EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Measurement of the Ta(n,γ) cross-section at EAR1

September 26, 2022

V. Alcayne[1], D. Cano-Ott[1], E. González-Romero[1], T. Martínez[1], E. Mendoza[1], A. Sánchez-Caballero[1], J. Balibrea-Correa[2], F. Calviño[3], R. Capote[4], A. Casanovas[3], C. Domingo-Pardo[2] and J. Lerendegui-Marco[2].

[1]*Centro de Investigaciones Energéticas Medioambientales y Tecnológicas (CIEMAT), Spain*
[2]*Instituto de Física Corpuscular, CSIC - Universidad de Valencia, Spain*
[3]*Universitat Politècnica de Catalunya, Spain*
[4]*International Atomic Energy Agency, Vienna-A-1400, PO Box 100, Austria*

Spokesperson: Victor Alcayne (victor.alcayne@ciemat.es) and Emilio Mendoza (Emilio.mendoza@ciemat.es)
Technical coordinator: Oliver Aberle (Oliver.aberle@cern.ch)

Abstract:

Metallic alloys or metals with high melting points such as tantalum are being considered for the development of nuclear reactors for space. In recent critical experiments using highly enriched uranium or plutonium fuels, moderators and tantalum, large discrepancies have been found between the predicted and measured k_{eff} (i.e. needed critical masses). These observed discrepancies have been attributed to larger than reported uncertainties in the nuclear data of the materials involved, mainly tantalum, plutonium and graphite. The Ta(n,γ) cross section has also been reported as an important contributor to the uncertainty in the activation and heating of magnets used in large fusion reactors. The different measurements of the Ta neutron capture cross sections used in the evaluations are discrepant and affected by important experimental corrections like the self-shielding or angular correlations between γ-rays. For these reasons, a new measurement of Ta(n,γ) cross section with thin samples in the energy range from 0.1 eV to 500 keV is proposed at n_TOF Experimental ARea 1 (EAR1). The lower self-shielding corrections and an adequate angular distribution of the C_6D_6 detectors around the samples will result in an accuracy below 5%.

Requested protons: $2.0 \cdot 10^{18}$ protons on target
Experimental Area: EAR1

1 Introduction and motivation

The absence of hydrocarbon power sources in space and the limitations of batteries have led, since the beginning of space exploration in the late 1950s, to the development of photovoltaic and nuclear power devices. For interplanetary exploration beyond the Earth distance from the Sun, solar power quickly reaches its limits. In this case, nuclear devices are the most suitable power sources for a whole range of missions, including space missions to Jupiter and beyond and surface missions on Mars [1].

Different nuclear devices have been used in space missions. Radioactive Power Sources (RPS) are passive devices using the decay heat of radioisotopes. Also, small fission reactors similar to the ones used on the earth have been used and are under investigation for space missions. Fission power systems may be utilized to power a spacecraft's heating or propulsion systems. In terms of heating requirements, when spacecrafts require more than 100 kW power, nuclear reactors are much more cost-effective than RPSs.

For all these reasons, NASA is interested in developing a nuclear reactor for space and has carried out projects like the SP-100 [2] and, more recently, the Prometheus project [3] and the Kilopower project [4]. The Kilopower project has already built and operated a full-powered reactor on earth matching the required operational parameters [5]. In Europe, the European Space Agency (ESA) exploration programmes include future missions to the outer planets. The ESA has already used RPSs in the ExoMars project and has a drafted framework for nuclear power sources for space [1, 6].

The efficient generation of electrical power for space missions may require nuclear reactors that operate at high temperatures. Refractory metals such as Mo, W, Rh, and Ta are considered for use in space reactors due to their high melting point. Because these metals are relatively strong neutron absorbers, it is important to know their neutron cross sections accurately [7]. In general, Ta compared to Mo and W offers higher strength, readily suppliers and previous hardware programs. However, for future space fission reactors using refractory metals is needed to investigate which is the most suitable [8, 9].

