International Conference on Nuclear Data for Science and Technology 2007
DOI:

Delayed neutron yields and spectra from photofission of actinides: data and calculations with Bremsstrahlung photons below 20 MeV

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Abstract. Photonuclear reactions, especially photofission delayed neutron emission, play an important role for applications involving nuclear material detection. In order to provide good quality data for evaluations and data libraries, an experimental programme of delayed neutron measurements has been undertaken for actinides with Bremsstrahlung energy from 12 up to 20 MeV. In this paper, we report on delayed neutron yields and time spectra characteristics from the photofission of ^{238}U and ^{235}U for several endpoint energies between 12 and 18.5 MeV. The associated modelling efforts will be described including some calculations compared to experimental data.

1 Introduction

A renewed interest in photonuclear reactions was stimulated by applications as radioactive ion beam production, shielding of electron accelerators, production of intense neutron beams, etc. Today, a particular attention is paid to the non-destructive characterization of waste barrels and detection of nuclear materials, both based on photo-fission process and the associated delayed neutron (DN) emission. Need of accurate and complete data for DN yields and time characteristics of actinides was the motivation for the experimental campaign, started in 2004. The experimental effort is supported by a complete modelling of the process.

The contributions of around 200 DN precursors are generally lumped into 6 groups according to their half-lives $t_{1/2}$ going from 50 ms to 55 s. For each group i, two parameters are determined: the DN contribution a_i and the mean decay constant of the group λ_i ($\ln 2/t_{1/2}$) leading to a simplified description of the DN decay curve through the relation

$$Y_{DN}(t) = \sum_{i=1}^{6} a_i e^{-\lambda_i t}(1 - e^{-\lambda_i T_{irr}})$$

where T_{irr} is the irradiation period. The sum of all a_i is called ν_d and is characteristic for a fissionable nucleus. For photo-fission experiments, a_i, λ_i and ν_d are extracted from time dependent DN measurements. T_{irr} and T_{dec} (decay time) are varied to enhance alternatively each group contribution with respect to others. From 2004 to 2006, four experiments have been performed with different actinides (^{238}U, ^{232}Th, ^{235}U) at different electron energies (from 12 to 18.5 MeV) [ref. 1-3] and with three different experimental setups. In this paper, we describe 2006's experiments and show preliminary data for absolute yield determination and group parameter extraction. Modelling work [ref. 3-4] is also briefly presented and results compared to experimental ones. Finally, importance of precise measurements for group parameters is emphasized with an example aiming at the isotopic identification of a mixed sample.

2 Experimental setup and method

The ELSA electron accelerator of CEA/DIF/DPTA was used to provide end-point Bremsstrahlung photons with energies from 12 to 19 MeV. A simplified scheme of the experiment is shown on Fig. 1. The electron beam, with intensity around 1 µA and energy resolution ~100 keV, impinges on the 2 mm thick Ta target. The Al cylinder stops the residual electrons. A Pb collimator placed at 0 degree defines the photon beam opening angle. At 2.34 m (target position), the photon beam spot has a diameter of 6.7 cm and the beam intensity is around 10^8 γ/cm^2.

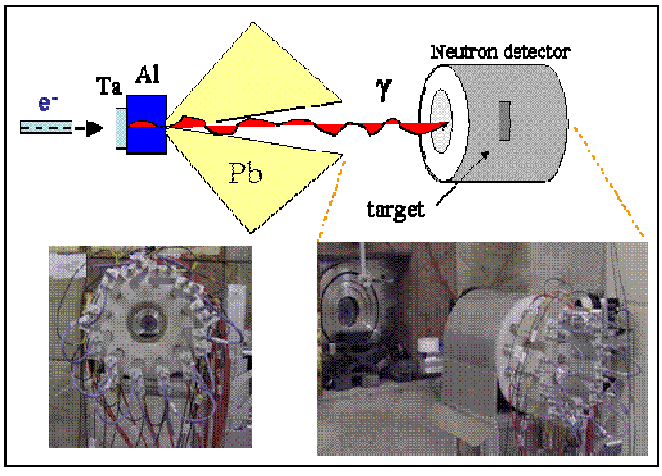

Fig. 1. Top**:** experimental setup for DN measurements.
Bottom: photos of the DN detector..

