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Measurements of delayed neutron yields from neutron induced fission on ^{232}Th

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Abstract. The total delayed neutron (DN) yields from neutron induced fission on ^{232}Th were measured for neutron energies of 2.0, 2.4 and 16.0 MeV. In this paper, the experimental results are presented and compared to existing experimental data and evaluated data libraries. The future measurements are outlined.

1 Introduction

According to the last recommendations of the 6th subgroup of OECD/NEA NSC working party on International Evaluation Cooperation, the measurements of delayed neutron (DN) yields and time spectra relative to innovative critical reactors are needed for some of the Pu isotopes, minor actinides and nuclei contributing to the Th fuel cycle with 5 % data accuracy [1]. For the moment, a number of experiments have been performed to determine the total DN yields from neutron induced fission on ^{232}Th. The results of most of these experiments are presented in fig.1 [2,3].

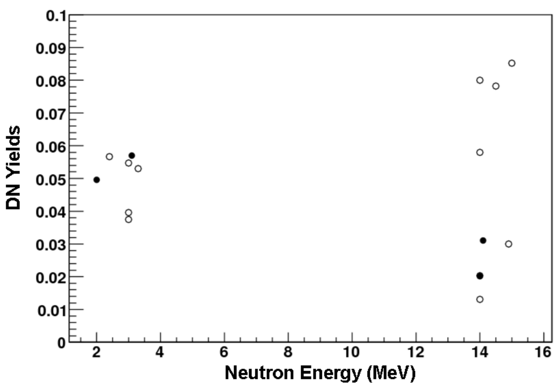

Fig. 1. Delayed neutron yield, υ_d, data (in delayed neutrons per fission) for neutron induced fission on ^{232}Th.

We can notice significant discrepancies of data for the incident neutron energies below 4 MeV. Above 14 MeV, the data are even more dispersed. In consequence, data retained for evaluations [4] are scarce (solid dots) and their dependence on incident neutron energy is not known precisely. Indeed, as presented in fig. 1, in the neutron energy range between 4 and 14 MeV up to now there are no experimental data available. Therefore, a DN yield measurement programme has been undertaken in order to measure DN yields from ^{232}Th with incident neutron energies from 4 to 8 MeV. First experiments were performed for several neutron energies, namely at 2.0, 2.4 and 16.0 MeV in order to compare our results to previous data. We note separately that new measurements were also made with neutrons between 3.8 and 5.0 MeV but suffered from severe background and were not suitable for final analysis.

In this paper, our experimental set-up is presented, the measurement strategy is explained, including the description of the data analysis. Finally, preliminary experimental results are presented and compared to previous experimental data and evaluated data libraries.

2 Experimental procedure

2.1 Experimental set-up

The experiments were performed using the 4 MV Van de Graaff Accelerator at Bruyères-le-Châtel of CEA/DIF/DPTA. This accelerator was used to deliver continuous proton and deuteron beams. These beams impinged on titanium-tritium target (TiT) in order to produce mono-energetic neutrons. To produce irradiation-decay cycles (see next section) the beam was stopped periodically by a shutter placed several meters upstream of the production target. The neutron flux was measured by a reference neutron detector BF3. This flux was about $9.5 \cdot 10^6$ n/s for neutron energy of 2 MeV and at the position where the ^{232}Th sample (300 g) was placed. Figure 2 presents a schematic view of the experimental arrangement. The thorium target was a cylinder of 2.6 cm diameter and 5 cm thickness. It was placed at 20° from the accelerator beam axis (like the BF3 monitor detector) and its distance to the neutron production

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target was 10 cm as shown in fig.2. DN detection system, placed at 10 cm from actinide sample, was composed of four ^{3}He counters under pressure of 4 atmospheres. The principle of neutron detection is based on gas ionization via (n,p) reaction. The ^{3}He counters of 30 cm long and 2.5 cm in diameter were placed in two polyethylene blocks in order to increase the neutron detection efficiency in terms of neutron moderation. Finally, to avoid the background due to the thermal neutrons reflected from the concrete walls, both polyethylene blocks were surrounded by 1 mm Cadmium foils. Optimization of the polyethylene thickness, done with MCNPX [5], aimed to obtain a constant efficiency in the range of DN energy from 0.1-1.2 MeV [6,7]. Our calculations showed that 5 cm of thickness allow a good DN moderation leading in the detection efficiency of 2.4 %. Model calculations were also verified using detector efficiency measurements with the standard Cf and AmBe neutron sources.

