsstrahlung* photons below 9 MeV. First, we show the experimental setup, determine the irradiation times and describe our nuclear data extraction algorithm. Then, we present and discuss the results obtained. In addition to the description in six delayed neutron groups, based on the consistent set of half-lives proposed by Spriggs *et al.* as early as 1999 for neutron-

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induced fission [11, 12], our description in eight groups for  $^{238}\text{U}$  at 9 MeV is the first published for photofission.

## 2 Materials and methods

### 2.1 Experimental setup

The experiments were conducted at the SAPHIR platform located at CEA Paris-Saclay, France. This facility houses a linear electron accelerator, Linatron® M9A manufactured by *Varex Imaging Corp.*, which was operated at 9 MeV and 200 Hz. The neutron detection block, designed and optimized beforehand by the authors of this paper using Monte Carlo simulation with the MCNP6 code [13], consists of ten  $^3\text{He}$ -filled gas proportional counters (150NH100, 4 bars) embedded in high-density polyethylene surrounded on its outer and inner surfaces by a 1 mm-thick cadmium layer. A 241 g reference sample of depleted uranium was positioned and laser-aligned at the centre of the neutron detection block. The delayed neutron signal was acquired using new digital readout electronics developed at our laboratory, which will be presented in detail in a dedicated paper. Fig. 1 shows an overview of the experimental setup whereas Fig. 2 focuses on the neutron detection block.

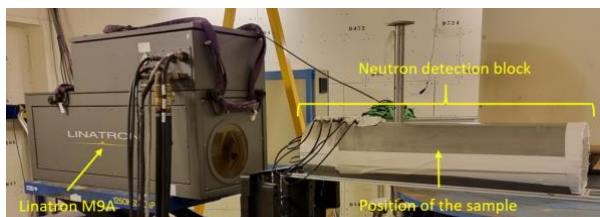


Fig. 1. Overview of the experimental setup.

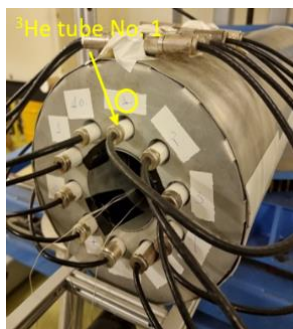


Fig. 2. The neutron detection block.

### 2.2 Irradiation times

The six or eight groups being characterized by half-lives ranging from  $\sim 50$  s to a few tens of ms, the irradiation time can be adapted to the delayed neutron group whose parameters are to be extracted. In order to determine the most adapted irradiation times to be used in our measurement protocol, we plotted the relative delayed neutron yields of each group as a function of irradiation time  $T_{\text{irr}}$  thanks to Eq. 2.

$$\int_0^{T_{\text{max}}} [Y_{\text{DN}}(t)]_i dt = a_i (1 - e^{-\lambda_i T_{\text{irr}}}) (-\lambda_i e^{-\lambda_i T_{\text{max}}} + \lambda_i) \quad (2)$$

As a first approach, the relative delayed neutron yields  $a_i$  and the decay constants  $\lambda_i$  for  $^{238}\text{U}$  were taken from Nikotin *et al.* [2] and the maximum counting time  $T_{\text{max}}$  was set at 300 s. Fig. 3 presents the results obtained. However, for this type of study, the delayed neutron groups are usually matched in pairs and three different irradiation times are chosen [6-10]. Fig. 4 presents the updated results. Finally, the irradiation times chosen to “stimulate” preferentially groups two by two are: 300 s, 10 s and 1 s. Whatever the irradiation time, as soon as an irradiation sequence is completed, the neutron detection system is switched on and acquires the signal for a duration of 300 s. For statistics purposes, these measurements – based on 300 s, 10 s and 1 s irradiation times – were repeated respectively: 5, 50 and 100 times. Regarding groups 5 and 6, irradiation times below 1 s raise questions about beam stability and measurement reliability, which from the point of view of the authors of this paper is the case for experimental protocols followed in previous studies [6-10].

