EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Measurement of the ^{238}U(n,γ) cross section at n_TOF

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Abstract

The ^{238}U(n,γ) cross section is one of the key reaction channels for nuclear applications, and small changes in it have a major impact on the results of many neutronic calculations. In recent years, an effort has been made to improve knowledge of this cross section, and four new measurements (two of them at n_TOF) and new evaluations have been performed. Despite this, there is still inconsistent data and sizeable differences between evaluated cross sections, and new evaluations are currently being worked on. Here we propose a new measurement at n_TOF with a new experimental setup focused on resolving some of the existing inconsistencies, thereby being able to improve the evaluations.

Requested protons: $3.2 \cdot 10^{18}$ protons on target
Experimental Area: EAR1

1 INTRODUCTION

Neutron capture on ^{238}U is one of the key reaction channels for nuclear applications [1]-[4]. In particular, ^{238}U is the major component of the light water reactor fuels, so the ^{238}U(n,γ) cross section is one of the most relevant to perform calculations of many kinds: inventory, reactivity, criticality, etc. for both present and advanced concept reactors.

Due to the importance of ^{238}U in nuclear applications, recent efforts have been made to improve existing evaluations. In particular, new neutron capture measurements have been performed at Los Alamos Neutron Science Center [5], GELINA [6] and n_TOF [7][8]; and two new evaluations of ^{238}U have been released: JEFF-3.3 [9] and IAEA-CIELO [10], both using the mentioned new datasets. The IAEA CIELO evaluation was then adopted by ENDF/B-VIII.0.

Despite all these efforts, the work is currently continuing to improve the evaluations of ^{238}U. Proof of this is the new JENDL-5.0 evaluation [11], which is based on the IAEA-CIELO but with some corrections; and the existence of the IAEA INDEN project [12], a continuation of IAEA-CIELO which continues studying some of the most important isotopes for nuclear applications, including ^{238}U. Moreover, the JEFF-3.3 and the IAEA-CIELO ^{238}U evaluations present sizeable differences [13].

Regarding the ^{238}U(n,γ) measurements carried out at n_TOF, both were made with the same sample and in the same experimental area (EAR1), but different detectors were used. One of the measurements was performed with two C_6D_6 detectors [7], and the other with the n_TOF Total Absorption Calorimeter (TAC) [8]. Although the ^{238}U(n,γ) has already been measured twice at n_TOF, we propose to perform an additional measurement mainly due to the following two reasons.

The first one is related with the ^{238}U(n,γ) cross section in the lower energy part of the Resolved Resonance Region (RRR). Both the JEFF-3.3 and the IAEA-CIELO evaluations take the resonance parameters up to 1200 eV from Kim et. al [6], which suggests a new average radiation width of 22.5 meV, which is a bit lower than the previous evaluated value of 23.0 meV. This apparently causes problems in the interpretation of some integral benchmarks. A collaborative effort was made between JRC-Geel and PSI to verify this problem, and it was found that by adopting a radiation width of about 22.7 meV for all resonances a consistent description of the benchmark is obtained. Additional ^{238}U(n,γ) experimental data will help to verify the average radiation width of 22.7 meV. The sample used for the two ^{238}U(n,γ) measurements at n_TOF was rather thick (0.375(2) g/cm^2, i.e. 9.56(5) $\cdot 10^{-4}$ atoms/barn) so the resonances at low energies were saturated; and saturated resonances are not the most appropriate for measuring resonance widths, since the fitted values become very dependent on the resolution function and multiple interaction corrections. A new measurement with a thin sample would solve these problems and allow the parameters of the largest low-energy resonances to be measured much more precisely.

The second reason is related with the cross section above 100 keV. Only two of the four new neutron capture measurements mentioned at the beginning of this document extends above 100 keV: the ones from Ullmann et al. [5] (Los Alamos) and Mingrone et al. [7] (n_TOF). Both of them have reported cross sections larger than the evaluated ones above 100 keV. This is discussed in the IAEA-CIELO evaluation publication (see Fig. 15 of [10] and associated text and Fig. 2 of [14] and associated text), where they conclude that both the data from Los Alamos and n_TOF are probably wrong due to missing corrections. We show Fig. 2 of [14] in Figure 1, which illustrates this situation.

