

Delayed neutron measurements for ^{232}Th neutron-induced fission

X. Ledoux¹, D. Doré², M. Mosconi³, R. Nolte³, and S. Roettger³

¹ CEA/DAM/DIF, 91297 Arpajon, France

² IRFU/SPhN, CEA/Saclay, 91191 Gif-sur-Yvette, France

³ PTB, 38116 Braunschweig, Germany

Abstract. Delayed neutrons (DN) play an important role in nuclear reactor physics. Innovative critical reactor studies bring to light the need of new DN yields data. For the Th fuel cycle, according to the OECD recommendation, DN of the ^{232}Th is needed with an accuracy of 5%. In the literature, significant discrepancies were observed for energies below 4 MeV and data are dispersed around 14 MeV. Therefore, a programme has been undertaken by CEA in order to measure DN yields from ^{232}Th with incident neutron energies from 2 to 16 MeV. In this paper, the experimental setup will be described and preliminary results obtained at the PTB Ion Accelerator Facility of Braunschweig for incident neutron beam energies of 2, 3, 4, 6, 7, 10 and 16 MeV will be presented.

1 Motivations

Delayed neutrons (DN) emitted after fission are of prime interest for several topics. Among others, they allow the control of a nuclear reactor. With the development of reactors of new generation and/or new fuels, additional requirements appear in determining DN yields or to measure more accurately existing data. For the innovative thorium cycle, data on DN yields of ^{232}Th are needed. Unfortunately, available data in the literature are scarce and important discrepancies are observed as can be seen in Figure 1. Moreover, inside the recent ISTC project, results obtained between 3 and 5 MeV seem to indicate a quasi constant yield in this energy region which deviates from the energy dependence predicted by Yoshida et al. [1]. These observations have motivated physicists of CEA (Commissariat à l'énergie atomique et aux énergies alternatives) to undertake an experimental program for measurements of DN yields of ^{232}Th for the incident neutron energy range from 2 MeV up to 16 MeV.

The experimental procedure, the neutron production facility and the detection setup will be described in the section 2. In section 3, analysis and simulations will be presented. Preliminary results will be shown in section 4.

2 Experiment

2.1 Methodology

Emission of delayed neutrons follows the beta decay of some fissions fragments called precursors. There are more than 200 different fragment isotopes involved which are usually lumped in 6 groups

This is an Open Access article distributed under the terms of the Creative Commons Attribution-Noncommercial License 3.0, which permits unrestricted use, distribution, and reproduction in any noncommercial medium, provided the original work is properly cited.

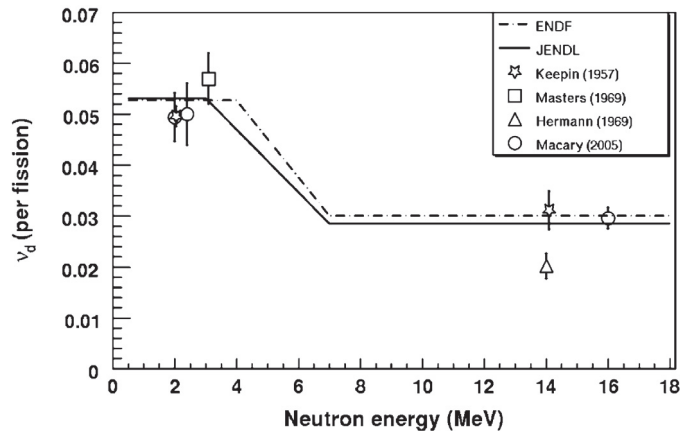


Fig. 1. Experimental [2] and evaluated [1] data of absolute DN yield in the ^{232}Th neutron induced fission.

Table 1. Characteristics of the six groups of Thorium neutron induced fission [2].

Group	$T_i(\text{s})$	$a_i(\%)$
1	56.03 ± 0.95	3.4 ± 0.2
2	20.75 ± 0.66	15.0 ± 0.5
3	5.74 ± 0.24	15.5 ± 2.1
4	2.16 ± 0.08	44.6 ± 1.5
5	0.571 ± 0.042	17.2 ± 1.3
6	0.211 ± 0.019	4.3 ± 0.6

according to their half-lives T_i . After a finite irradiation of duration t_{irr} , the DN time dependence can be defined by the function:

$$Y(t) = \nu_d \sum_{i=1}^6 a_i \exp(-\lambda_i t) (1 - \exp(-\lambda_i t_{\text{irr}})) \quad (1)$$

where

ν_d = number of delayed neutrons per fission

a_i = the relative abundance of the group i

$\lambda_i = \ln(2)/T_i$, T_i the period of the group i .