In the framework of the Prometheus Project, critical experiments in refractory metals were performed [10, 11]. The configurations of the experiments consist of a cylindrical core containing plates of Highly Enriched Uranium (HEU), refractory metals, and graphite or polyethylene. In the experiment, the mass needed to make the system critical is obtained and compared to the critical mass obtained with MCNP and ENDF/B-VI. Considerable differences were observed in the comparison of the masses for different Ta experiments, as it is shown in Table 1. The experiment with the harder spectrum (Ta-2.5W-1) has a small discrepancy of less than 1%, whereas the rest of the experiments (Ta-2.5W-2,-3,-4) with softer spectrum show discrepancies from 7 to 9% that may indicate a need for additional precise measurements in the keV region and below for Ta.

In 2015, the first phase of Thermal Epithermal eXperiments (TEX) [12] was completed. TEX is a project to perform critical experiments that span a wide range of fission energy

Uncertainty	Energy spectrum in the experiment			Mass differences in percentage
	<0.625 eV	0.625 eV-100 keV	<100 keV	
Ta-2.5W-1	0.0%	14.0%	86.0%	0.17
Ta-2.5W-2	0.0%	20.7%	79.3%	9.25
Ta-2.5W-3	0.0%	31.1%	68.9%	7.67
Ta-2.5W-4	3.7%	43.4%	52.9%	7.48

Table 1: Energy spectrum in the Loaiza *et al.* critical experiments and the differences between the measured and calculated critical masses [10, 11].

spectra, from thermal (below 0.625 eV), through the intermediate (0.65 eV to 100 keV), and to fast energies (above 100 keV). One of the first elements measured was tantalum, because Ta showed the highest cross section sensitivity when used as a diluent. In 2018, the experiments with Ta and Pu ZPPR plates were performed, and the preliminary analysis points to issues with the Ta cross section in the intermediate energy region [13].

Tantalum targets have been considered for producing neutrons in Accelerator Driven Systems (ADS) [14, 15, 16]. The main problem of the use of tantalum as a target is its high thermal neutron absorption and poor oxidation resistance, however other materials have also various constraints. Tantalum has been also proposed to be used as a structural component for liquid ADS liquid lead targets [14] due to its high melting point. Moreover, tantalum is proposed as a candidate for the first wall and blanket structural components of fusion reactors [17, 18, 19]. Tantalum is considered as one of the high-priority elements for which well-qualified evaluated data sets are required for the ITER and IFMIF fusion projects [20]. In particular, it is claimed that an uncertainty lower than 10% is needed in the capture cross section of Ta in the energy region from 0.01 eV to 1 keV. This energy region is important due to the activation of Ta by thermalized neutrons [21].

2 Previous measurements and evaluations of ^{181}Ta

In the region below 100 eV the most precise measurements are the ones performed by Harvey et al. [22] and Belanova [23], these measurements were used in the compilation of S. F. Mughabghab [24]. Recently there are two new measurements by Meaze et al. [25] and McDermott et al. [26] (the Resonance Parameter (RP) obtained in this measurement are not publicly available). In Figure 2, it is observed how the radiative kernels ($\Gamma_\gamma \cdot \Gamma_n/\Gamma$) of the Meaze measurement are not compatible with the Mughabghab compilation. The Γ_γ parameters obtained in this measurement are on average four times larger than the values of Mughabghab. At energies higher than 100 eV the works considered in the evaluations are the ones of Macklin et al. [27], Tsubone et al. [28] and Yamamuro et al. [29]. In this energy region differences were observed in the critical experiments. For this reason, Brown *et al.* performed a new measurement, published in 2020 [30, 31]. However, as mentioned by the authors, the measurement has considerable limitations. All the detectors were placed at the same angles and thus the measurement

