

BNL-78163-2007-CP

Measurement of the average energy and multiplicity of prompt-fission-neutrons from ²³⁸U(n,f) and ²³⁷Np(n,f) from 1 to 200 MeV

J. Taieb, T. Granier, T. Ethvignot, M. Devlin, R.C. Haight, R.O Nelson, J.M. O'Donnell, D. Rochman

¹ CEA Bruyeres le Chatel, DPTA/SPN, Domaine du Grand Rué, BP12, 91680 Bruyères le Châtel, France ² LANSCE, Los Alamos National Laboratory, MS H855, Los Alamos, New Mexico 87545, USA

> Energy Sciences & Technology Department National Nuclear Data Center Brookhaven National Laboratory P.O. Box 5000 Upton, NY 11973-5000 www.bnl.gov

Notice: This manuscript has been co-authored by employees of Brookhaven Science Associates, LLC under Contract No. DE-AC02-98CH10886 with the U.S. Department of Energy. The publisher by accepting the manuscript for publication acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or any third party's use or the results of such use of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof or its contractors or subcontractors. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

Measurement of the average energy and multiplicity of prompt-fissionneutrons from ²³⁸U(n,f) and ²³⁷Np(n,f) from 1 to 200 MeV

J. Taieb^{1,a}, T. Granier¹, T. Ethvignot¹, M. Devlin, R.C. Haight, R.O Nelson, J.M. O'Donnell, D. Rochman

¹ CEA Bruyeres le Chatel, DPTA/SPN, Domaine du Grand Rué, BP12, 91680 Bruyères le Châtel, France ² LANSCE, Los Alamos National Laboratory, MS H855, Los Alamos, New Mexico 87545, USA

Abstract. Taking advantage of the neutron source of the LANCSE, it has been possible to obtain a measure of the velocity distribution and the number of prompt-neutrons emitted in the neutron-induced fission of ²³⁸U and ²³⁷Np over a broad incident neutron energy range. The mean kinetic energy was extracted and is shown as the function of the incident-neutron energy. We confirm here the observation, for both reactions, of a dip around the second-chance fission which is explained by the lower kinetic energy of the pre-fission neutrons. Such a observation is reproduced by Los Alamos model as implemented at Bruyeres le Chatel and by the Maslov model. As far as the neutron multiplicity is concerned, a similar dip is observed. However, such a behaviour is not present in data measured by other groups.

1 Introduction

On one side, a precise knowledge on prompt-fission neutron properties is of major importance for simulating nuclear reactors. Indeed, the number of emitted neutrons during the fission process as well as their spectra have been consequently theoretically and experimentally studied for 60 years. These studies mainly concern a few actinides of importance in commonly exploited nuclear systems. The forthcoming generation of nuclear reactors (GEN4) will probably run, taking advantage of a fast neutron spectrum to incinerate minor actinides like ²³⁷Np or various americium and curium isotopes. Moreover, high-energy neutron induced fission must be specially modelled in the so-called accelerator-driven systems (ADS) which are currently designed. The latter may be used in the future to incinerate specific waste in dedicated facilities. On the other side, new knowledge on properties of neutrons produced in fission of actinides at moderate excitation energies contributes for instance to the understanding of the competition between neutron and Light Charged Particle statistical emission. Several groups (refs. [1,2]) have pursued some efforts to develop theoretical descriptions in order to estimate the fission prompt-neutron properties. Those descriptions are based on the energy conservation considerations and statistical calculations for estimating the competition between the open deexcitation channels.

Our collaboration already published (see refs. [3,4]) recent experimental results concerning prompt fission neutron spectra (PFNS) emerging from the neutron induced fission of ²³⁸U and ²³⁵U. Those results have been obtained taking advantage of a similar experimental set-up. We decided to measure again fission neutron properties for neutron induced fission of ²³⁸U and study in addition the fission of ²³⁷Np.

2 The experiment

2.1 The set-up

It was performed at the LANSCE using the fast neutrons produced by the WNR spallation source. The 800 MeV proton beam impinges on a tungsten target producing highenergy neutrons. Even if most of the neutrons are produced in the 1-10 MeV range, the high energy tail allows to study neutron induced reactions up to 300 MeV incident energy. The proton-beam structure consists of 300ps pulses spaced every 1.8µs.

The FIGARO experimental set-up (see ref. [5] for an extensive description of the set-up) is located 22m away from the spallation target allowing a precise determination of the kinetic energy of the incoming neutrons in measuring their time-of-flight (ToF). A time signal is provided by a 10cm long ionisation chamber containing 94 plates where 380mg of pure actinide (resp. ²³⁸U or ²³⁷Np) were deposited. The neutrons emitted in coincidence with the detection of fission fragment were detected thanks to 20 scintillation neutron detectors located 1 meter far from the fission chamber covering angles ranging from 45 to 135 degrees. A pulse-shape analysis permitted to fully disentangle gammas from neutrons. In the previously reported experiment (refs. [3] and [4]), only 3 neutron detectors were available. This improvement made possible a better angular coverage and allowed for a higher statistics. Thus, an additional ToF signal was recorded, the start signal being given by the fission chamber as the stop was obtained from the touched neutron detector. The kinetic energy of the fission neutrons were consequently recorded providing us with the prompt fission

^a Email address : julien.taieb@cea.fr

International Conference on Nuclear Data for Science and Technology 2007

neutron spectra (PFNS). The ToF resolution of the fission chamber was of 6ns. The energy resolution of the latter allowed for disentangling alpha emission from real fission events. The 20 EJ301-type neutron detectors were made up with cylindrical cell filled with a scintillating liquid.

