EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

# Measurement of the α-ratio and (n,γ) cross section of $^{239}$Pu at n_TOF

September 22, 2020

V. Alcayne[1], F. Álvarez[1], J. Andrzejewski[2], J. Balibrea[3], M. Caamaño[4], F. Calviño[5], D. Cano-Ott[1], C. Domingo[3], I. Durán[4], A. Gawlik[2], C. Guerrero[6], B. Fernández[4], E. Gónzalez-Romero[1], J. Heyse[7], T. Martínez[1], J. Lerendegui[3], E. Mendoza[1], J. Perkowski[2], A. Plompen[7], J.M. Quesada[7], A. Sánchez-Caballero[1], P. Schillebeeckx[7], A. Tarifeño[5], G. Sibbens[7] and the n_TOF collaboration

[1]CIEMAT, Spain
[2]University of Lodz, Poland
[3]IFIC-CSIC, Spain
[4]Universidad de Santiago de Compostela, Spain
[5]Universidad Politécnica de Cataluña, Spain
[6]Universidad de Sevilla, Spain
[7]JRC Geel, Belgium

Spokesperson(s): D. Cano-Ott (daniel.cano@ciemat.es), Jaroslaw Perkowski (jaroslaw.perkowski@uni.lodz.pl)
Technical coordinator: O. Aberle (Oliver.Aberle@cern.ch)

## Abstract

More accurate $^{239}$Pu capture and fission cross section data are required for the design of new nuclear devices and the optimization of nuclear waste, as they have been listed in the NEA/OCDE High Priority Request List. We propose to measure the capture and fission rates of $^{239}$Pu in the n_TOF EAR-1 in order to obtain the capture-to-fission ratio (α-ratio), the capture, and the fission cross sections. The measurement is part of the scientific program of the EURATOM H2020 SANDA project. The measurement is very challenging since $^{239}$Pu is a fissile isotope and also because it has a high α-activity. We will apply our expertise acquired during the $^{235}$U(n,γ) n_TOF measurement and analysis to perform the measurement with an improved experimental setup. It will be performed with two complementary experimental setups. In the first one we will use 10 samples of 1 mg $^{239}$Pu each, placed in the center of the n_TOF Total Absorption Calorimeter (TAC), which will detect the γ-rays emitted after the (n,γ) reactions. The samples will be located inside an ionization chamber which will operate in coincidence with the TAC and will be used as a fission tagging detector to strongly reduce the background in the TAC due to fission reactions. The second experimental setup will use an encapsulated 50-100 mg sample, and not the ionization chamber, and will extend the measurement up to higher neutron energies. We expect to obtain the α-ratio and the capture cross section of $^{239}$Pu with an uncertainty of 3% from thermal up to 100 eV, and with 3-4% between 100 eV and 10 keV.

**Requested protons**: $5 \cdot 10^{18}$ protons on target
**Experimental Area**: EAR1

# 1 INTRODUCTION AND MOTIVATION

The reduction of the uncertainties on neutron capture and fission cross-sections of major and minor actinides has a direct impact on the improvement of the safety and economy of our actual nuclear reactors (especially in life extension and higher burnup scenarios), the design of new critical nuclear systems such as Gen IV reactors and the optimization of nuclear waste management strategies, also including the transmutation of minor actinides [1] [2] [3]. In particular, $^{239}$Pu plays a central role in the operation of fast reactors and is also very important for the operation of thermal reactors, especially when loaded with MOX fuels or when considering higher burnups. More accurate $^{239}$Pu capture and fission cross section data are needed, as stated in the NEA/OECD High Priority Request List [4].

Due to the strong interest in obtaining new cross section data on $^{239}$Pu, the measurement proposed is part of the scientific program approved by the EURATOM H2020 Supplying Accurate Nuclear Data for energy and non-energy Applications – SANDA project [5].

Until recently, there was only one *high energy resolution* (with enough data points to perform a reasonable resonance analysis) capture measurement in the EXFOR database [6] in the Resolved Resonance Region [7], published in 1971. In 2014, a new measurement was performed at LANSCE (Los Alamos, USA) [8] [9] [10], in the neutron energy range between 10 eV and 1.3 MeV. Only the shape of the cross section was measured in this case, and the results were normalized to the ENDF/B-VII.1 [11] cross section at 17-18 eV. Figure 1 shows the comparison of the two sets of data with the cross sections taken from the recent JEFF-3.3 [12] and ENDF/B-VIII.0 [13] evaluated nuclear data libraries. The effect of the resolution broadening increasing with the neutron energy is clearly visible in the experimental data, especially in Mosby et al. [8]. It can be noticed that the $^{239}$Pu(n,$\gamma$) cross section in JEFF-3.3 is significantly larger (~30%) than the one in ENDF/B-VIII.0 in the 40-150 eV range, as  partially shown in the left panel.