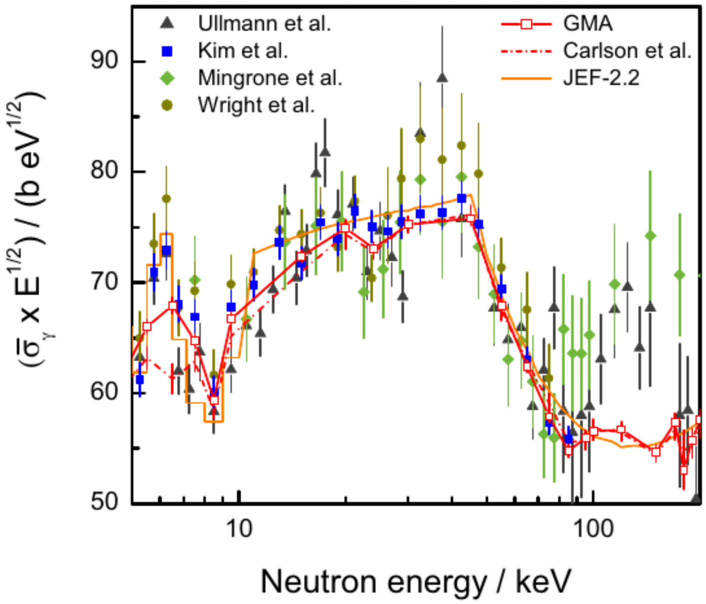

Figure 1 Average capture cross section for ^{238}U(n,γ) as a function of neutron energy. The recent measurement at Los Alamos (Ullmann et al. [5]), GELINA (Kim et al. [6]) and n_TOF (Mingrone et al. [7] with C_6D_6 and Wright et al. [8] with the TAC) are compared to three different evaluations: GMA, Calrson et al, and JEFF-3.2. The figure has been obtained from [14].

In addition to the thin sample, we propose to use a thick sample to perform a new measurement of the ^{238}U(n,γ) cross section in order to solve these discrepancies. One of the most important corrections in the keV region at n_TOF is the one related to the background produced by in-beam γ-rays (~40% of the background, according to Fig. 2 of [7]). The correction is performed by measuring a ^{nat}Pb sample and using black-resonance filters in the neutron beam. As mentioned in [7], it was possible to use the filters to determine the background only below 100 keV, and the determination of the background above 100 keV relies in the γ-ray energy spectra obtained from Monte Carlo calculations. The new measurement we propose will improve this situation for three reasons:

1. The new n_TOF spallation target produces less γ-rays (~50% less according to Monte Carlo calculations performed during the design phase) [15].
2. We will use a different experimental setup, consisting also in C_6D_6 detectors, but located at different forward and backward angles with respect to the neutron beam. The previous measurement used two detectors located at 125° with respect to the beam. For the same threshold in the detectors, such a setup more sensitive to scattered γ-rays than one made of detectors placed backwards. With the new setup proposed, the contribution of the in-beam γ-rays to the background will be different for each detector, thus allowing the determination of this background component better estimated and corrected for.
3. We will use additional filters in the neutron beam.

2 EXPERIMENTAL SETUP

The measurement will be performed at EAR1, since it has a better resolution function than EAR2.

For the detection system, we are considering two different possibilities. The first one is to use the same setup as in the ^{181}Ta(n,γ) [16] and ^{nat}Er(n,γ) [17] measurements performed in May-June 2023, which is shown in the left panel of Figure 2. This setup consists in three C_6D_6 BICRON detectors, one of them located at 90° with respect to the neutron beam and the other two at 125°; and three sTED small C_6D_6 detectors located at 90 °, 110 ° and 130 °. According to the analysis work carried out to date, the results seem excellent. The detection efficiency of this setup for ^{238}U(n,γ) cascades is expected to be ~17%.