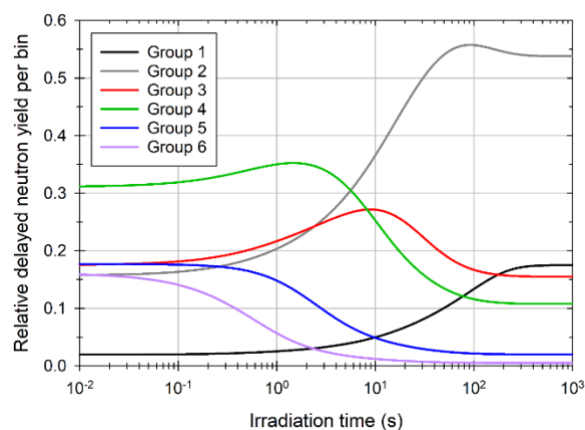


Fig. 3. Relative yields for each of the six delayed neutron groups as a function of irradiation time (integration over the counting time from 0 to 300 s).

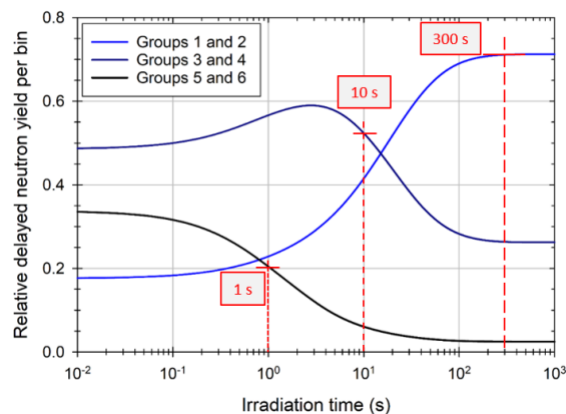


Fig. 4. Relative delayed neutron yields as a function of irradiation time (integration over the counting time from 0 to 300 s): groups matched in pairs.

### 2.3 Nuclear data extraction algorithm

To determine the twelve parameters of delayed neutrons from photofission of  $^{238}\text{U}$  at 9 MeV according to the six-group model:

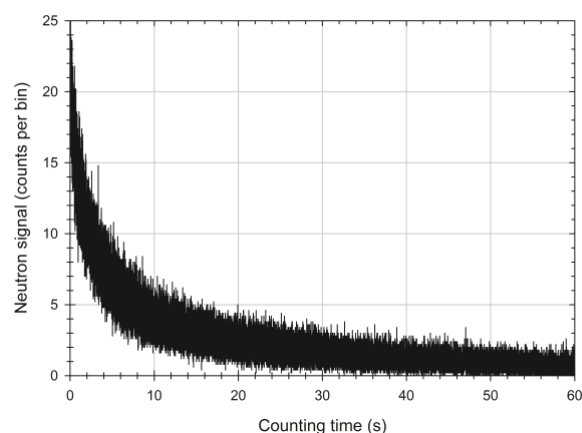
- the first step consists in extracting  $\lambda_1$  and  $\lambda_2$  from the 300 s-irradiation data;
- the second step consists in extracting  $\lambda_3$  and  $\lambda_4$  from the 10 s-irradiation data;
- the third step consists in extracting  $\lambda_5$  and  $\lambda_6$  from the 1 s-irradiation data;
- running again steps 1 to 3 through an iterative process enables to fine-tune all the parameters  $\lambda_i$ ;
- the last step consists in taking the 300 s-irradiation data (all groups at equilibrium), fixing the six decay constants  $\lambda_i$  to their previously determined values, and extracting the six relative yields  $a_i$ .

To determine the parameters of delayed neutrons from photofission of  $^{238}\text{U}$  at 9 MeV according to the eight-group model, we took the consistent set of eight half-lives proposed by Spriggs *et al.* for neutron-induced fission [11, 12], which following the recommendations from the NEA/WPEC-6 report [14] was later used by the JEFF evaluation, since version JEFF-3.1 [15]. The eight remaining relative yields to be determined are obtained using the 300 s-irradiation data fitted with an eight-group multi-exponential.

Furthermore, whatever the number of delayed neutron groups (six or eight), the uncertainties associated to the parameters determined experimentally are evaluated using the series of repeated measurements and under the assumption of a Poisson distribution.

