Energy measurement of prompt fission neutrons in ²³⁹Pu(n,f) for incident neutron energies from 1 to 200 MeV

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Abstract

Prompt fission neutron spectra in the neutron-induced fission of ²³⁹Pu have been measured for incident neutron energies from 1 to 200 MeV at the Los Alamos Neutron Science Center. Preliminary results are discussed and compared to theoretical model calculation.

1 Introduction

An experimental campaign was started in 2002 in the framework of a collaboration between CEA/DAM of Bruyères-le-Châtel and the Los Alamos National Laboratory in order to measure the prompt fission neutron spectra (PFNS) for incident neutron energies from 1 to 200 MeV. The prompt neutron spectra in 235 U(n,f), 238 U(n,f) and 237 Np(n,f) were already studied successfully (Refs. [1–4]). This paper reports on new results obtained for 239 Pu(n,f) during two experiments performed in 2007 and 2008 and on results obtained for 235 U(n,f) in 2007.

2 Experimental Setup

2.1 Neutron beam at the WNR facility

The WNR facility at the Los Alamos Neutron Science Center (LANSCE) provides white and pulsed incident neutron beams with an energy distribution spread from one to several hundreds of MeV with a maximum at ~ 2 MeV. Neutrons are produced via the spallation reactions induced by the LANSCE 800 MeV proton pulsed beam impinging a tungsten target. Our experiments were set on the 30° right flight path (see Fig. 1, left), 22.7 m downstream from the spallation target and with a collimation of 2.8 cm diameter at the ²³⁹Pu target position.

Thanks to this spallation neutron source, our experiments provide a large set of data with a consistent systematic uncertainty over the whole energy range.

2.2 Fission Chamber

The target consists of a multi-layer ionization fission chamber. Each layer consists of actinide material deposited on a platinum backing. Two different fission chambers were used respectively for the two experiments: a multiple actinide chamber containing 109 mg of 235 U and 92 mg of 239 Pu in 2007, and another chamber containing 100 mg of 239 Pu in 2008. The signal of the fission chamber is used to trigger the data acquisition system.

2.3 Prompt fission neutron detection

2.3.1 FIGARO detector

The multidetector FIGARO (see Fig. 1 right, Ref. [5]) was used in order to measure the prompt fission neutrons (PFN) in coincidence with the fission chamber signals. In its basic configuration (as used in 2007) FIGARO consists of twenty EJ301 organic liquid scintillators located about one meter from the fission chamber and on seven different detection angles (45° , 60° , 75° , 90° , 105° , 112° and 135° with

respect to the beam direction). During our experiment in 2008, three detectors were replaced by one stilbene and two paraterphenyl detectors having a higher efficiency at low energy (below 500 keV). The EJ301, stilbene and paraterphenyl detectors are sensitive to both, neutrons and gamma-rays, but with different responses. Taking advantage of this feature, a discrimination on the pulse shape based on the charge integration was performed in order to reject offline the events due to gamma rays.



Fig. 1: Left side: The WNR facility at the LANSCE. The FIGARO set-up is located on the 30° right flight path. Right side: Artistic view of the FIGARO neutron set-up. The neutron beam is represented by the straight line.

2.3.2 Double time-of-flight method

For each triggered event, two times of flights were measured (with a resolution of \sim 3.5 ns FWHM): first, for the incident neutrons from the spallation target to the fission chamber, and second, for the prompt fission neutrons from the fission chamber to the hit detector. Knowing the exact flight path of the neutrons, we calculated their velocity and therefore their kinetic energy event by event.

PFNS were determined for 31 incident neutron energy groups ranging from 1 to 250 MeV. Afterwards, the spectra were corrected for the detector efficiency using data taken with a spontaneous fission source of 252 Cf, placed at the position of the fission chamber. The trigger for these 252 Cf runs was a gamma-ray detector placed at 20 cm from the source. The comparison of the well known prompt neutron spectrum in 252 Cf(sf) with the measured spectra gives the efficiency of each neutron detector.

2.4 Background subtraction

The incident neutrons not only can induce fission, but also can scatter on the fission chamber structure (windows and samples' backings). These scattered neutrons are not correlated with fission and create background in the neutron detectors by random coincidences. During both experiments this source of background was monitored using an additional trigger. Spectra of scattered neutrons were determined as a function of the incident neutron energy, and subtracted offline from the spectra measured in coincidence with the fission chamber.

The body of the fission chamber used during the first experiment was made of stainless steel and the backings of platinum. The signal to background ratio was around 1. In addition, due to experimental difficulties, prompt fission neutron energy spectra were obtained only with poor statistics. For the second experiment, the body of the fission chamber used was thinner and made of Aluminum, and the signal to background ratio was increased to 2:1. From this experiment the data are more precise thanks to better statistics and a better monitoring of the background noise due to scattered neutrons.

3 Results

In this section the mean energy of the measured prompt fission neutron spectra after background subtraction is discussed and compared to data evaluations. The preliminary results obtained from the 2008 experiment are shown in Figs. 2 and 3.

The neutron detectors were used from 600 keV to 7 MeV. Above 7 MeV the statistics is really poor. Below 600 keV the limit of the pulse shape discrimination is reached and the efficiency of the detector decreases strongly. In Fig.2 the mean energy of the experimental spectra from 600 keV to 7 MeV is represented as a function of the incident neutron energy. The same energy cut is applied to the model calculation represented in the figures with the dashed lines. The evaluated results are based on the Los Alamos model [6] in its improved form, following the prescription of A. Tudora and G. Vladuca [7], and implemented at CEA/DAM of Bruyères-le-Châtel by B. Morillon.