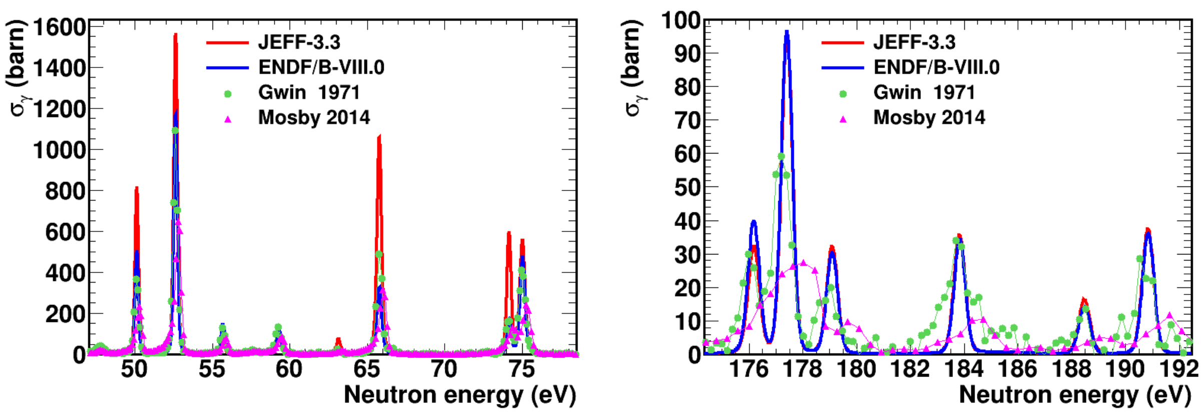


Figure 1 $^{239}$Pu(n,$\gamma$) cross section in the JEFF-3.3 and ENDF/B-VIII.0 data libraries, together with the data from Gwin et al. [7] and Mosby et al. [8], in two different neutron energy ranges.

The existence of only two measurements and the discrepancies between the evaluated cross sections are originating in the difficulty of measuring capture cross sections of fissile isotopes, due to the strong competing $\gamma$-ray background from fission. To overcome this difficulty, we propose to measure the neutron capture-to-fission ratio ($\alpha$-ratio), the fission and the capture cross sections of $^{239}$Pu at the n_TOF EAR-1 flight path in one single measurement. The goal is to measure simultaneously the neutron-induced capture and fission rates by means of the n_TOF $BaF_2$ Total Absorption Calorimeter (TAC), used for detection of γ-rays, in combination with fast ionization chambers used as fission tagging detectors. In this way, it will be possible to separate and subtract with high accuracy the fission background. We will apply our world-leading experience, methodologies and analysis techniques developed ad-hoc for previous capture and fission measurements at n_TOF [14][15][16] on the fissile $^{235}$U and high activity α–emitters like $^{241}$Am and $^{244,246}$Cm, to perform a high accuracy measurement with an improved experimental setup.

This measurement will improve significantly the previously existing data by:

- Providing a capture yield with an overall uncertainty ~3% in the range from thermal energies to 10 keV. The capture yield will not be normalised arbitrarily to the 17-18 eV resonance, as for the case of the LANSCE data, but normalised to the well-known $^{239}$Pu(n,f) cross section measured simultaneously.
- Providing an absolute α–ratio, thanks to the accurate determination of the fission and capture detection efficiencies. This methodology was developed and applied very successfully for the $^{235}$U(n,γ) measurement.
- Providing data with significantly better energy resolution, measured with a flight path 10 times longer than in [7] and [8], which will improve significantly the resonance analysis.
- Solving the existing discrepancies between the recent JEFF-3.3 and ENDF/B-VIII.0 evaluations.
- The measurement will provide also very valuable information on the distribution of the γ-rays cascades emitted in $^{239}$Pu(n,γ) and $^{239}$Pu(n,f) reactions [17], as has been the case in previous experiments performed with the TAC [18] [19] [20] .

## 2 EXPERIMENTAL SETUP

The measurement will be performed with two different types of samples produced at JRC-Geel. The first type will consist in ten samples of 1 mg $^{239}$Pu each. They will be placed inside the fission chamber, in the centre of the TAC. With this experimental setup most of the fission reactions will be detected in coincidence with both detection systems, and thus the background in the TAC due to fissions will be strongly supressed. The implementation in Geant4 [21] of this geometry, used in the calculations performed to prepare this measurement, is presented in the left panel of Figure 2.