First experiments were performed with a basic neutron

detector (low efficiency) and important target masses (400 g). Since 2006, due to the low mass of actinide available and limited beam intensity (due to radioprotection constraints), a new detector has been designed and constructed (fig. 1). Twelve ^{3}He gas counters were uniformly distributed inside a CH_2 ring of 37 cm long with inner and outer radius of 6 and 16 cm respectively. The ensemble was wrapped in a 1 mm thick Cd foil to avoid low energy background neutrons to be detected. CH_2 thickness has been optimized to obtain a constant efficiency over the expected DN energy range (0.1 to 1.0 MeV). Within this configuration an efficiency of ~20% is obtained, a gain of a factor 10 compared to our previous DN detectors.

The experimental method consists of repetitive cycles of irradiation and decay periods. To extract group parameter three different combinations of irradiation-decay times are performed: 300 s - 300 s; 10 s - 100 s and 100 μs - 40 s. For absolute DN yield measurements, the short irradiation is used.

2.1 Absolute DN yields

In Novembre 2006, the whole range of electron energy between 12 and 18.5 MeV has been covered to measure absolute DN yields for ^{238}U (400 g) and ^{232}Th (300 g). For ^{235}U (11 g), only few points have been studied. A typical decay spectrum of ^{238}U DN yields, obtained after 70 repetition cycles with 100 μs - 40 s irradiation-decay sequences, is presented in fig. 2.

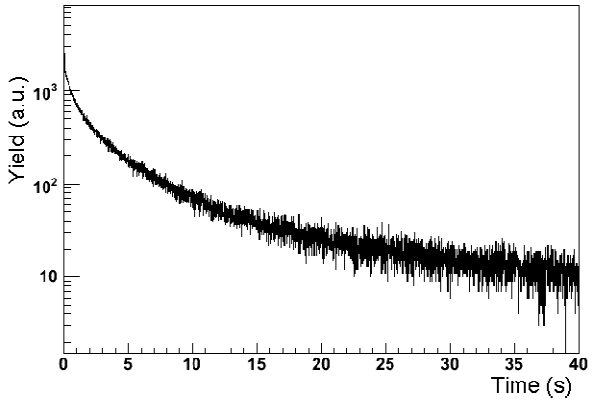

Fig. 2. DN time dependent yield for ^{238}U after 70 cycles with T_{irr}=100 μs.

For a T_{irr} << shortest $T^i_{1/2}$, the absolute yield (ν_d) is the ratio of the emitted DN over the number of fissions. The number of DN is the integral of the curve on fig. 2 corrected for background, detector efficiency and contribution of (n,fiss) reactions. The number of fissions (induced by photon and neutrons) is evaluated using the MCNPX transport code. The total beam charge was recorded during the irradiations. The exact geometrical description of the set-up (converter; collimator and sample) is taken into account in the simulation. Figs. 3 and 4 show preliminary results of total DN yields (full symbols) for the ^{238}U and ^{235}U targets. Error bars are only statistical. The systematic uncertainty due to the electron charge measurements is difficult to evaluate and has not yet been taken into account. Previous results of Nikotin et al. [ref. 5] at 15 MeV are also shown in figs. 3 and 4 (open circles). As can be seen from these comparisons, our data are coherent with Nikotin's data giving the uncertainties which are reduced. Some other experimental data are also plotted on figs. 3 and 4 [ref.6-7]. Here again, within the error bars, all data are consistent with our preliminary results. We note separately that in the case of the ^{235}U target, containing 7% of ^{238}U, the contribution of ^{238}U(γ,f) had to be subtracted. This will be done after complete analysis of the present ^{238}U data.