The groups 1 to 6 are defined with period from 56 s to 200 ms respectively (see Table 1).

After a long irradiation time, $t_{\text{irr}} \gg T_1$, all the precursors are at equilibrium and the DN decay time distribution after beam switch off at $t = 0$ is:

$$Y(t) = \nu_d \sum_{i=1}^6 a_i \exp(-\lambda_i t) \quad (2)$$

and

$$\text{at } t = 0 : Y(t = 0) = \nu_d \quad (3)$$

The number of DN detected at $t = 0$ is $F_{\text{det}}(0) = \nu_d N_{\text{fission}} \varepsilon$ with N_{fission} , and ε denote the fissions rate in the sample and the detection efficiency, respectively. The average number of DN per fission ν_d is then deduced:

$$\nu_d = \frac{F_{\text{det}}(0)}{\varepsilon N_{\text{fission}}} \quad (4)$$

A single irradiation-counting sequence does not allow to reach sufficient statistic. Therefore, the measurement consists of a 5 minutes continuous irradiation period to reach the equilibrium, followed by a large number of cycles composed of irradiation sequences with $T_{\text{irr}} = 7$ s and counting sequences with $T_{\text{counting}} = 1$ s. An irradiation time T_{irr} of 7 s is necessary to reach the equilibrium after each decay period.

2.2 Experimental setup

2.2.1 Facility

The experiment was performed in the “Low-Scatter Hall” at the PTB Ion Accelerator Facility at Braunschweig (Germany). Neutrons were produced by $^3\text{H}(p,n)^3\text{He}$, $^2\text{H}(d,n)^3\text{He}$ and $^3\text{H}(d,n)^4\text{He}$ reactions. Proton and deuteron beams were delivered by the Van De Graaff accelerator and the cyclotron CV28 depending on the energy of the neutrons [6, 7]. A fast steering magnet, controlled by a clock, was used to switch the beam according to the time structure described in section 2.1.

2.2.2 Detection setup

The delayed neutron detector was already used for DN photofission studies [8]. It is a hollow cylinder ($\Phi_{\text{int}} = 12$ cm, $\Phi_{\text{ext}} = 32$ cm, $L = 37$ cm) of polyethylene. In this cylinder 12 gas detectors filled with ^3He are inserted. The neutrons are slowed down in the CH_2 before being detected by $^3\text{He}(n,p)^3\text{H}$ reaction in one of the 12 tubes. The detector design has been optimized (thickness of the CH_2 and position of the ^3He tubes) to have a constant detection efficiency to neutrons in the 100 keV–1 MeV range [5]. When the sample is placed in the centre of the detector the DN detection efficiency is approximately 22%. For the present experiment, however, the cylindrical thorium sample (2.55 cm in diameter and 3.64 cm in length) with a mass of 182.23 g was placed inside the neutron detector at zero degree with respect to the ion beam direction at a distance of 75 mm from the production target (at 24 mm from the entrance of the DN detector). This distance is a compromise between reduction of the statistical uncertainty and the minimization of the effect of scattered incident neutrons to the sample. At the present sample position the efficiency was around 12%. The detection efficiency to neutrons (between 100 keV and 1 MeV) emitted in the thorium sample is simulated with the MCNPX [9] transport code. Since the ^3He pressure in the tubes being not well known, the results of simulations had to be normalized to the efficiency for a ^{252}Cf neutron spectrum measured using two calibrated ^{252}Cf sources at different positions.

2.2.3 Normalisation

Two detectors were used to determine the neutron fluence: a proton recoil telescope (PRT) and a fixed long counter placed around 6 m from the neutron source at 98 degrees with respect to the ion beam line. During dedicated runs, the PRT was placed at 0 degree with respect to the ion beam line and the DN detector was moved far away from the neutron source at backward angle. The fluence is measured relatively to the (n, p) elastic cross-section. The fluence during the DN measurement runs was obtained from the PRT measurement using the long counter or the integrated accelerator current as monitors. The long counter is sensitive to neutrons scattered off the DN detector into the long counter. This effect was measured during runs with and without DN detector in place and a correction factor (approximately 7%) is applied.