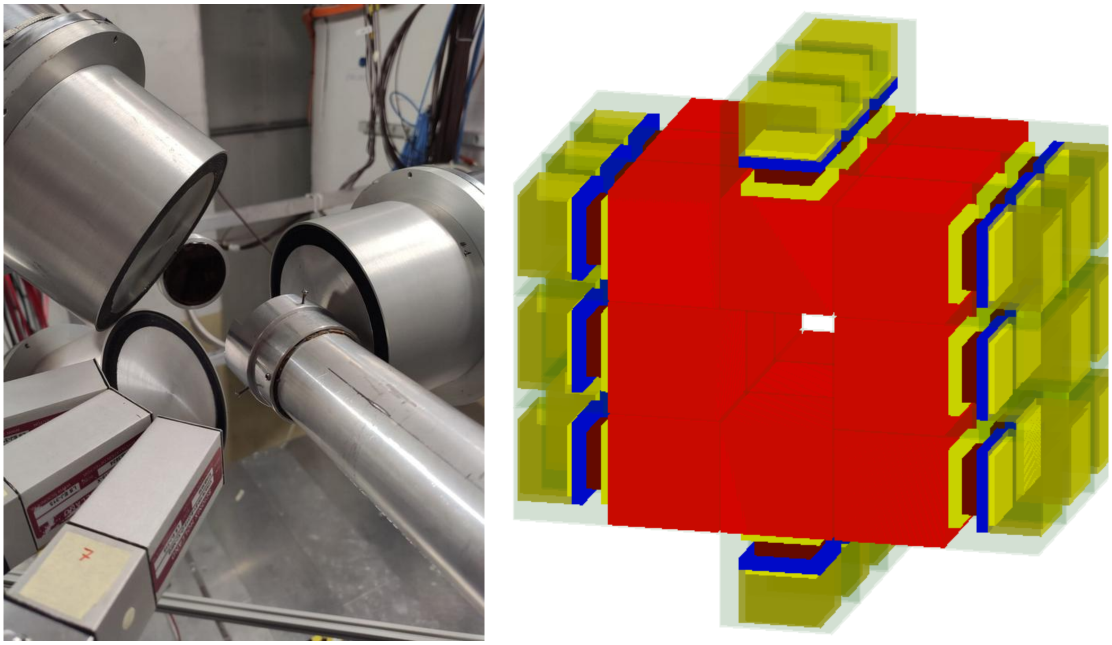

Figure 2 On the left, experimental setup used in the ^{181}Ta(n,γ) and ^{nat}Er(n,γ) measurements in May-June 2023. On the right, geometry of a cluster of 24 C_6D_6 *sliceTED* modules implemented in the Geant4 Monte Carlo transport code, an experimental setup which is at the moment in the design phase.

The second possibility for the experimental setup would be to use a new detector, for the moment called *sliceTED*, which is still in the design phase. It consists in a high efficiency C_6D_6 setup made of several C_6D_6 modules. An illustration of a possible sliceTED detector is presented in the right panel of Figure 2. The operational principle of this detector, described in detail in [18], is similar to the standard C_6D_6 total energy detectors, but with a larger (n,γ) detection efficiency. The design presented in Figure 2, made of 24 C_6D_6 modules with 3''x3''x3'' dimensions each, is expected to have a detection efficiency of ~60% for ^{238}U(n,γ) cascades. In addition to the high efficiency, it has the advantage of detecting γ-rays emitted at different angles, making the results little sensitive to anisotropies in the γ-ray emission. The main drawback will be probably an increase in the *neutron sensitivity*.

Concerning the samples, we will use a thin and a thick sample. For the thick sample, we are planning to use the same as in the previous measurements performed at n_TOF [7][8], which is a 99.999% pure ^{238}U metallic sample provided by EC-JRC-Geel with 6.125(2) grams and 53.90 × 30.30 mm^2 ($9.56(5)\cdot10^{-4}$ atoms/barn). For the thin sample, we estimated that a ^{nat}U sample with an areal density of $1.8\cdot10^{-5}$ atoms/barn (7.07 mg/cm^2) will be appropriate for our purposes.

