EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Measurement of the ^{238}Pu(n,γ) cross-section at EAR2

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Abstract:
The neutron capture cross-section of ^{238}Pu is a key parameter for applications in space exploration and nuclear reactor safety. In space exploration, ^{238}Pu is the primary fuel for Radioisotope Thermoelectric Generators (RTGs), while in nuclear reactors, its accurate characterization is important for predicting decay heat after shutdown. Despite its significance, only two previous measurements of the ^{238}Pu(n,γ) cross-section exist, both of which are affected by substantial limitations. Current nuclear data libraries, including JEFF-3.3 and JENDL-5.0, heavily depend on these outdated datasets, highlighting the need for improved experimental data. This proposal outlines a measurement of the ^{238}Pu(n,γ) cross-section at the n_TOF EAR2 facility at CERN in an energy range of 2–500 eV. Utilizing the high-intensity neutron flux of EAR2 and the sTED detector array, this measurement will provide high-precision resonance data, reducing uncertainties in nuclear data libraries. The expected results will significantly improve nuclear modeling, benefiting reactor safety analysis and enabling the production of ^{238}Pu for RTGs for deep-space missions.

Requested protons: $4.0{\cdot}10^{18}$ protons on target
Experimental Area: EAR2

1 Introduction and motivation

All space exploration missions traveling beyond Jupiter require Radioisotope Thermoelectric Generator (RTG) power sources to provide electrical power. The most suitable isotope for these power sources is plutonium-238 (^{238}Pu), an alpha-emitter with a half-life of 87.7 years, offering sufficient decay heat and characterized by low radiation background. For instance, the Curiosity and Perseverance Mars rovers were powered by 4.83 kg of plutonium oxide [1].

Following the Cold War, the majority of ^{238}Pu was produced in Russia. However, to meet future demands, the U.S. Department of Energy (DOE) has restarted the production of this isotope. Currently, ^{238}Pu is produced in the United States at research reactors located at the Idaho and Oak Ridge National Laboratories [2], with a production scale of approximately 400 grams per year. Nonetheless, the anticipated demand for ^{238}Pu is several times higher than the current production rate, with plans to scale up to 1.5 kg per year [3]. In Europe, the European Space Agency (ESA) has conducted studies on ^{238}Pu production at the BR2 reactor at SCK-CEN [4]. These studies demonstrate the technical feasibility of producing this isotope within Europe, reducing reliance on non-European partners [5].

The majority of ^{238}Pu production currently takes place in nuclear reactors through the neutron irradiation of highly enriched ^{237}Np samples. Upon neutron capture, ^{237}Np is transformed into ^{238}Np, which decays into ^{238}Pu with a half-life of 2.1 days. A schematic view of this process is shown in Fig. 1.

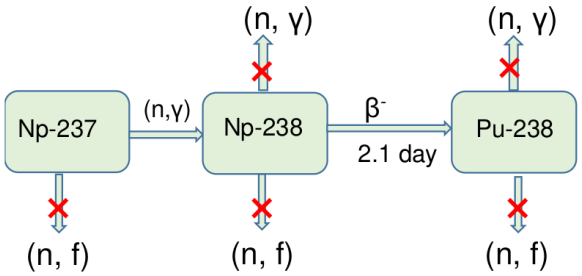

Figure 1: Chain of isotopic transitions during irradiation of Np-237. The "X" indicates an unwanted channel in the ^{238}Pu production process. The figures was extracted from Ref. [6].

To maximize the production of ^{238}Pu and minimize the formation of undesirable isotopes, the preferred neutron energy range for irradiation is the resonance absorption region of ^{237}Np, spanning from 1 to 600 eV [6]. One undesired reaction in the production process is the neutron capture by ^{238}Pu itself, which produces ^{239}Pu. This isotope cannot be chemically separated and is fertile, meaning it could compromise reactor safety by reducing the thermal safety margin [1]. Therefore, a deeper understanding of these reaction could enable more efficient ^{238}Pu production. In particular, the Resolved Resonance Region (RRR) is crucial, as it represents the most favorable energy range for ^{238}Pu production.