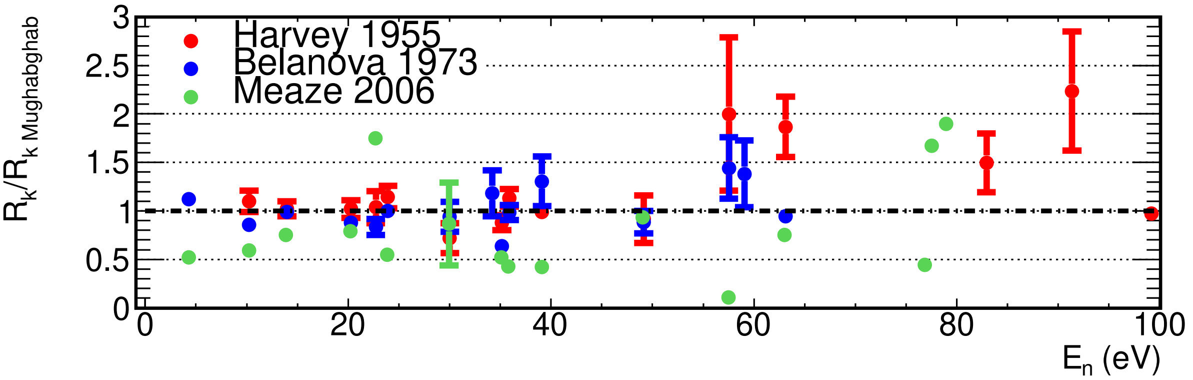


Figure 1: The radiative kernels ($\Gamma_\gamma \cdot \Gamma_n/\Gamma$) of Ta obtained in previous experiments [22, 23, 25] divided by the values of Mughabghab [24].

was not very sensible to anisotropies and angular correlations of the emitted γ-rays. The measurement was done with samples of more than 1 mm thickness, so corrections as high as 30% have to be applied in the analysis due to multiple scattering and photon attenuation. The measurements of Macklin, Tsobune and Yamamuro also used samples thicker than 1 mm.

The ENDF/B-VIII.0 library [32] is based on the work of Mughabghab and Macklin, whereas the JEFF-3.3 [33] and JENDL-4 [34] libraries take the JENDL-3.3 [35]. The JENDL-3.3 library is based on the work of Mughabghab, Macklin, Tsubone and Yamamuro. The ENDF/B-VIII.0 only reports RP until 300 eV and JENDL-3.3 reports until 2.4 keV. JENDL-5 released in 2022 includes data from an unpublished work by S. Endo at J-PARC [36]. The RP below 150 eV in JENDL-5 are taken from this work showing discrepancies with previous evaluations (Figure 2). Concerning the Unresolved Resonance Region (URR) there are differences between the three evaluations as high as 20% (Figure 2). There is also a recent work in the Lead Slowing-Down Spectrometer at Rensselaer Polytechnic Institute showing that at energies higher than 300 eV, the preliminary results obtained are not matching the evaluations [37].

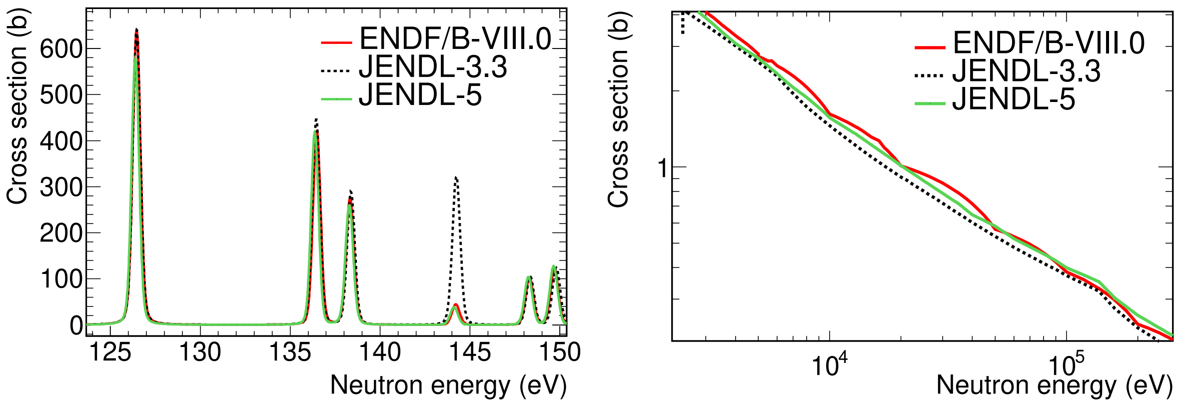


Figure 2: Capture cross section for different evaluations of ^{181}Ta.