The number of protons requested for this measurement is $3\cdot10^{18}$, which is broken down in the different measurement configurations in Table 1. The number of protons for the thin sample measurement has been estimated to achieve sufficient statistics at the largest resonances so that the uncertainties in the resonance parameters are dominated by systematic effects. An example of the expected results for two resonances is shown in Figure 3. For the thick sample measurement, the number of requested protons leads to ~3000 counts per bin in the keV region when using 100 bins per decade. This will lead to uncertainties due to counting statistics of ~3% in each bin after subtracting the background (~2% before the subtraction). The expected number of counts in the measurement when measuring the thick target is shown in Figure 4. Additional measurements with ^{197}Au, ^{nat}Pb and ^{nat}C samples and with filters, needed as a reference measurement and for subtracting the background, have been also considered.

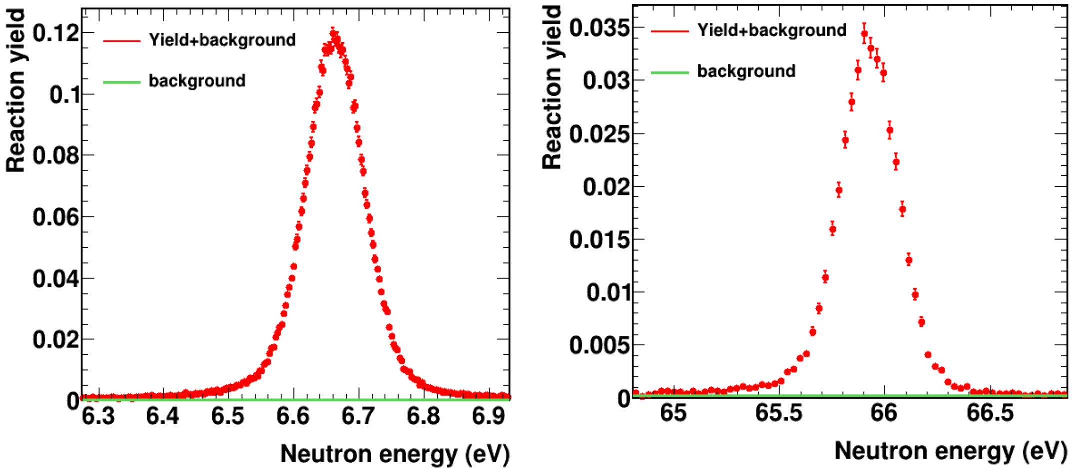

Figure 3 Expected ^{238}U(n,γ) yield when measuring the thin sample, for the first (left) and fourth (right) strongest resonances. The statistical fluctuations have been modelled assuming $0.5 \cdot 10^{18}$ protons on target when using the same experimental setup as in the ^{181}Ta(n,γ) and ^{nat}Er(n,γ) measurements. The background also comes from the same experiment.