In nuclear reactors, it is crucial to characterize the decay heat, particularly after the reactor has been shut down. This parameter is essential for the transport, transmutation, and storage of spent nuclear fuel. In recent years, studies have been conducted to identify the main isotopes

contributing to decay heat. Specifically, a series of investigations have analyzed how uncertainties in nuclear data propagate to uncertainties in decay heat estimation [7, 8, 9]. For Pressurized Water Reactors (PWRs) and Boiling Water Reactors (BWRs), the decay heat during the first 50 years is primarily dominated by the decay of fission products. However, beyond this period, actinides become increasingly significant. Notably, ^{238}Pu contributes approximately 10% of the decay heat after ten years of cooling and continues to play a significant role for hundreds of years.

In the specific case of a BWR, after 15.6 years of cooling, uncertainties in nuclear data propagate to a 4% uncertainty in the estimation of ^{238}Pu content in the spent nuclear fuel [7]. At this cooling time, approximately 10% of the decay heat is produced by this isotope, meaning that a 0.4% uncertainty in decay heat originates from nuclear data uncertainties related to ^{238}Pu.

Due to the significance of the capture cross-section of ^{238}Pu, dedicated integral measurements were performed as part of the PROFIL and PROFIL-2 experiments [10]. These experiments, conducted in the Phénix fast reactor [11], involved the irradiation of nearly pure isotope samples to obtain precise data on integral capture cross-sections and branching ratios for various actinides and fission products. For ^{238}Pu, the study reported an overestimation of its integral capture cross-section by approximately 2.5% based on the analysis of the JEFF-3.1 nuclear data library [12]. While this library is not the most recent, as discussed in the next section, there have been no significant updates to the evaluation of this isotope due to the lack of new experimental measurements.

MYRRHA, a multi-purpose fast spectrum research reactor [13], depends on accurate nuclear data for its criticality calculations and safety assessments. As a lead-bismuth-cooled system using MOX fuel containing 30% plutonium, precise evaluations of cross-sections are essential for its performance and licensing.

Among the isotopes of interest, ^{238}Pu plays a role due to its capture and fission reactions, which significantly influence the neutron economy. However, current nuclear data libraries exhibit discrepancies, particularly in the ^{238}Pu(n,γ) and ^{238}Pu(n,f) cross-sections [14]. These differences highlight the need for updated evaluations and new measurements for this isotope. The inconsistencies directly impact the calculated multiplication factor (k_{eff}) and emphasize the necessity of new experimental data to improve nuclear data accuracy and enhance the reliability of simulations for advanced reactors like MYRRHA.

2 Previous measurements and evaluations of ^{238}Pu

Despite its importance, there are relatively few measurements of the various reaction channels of ^{238}Pu with neutrons. The most extensively studied channel is the fission reaction, with a considerable number of measurements available [15, 16, 17, 18]. However, even for this channel, additional nuclear data is needed, as highlighted in the High Priority Request List (HPRL) [19]. Regarding the capture cross-section at thermal energies, there are two older measurements [20, 21] and a more recent one performed at the ILL in 2009 [22]. However, the most significant gap in the data lies in the capture cross-section of ^{238}Pu. As summarized in Table 1, only two capture measurements and one transmission measurement have been reported.
The first measurement was a transmission experiment conducted in 1967 by Young *et al.* [23] at the Materials Testing Reactor (MTR). This work determined resonance parameters up to

	Type	Sample mass	Energy Range
T. E. Young (1967) [23]	Transmission	∼0.1 mg	Thermal- 200 eV
M. G. Silbert (1973) [24]	Capture	1.24 g	18.6- 490 eV
A. Chyzh (2013) [25]	Capture	∼0.4 mg	Thermal- 410 eV

Table 1: List of experimental ^{238}Pu transmission and capture measurements to obtain the neutron cross and their energy ranges.

200 eV, but the data are not available in the EXFOR database. The second measurement was performed in 1973 by Silbert and Berreth [24], where the capture cross-section was measured using a 1.25 g sample and a nuclear explosion as the neutron source. This unique experiment was a single-shot measurement,with considerable difficulties. The uncertainties in the radiative kernels ($R_k = g \cdot \Gamma\gamma \cdot \Gamma_n/\Gamma$), equivalent to the resonance areas, were on the order of 20%. The third and most recent measurement was carried out in 2013 at the Los Alamos Neutron Science Center using the DANCE calorimeter [26] by Chyzh *et al.* [25]. In this experiment, a high-purity sample with a mass of less than 0.4 mg was used. Thermal and resonance parameters were measured up to 410 eV. For the first resonances, the uncertainties in R_k were a few percent; however, they increased to around 20% for resonances at higher energies. In this measurement, the high α-activity of ^{238}Pu produced a considerable background. To improve this situation, a measurement with a higher instantaneous flux is needed.