3 Ta measurement at n_TOF EAR1

We propose to perform a new measurement at EAR1 of natural Ta (99.99% of ^{181}Ta and 1.2·10^{-4} ^{180m}Ta [38]) in the range from 0.1 eV to 500 keV with an accuracy better than 5%. We propose to use a set of carbon fibre C_6D_6 detectors [39] at 125º with respect to the beam and a complementary set of sTED detectors [40] at various angles for the determination of the angular dependence of the γ-ray emission, see Figure 3. The efficiency to detect the Ta(n,γ) cascades with one carbon fibre detector at 9 cm from the sample is ~2%, whereas the one of one sTED detector is ~0.2%. The estimated efficiency of the setup is ~7%. For the analysis, the Total Energy Detection (TED) in combination with the Pulse Height Weighting Technique (PHWT) would be used, which will led to an estimated uncertainty of 2% in the cross section associated with the techniques [41].

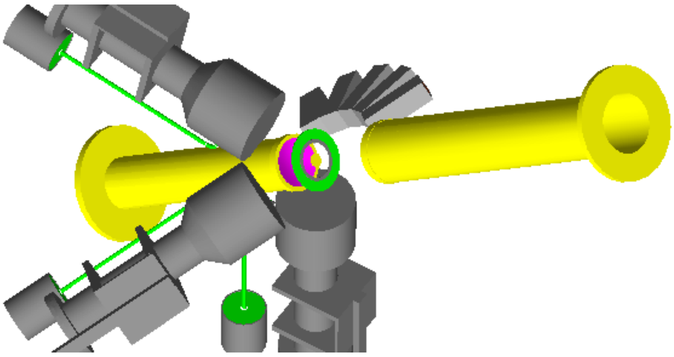


Figure 3: Schema of the possible setup for the measurement in EAR1.

In the experiment, two natural metallic samples of Ta would be used. These samples are commercially available with a purity of 99.999% [42]. The *thick* sample would be of 0.1 mm and the *thin* sample would be of 0.01 mm. In previous measurements, they use very thick samples (thickness>1 mm) and, considerable corrections were needed. For this reason, in this experiment, thinner samples are used. The *thick* sample aims to measure the keV region and the URR. As presented in Figure 4, the capture yield at low energies for this sample are close to 1 so considerable corrections will be needed. For this reason, the *thin* sample will be used aiming to measure the region below 200 eV with small corrections.