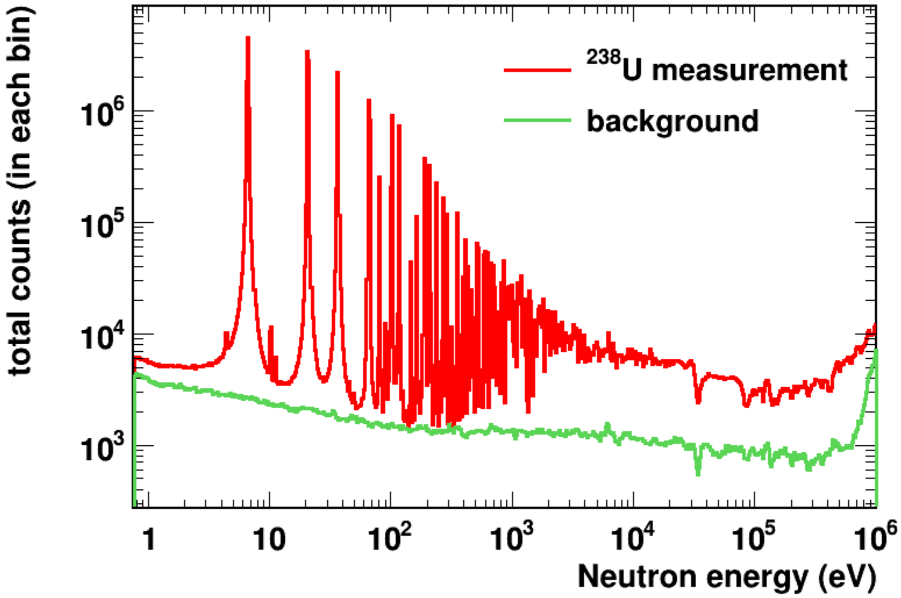

Figure 4 Expected total number of counts in the ^{238}U(n,γ) measurements (100 bpd) when delivering $0.9 \cdot 10^{18}$ protons on target. The background comes from the ^{181}Ta(n,γ) and ^{nat}Er(n,γ) measurements.

Summary of requested protons:

Measurement	Protons
Thin ^{238}U sample	$0.5 \cdot 10^{18}$
Thin ^{238}U sample – empty	$0.2 \cdot 10^{18}$
Thick ^{238}U sample	$0.9 \cdot 10^{18}$
Thin ^{238}U sample – empty	$0.4 \cdot 10^{18}$
^{197}Au sample	$0.3 \cdot 10^{18}$
^{nat}Pb sample	$0.2 \cdot 10^{18}$
^{nat}C sample	$0.2 \cdot 10^{18}$

Measurements with filters	$0.5 \cdot 10^{18}$
Total	$\mathbf{3.2 \cdot 10^{18}}$

Table 1 – Summary of the number protons on target requested for each of the configurations to be measured.

References:

[1] OECD, Nuclear Energy Agency, Collaborative International Evaluated Library Organisation (CIELO) Pilot Project, WPEC Subgroup 40 (SG40). https://www.oecd-nea.org/science/wpec/sg40-cielo/

[2] M.B. Chadwick, E. Dupont, E. Bauge et al., The CIELO collaboration: neutron reactions on ^{1}H, ^{16}O, ^{56}Fe, ^{235,238}U, and ^{239}Pu, Nucl. Data Sheets 118, 1 (2014). https://doi.org/10.1016/j.nds.2014.04.002

[3] R. Capote, A. Trkov (coordinators), IAEA CIELO Data Development Project within the International Pilot Project of the OECD/NEA, 235U and 238U files released December 1st, 2017, https://www-nds.iaea.org/CIELO/

[4] M.B. Chadwick, R. Capote, A. Trkov et al., CIELO collaboration summary results: international evaluations of neutron reactions on Uranium, Plutonium, Iron, Oxygen and Hydrogen, Nucl. Data Sheets 148, 189 (2018). https://doi.org/10.1016/j.nds.2018.02.003

[5] J. L. Ullmann et al., Cross section and γ-ray spectra for ^{238}U(n,γ) measured with the DANCE detector array at the Los Alamos Neutron Science Center, Phys. Rev. C 89, 034603 (2014). http://dx.doi.org/10.1103/PhysRevC.89.034603

[6] H. I. Kim et al., Neutron capture cross section measurements for ^{238}U in the resonance region at GELINA, Eur. Phys. J. A 52, 170 (2016). https://doi.org/10.1140/epja/i2016-16170-6

[7] F. Mingrone et al., Neutron capture cross section measurement of ^{238}U at the CERN n_TOF facility in the energy region from 1 eV to 700 keV, Phys. Rev. C 95, 034604 (2017). https://doi.org/10.1103/PhysRevC.95.034604

[8] T. Wright et al., Measurement of the ^{238}U(n,γ) cross section up to 80 keV with the Total Absorption Calorimeter at the CERN n_TOF facility, Phys. Rev. C 96, 064601 (2017). https://doi.org/10.1103/PhysRevC.96.064601