Fig. 2: Mean energy of the prompt neutrons emitted during the neutron induced fission of 239 Pu calculated over the range 600 keV to 7 MeV. Experimental data (solid line) are compared with the BRC09 evaluation (dashed line). Top panel: for the whole energy range. Bottom panel: for incident neutron energies below 25 MeV.

Alternatively, the spectra have been fitted using a Maxwellian functional form:

$$N(E) = 2A\sqrt{\frac{E}{\pi T_m^3}}exp(-\frac{E}{T_m}),\tag{1}$$

where A and T_m are the two free parameters of the fit. A is the integral of the function and T_m is the so-called "fission temperature" which is related to the emitter temperature, and is directly proportional to the total mean energy:



$$\langle E \rangle = \frac{3}{2} T_m. \tag{2}$$

Fig. 3: Average energy of the prompt neutrons emitted during the neutron induced fission of ²³⁹Pu. Experimental data (solid line), deduced from the Maxwellian fit, are compared with evaluations of V. Maslov (dotted line) and B. Morillon (dashed line). Top panel: for the whole energy range. Bottom panel: for incident neutron energies below 25 MeV.

This approach is valid under the assumption that the spectra follow a Maxwellian distribution also in

its lower part (below 600 keV) and its higher part (above 7 MeV). This total mean kinetic energy is increasing from 2.1 MeV at 1 MeV neutron beam energy, up to more than 2.6 MeV for very fast incoming neutrons, as shown in Fig.3. Experimental results are compared to the BRC09 evaluation and to the model calculations of V. Maslov [8], where no cut in fission neutron energy was applied. The evaluated data are obtained apart from the experimental results presented in this paper.

Finally in Fig.4 the mean energy of the PFNS in 235 U(n,f), obtained for the 2007 experiment, is represented for incident neutron energies from 1 to 25 MeV. In this case, the mean energy is calculated from experimental spectra from 1 to 7 MeV. The same cut is applied to the BRC09 calculation represented in dashed line. The results are consistent with the previous experimental results obtained by our collaboration (see Ref. [2]).



Fig. 4: Average energy of the prompt neutrons depending on the incident neutron energy for the neutron induced fission of 235 U. Experimental data are calculated within the energy cut from 1 to 7 MeV and compared with evaluation of B. Morillon (dashed line), where the same cut was applied. Results are from the 2007 experiment.

4 Discussion

Depending on the incident neutron energy, the prompt fission neutrons can be emitted from different sources. Mainly they are evaporated by the fission fragments, but they can also be evaporated by the compound nucleus before fission when the incident neutron energy is higher than the separation energy of one neutron, at 6-7 MeV depending on the actinides. At this point the competition starts between the fission of first chance, (n,f) reaction, and the fission of second chance, (n,nf) reaction. At higher neutron beam energy (already above 14 MeV), pre-equilibrium contribution starts to increase and is not anymore negligible compared to the contribution of evaporation.

The general trend is an increase of about 30 % of the mean kinetic energy of the PFN with the incoming neutron energy, going from 1 MeV to 200 MeV, except at 7 MeV and 13 MeV where it is strongly dropping.

On average the temperature of the compound nucleus is increasing with the beam energy, so the prompt neutrons will globally be emitted with a higher kinetic energy for higher incident neutron energies.

The two dips at 7 MeV and 13 MeV have already been seen experimentally for other actinides and were

predicted by the models (see for example Refs. [1–4]). These two decreases are understood as the openings of the fission of second and third chances and they show the weight of the pre-fission neutrons. The agreement between the experimental values and the calculation is fair, especially with the BRC09 evaluation (below 4%) that predicts the position of the openings of the second and third chances more accurately than V. Maslov. But the amplitude of the third chance is better reproduced by the model of V. Maslov, whereas BRC09 seems to underestimate it. Above 18 MeV for BRC09 and 13 MeV for V. Maslov, the mean kinetic energy seems overestimated by both models.

It should be noted that pre-equilibrium neutrons are emitted along the beam axis at forward angle and with an energy up to the incident neutron energy, whereas the neutron emissions by evaporation are isotropic and follow a Maxwellian energy distribution. But since the neutron detectors are located at side angles and are efficient from 600 keV to 7 MeV, the set-up is not so sensitive to the pre-equilibrium. As a consequence, from 14 MeV incident neutron energy, the data do not include all the contributions to prompt neutron spectrum and the experimental mean energy values are below the real ones, as pre-equilibrium neutrons are more energetic than the evaporated neutrons.

5 Conclusion

Two experiments were performed with the FIGARO set-up of the LANSCE, in 2007 and 2008, to measure the kinetic energy spectra of the prompt neutrons emitted in 239 Pu(n,f) as a function of the energy of the incident neutrons.

Preliminary results from the experiment of 2008 are promising. The prompt fission neutron average energy was deduced for a large range of incident neutrons energy. The positions of the openings of the fission of second and third chances were measured at 7 MeV and 13 MeV, respectively. These experimental values as well as the global trend are well reproduced, below 4%, by the evaluations of B. Morillon and collaborators.

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