## 3 Results and discussion

An example of time spectrum of delayed neutrons from photofission of  $^{238}\text{U}$  at 9 MeV, acquired following a 300 s-irradiation, is shown in Fig. 5. Although the total counting time is equal to 300 s, this figure focuses on the first 60 s of counting.



**Fig. 5.** Example of time spectrum of delayed neutrons from photofission of  $^{238}\text{U}$  at 9 MeV, acquired following a 300 s-irradiation.

### 3.1 Six-group model

The parameters obtained for the six-group model of delayed neutrons from photofission of  $^{238}\text{U}$  at 9 MeV are gathered in Table 1. We can note in particular that the half-life of *group 1* is lower than that assessed by Nikotin and Petrzhak for  $^{238}\text{U}$  at 15 MeV ( $56.2 \pm 0.8$  [6]), while the relative yield of *group 6* is more in agreement with Nikotin and Petrzhak ( $16.1 \pm_{5.0}^{2.0}$  % [6]) than Doré *et al.* ( $8.5 \pm 0.8$  % [8]). As both the half-lives  $[t_{1/2}]_i$  and the relative yields  $a_i$  vary simultaneously through the nuclear data extraction algorithm, it is however difficult to be conclusive when comparing with parameters determined by other authors working on photofission delayed neutrons with a six-group model.

**Table 1.** Parameters of the six-group model for delayed neutrons from photofission of  $^{238}\text{U}$  at 9 MeV.

Group	$[t_{1/2}]_i$ (s)	$a_i$ (%)
1	$50.6 \pm 0.5$	$2.1 \pm 0.1$
2	$21.3 \pm 0.3$	$14.8 \pm 0.1$
3	$6.42 \pm 0.15$	$12.4 \pm 0.5$
4	$2.28 \pm 0.06$	$40.6 \pm 0.6$
5	$0.639 \pm 0.047$	$15.6 \pm 1.6$
6	$0.210 \pm 0.037$	$14.5 \pm 1.6$

### 3.2 Eight-group model

Table 2 gathers the parameters obtained for the eight-group model of delayed neutrons from photofission of  $^{238}\text{U}$  at 9 MeV. As a reminder, the consistent set of half-lives comes from neutron-induced fission [11, 12]. These values could be used as a comparison basis for future evaluations of delayed neutrons from photofission according to an eight-group model.

**Table 2.** Parameters of the eight-group model for delayed neutrons from photofission of  $^{238}\text{U}$  at 9 MeV (consistent set of half-lives taken from [11, 12]).

Group	$[\lambda_{1/2}]_i$ (s)	$\alpha_i$ (%)
1	55.6	$1.6 \pm 0.1$
2	24.5	$9.2 \pm 0.6$
3	16.3	$7.9 \pm 0.8$
4	5.21	$12.8 \pm 1.1$
5	2.37	$33.5 \pm 1.0$
6	1.04	$13.1 \pm 1.1$
7	0.424	$9.9 \pm 3.2$
8	0.195	$12.1 \pm 3.0$

#### 4 Conclusions and perspectives

In this paper, we reported a six-group time model for delayed neutrons from photofission of  $^{238}\text{U}$  induced by a *Bremsstrahlung* photon spectrum with an end-point energy at 9 MeV. Moreover, based on the consistent set of half-lives proposed by Spriggs *et al.* as early as 1999 for neutron-induced fission, we also provided a description in eight groups, which is the first one published for photofission. These results should constitute a valuable contribution considering the scarce data available in the literature for time models of delayed neutrons from photofission, i.e., three peer-reviewed articles and two proceedings papers in more than 50 years.

The next step of this study would consist in carrying out additional measurements at both 6 MeV – i.e., just above the energy threshold of the photofission reaction – and 9 MeV as well as for other radionuclides such as  $^{235}\text{U}$  and  $^{239}\text{Pu}$ . Furthermore, complementary Monte Carlo simulations would enable to quantify the impact of neutron-induced fission reactions in the samples during these photofission measurements. The logical follow-up of this work would also include the determination of the absolute yields for delayed neutrons from photofission for the various radionuclides of interest at the two *Bremsstrahlung* spectrum end-point energies. These new measurements would allow comparison between the different results obtained and in particular enable to draw conclusions regarding the influence of the *Bremsstrahlung* photon spectrum end-point energy on the absolute and relative yields.

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