3 Data analysis

3.1 Data reduction and analysis

During measurement, each ^3He analogue signal was registered by an ADC (amplitude to digital converter) module after amplification. A 1 kHz clock was used to determine the time of the event with

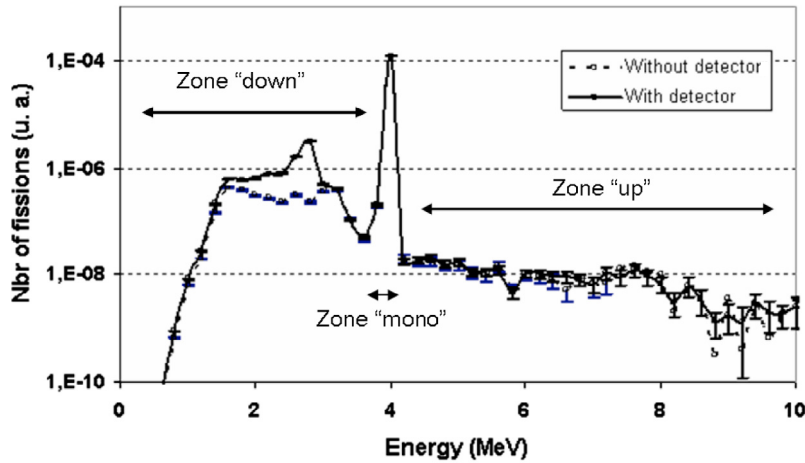


Fig. 2. Simulated number of fissions in the sample as a function of the energy of the neutrons for 4 MeV incident neutrons.

respect to the beam switch off time. The first step of the analysis is the selection of neutron-induced events by placing a cut on the energy spectrum of each ^3He tubes. Secondly, the T_0 of each time spectrum is determined cycle by cycle before summing the time spectra from all counting sequences. Time spectra measured during runs without thorium sample are then subtracted (after normalisation) to take into account the background contribution. The number of delayed neutrons at $t = 0$ (beam-off switching time) is determined by fitting the decay time spectrum with the function defined in Eq. (1) and the parameters listed in Table 1. Only the contributions of the groups 4, 5 and 6 have been considered to fit the curve since their contribution is predominant.

3.2 Simulations

As shown in Eq. (4), the ν_d determination requires the knowledge of the number of fissions in the sample. It is obtained from the fluence measurement combined with MCNPX transport code calculations using a detailed model of the set-up containing the thorium sample, the DN detector and the neutron source. The neutron source angular and energy distributions have been calculated with the DROSG code [10]. The Figure 2 represents the simulated number of fissions in the sample with and without (dashed line) the neutron detector. Three energy domains appear clearly: at the incident energy (mono), above (up) and under (down) this value. The “mono” zone corresponds to fissions induced by neutrons coming directly from the source. The fissions in the “up” zone are induced by fission neutrons. The fissions in the zone “down” are due to neutrons slowed down in the thick thorium sample or to neutrons scattered in the CH_2 of the detector. The influence of the scattering of neutrons on the detector is shown by the difference between the two curves. Whatever the incident energy, the additional number of fissions due to the detector presence is never larger than 5%. This low contribution is obtained thanks to the optimized sample position close to the “entrance” of the detector.

Since the detected DN originate from the fissions in these 3 zones, then:

$$F_{\text{det}}(0)/\varepsilon = DN^{\text{mono}} + DN^d + DN^u$$

The average number of DN at neutron source energy (zone “mono”) is:

$$\nu_d^{\text{mono}} = \frac{F_{\text{det}}(0)/\varepsilon - \nu^u N_{\text{fission}}^u - \nu^d N_{\text{fission}}^d}{N_{\text{fission}}^{\text{mono}}}$$

where $N_{fission}^{mono}$, $N_{fission}^u$ and $N_{fission}^d$ are calculated with MCNPX for the energy ranges previously defined. ν^u and ν^d are taken from the Yoshida evaluation (see Figure 1) averaged on zones “up” and “down”. The total corrections due to the fissions in the “up” and “down” zones depend on the energy but are in any case (except at 10 MeV) smaller than 7%.

Contrary to the other energies, the measurement at 10 MeV was not performed with purely monoenergetic neutrons. In addition to the monoenergetic neutrons from the $D(d, n)^3He$ reaction, the deuteron break-up reactions produces a low energy continuous component which has to be taken into account. The same procedure as previously described, was used including the low energy neutron component in the source. This component was simulated with the energy distribution measured at zero degree and the angular distribution of the $^2H(d, n)^3He$ reaction. In that case the correction due to the break-up component and the target thickness is around 10%.

4 Preliminary results

The Figure 3 represents the preliminary results for the absolute DN yield as a function of the incident neutron energy. A coherent set of measurements is obtained between 2 and 16 MeV. To our knowledge, the points between 4 and 10 MeV were measured for the first time. The uncertainties take into account the flux measurement (2%) and the detection efficiency (5%). A more detailed treatment of the uncertainties is still needed particularly by adding the fission rate calculations uncertainties.