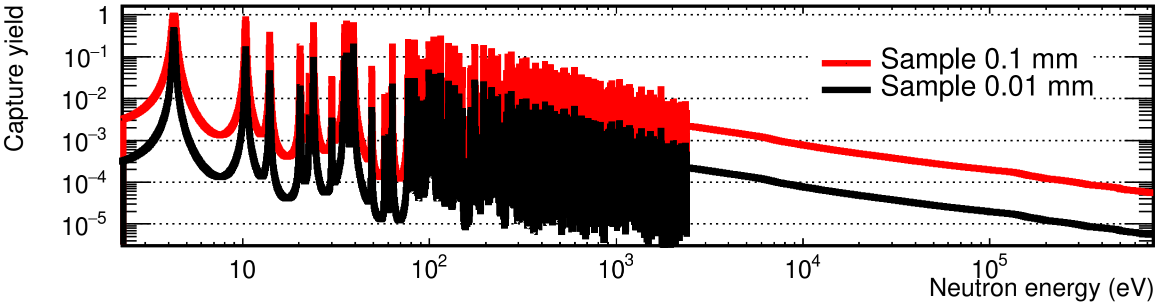
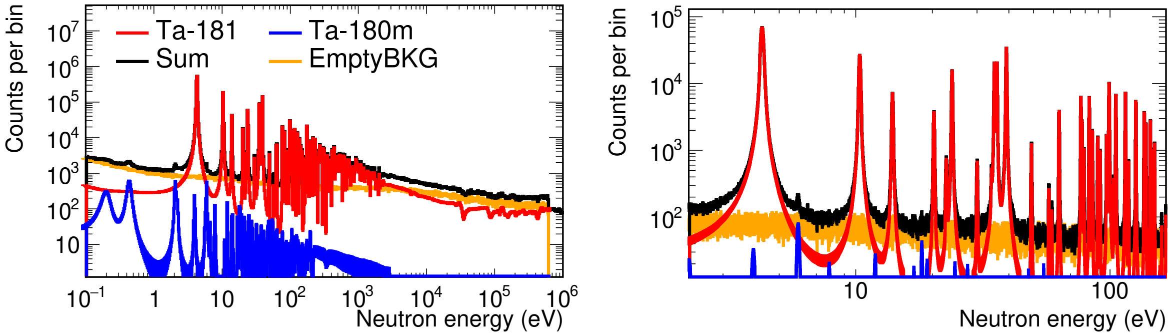
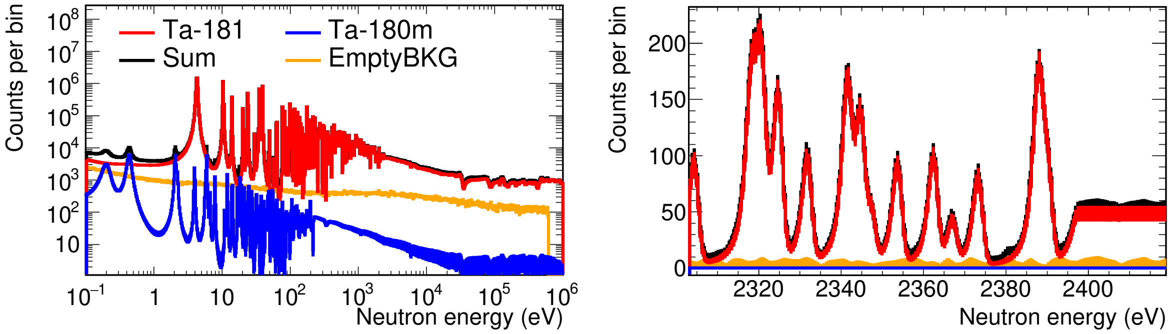


Figure 4: Estimated capture yields ^{181}Ta for the two samples.

The aimed accuracy is 5%. The uncertainty due to counting statistics has to be low in order to fullfil this requierement, considering also the ∼2% uncertainty in the detection method and the uncertainties in the sample characterization. In Figure 5, the counting rate estimates for the two samples with $7{\cdot}10^{17}$ protons each are presented. For the *thin* sample there are enough statistics to perform precise fits of the RP, with at least 2000 counts per resonance. The uncertainties due to counting statistics with the *Thick* sample are ∼3% in the URR and at lower energies ∼1% with 100 bins per decade. The background will be determined from beam-off and beam-on runs, while the normalization will be done with the saturated resonance method using the saturated resonances of the Ta *Thick* sample and the one of a ^{197}Au sample. The background and normalization measurements correspond to $6{\cdot}10^{17}$ protons.



(a) *Thin* sample (0.01 mm), 100 bins per decade (b) *Thin* sample (0.01 mm), 1000 bins per decade



(c) *Thick* sample (0.1 mm), 100 bins per decade (d) *Thick* sample (0.1 mm), 10000 bins per decade

Figure 5: Counting rate estimations for the Ta samples and $7{\cdot}10^{17}$ protons in each sample. In the plot the expected counts produced by each isotope (Ta-181 and Ta-180m), the empty beam-on background (EmtpyBKG) and the sum of all the components (Sum).

Sample	*Thin* sample	*Thick* sample	Backg. and Norm.	**Total**
Protons	$7{\cdot}10^{17}$	$7{\cdot}10^{17}$	$6{\cdot}10^{17}$	$\mathbf{2.0{\cdot}10^{18}}$

Table 2: Beam time request and distribution.

References

[1] L. Summerer et al., Proceedings of ICAPP France, 7325 (2007).

[2] V. C. Truscello, et al., SP-100, the US Space Nuclear Reactor Power Program (1983).

[3] Prometheus Project Final Report (Report). NASA/JPL. p. 191 (2005).

[4] M. A. Gibson et al., NASA TN, ID 20170002010 (2017).

[5] P. R. McClure et al., Nuc. Tech., 206 (2020).

[6] K. Stephenson et al., Proceedings of the 8th European Space Power Conference, 108 (2007).