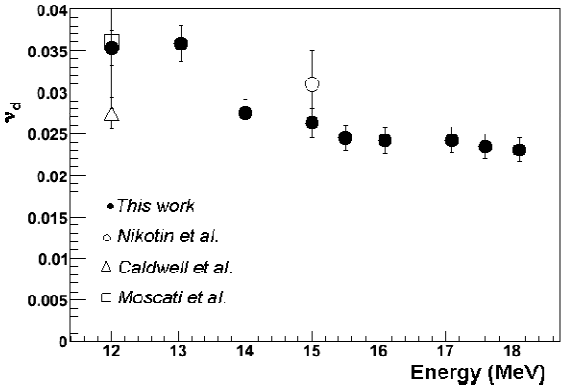

Fig. 3. Absolute DN yields for ^{238}U as a function of electron energy.

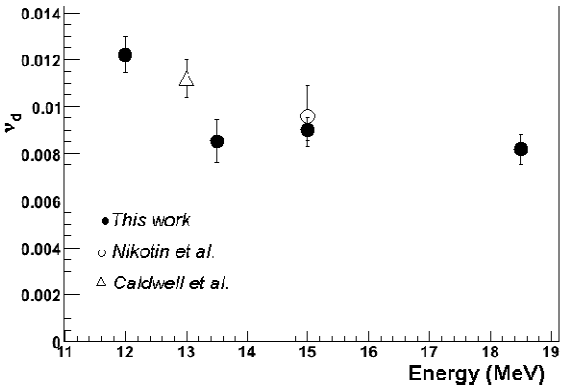

Fig. 4. Same as Fig. 3 but for ^{235}U.

The behaviour of ν_d as a function of the electron energy is also of our primary interest. Trends are similar for ^{235}U and ^{238}U with a decrease of ν_d around 13 MeV. This decrease is generally interpreted as the opening of the 2nd chance fission channel (E_{thr},~12 MeV) but more precise interpretation has still to be studied.

2.2 Group parameters

Available data for ^{238}U DN six group parameters show important discrepancies. For example, Kull et al. [ref 8] have reported an important difference for contribution of groups 4 and 6 in respect to Nikotin's data. It can be seen in

fig. 5, where there is a factor of two between Kull's and Nikotin's data for these groups. Kull et al. have attributed these discrepancies to the opening of the 2nd chance fission around 15 MeV.

We have determined group parameters for ^{238}U at 15 and 18 MeV previously. As mentioned earlier, in order to extract a_i and λ_i for all groups, three types of irradiation are used. First, for the 300-300 s combination, the parameters for groups 1 and 2 have been evaluated. Then, these parameters have been fixed and from the second type of irradiation (10-100 s), parameters of groups 3 and 4 have been extracted. Finally, using the short irradiation spectrum, 5th and 6th group parameters have been determined. Results shown on fig. 5 for 15 MeV (black squares) and 18 MeV (black points) have confirmed Kull's data. It seems that there is no evolution of a_i between 8 and 18 MeV. This observation concerning group 5 and 6 is particularly important for applications and will be discussed in section 3.

For ^{235}U (fig. 6), Kull's data at 8 and 10 MeV (open squares and triangles) are rather close except for group 3 and 5. However, discrepancies with Nikotin's data appear for almost all groups. Our analysis for ^{235}U is still in progress but already seems to indicate that here again Nikotin's data overestimates the 6th group contribution.

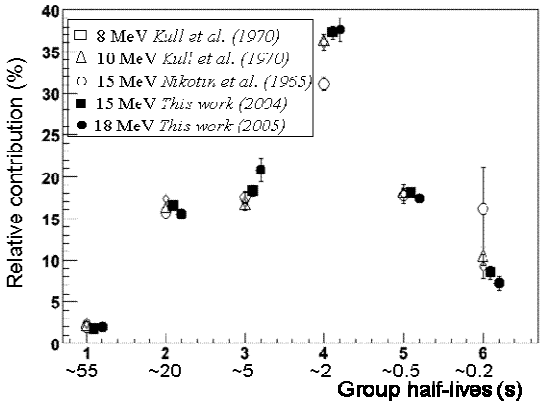

Fig. 5. Relative DN group contributions for photofission of ^{238}U.