After a very detailed study involving data from previous measurements with the TAC and Monte Carlo simulations, we have concluded that with this experimental setup we can perform a high precision measurement up to 0.5-1 keV. Above this energy the expected level of the background in the TAC not related with fission will become large compared with the expected capture rates. In order to improve the capture-to-signal ratio we will measure with a second experimental setup, using a 50-100 mg encapsulated $^{239}$Pu sample and without the fission chamber. With this second measurement we will be able to extend the measurement up to ~10 keV, similar to [20]. The shape of the deposited energy spectra in the TAC due to capture and fission reactions will be determined with the first experimental setup, and they will be used, in combination with the measured fission rates, for subtracting the background in the TAC due to fissions in this second measurement. In both experimental setups, a neutron absorber made of borated polyethylene will be placed between the sample and the TAC, in order to reduce the background due to elastically scattered neutrons.

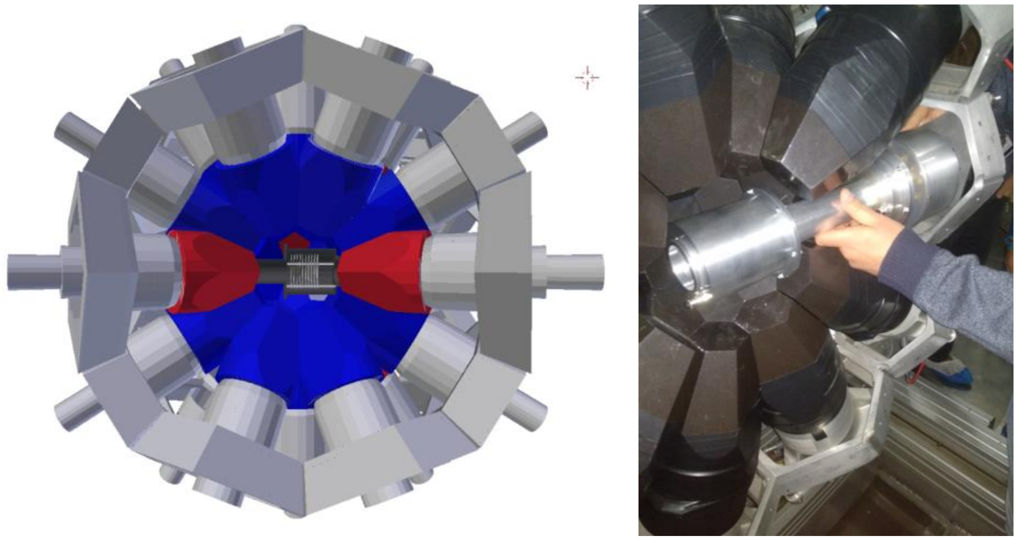

Figure 2 Schematic view (left) and picture (right) of the n_TOF TAC with the fission chamber in the centre.

The fission chamber has been recently designed and built at the University of Lodz, and a picture of it, placed in the centre of the TAC, is presented in the right panel of Figure 2. Its design has been optimised taking into account three important requirements: (i) to have a low mass intercepting the neutron beam, to minimize the background in the TAC due to capture and elastically scattered neutrons; (ii) to have good discrimination between alphas and fission fragments; and (iii) to minimize and process adequately pile-up effects [22] [23].

One of the main difficulties of the measurement, apart from the competing fission γ–ray background, is the large α–activity of $^{239}$Pu (~2 MBq/mg). After a testing phase with α–sources, the fission chamber will be commissioned in a $^{239}$Pu(n,f) measurement at JRC-Geel before the measurement at n_TOF.

## 3 OBJECTIVES AND BEAM TIME REQUEST

The beam time request follows a very detailed analysis of the uncertainties in the α–ratio and in the capture cross section. In the study we have propagated to the final results the uncertainties due to the capture and fission efficiencies, the subtraction of the beam related background, the normalization and the counting statistics. These estimations have been validated with the results of the $^{235}$U(n,γ) measurement.

Together with the measurements performed with the $^{239}$Pu samples using the two experimental setups (10 samples of 1 mg using fission detectors and the 50-100 mg sample without fission detectors), it is necessary to make additional measurements to determine the backgrounds and to validate the obtained results. For the background we plan to measure with (i) dummy samples (a dummy fission chamber and a dummy canning), with nothing else in the beam (*sample out*), (ii) activity without the neutron beam, (iii) and with a graphite sample. The purpose of these measurements is to determine the different components of the background: i) due to the interaction of the neutron beam with the different materials, ii) due to the activity of the $^{239}$Pu sample, iii) and due to the elastic scattered neutrons in the $^{239}$Pu sample.

