Measurement of ^{242}Pu(n,f) in the [1;2MeV] energy range

Ludovic Mathieu, Mourad Aiche, Paola Marini, Olivier Bouland, Carole Chatel, Serge Czajkowski, Deby Kattikat-Melcom, Teresa Kurtukian-Nieto, Stephan Oberstedt, Igor Tsekhanovich

Measurement of ^{242}Pu(n,f) in the [1;2MeV] energy range

Ludovic Mathieu[1*], *Mourad* Aïche[1], *Paola* Marini[1], *Olivier* Bouland[2], *Carole* Chatel[3], *Serge* Czajkowski[1], *Deby* Kattikat-Melcom[1], *Teresa* Kurtukian[1], *Stephan* Oberstedt[4], *Igor* Tsekhanovich[1]

[1]Univ. Bordeaux, CNRS, LP2I, UMR 5797, F-33170 Gradignan, France
[2]CEA, DES, IRESNE, DER, SPRC, Physics Studies Laboratory, Cadarache, F-13108 Saint-Paul-lez-Durance, France
[3]Université de Strasbourg, CNRS, IPHC/DRS UMR 7178, 23 Rue du Loess, F-67037 Strasbourg, France
[4]European Commission, Joint Research Centre, 2440 Geel, Belgium

Abstract. The design of new generation fast nuclear reactors requires highly accurate cross-section measurements in the MeV energy region. The ^{242}Pu fission cross section is of particular interest for Pu incineration and nuclear waste production. There are discrepancies around 1 MeV incident neutron energy between libraries and among experimental data. Some data suggest the presence of a strong structure between 1 and 1.2 MeV whereas it is barely visible on some other data and its shape is very different among evaluations. The large majority of the ^{242}Pu(n,f) measurements have been carried out with respect to the ^{235}U(n,f) secondary-standard cross section. This introduces a strong correlation between independent measurements and this cross section exhibits structures, in particular a steep increase of +10% at 1 MeV. Therefore, we aim to re-measure the ^{242}Pu(n,f) cross section relative to the primary-standard ^{1}H(n,n)p cross section, by using a proton recoil detector. This standard has a very high accuracy (0.4%), is not used for of other ^{242}Pu measurements, and is structureless. An experiment has been carried out in October 2022 at the MONNET facility in JRC Geel, with incident neutron energies from 0.9 MeV to 2.0 MeV. The experimental setup will be presented, and the analysis procedure will be detailed.

1 Introduction

The maximum of the neutron flux in a nuclear reactor is around 1-2 MeV for neutrons are produced at these energies. In thermal reactors, and due to the 1/v cross section behaviour, the thermal part of the neutron spectrum is predominant in the fission rate. However, in fast reactors, this thermal part is negligible, and the majority of the fissions are due to the 1-2 MeV neutrons. The knowledge of fission cross sections in this energy region is then of key importance for such reactors.

One of the goals of nuclear fast reactors could be to incinerate nuclear wastes and nuclear materials. The majority of waste radiotoxicity is due to Pu isotopes. With a half-life of 375.000 years, the ^{242}Pu is the main contributor to the long-term radiotoxicity. A precise knowledge of its behaviour in reactor is then mandatory.

The ^{242}Pu fission cross section has been measured several times in the past decades. There was a renewed interest since the 2000's when it was included in the HPRL [1,2], with a target uncertainty of 5% in the 1-2 MeV energy range. However, there are some discrepancies between experimental dataset and among evaluations. In particular, a structure at 1.2 MeV is visible on ToF data of Weigman [3], Tovesson [4] and Koegler [5] but do not appear as clearly in quasi mono-energetic data of Salvador-Castiniera [6]. This feature cannot be explained by the nuclear structure in ^{243}Pu* compound nucleus, but could be linked to the ^{242}Pu structure itself [7]. Major evaluations are based on ToF data and then all show the same structure at 1.2 MeV. They are however in disagreement on the average value, with JEFF-3.3 being the highest (+3%) and JENDL-5 the lowest (-4%).