3 Modeling

The modeling work is an important step in order to provide group parameter for all actinides. The whole process has been taken into account in the developed calculations. First of all, the Bremsstrahlung photon spectrum inside the actinide sample is calculated with the MCNPX code [ref. 9] taking into account the exact experimental set-up. In the γ-excitation of nucleus we considered only the Giant Dipole Resonance contribution, where an empirical systematic based on the sum of two Lorenzian distributions seems to describe this process properly below 20 MeV.

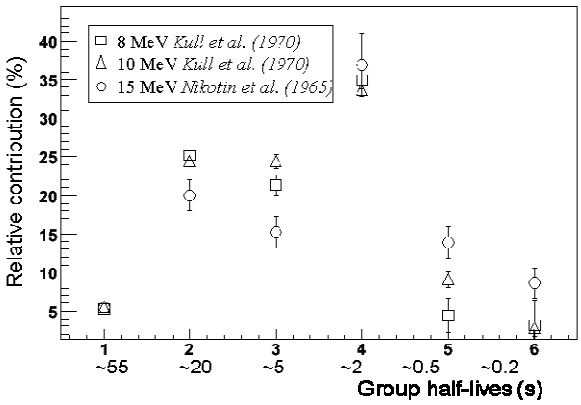

Fig. 6. Same as Fig. 5 but for ^{235}U.

The de-excitation of the nucleus is treated by the ABLA model developed at GSI [10] known to give good results in the case of high energy spallation reactions. Competition between particle emission and fission is treated by a statistical model. In other words, the complete code provides neutron emission, fission cross section and also fission yields. Note that the multi-chance fissions are taken into account as well. At this step, some experimental fission mass distributions are available to be compared to predicted ones. In fig. 7, ^{235}U mass yields show a good agreement with Jacobs and Thierens data [ref. 11-13]. Similar quality is also obtained for ^{238}U, ^{237}Np and ^{239}Pu. Isotopic yields, closely related to our observable, i.e. DN yields, are presented in [ref. 3].

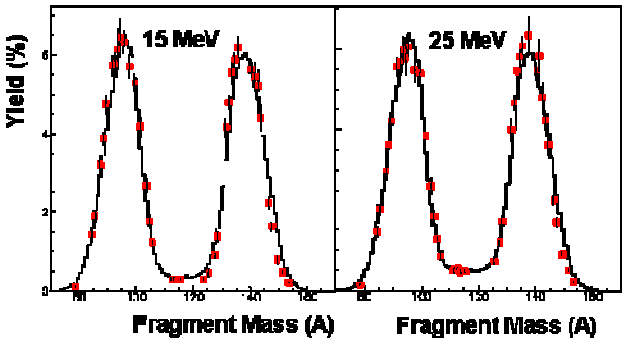

Fig. 7. Predicted and measured mass distributions for photofission of ^{235}U at 15 and 25 MeV.

To treat the calculation of DNs, the independent fission yields predicted by ABLA are transferred into cumulative yields using the CINDER'90 transmutation-decay code [14]. The DN precursors are identified and selected according to the nuclear data tables. Using tabulated DN emission probabilities and half-lives, all precursors are merged into six DN groups according to their half-lives. In this way the model provides a full set of DN parameters (total yield ν_d, six group yields a_i and averaged group time constants λ_i).

Calculated group yields (stars) for ^{235}U are compared to experimental data (triangles) in Fig. 8. The general agreement is good and especially for group 5 and 6, which have an important role for applications such the one

described below. Discrepancies for groups 3 and 4 seem to be systematic since it has also been observed for ^{238}U and ^{232}Th. This shift has to be investigated.