A comparison of the radiative kernels from the three measurements is shown in Fig. 2. The recent measurement by Chyzh reports values for the first three resonances that are 10–20% lower than those from earlier measurements, and these results are not compatible. For the remaining resonance areas, significant differences are observed. However, due to the large uncertainties in the Silbert measurement, most of the areas are compatible.

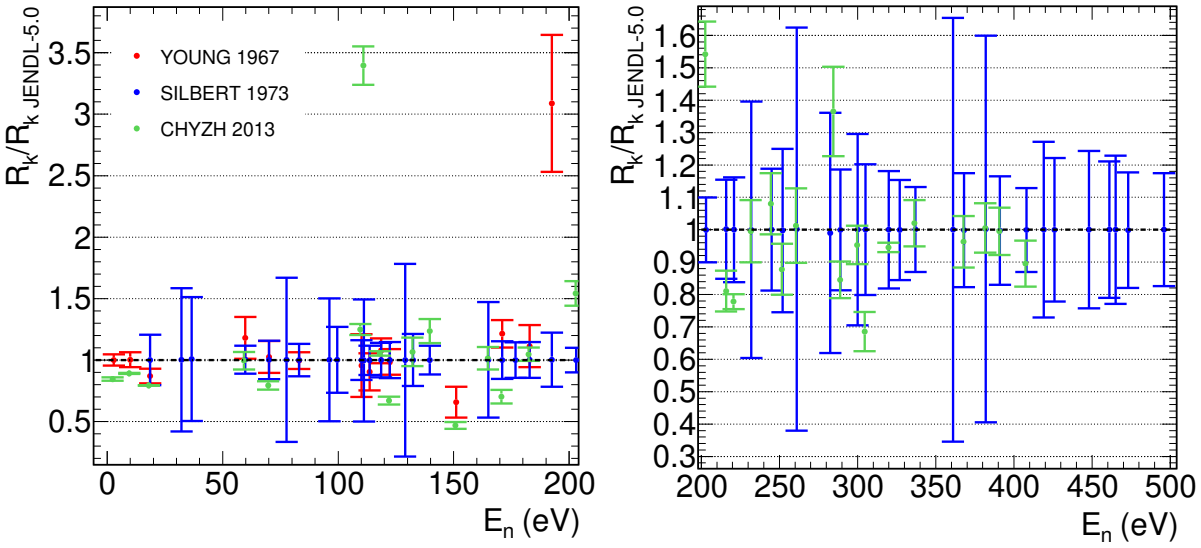

Figure 2: The ratios of the radiative kernels for the resonances of ^{238}Pu obtained in the three previous measurements [23, 24, 25] compared with the evaluation JENDL-5 [29].

Regarding the evaluations of the resonance parameters, the most recent libraries—JEFF-3.3 [27], JENDL-4 [28], JENDL-5 [29], and ENDF-VIII [30]—use the values, that are taken from the evaluation performed by Maslov in 1997 [31], which combines the measurements by Silbert and Young. For the JENDL-5.0 library, as can be seen on page 8 of Ref. [29], they have modified the negative resonances to match the new thermal values obtained from the measurements by Chyzh and Letourneau. The thermal capture cross section have been increased a 30% in this new evaluation. However the resonances parameters of the RRR have not been modified.

3 Measurement of ^{238}Pu at n_TOF EAR2

As seen in the previous sections, there is a need for new data on ^{238}Pu. Therefore, we propose to perform a capture measurement at n_TOF EAR2 in the energy range from 2 to 500 eV. In order to perform the measurement for this isotope, due to its short half-life ($T_{1/2}$ = 87.7 years) and high α-particle emission, a strong instantaneous neutron flux is required. Therefore, EAR2 [32] is the ideal area for this measurement. Another main difficulty in measuring the capture cross-section of this isotope is obtaining a sample sufficiently large and with the necessary enrichment. In this case, the samples will be loaned from CEA Cadarache and consist of a set of pellets of $\sim$2 mg each in a stainless steel container. Finally, to perform the measurements, the sTED detectors [33] will be used with the Pulse Height Weighting Technique (PHWT) [34].