2.2 The data reduction

The main issue is here to determine the efficiency of the neutron detectors. It strongly varies with the kinetic energy of the detected neutrons. It yields to 0 below 0.5 MeV and increases to reach a plateau around 2 MeV. Then, it slowly decreases. We calibrated individually all 20 detectors using the ²³⁸U run. Boykov et al. (ref. [6]) measured accurately the PFNS in the 2.9MeV neutron induced fission of ²³⁸U. Therefore, we gated on the incident neutron spectrum to select only 2.9±0.1 MeV neutrons and obtained with that condition raw fission neutron spectra for all neutron detectors. The raw spectra were divided energy bin per energy bin by the Boykov spectrum to get the efficiency function for every detector. The neutron detector were solely exploited in the 0.7-7 MeV range. Below 0.7 MeV, their response was to poor to correct properly for it. Above 7 MeV, we acquired too few events to get a reasonable statistical uncertainty. All PFNS were therefore truncated due to the lack of efficiency of the neutron detectors below 0.7 MeV and above 7 MeV. Consequently, we had to fit the measured spectra by a Watt function to recover the missing parts. The integration of the latter amounts to $21\pm1\%$ of the entire PFNS.

The overall efficiency of the neutron detectors was estimated in a similar way. The neutron multiplicity was also precisely determined in the Boykov paper. We again gated on the kinetic energy of the incoming neutrons in the range 2.9 ± 0.1 MeV, fitted the corrected PFNS by a Watt spectrum and finally estimated the overall efficiency of every neutron detector assuming a 2.76 neutron multiplicity.

The incoming neutron energy spectrum was divided into 41 energy bins from 1MeV to 200 MeV. The fission cross-section being low for neutron energy below 1 MeV, no reliable result could be obtained. Above 200 MeV, the incident neutron flux was too little to allow for an accurate measurement.

3 Results and discussion

We produce here consecutively the results for 238 U and 237 Np neutron induced fission. We fitted the PFNS by Watt spectra (see eq.(1)) for accounting for the non-measured low-energy part and high-energy tail of the spectra.

$$N_{w}(E) = Amp \times \frac{2A^{2/3}}{\sqrt{\pi B}} \exp\left(-\frac{B}{4A}\right) \exp\left(-AE\right) \times \sinh\left(BE\right)^{1/2}.$$
 (1)

Where *Amp* stands for the amplitude of the fitted distribution, A=1/T, the inverse of the temperature of the emitting fragments. $B=4E_f/T$, where E_f is the recoil energy of the fragments.

The mean kinetic energy of the neutrons is given by eq. (2)

$$\langle E \rangle = \frac{3}{2}T + E_f \cdot$$
 (2)

The results of the measurement are produced in the following section. They are still preliminary. They are subject to improvement, especially concerning the estimation of the error bars. The FIGARO experimental set-up does not permit to measure the high energy neutrons which could emerge from pre-equilibrium emission or direct interaction. Such a component is peaked in the forward direction where no detector could be positioned. It is supposed to be negligible below 40 MeV incident energy. Above that value, the lack of detection of those high energy neutrons induces a bias mainly on the estimated mean kinetic energy of the emitted neutrons. The measured multiplicity is affected to a lower extent. The reader should keep in mind that all evaporated neutrons were accounted for, the high energy neutrons being not detected.

3.1 The ²³⁸U results

We chose to measure again ²³⁸U in order to test our new setup. We could compare our new measurement with other experimental results.

3.1.1 The mean kinetic energy

The mean value of the measured PFNS is shown, **Figure 1** as a function of the kinetic energy of the incoming neutron. All available other experimental results are also plotted on the same graph. The uncertainty is currently estimated to be of 2%.



Figure 1. Mean value of the PFNS in the 238 U(n,f) reaction as a function of the neutron energy. Other experimental results are also shown

The amount of new experimental data provided by our recent measurement strongly contrasts with the scarce other experimental results available until now. Our data allows to Measurement of the average energy and multiplicity of prompt-fission-neutrons from 238 U(n,f) and 237 Np(n,f) from 1 to 200 MeV extend the measured domain from 20 to 200 MeV.