[9] A. J. M. Plompen et al., The joint evaluated fission and fusion nuclear data library, JEFF-3.3, Eur. Phys. J. A 56, 181 (2020). https://doi.org/10.1140/epja/s10050-020-00141-9

[10] R. Capote, A. Trkov, M. Sin et al., IAEA CIELO evaluation of neutron-induced reactions on ^{235}U and ^{238}U targets, Nucl. Data Sheets 148, 254 (2018). https://doi.org/10.1016/j.nds.2018.02.005

[11] O. Iwamoto et al., Japanese evaluated nuclear data library version 5: JENDL-5, Journal of Nuclear Science and Technology, 60:1, 1-60 (2023). https://doi.org/10.1080/00223131.2022.2141903

[12] IAEA, INDEN - International Nuclear Data Evaluation Network, https://www-nds.iaea.org/INDEN/

[13] R. Capote, A. Trkov, Critical review of CIELO evaluations of n+^{235}U, ^{238}U using differential experiments, EPJ Nuclear Sci. Technol. 4, 27 (2018). https://doi.org/10.1051/epjn/2018029

[14] I. Sirakov et al., Evaluation of cross sections for neutron interactions with ^{238}U in the energy region between 5 keV and 150 keV, Eur. Phys. J. A 53, 199 (2017). https://doi.org/10.1140/epja/i2017-12394-2

[15] V. Vlachoudis, Physics reach of Target#3, n_TOF Collaboration Meeting Nov. 2018. https://indico.cern.ch/event/767271/contributions/3187297/attachments/1761233/2857872/Spallation_Target_No3_2018-11-28.pdf

[16] V. Alcayne et al., Measurement of the Ta(n,γ) cross-section at EAR1, CERN-INTC-2022-038, INTC-P-640 (2022). https://cds.cern.ch/record/2834505

[17] V. Alcayne et al., Measurement of the ^{166,167,168,170}Er(n,γ) cross-section at EAR1, CERN-INTC-2023-015, INTC-P-656 (2023). https://cds.cern.ch/record/2845947

[18] E. Mendoza et al., Neutron capture measurements with high efficiency detectors and the Pulse Height Weighting Technique, Nucl. Instrum. Methods A 1047, 167894 (2023). https://doi.org/10.1016/j.nima.2022.167894

Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

Please describe here below the main parts of your experimental set-up:

Part of the experiment	Design and manufacturing
C_6D_6 setup at EAR1 with sTED detectors (main option) or new sliceTED C_6D_6 detector.	☒ To be used without any modification ☐ To be modified
Uranium samples	☒ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT

Additional hazard from <u>flexible or transported</u> equipment to the CERN site:

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Pressure	☐	[pressure] [bar], [volume][l]
	Vacuum	☒	n_TOF beam pipes on vacuum
	Machine tools	☐	
	Mechanical energy (moving parts)	☐	
	Hot/Cold surfaces	☐	
Cryogenic Safety	Cryogenic fluid	☐	[fluid] [m^3]
Electrical Safety	Electrical equipment and installations	☒	[voltage] [V], [current] [A]
	High Voltage equipment	☒	[voltage] [V]
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	☐	[fluid], [quantity]
	Toxic/Irritant	☐	[fluid], [quantity]
	Corrosive	☐	[fluid], [quantity]
	Oxidizing	☐	[fluid], [quantity]
	Flammable/Potentially explosive atmospheres	☐	[fluid], [quantity]
	Dangerous for the environment	☐	[fluid], [quantity]
Non-ionizing radiation	Laser	☐	[laser], [class]
	UV light	☐	

Safety	Magnetic field	☐	[magnetic field] [T]
Workplace	Excessive noise	☐	
	Working outside normal working hours	☒	The measurement will be running 24h per day
	Working at height (climbing platforms, etc.)	☐	
	Outdoor activities	☐	
Fire Safety	Ignition sources	☐	
	Combustible Materials	☐	
	Hot Work (e.g. welding, grinding)	☐	
Other hazards			