Nearly all experimental measurements have been performed with respect to the ^{235}U(n,f) cross section, apart from Salvador-Castiniera et al. where ^{235}U(n,f), ^{238}U(n,f) and ^{237}Np(n,f) cross sections were used. Every measurement performed using ^{235}U(n,f) standard are then strongly correlated. These standard cross sections are well confirmed by integral measurements, but discrepancies can be seen here and there. For instance, the ^{235}U(n,f) cross section show a "threshold-like" structures around 1 MeV, with a +9% increase within 150 keV, and discrepancies between experimental data set can reach up to 5%.

Another method is to use the ^{1}H(n,n)p cross section as reference, known as proton recoil method. This cross section is a primary standard, with an uncertainty of less than 0.4% on a large energy range [8], and without any structure. Measurements using this standard can add precious accurate and uncorrelated values to help to improve the cross section knowledge. In 2012 a ^{242}Pu(n,f) measurement was carried out by our team using this standard [9]. Technical issues prevented us from obtaining enough energy points to bring new information about the structure at 1.2 MeV. Nevertheless, these results are compatible with the lower JENDL-5 evaluation, and much less with JEFF-3.3 or ENDF/B-VIII.

A new experiment has been carried out in 2022 to re-measure the ^{242}Pu(n,f) in the 1-2 MeV energy range. This paper present the experimental setup and the main stages of the ongoing analysis.

2 Experimental setup

2.1 Facility and sample

The experiment took place in October 2022 at the new MONNET facility in JRC Geel [10]. This facility had two major advantages that justified this choice:

- it allows the use of ^{3}H beam target, which enables to produce neutrons with energies covering the energy range of interest ;
- the JRC has a radioactive sample laboratory, and the ^{242}Pu target made there for previous experiments was still available. This ^{242}Pu sample has an activity of 98.4 kBq for a diameter of 29.95 mm and is electroplated on a 0.25mm-thick Al-backing.

The neutron production reaction was ^{3}H(p,n)^{3}He, producing neutrons with an energy from 0.9 to 2.0 MeV. Two different TiT targets were used with thicknesses of 930 and 260 µg/cm². The beam intensity varied from 15 to 40 µA. A very light water-cooling device was used on the TiT target, ensuring a low quantity of scattered neutrons.

The first element of the experimental setup was placed 90 mm away from the neutron production point. A closer distance of 60 mm was used with the thin production target.

2.2 Fission setup

Fission events were measured via the detection of a single Fission Fragment (FF) by two photovoltaic cells of 40x20 mm². The Pu target is located in a compact fission device, with two photovoltaic cells placed in front of it at a distance of 5.4 mm. The cells frame is used as a collimator with a square aperture of 34x34 mm², and cells are 0.5 mm apart.

The interaction energy on the ^{242}Pu target depends on two factors: the TiT target thickness and the angle of aperture of the Pu target. The further setup distance of 90 mm was used to reduce the energy spread when using the thick TiT target.

2.3 Proton recoil setup

The proton recoil technique consists in irradiating an H-rich material with neutron, in order to convert a small part of the neutron flux into easy-to-detect protons. For this purpose, a polypropylene (PP) foil was placed downstream the fission setup. The recoil protons were emitted by neutron scattering in every direction downstream. A silicon detector was placed 95 mm further, with a 15 mm diameter collimator, in order to collect protons emitted with a very small angle. The proton energy is given by:

$$E_p = \cos^2\theta \times E_n \quad (1)$$

with θ the proton scattering angle with respect to the incident neutron direction. The protons also lose energy going through the PP foil itself, depending of their starting point. To reduce this phenomenon, the PP foil thickness had to be kept small. A 4 µm-tick PP foil was used, ensuring a proton energy loss lower than 15% in the worst case: for the lowest neutron energy (0.9 MeV) and a recoil proton emitted at the beginning of the PP foil.

The detected protons energy is then close to that of incident neutrons, and can be more easily discriminated from the background. In particular, recoil protons from scattered neutrons will have a lower energy due to a lower incident neutron energy and a different incoming direction (see equation 1).