[7] D. Bogart et al., NASA TN, ID 19700007991 (1970).

[8] R. R. Hickman et al., NASA TN, ID 20100021082 (2010).

[9] P. J. Ring and E. D. Sayre. AIP Conference Proceedings 699, 806 (2004).

[10] M. Zerkle et al., Tech. Rep. B-TM-1639, Bettis Atomic Power Laboratory (2006).

[11] D. Loaiza et al., Nucl. Sci. Eng. 160, 217 (2008).

[12] C. Percher et al., “Final design for (TEX)“ 13592-US-ANS (2015).

[13] C. Percher et al., “Thermal Epithermal eXperiments (TEX)”, Proceedings 11th ICNC (2019).

[14] OCED-NEA. ADS and FR in advanced nuclear fuel cycles. ISBN: 92-64-18482-1 (2002).

[15] L. Yang and W. Zhan Sci. China Technol. Sci. 58, 1705–1711 (2015).

[16] A. I. Dubrouski and A. I. Kiyavitskaya Phys. Part. Nuclei Lett. 17, 19–26 (2020).

[17] F. Brossa et al., Jour. Nuc. Mat., 103, 261-265 (1981).

[18] E. Tejado et al., Jour. Nuc. Mat., 467, 2, 949-955 (2015).

[19] V. A. Makhlai et al., Jour. Mat. Ene., 9, 116-122 (2016).

[20] U. Fischer et al. AIP Conf. Proceedings, 769, 1478–1485, (2005).

[21] E.T. Cheng et al., JAERI-Conf–2000-005, (2000).

[22] J. A. Harvey, D. J. Hughes, R. S. Carter, and V. E. Pilcher. Phys. Rev. 99, 10 (1955).

[23] T. S. Belanova et al., Sov. Atomic Energy, vol. 38, no. 6, pp. 553–554 (1975).

[24] S. Mughabghab and D. Gader, BNL, Upton, NY, USA, Tech. Rep. BNL325 (1973).

[25] AKM. M. H. Meaze et al., Jour. Kor. Phys. Soc., 48, 4, 827834 (2006).

[26] B. J. McDermott et al., Phys. Rev. C 96, 014607 (2017).

[27] R. L. Macklin, Nucl. Sci. Eng. 86, 362 (1984).

[28] I. Tsubone et al., J. Nuc. Sci. Tech., vol. 24, no. 12, pp. 975–987 (1987).

[29] N. Yamamuroet et al., J. Nuc. Sci. Tech., vol. 17, no. 8, pp. 582–592 (1980).

[30] J. M. Brown et al., Nucl. Sci. Eng. 194:3, 221-231 (2020).

[31] J. M. Brown et al., PhD Thesis (2019).

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

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

[34] K. Shibata et al., J. Nuc. Sci. Tech. 48:1, 1-30 (2011).

[35] K. Shibata et al., J. Nucl. Sci. Technol., vol. 39, no. 11, pp. 1125–1136 (2002).

[36] O. Iwamoto et al., "Japanese Evaluated Nuclear Data Library 5", to be published.

[37] N. W. Thompson et al., "Progress on Using a LSDS", AccApp 15 (2015).

[38] M. Weiser et al., Pure Appl. Chem., vol. 85, no. 5, pp. 1047–1078 (2013).

[39] P. F. Mastinu et al.; New C6D6 detectors,CERN-n TOF-PUB-2013-002 (2013).

[40] V. Alcayne et al. The sTED detector. Talk in the n_TOF CM (2022).

[41] U. Abbondanno et al, Nucl. Instrum. Methods A, 521 (2004).

[42] Webpage of Goodfellow. Retrieve August 2022.

Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

Part of the experiment	Design and manufacturing
C6D6 setup at EAR1 with 5 sTED detectors	To be used without any modification
Two stable natural metallic samples of tantalum with a total mass of ~1 g	Standard equipment supplied by a manufacturer

HAZARDS GENERATED BY THE EXPERIMENT
Additional hazard from flexible or transported equipment to the CERN site:
Only the two stable samples of Ta would be transported to CERN the detectors are already at the n_TOF facility.