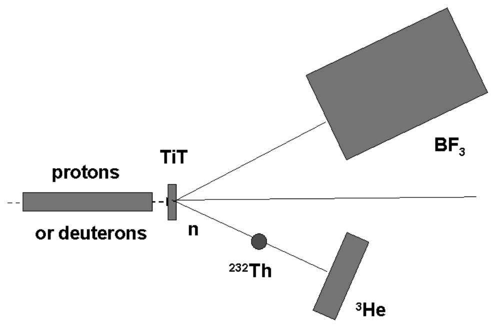

Fig. 2. Schematic view of the experiment for DN measurements from neutron induced fission.

2.2 Measurement strategy

It is well known that DN emissions follow the beta decay of some fission fragments which are called precursors. The precursors are generally lumped in 6 groups according to their half-lives [3]. Then, the time dependence of DN yields ($Y_{DN}(t)$) after an infinite irradiation, i.e. the irradiation duration much longer compared to the longest precursor period, can be represented as a sum of six exponentials:

$$Y_{DN}(t) = \nu_d \sum_{i=1}^{6} F_i e^{-\lambda_i t} \ , \tag{1}$$

where $\lambda_i = \ln 2/T^i_{1/2}$ is the decay constant with $T^i_{1/2}$ the half-life of the ith group, F_i the contribution of ith group and ν_d the total DN emitted per fission. At the very first stage, our measurement strategy was to irradiate the sample for five minutes, in order to reach the equilibrium level even for the longest lived DN precursors. Afterwards, this long continuous irradiation is followed by a number of periodic irradiation-decay cycles, (7s-1s), in order to keep precursors at equilibrium. Finally, the DN yield was determined from the detection system counting rate at t = 0, i.e. the DN yield ν_d can be written according to the following equation:

$$\nu_d = \frac{N_{^3He}(0)}{\varepsilon \cdot N_{fiss}} . \tag{2}$$

In this equation $N_{^3He}(0)$ is the total accumulated neutron counts in detectors at t = 0 s, i.e. right after the beam is switched off. Note that this value had to be corrected for the background, obtained from equivalent measurements but without thorium sample. ε is the system detection efficiency and N_{fiss} is the accumulated number of fissions in the sample during the irradiation. The number of fissions was evaluated using the MCNPX code calculations normalized to the neutron fluence recorded with the online BF_3.

3 Measurements and results

3.1 Measurements and analysis

We performed the DN yield measurements for neutron energy of 2.0 MeV in order to test the measurement strategy and to have a measurement which could be compared to previous data. Similar quality measurements were also done at 2.4 MeV and 16.0 MeV. Our try to obtain the DN measurements in the incident neutron energy range of 3.5-5.0 MeV was not possible due to significant background (more than 50 %), generated by the deuteron beam interaction on the shutter. Fig.3 presents a typical DN time spectrum for the measurement with neutron energy of 2.0 MeV. Note that only the very last part of the spectra with beam on was recorded.

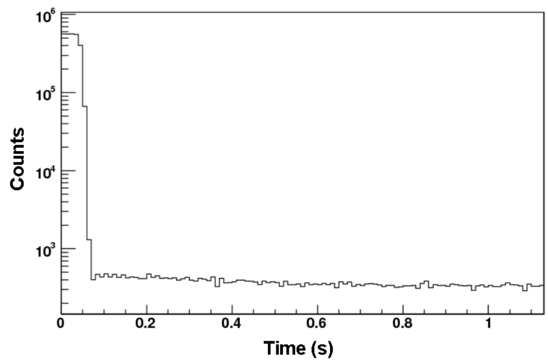

Fig. 3. Experimental DN spectrum for the 2 MeV incident neutrons on ^{232}Th.

