CERN-INTC-2024-032 / INTC-P-569-ADD-1
09/04/2024

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Addendum to the Proposal INTC-P-569 for the ISOLDE and Neutron Time-of-Flight Committee

Measurement of ^{92,97,98,100}Mo(n,γ) relevant to Astrophysics and Nuclear Technology

April 9, 2024

M. Busso[1,2], D. M. Castelluccio[1,3], P. Console Camprini[1,3], N. Colonna[1], S. Cristallo[1,4], C. Domingo-Pardo[5], A. Guglielmelli[3,6], J. Heyse[7], S. Kopecky[7], C. Lederer-Woods[8], A. Manna[1,9], C. Massimi[1,9], P. Mastinu[10], A. Mengoni[1,3], P. M. Milazzo[1], R. Mucciola[1], C. Paradela-Dobarro[7], I. Porras[11], T. Rauscher[12], F. Rocchi[3], P. Schillebeeckx[7], N. V. Sosnin[6,13], N. Terranova[1,3], G. Vannini[1], and the n_TOF Collaboration[14]

[1]*Istituto Nazionale di Fisica Nucleare, INFN, Italy*
[2]*University of Perugia, Italy*
[3]*Agenzia nazionale per le nuove tecnologie, l'energia e lo sviluppo economico sostenibile (ENEA), Italy*
[4]*Istituto Nazionale di Astrofisica - Osservatorio Astronomico d'Abruzzo, Italy*
[5]*Instituto de Física Corpuscular, CSIC - Universidad de Valencia, Spain*
[6]*European Commission, Joint Research Centre (JRC), Ispra, Italy*
[7]*European Commission, Joint Research Centre (JRC), Geel, Belgium*
[8]*School of Physics and Astronomy, University of Edinburgh, United Kingdom*
[9]*Dipartimento di Fisica e Astronomia, Università di Bologna, Italy*
[10]*INFN Laboratori Nazionali di Legnaro, Italy*
[11]*Departamento de Fisica Atómica, Molecular y Nuclear - Universidad de Granada, Spain,*
[12]*Department of Physics, University of Basel, Switzerland*
[13]*University of Manchester, United Kingdom*
[14]*www.cern.ch/n_TOF*

Spokesperson: Riccardo Mucciola (riccardo.mucciola@cern.ch), Cristian Massimi (cristian.massimi@bo.infn.it)
Technical coordinator: Oliver Aberle (oliver.aberle@cern.ch)

Abstract: We propose to perform neutron capture cross section measurements at the n_TOF facility for the stable ^{92}Mo, ^{97}Mo, ^{98}Mo, and ^{100}Mo isotopes, which are relevant for nuclear astrophysics and nuclear technologies. The neutron capture cross section for these isotopes are currently known with large uncertainties. The measurements will be

carried out at EAR1 and EAR2 under similar conditions to previously performed measurements on ^{94,95,96}Mo, using optimized detection setup consisting of detectors specifically designed for accurate (n,γ) cross section determination. Highly isotopically enriched samples will be used to reduce the background induced by the other molybdenum isotopes allowing for a better assignment of the resonances.

Requested protons: 7.5×10^{18} in EAR1 and 2.5×10^{18} in EAR2
Experimental Area: EAR1 and EAR2

1 Introduction

Molybdenum is a very favorable element to study stellar nucleosynthesis because of the variety of processes involved in the creation of its stable isotopes. In fact, the formation of its seven stable isotopes can be traced back to the $p-$process (^{92}Mo and mostly ^{94}Mo), mixed $s-$ and $r-$process(^{95}Mo,^{97}Mo, and ^{98}Mo) $s-$process only (^{96}Mo), and $r-$process only (^{100}Mo). For example, studies aimed at the interpretation of $s-$process pollution traces in SiC grains and providing strong constraints on the main neutron source in AGB stars [1], are strongly dependent on the nuclear reaction network used, where accurate neutron capture rates are the key nuclear data input parameters. The recent disagreement found between stellar models yields and molybdenum abundances in presolar grain laboratory measurements [2], enforces the need for a new determination of molybdenum neutron capture cross sections, in particular for ^{97}Mo and ^{98}Mo. The accurate knowledge of neutron capture cross section for molybdenum isotopes plays a role also outside the $s-$process regime. The ^{92}Mo(n,γ) reaction cross section is important for the production of ^{92}Mo via the reverse reaction (γ,n), which is significant for stars with a temperature below 3×10^9K. In addition, the neutron capture cross section for ^{100}Mo is important for its production and destruction through the $n-$process in explosive He burning conditions in stars with a temperature around 1×10^9K.