During the PROFIL and PROFIL2 experiments [10], a considerable set of pure isotopic samples were produced. Among these, four cylindrical ^{238}Pu samples are particularly interesting, with a height of 4 mm and a diameter of 3.75 mm. After irradiation in the Phenix reactor [11], these samples contain approximately 2 mg of ^{238}Pu with an enrichment of 70%. In addition to ^{238}Pu, the samples also contain ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{234}U. In particular, the amount of ^{234}U is currently $\sim$10% due to the fact that this isotope is the daughter of ^{238}Pu. In addition to these isotopes, the sample also contains ^{137}Cs, which is responsible for the majority of the γ-rays emitted from the sample. The samples are inside a stainless steel case.

The development of the sTED detector [33] has enabled the performance of capture measurements at EAR2 in the energy range from thermal to hundreds of keV. The sTED has been used to perform many capture measurements in an array consisting of 9 modules, as seen in the left panel of Fig. 3. There are plans to upgrade the setup by using 27 modules [35], as shown in the right panel of Fig. 3. The new detectors are scheduled to be commissioned in 2025. In principle, we would use the configuration with 9 sTEDs; however, if the 27-module configuration shows good performance, we may switch to it.

In order to estimate the number of protons and the feasibility of the measurement, the expected counts in the experiment have been calculated for a total of $2{\cdot}10^{18}$ and are presented in Fig. 4. The efficiency used for the setup with 9 sTEDs is 6%. The counting rates account for the different backgrounds: background produced by capture reactions in the isotopes of the pellet or in the stainless steel case (BKG Cap), background produced by fission reactions in the actinides of the sample (BKG Fis), the empty background measured in previous experimental campaigns (BKG Empty), and finally, the background produced by the radioactivity of the sample (BKG Beam Off), mainly caused by ^{137}Cs.

The main sources of uncertainty in the measurement are due to counting statistics and the

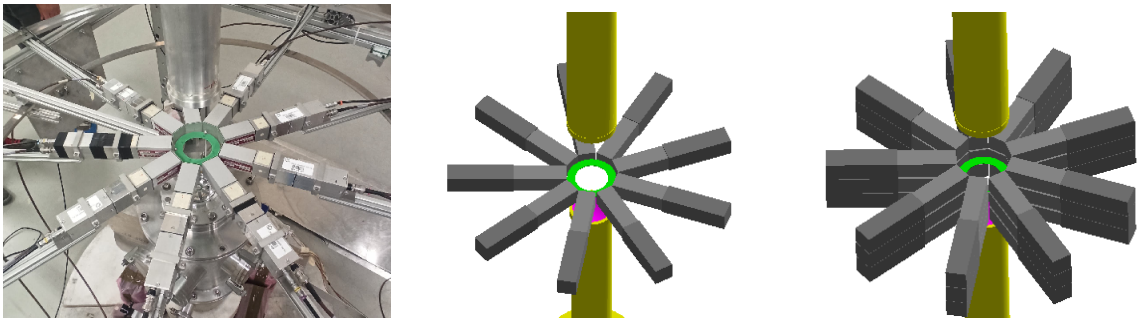

Figure 3: Left panel picture of the sTED setup with 9 modules. Central and right panel simulation of a configuration of sTEDs with 9 and 27 modules respectively.