To subtract recoil protons coming from other sources, a tantalum screen could be remotely placed downstream the PP foil to stop all recoil protons coming from the foil.

3 Data Analysis

3.1 Cross section calculation

The ^{242}Pu fission cross section is measured relative to the ^{1}H(n,n) cross section, with several correction factors:

$$\sigma_{^{242}Pu(n,f)} = \frac{N_{FF}}{N_p} \times \frac{\varepsilon_p}{\varepsilon_{FF}} \times \frac{\Omega_{PP}}{\Omega_{^{242}Pu}} \times \frac{N_{PP}}{N_{^{242}Pu}} \times \sigma_{H(n,n)} \quad (2)$$

with:

- N_{FF} and N_p the numbers of fission fragments (FF) and recoil protons detected respectively;
- ε_p and ε_{FF} the detection efficiencies of recoil protons and FF respectively;
- Ω_{PP} and Ω_{242Pu} the solid angles of the PP foil and ^{242}Pu target respectively;
- N_{PP} and N_{242Pu} the number of hydrogen atoms in the PP foil and ^{242}Pu atoms in the Pu target respectively;
- $\sigma_{H(n,n)}$ the neutron elastic scattering cross section on ^{1}H.

All these correction factors take into account the differences in counting rates, efficiencies, irradiation and atom quantities between the ^{242}Pu target and the reference PP foil.

3.2 Counting rates

Fission spectra in photovoltaic cells show less background and a clear alpha/FF discrimination, as can be seen in Figure 1. However, ^{242}Pu undergoes spontaneous fission that has to be subtracted (see following sub-section). In addition, there is also a small contribution of a constant background, which has been measured without Pu target at the end of the experiment.

Due to the amount of structure materials in the neutron production target, the reaction chamber and the structure materials, some scattered neutrons may irradiate the fission target and induce parasitic fissions. The scattered neutron energy is usually much lower than the one of direct neutrons, due to the production angle and the scattering process. An MCNP simulation has to be carried out to subtract this contribution. In the 2012 experiment, this represented 5 to 15% of the fission rate, depending on the neutron energy and the cooling system used. A low value of about 5% is expected for this experiment, as the neutron production device and cooling system was very light.

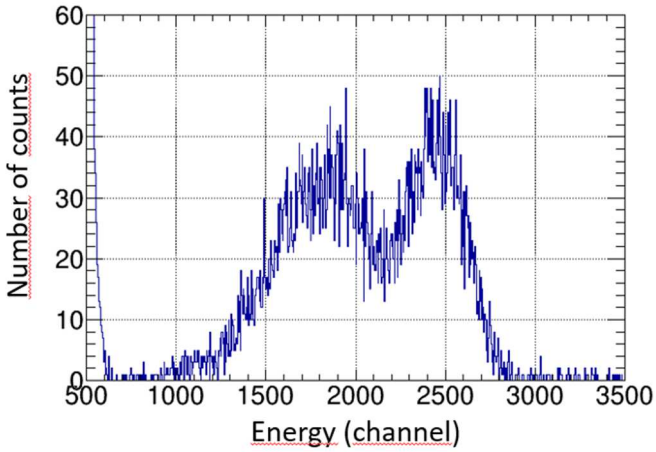

Fig. 1. Fission spectrum in one photovoltaic cell.

Proton recoil spectra are polluted with a strong low energy background coming from electrons generated by the neutron production process [11]. When the proton energy is high enough, the proton peak can easily be discriminated from the background. However, this discrimination is trickier at low energies such as 1 MeV.

In addition, some hydrogen contaminations on structure materials can emit recoil protons up to the maximum proton energy. To reduce such contributions, efforts have been made to minimize and clean hydrogen contamination of the experimental device. A measurement with a tantalum screen downstream the PP foil enables to measure most of this background. Typical spectra are shown in Figure 2. Measurement runs and tantalum screen runs have to be normalized one to each other by a combination of fission number and neutron flux (via MONNET long-counters).