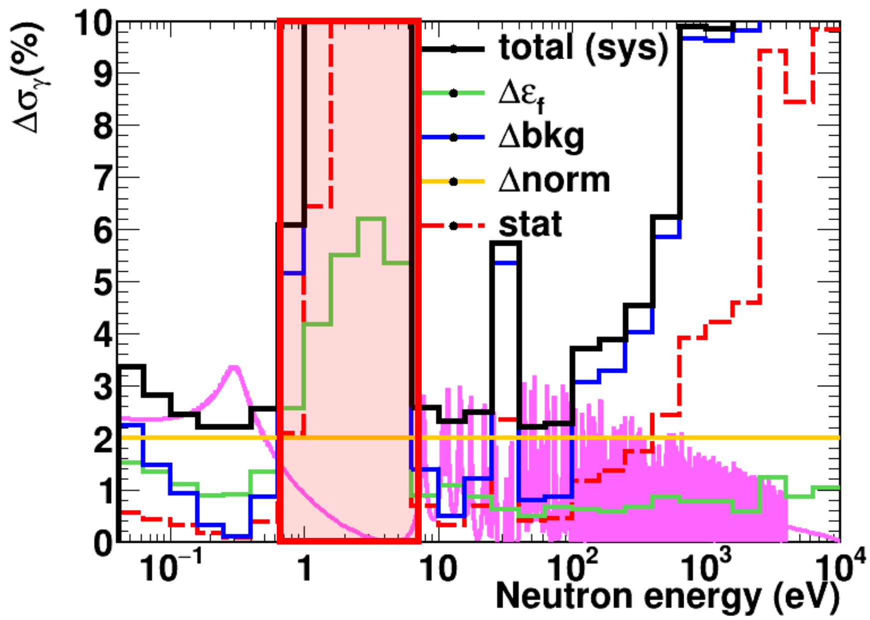


Figure 3 Estimated uncertainties (5 bins/decade) in the $^{239}$Pu capture cross section measured with the 10 x 1 mg samples setup. The shape of the JEFF-3.3 $^{239}$Pu(n,γ) cross section is plot in magenta (Log10($\sigma_\gamma$/barn)). The uncertainties in the 0.8-7 eV neutron energy range (shadowed area) are large because of the valley in the cross section.

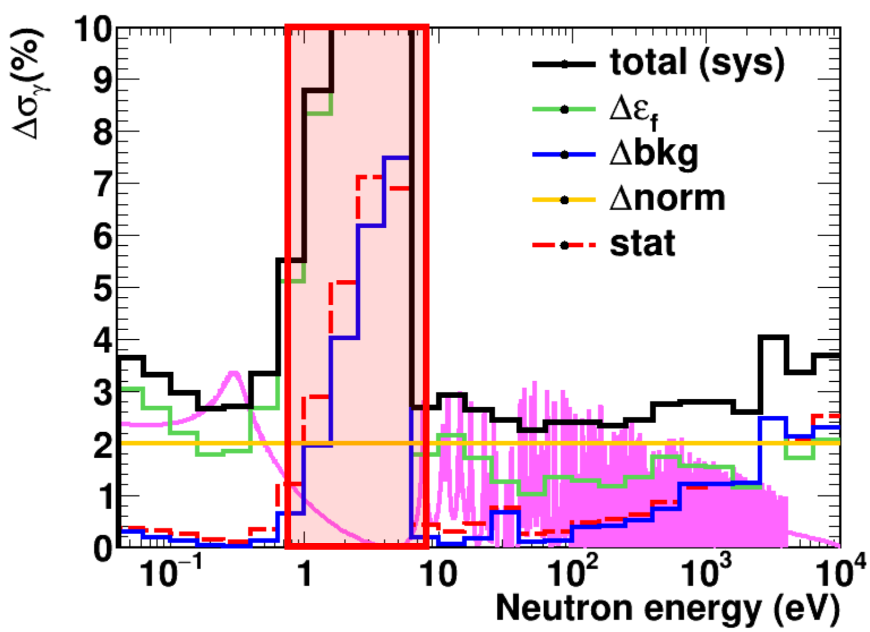


Figure 4 Same as Figure 3, but for the measurement performed with an 80 mg sample.