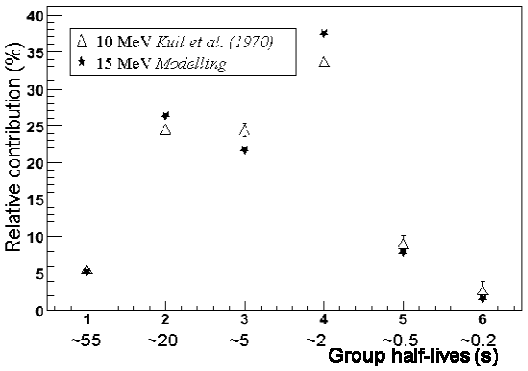

Fig. 8. Relative DN group contributions for photofission of ^{235}U.

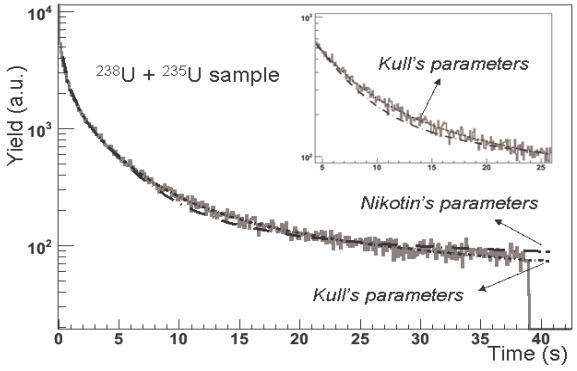

Fig. 9. DN time dependent yield for a mixture of ^{235}U and ^{238}U at 15 MeV.

4 Isotopic Identification

Irradiation of actinide mixtures was also performed in order to test the possibility to determine the composition and the mass of unknown composite samples. Precise measurements for the 5th and 6th group parameters are very important for such identification since short irradiation is used, as it would be probably the case for an eventual application. In fig. 9, DN time dependent yield of a sample composed of 11 g of ^{235}U and 10 g of ^{238}U is shown. Hundred cycles of Tirr=100 μs – T_{dec}=40 s were performed. Our analysis shows that using only one baseline (one actinide) it is impossible to reproduce the experimental curve. Taking into account both actinides, the experimental time dependence is obtained by fitting only the background and mass of both actinides. Using parameters of Nikotin for ^{238}U and ^{235}U, the fit (dashed line) does not reproduce the experimental curve since the ^{238}U contribution is set to a negative value. Fortunately, with Kull's parameter (dotted line) data are well reproduced. This result argues in favour of Kull's data analysis both for ^{235}U and ^{238}U. The next step, still in progress, will be the mass evaluations.

5 Conclusions

An experimental campaign launched in 2004 for actinide DN measurements from photofission below 20 MeV is giving interesting results. Absolute DN yields for ^{238}U, ^{232}Th and ^{235}U have been obtained for several Bremsstrahlung endpoint energies. Data are precise and energy dependence becomes clearer. Our experimental values will be included in the evaluated files obtained by E. Dupont et al. (in this conference) and to be provided to the JEFF community. For group parameters, the situation for the 6th group of ^{238}U and ^{232}Th was clarified and old values at 15 MeV were corrected. Preliminary results for ^{235}U seem to indicate the same feature. These new parameters allow determining the actinide content of a mixed actinide sample. Many mixtures with different proportions have been studied and analysis is in progress. Modelling has also been improved and isotopic yield as well of mass distributions are rather well reproduced by the calculations. Systematic differences still have to be explained.

In 2007 and 2008, ^{237}Np and ^{239}Pu targets will be available, and therefore absolute yields and group parameters will be measured for some Bremsstrahlung endpoint energies. These measurements will be done in parallel with detection of photofission delayed photons (DP). These experiments aim providing the DP/DN ratios in the frame of the INPHO project presently in progress at CEA.

Acknowledgements

Authors thank the staff of the ELSA accelerator for their support and fine working conditions during the experiments

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