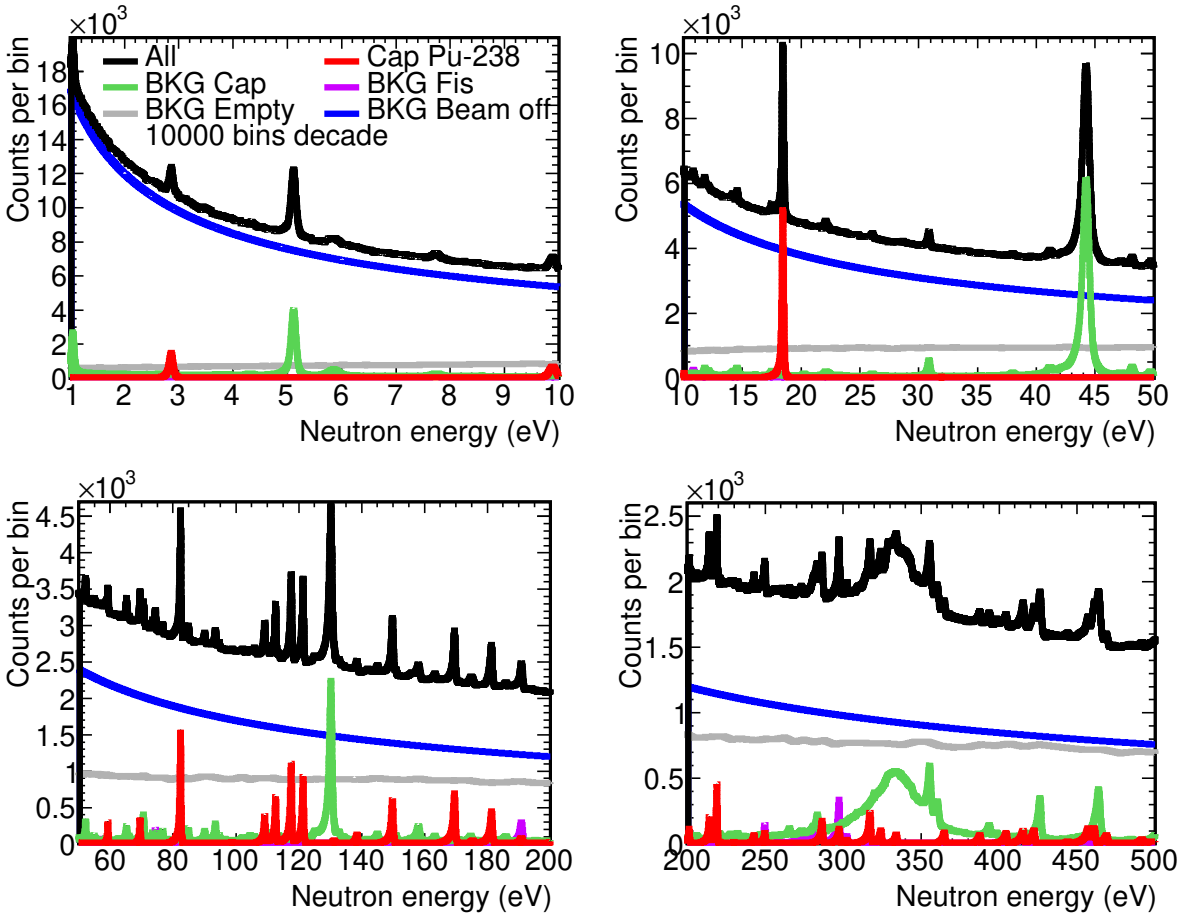

Figure 4: Counting rate estimations for a ^{238}Pu sample of 2mg and 0.375 cm diameter. The number of protons are $2{\cdot}10^{18}$. The different background included in the figure are detailed in the text.

subtraction of the backgrounds. The uncertainty in the determination of the background has been estimated to be 0.5%, as in previous works [36]. This uncertainty has been propagated to the area of the resonances, together with the uncertainty due to counting statistics. The estimated number of protons, $2{\cdot}10^{18}$, has been chosen to have uncertainties from both the

subtraction of the background and statistics that are similar. With this number of protons, we can measure a total of 35 resonances, with half of them having an uncertainty lower than 10%. In addition to the measurement of ^{238}Pu, $1{\cdot}10^{18}$ protons are requested to measure the dummy, and $1{\cdot}10^{18}$ to estimate the remaining backgrounds and for normalization.

Measurement	Protons
Pu^{238}sample	$2{\cdot}10^{18}$
Dummy sample	$1{\cdot}10^{18}$
Auxiliary and Normalization	$1{\cdot}10^{18}$
Total	$4{\cdot}10^{18}$

Table 2: Beam time request and distribution.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

Part of the experiment	Design and manufacturing
sTED detectors made of C_6D_6 liquid	To be used without any modification
^{238}Pu sample with a mass of $\sim$2 mg	Standard equipment supplied by a manufacturer

HAZARDS GENERATED BY THE EXPERIMENT
Additional hazard from flexible or transported equipment to the CERN site:
Only the ^{238}Pu samples would be transported to CERN the detectors are already at the n_TOF facility.