The absolute DN yield is stable between 2 and 4 MeV, it decreases between 4 and 6 MeV and is stable up to 16 MeV. This behaviour is comparable to the one observed for ^{235}U , ^{238}U and ^{239}Pu neutron induced fission [11]. The decrease of the DN yields around 4 MeV is not yet well understood. It could be explained by the opening of the second chance fission channel. Actually, above the 2nd chance fission threshold, the fissioning nucleus is less neutron-rich. The fission fragments distribution is modified and less (β, n) emitters are produced. There are other attempts based on even-odd effects or sharing on the excitation to explain this decreasing yield [12, 13] but nevertheless this is still an open question.

Our data are in quite good agreement with published data between 2 MeV and 3 MeV and are a little bit lower at 16 MeV. We observe that the predictions of the ENDF and the JENDL library are in good agreement with our data.

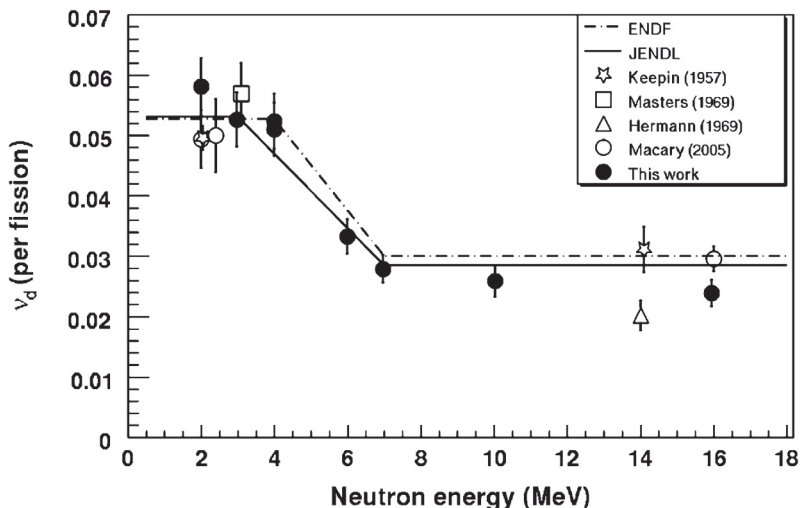


Fig. 3. Absolute DN yield as a function of the incident neutron energy. Data of this work are represented by full symbols, other works by open symbols and evaluations by full and dashed lines.

5 Conclusions

The absolute DN yield emitted in the neutron induced fission of ^{232}Th has been measured at 2, 3, 4, 6, 7, 10 and 16 MeV. The energy range between 3 and 10 MeV is particularly interesting since no data have been published in this range yet. The experimental setup composed of twelve ^3He counter embedded in a polyethylene cylinder has been described. The analysis method and the normalisation procedure have been explained. Sources of uncertainties have been mentioned.

The energy dependence of DN yields presented here show the same behaviour than the one observed for uranium and plutonium isotopes: a plateau for incident neutron energies lower than 4 MeV, a decreasing slope between 4 and 7 MeV and a second plateau between 7 and 16 MeV. This behaviour and the absolute values are in quite good agreement with the ENDF and JENDL libraries. Moreover data presented in this paper are in agreement with published data for the low energy part.

This work was supported by the EURATOM 6. Framework Programme “European Facilities for Nuclear Data Measurements” (EFNUDAT). We also would like to thank the PTB Ion Accelerator Facility teams.

References

1. T. Yoshida, private communication
2. G.R. Keepin et al., *Phys. Rev.* **107** (1957)
3. C.F. Masters et al., *Nuclear Science and Engineering*, **36**, 202–208 (1969)
4. S.A. Cox, Proc. Panel Delayed Fission Neutrons, International Atomic Energy Agency, Vienna (4968)
5. V. Macary, Thèse de doctorat, Université Pierre et Marie Curie, 2008, report IRFU-08-02-T
6. H.J. Brede et al., *Nucl. Instr. and Methods* **169** (1980) 349–358
7. R. Nolte et al., *Radiat. Prot. Dosim.* **110** (2004) 97–102
8. D. Doré et al., *Proceeding of the International Conference on Nuclear Data for Science and Technology*, Nice, France, April 22–27, 2007
9. J. Hendricks, MCNPX extensions : versions 2.5.0, LANL, USA
10. DROSG-2000, IAEA, <http://www-nds.iaea.org/drosg2000.html>
11. T. Yoshida et al., *Journal of Nuclear Science and Technology*, Supplement 2, p. 136–139 (2002)
12. D.R. Alexander et al., *Nuclear Science and Engineering*, **62**, 627 (1977)
13. T. Ohsawa et al., *Proceeding of the International Conference on Nuclear Data for Science and Technology*, Nice, France, April 22–27, 2007