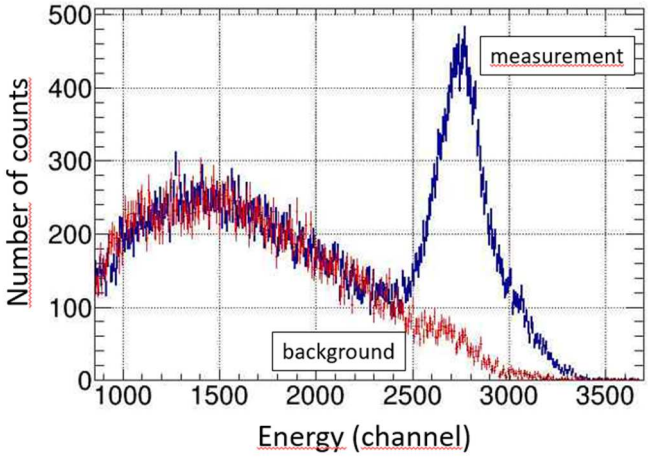

Fig. 2. Proton recoil spectra with (measurement) and without (background) PP foil, for an incident neutron energy of 2 MeV.

3.3 Efficiencies

The fission efficiency is quite difficult to infer: there is a non-sensitive collection grid in the front face of the cell, and polycrystalline cells may have some unconnected grains which won't participate to the energy collection. The fission efficiency is then deduced from the spontaneous fission (SF) emissions. Thus, SF runs have been performed almost every night or weekend. The SF efficiency has the advantage to also include the real target features (thickness and homogeneity). The results are presented in Figure 3. Some runs are still to be analysed to fill the gaps, and to conclude about the general trend of this efficiency.

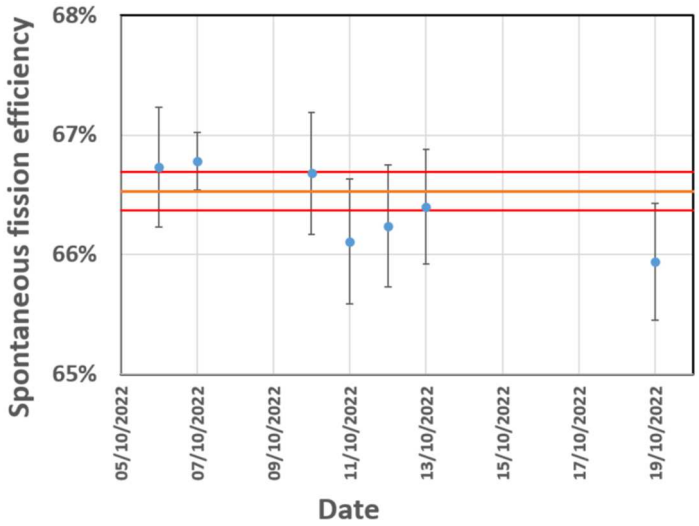

Fig. 3. The spontaneous fission efficiency for different SF runs is shown. Still several runs has to be analysed. The mean value with its uncertainty is indicated by the orange and red lines respectively.

To obtain the fission efficiency from the SF efficiency, one has to take into account several physical effects: irradiation profile in flux and energy, kinematic effect and fission

anisotropy. These corrections are calculated with a simulated efficiency, and then applied on the experimental SF efficiency.

The proton detection efficiency is determined via simulations, considering source characteristics, PP irradiation profile in flux and energy, and kinematic effects.

3.4 Irradiation

The third term in equation 2 deals with the amount of neutron passing through the fission target and the PP foil. For an isotropic neutron emission, this term equals the ratio of respective solid angles. The accuracy of this ratio strongly depends on the uncertainties on the distances, lengths and radii knowledge of the experimental setup. In particular, the larger distance used for the thick neutron production target helps to obtain a lower uncertainty on this term. As the neutron production is anisotropic, a simulation is carried out to calculate this ratio.

3.5 Material quantities

The fourth term in equation 2 corrects for the different number of nuclei between the fission target and the PP foil. As a radioactive isotope, the number of ^{242}Pu atoms is measured via alpha spectrometry with a high accuracy. Measuring the H content in the PP foil is trickier, relying either on a micro-weight measurement or on a thickness measurement. In any case, preventing H-contamination of the PP foil is mandatory as the subtraction of induced recoil protons is difficult and introduce an additional uncertainty [9,11].