The kinetic energy of the emitted neutrons vary moderately over the full dynamics, ranging from 1.8 to 2.3 MeV. Above 20 MeV, we observe a slow increase of the kinetic energy. The latter seems to stabilize above 150 MeV. However, some fluctuations are observed below 20 MeV. A zoom on the low energy part of the distribution is also shown Figure 1. We observe a very good agreement (within error bars) with all other measurements but the low energy Japanese data (see (Miura et al (ref. ⁷)(1996), see entry 22477 20010629 in the EXFOR database) at 1.9 and 2 MeV which are not consistent with the other experimental data. We observe a pronounced dip around 7 MeV. That dip is nicely described with 5 coherent experimental point between 6.5 and 11 MeV.

This dip could be explained by the weight of the pre-fission neutrons. Indeed, when the (n,nf) channel opens around 7 MeV, neutrons can be emitted prior to fission. Those neutrons exhibit a lower kinetic energy than the post-fission neutrons since they don't suffer from the fission boost.

Our data together with the results of other groups give an indication for the presence of a smaller second dip around 15MeV.



Figure 2. Our data compared to the Los Alamos model as implemented at Bruyeres le Chatel and to Maslov model

The Figure 2 shows the comparison of our data with the results of two models (see ref. [1,2]). Both models reproduce the dip around 7 MeV. The Maslov model seems to describe mo accurately the slope below and above the minimum. Moreover, the Maslov model predicts a second drop between 10 and 15 MeV. Such a slope is qualitatively observed in our data. Above 10 MeV for the Los Alamos model and 15 MeV for the Maslov one, the mean kinetic energy seems overestimated by both models.

3.1.2 The neutron multiplicity

The integration of the fitted Watt spectra allowed us to extract the fission neutron multiplicity.



Figure 3. Neutron multiplicity as a function of the kinetic energy of the neutron. Previously measured data are also shown

The neutron multiplicity measured in the neutron induced fission of ²³⁸U is plotted Figure 3. Recent and older other experimental data are also plotted on the same graph. Our data exhibit a constant increase up to 200 MeV. They are no fluctuation in our data. The uncertainty is estimated to be of 3%. We observe a change of slope in our data around 7 MeV. Such an behaviour is not seen in Fréhaut's data which exhibit a regular increase. We don't have any clear explanation for such a disagreement at the moment. However, our data seem to be in good agreement (within error bars) with the other experimental results even if our data are systematically higher than other measurements below 8 MeV and lower above 8 MeV.

3.2 The ²³⁷Np results

As ²³⁸U was extensively measured during the last 30 years, few measurement concerns ²³⁷Np. We only found 2 recent measurements for PFNS. We report here for the first time on a new measurement of both PFNS and multiplicity on the neutron-induced fission of ²³⁷Np over a broad energy range. The statistical uncertainty on the ²³⁷Np data is higher than what we obtained for the ²³⁸U run. The measurement time was shorter due to technical problems

3.2.1 The mean kinetic energy

The mean kinetic energy obtained from the PFNS measurement is plotted as a function of the neutron energy in **Figure 4**. Only 2 sets of other experimental data were found in the literature. Both measurement as well as the Maslov model are also plotted in **Figure 4**. It's impressive to see how close our data are from the Maslov simulation. Actually, the model is in better agreement with our results than the other experimental data. Especially Lovchikova's data give between 5 and 10% higher mean kinetic energy. Boykov data are however in agreement within uncertainty. Again, we observe a pronounced dip around 7 MeV. It fits nicely with the increase of the 2^{nd} chance fission cross-section. The

presence of a second dip around 13 MeV, thus corresponding to the 3^{rd} chance fission, seems to be confirmed by those new data.



Figure 4. Measured mean kinetic energy compared to other data and Maslov model.

Again, the predicted very fast increase of the mean kinetic energy above 15 MeV is clearly not seen our experimental results.

3.2.2 The neutron multiplicity

The neutron multiplicity in the neutron induced fission of ²³⁷Np is plotted in Figure 5. Experimental results obtained by other groups are also shown. Fréhaut's data provide the more complete set of comparable data since they cover the full 1-15 MeV range. The agreement between our data and all others is rather good. They are only 3 data points which are not compatible with fréhaut's results. They appear to be in same corner than for ²³⁸U. It seems that our data suffer from a systematic underestimation from 7 to 10 MeV.



Figure 5. Neutron multiplicity as a function of the kinetic energy of the neutron. Previously measured data are also shown

Nevertheless, for all other experimental points, we observe an impressive agreement, the discrepancy being often as low as 1%.

- D.G Madland et al., Nucl. Sci. Eng. 81 (1982) 213
- ² V.M. Maslov et al., Phys. Rev. C 69 (2004) 034607 , V.M.
- Maslov et al, These proceedings
- ³ T. Ethvignot et al., Phys. Lett. B575 (2003) 221-228
- ⁴ T. Ethvignot et al., Phys. Rev. Lett 94 (2005) 052701
- ⁵ D. Rochman et al., Nucl. Inst. Meth. Phys. Res. Sec. A523 (2004) 102
- ⁶ Boykov et al., Ann. Nucl Energy 21-10 (1994) 585, see also the EXFOR database for numerical results

⁷ Miura et al (1996) browse the EXFOR database (www.nea.fr/html/dbdata/x4) entry 22477 20010629