In order to keep systematic uncertainties well under control, sufficient statistics are required in the deposited energy spectra as a function of neutron energy, both in the measurement made with the isotope and also in the background measurements. Taking this into account, the protons requested for each measurement are summarized in Table 1. The values of the table have been optimized to minimize the uncertainties due to counting statistics and due to systematic effects. For the 10 x 1 mg samples at least 40.000 counts need to be obtained in the low-energy resonances. The deposited energy histograms constructed with 100 bins will be obtained with an average uncertainty of ~5% per bin. For the measurement performed with the thick sample, the number of protons has been calculated in order to have an uncertainty due to counting statistics in the keV region comparable to the uncertainties due to systematic effects when using 5 bins per decade.

The expected uncertainties in the measured capture cross section are presented in Figure 3 and Figure 4. The figures show the uncertainties due to counting statistics (stat) and to different systematic effects: the propagation of the uncertainty in the efficiency of the fission detectors ($\Delta\varepsilon_f$), the uncertainty in the determination of the background not related to fission (Δbkg), and the normalization uncertainty (Δnorm), mainly due to the uncertainty in the capture detection efficiency. These three will dominate over other sources of uncertainty. They have been added quadratically to obtain the total uncertainty due to systematic effects (total).

With the requested number of protons, we will determine the $^{239}$Pu capture cross section and α-ratio with an accuracy of ~3% below 100 eV and with 3-4% between 100 eV and 10 keV.

## Summary of requested protons:

| Measurement | Purpose | Protons |
|---|---|---|
| **$^{239}$Pu samples (10 x 1 mg)** | (n,γ) cross section measurement (up to 0.5 keV) | $1.2\cdot10^{18}$ |
| **Dummy chamber (10 x 1 mg)** | Canning related background | $0.8\cdot10^{18}$ |
| **$^{239}$Pu sample (50 – 100 mg)** | (n,γ) cross section measurement ( 0.01–10 keV) | $0.8\cdot10^{18}$ |
| **Dummy (50 – 100 mg)** | Canning related background | $0.5\cdot10^{18}$ |
| **Sample out** | Beam related background | $0.5\cdot10^{18}$ |
| **$^{197}$Au** | Validation of the measurement | $0.5\cdot10^{18}$ |
| **Graphite** | Background due to elastic scattering | $0.3\cdot10^{18}$ |
| **$^{239}$Pu without neutron absorber** | Determination of the $^{239}$Pu(n,γ) cascades | $0.4\cdot10^{18}$ |
| **$^{239}$Pu (no beam)** | Background without beam | 0 |
| **Total** | | **$5\cdot10^{18}$** |

Table 1 Number of protons requested for each measurement.

## References:


[1] G. Aliberti et al., Nucl. Sci. Eng. 146, 13 (2004).

[2] G. Aliberti et al., Ann. Nucl. Energy 33, 700 (2006).

[3] M. Salvatores et al., *Uncertainty and target accuracy assessment for innovative systems using recent covariance data evaluations*, Technical Report No. 6410, NEA/WPEC-26, OECD, Paris, France (2008).

[4] https://www.oecd-nea.org/dbdata/hprl/

[5] https://cordis.europa.eu/project/id/847552

[6] https://www-nds.iaea.org/exfor/

[7] Gwin et al., Nucl. Sci. Eng. 45, 25 (1971).

[8] S. Mosby et al., Phys. Rev. C 89, 034610 (2014).

[9] S. Mosby et al., Phys. Rev. C 97, 041601 (2018).

[10] S. Mosby et al., Nucl. Data Sheets 148, 312 (2018).

[11] M. B. Chadwick et al., Nucl. Data Sheets 112, 2887 (2011).

[12] A. J. M. Plompen et al., Eur. Phys. J. A 56, 181 (2020).

[13] D. A. Brown et al., Nucl. Data Sheets 148, 1 (2018).

[14] J. Balibrea-Correa et al., PhD Thesis. (https://cds.cern.ch/record/2729120)

[15] J. Balibrea-Correa et al., *Measurement of the* α*-ratio and (n,*γ*) cross section of* $^{235}$*U from 0.2 to 200 eV at n_TOF*, Phys. Rev. C (in press).

[16] J. Balibrea-Correa et al., EPJ Web Conf. 146, 11021 (2017).

[17] C. De Saint Jean, NEA/WPEC-34 report (2014).

[18] C. Guerrero et al., Phys. Rev. C 85, 044616 (2012).

[19] E. Mendoza et al., Phys. Rev. C 90, 034608 (2014).

[20] E. Mendoza et al., Phys. Rev. C 97, 054616 (2018).

[21] S. Agostinelli et al., Nucl. Instrum. Methods A 506, 250 (2003).

[22] E. Mendoza et al., Nucl. Instrum. Methods A 768, 55 (2014).

[23] C. Guerrero et al., Nucl. Instrum. Methods A 777, 63 (2015).