3.6 Preliminary results

Few steps of the analysis have been done so far, and several corrections still need to be applied. Nevertheless, the general shape of the ^{242}Pu cross section begins to appear, as can be seen in Figure 4.

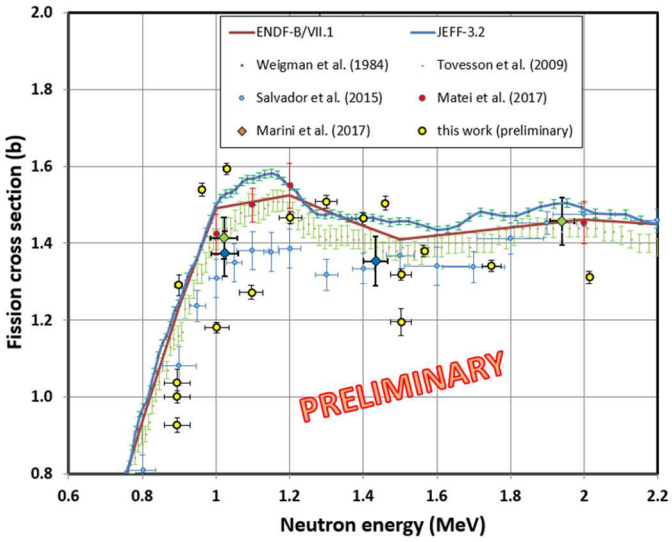

Fig. 4. Fission cross section of ^{242}Pu with preliminary result from this experiment is shown. Error bars of these points do not include systematic uncertainties.

As can be seen, a thorough check of each point is necessary. Indeed, an incorrect background subtraction in the proton recoil spectrum may lead to extremely high or low values. In addition, some corrections may change the global shape of the cross section. In particular, the scattered neutrons correction is different from one energy to the other, as well as the proton or fission efficiencies. Other corrections, like material quantities and clean background fission subtraction, influence mainly the absolute value of the cross section. At the end, the cross section will be obtained for about fifteen different energy points.

4 Conclusion

The ^{242}Pu fission cross section is an important nuclear data for the use of future nuclear fuel. Some discrepancies still exist among data and/or evaluations. This cross section has been re-measured at the MONNET facility at JRC Geel. The aim of this measurement is to provide a cross section value strongly independent from the existing data: different reference cross section, different measurement method and different type of detectors.

New data will bring valuable information about the structure around 1.2 MeV. The analysis is still ongoing and it is too early to draw any conclusion from the results.

Acknowledgement

We wish to acknowledge the MONNET team for providing support and a high quality beam during the experiment. This work is supported by the European Atomic Energy Community (EURATOM) under the European Union's Horizon 2020 SANDA, grant agreement No 847552.

References

1. NEA Nuclear Data High Priority Request List, http://www.nea.fr/html/dbdata/hprl
2. OECD/NEA Working Party on Evaluation and Co-operation (WPEC) Subgroup 26 Final Report, http://www.nea.fr/html/science/wpec/volume26/volume26.pdf
3. H. Weigman et al., Nucl. Phys. A, Vol. 438, Iss. 2 (1985)
4. F. Tovesson et al., Phys. Rev. C **79**, 014613 (2009)
5. T. Kögler at al., Phys. Rev. C **99**, 024604 (2019)
6. P. Salvador Castiñeira et al., Phys. Rev. C **92**, 044606 (2015)
7. C. Chatel, *Section efficace de fission du ^{242}Pu : progrès théoriques et expérimentaux*, PhD thesis (in French) (2021)
8. A.D Carlson et al., Nucl. Data Sheet (2018)
9. P. Marini et al., Phys. Rev. C **96**, 054604 (2017)
10. S. Oberstedt et al., Technical Report, IAEA (2014)
11. P. Marini, Nucl. Instr. Meth. A, Vol. 841 (2017)