Due to the quite long relaxation time of our detection system

(~30-40 ms) we could not obtain directly the DN counts at t = 0 s, i.e. $N_{^3He}(0)$. This value had to be extracted by performing the fitting procedure of the decay time spectrum (fig. 3). During the decay following irradiation, the decay of groups 4, 5 and 6 will be visible and the other groups will have a constant contribution (their half-lives being much longer then 1 s). Therefore, we fit the decay curve with expression (3) (least square fit method):

$$f(t) = a + be^{-\lambda t}, \qquad (3)$$

where *a* is the constant contribution of groups 1, 2 and 3, and $be^{-\lambda t}$ represents the total contribution of groups 4, 5 and 6. Then, the DN counts at t=0 is the value of *f(t)* at t=0.

3.2 Results

Results of this work, corresponding to the DN yield for ^{232}Th for incident neutron energy of 2.0, 2.4 and 16.0 MeV are shown in fig.4. In brief, the DN yield value measured at 2.0 MeV is in agreement with the one reported by Keepin et al. [3], our value being lower only by 9 % but within error bars. Note that there was no change of this value observed with energy increase from 2.0 MeV to 2.4 MeV (see our 2nd point at 2.4 MeV). The DN yield measured at 16.0 MeV is also consistent with the value retained by the evaluated data libraries (Yoshida et al.) as shown in fig.4.

Error propagation formula was used to calculate measurement errors. These errors arise from several causes. The most important is the statistical errors (75-80 % of the errors) due to the monitoring of the beam with BF_3 and to the DN counting. Other contributions of the errors arise from fission cross-section and system detection efficiency uncertainties.

In brief, our measurements support the evaluated data files at low and high energies but does not bring new information for the energy region, where DN yield decreases with energy, i.e. between ~3 and ~ 7 MeV (see fig. 4). See also the legend for details. Recent experiments have been performed in this energy domain but analysis is still in progress.

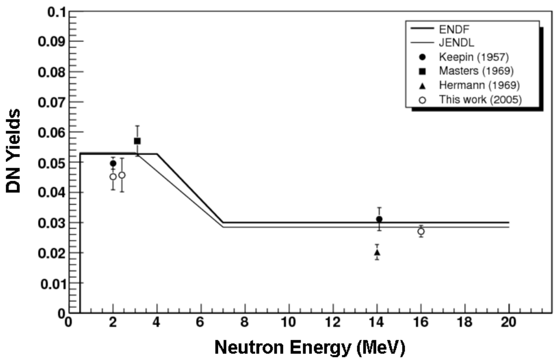

Fig. 4. Experimental data (points) and evaluations (solid lines) of DN yields as a function of neutron energy for ^{232}Th.

4 Summary and outlook

We launched an experimental programme to measure fission DN yields for ^{232}Th as a function of incident neutron energy. The first results at 2.0, 2.4 and 16.0 MeV confirm previous measurements and values selected for the data evaluation files as JENDL and ENDF. In addition, it proves the correctness of our experimental and data analysis procedure.

Recently we performed new measurements of DN yields of ^{232}Th for neutron energies of 4.2, 4.8, 5.2 and 7 MeV. The analysis of this new experiment is in progress. These measurements will help to complete the evaluated data file for ^{232}Th and should improve the uncertainty of the DN yield evaluations in the neutron energy region around the threshold of the 2nd chance fission.

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References

1. V.G. Pronyaev, *Meeting on Assessment of Nuclear Data Needs for Thorium and other Advanced Cycles*, IAEA Headquarters, Vienna, Austria, August 1999.
2. J. Meadows et al., *Evaluated Nuclear Data File for ^{232}Th,* Argonne National Laboratory, February 1978.
3. G.R. Keepin at al., *Delayed Neutrons from Fissionable Isotopes of Uranium, Plutonium and Thorium*, Physical Review 107, 1957.
4. T. Yoshida et al., *Evaluation of Delayed Neutron Data for JENDL-3.3*, Journal of Nuclear Science and Technology, Volume 1, Supplement 2, p. 136-139 (August 2002); T. Yoshida and T. Nakagawa, private communication (2004).
5. J. Hendricks, *MCNPX extensions : version 2.5.0*, LANL, USA, April 2005.
6. D.Doré et al., *Delayed neutron yields and spectra from photofission of actinides with Bremsstrahlung photons below 20 MeV*, EPS Journal of Physics: Conference Series 41 (2006) 241.
7. M.-L. Giacri-Mauborgne, Ph.D. thesis, University of Caen, 2005; CEA Saclay, report DAPNIA-05-14-T.