Mo isotopes are produced as fission products in nuclear reactors and are present in Mo-based alloys used to produce nuclear fuel for research, naval and space reactors [3, 4]. In addition, thanks to its thermo-mechanical characteristics the use of molybdenum is under study for the construction of future fusion power plants [5]. An accurate Mo reaction cross sections is required for criticality safety studies that are based on a burn-up credit approach including an extended list of fission products. Such studies are important for safety assessments of spent nuclear fuel transport, storage and final disposal and handling of spent nuclear fuel in reprocessing facilities (e.g. UPu-MoZr deposits in reprocessing plant equipment). Lastly, ^{98}Mo(γ,n) reaction is one of the main source of production for ^{99m}Tc [6], radioisotope vastly used in nuclear medicine for SPECT imaging [7]. Therefore, reliable nuclear data for naturally occurring Mo isotopes are relevant for safety assessments at the back-end of the nuclear fuel cycle.

Due to the importance of an accurate knowledge of the neutron capture cross section for all the Mo isotopes, we decided to submit an addendum to the proposal INTC-P-569 [8] to perform radiative capture measurements on the remaining four stable isotopes of molybdenum: ^{92}Mo, ^{97}Mo, ^{98}Mo, and ^{100}Mo.

2 State-of-the-art

Experimental data for neutron induced capture reactions on molybdenum isotopes reported in the literature suffer from large uncertainties. This is reflected in the status of the latest evaluated nuclear data libraries, as shown in Fig.1. This figure shows the relative uncertainties for the ^{92}Mo(n,γ), ^{97}Mo(n,γ), ^{98}Mo(n,γ), and ^{100}Mo(n,γ) reactions included in the ENDF/B-VIII.0 library [9]. In addition, the plot shows the uncertainty on the cross section at thermal energy compiled by Mughabghab [10] and on the Maxwellian-

Averaged-Cross-Section (MACS) at 30 keV in the KADoNIS database [11]. In the thermal energy region, the relative uncertainties reported in ENDF/B-VIII.0 [9] for neutron capture reactions are 25%, 9%, 5%, and 2% for ^{92}Mo, ^{97}Mo, ^{98}Mo, and ^{100}Mo respectively, while for energy range between 0.5 eV and 20 keV the values of the uncertainty are 10%, 20%, 5%, and 4%. For energies above 20 keV, the uncertainties are in the order of 20% for ^{92}Mo and ^{97}Mo, and around 8% for ^{98}Mo and ^{100}Mo.
The cross section data reported in the libraries are mainly based on the measurements of Weigmann et. al. [12] and Musgrove et. al. [13]. Weigmann et. al. [12] performed radiative capture measurements using the TOF technique at the 60 m station of the CBNM electron linac (Geel, BE) using Moxon-Rae detectors and isotopically enriched samples in all stable isotopes of molybdenum. The energy of the analyzed resonance parameters ranges from few eV up to 12 keV, 2 keV, 5 keV, and 5 keV for ^{92}Mo, ^{97}Mo, ^{98}Mo, and ^{100}Mo respectively. Musgrove et. al. [13], performed TOF measurements at the 40 m station of ORELA facility (US) using fluorocarbon liquid scintillators for all stable isotopes. In this case, the resonance parameters were obtained for energies above 3 keV up to 50 keV, 3.5 keV, 31 keV, and 12 keV for ^{92}Mo, ^{97}Mo, ^{98}Mo, and ^{100}Mo respectively. Additional studies were performed on individual isotopes by Wasson et. al., [14], Chrien et. al., [15], and Weigmann et. al., [16] using the time-of-flight technique at the ORELA facility (US). Wasson et. al., [17] carried out transmission and capture measurements on samples enriched in ^{92}Mo. The capture measurements were performed at the 40 m station using fluorocarbon liquid scintillators while the transmission measurements were carried out at the 78 m station using ^{6}Li glass scintillators. The resonance parameters obtained in this work cover the energy range up to 30 keV. A set of transmission measurements were performed by Chrien et. al., [15] at the 78 m station using ^{6}Li glass scintillators on powdered oxide samples enriched in ^{98}Mo, obtaining resonance parameters for energies up to 10 keV. Lastly, Weigmann et. al., [16] performed transmission measurements on samples enriched in ^{100}Mo at the 78 m flight-path station using ^{6}Li glass scintillators. From the transmission measurements the resonance parameters of ^{100}Mo were derived up top 3.5 keV. The DANCE γ calorimeter array was used by Walker et. al., [18] to study the property of γ-rays emitted after ^{97}Mo(n,γ) reactions in the resonance region. No capture yield were derived, however the spin assignment for resonances up to 1.7 keV was performed in this experiment.

3 Proposed experimental setup

We propose to perform time-of-flight measurements to determine the cross section for capture reaction on ^{92}Mo, ^{97}Mo, ^{98}Mo, and ^{100}Mo. The total energy detection principle in combination with the pulse height weighting technique will be applied using an array of C_6D_6 scintillators in EAR1 [19] and the sTED array in EAR2 [20]. These setup were optimized by the n_TOF collaboration to minimize their neutron sensitivity. In addition, the procedures to derive accurate weighting functions and capture yields are well defined [21]. The measurements are proposed at both EAR1 and EAR2 experimental areas in order to accurately estimate the neutron capture cross section from thermal energies (25.3 meV) up to 100 keV. From previous experiments performed at n_TOF, it was demonstrated that

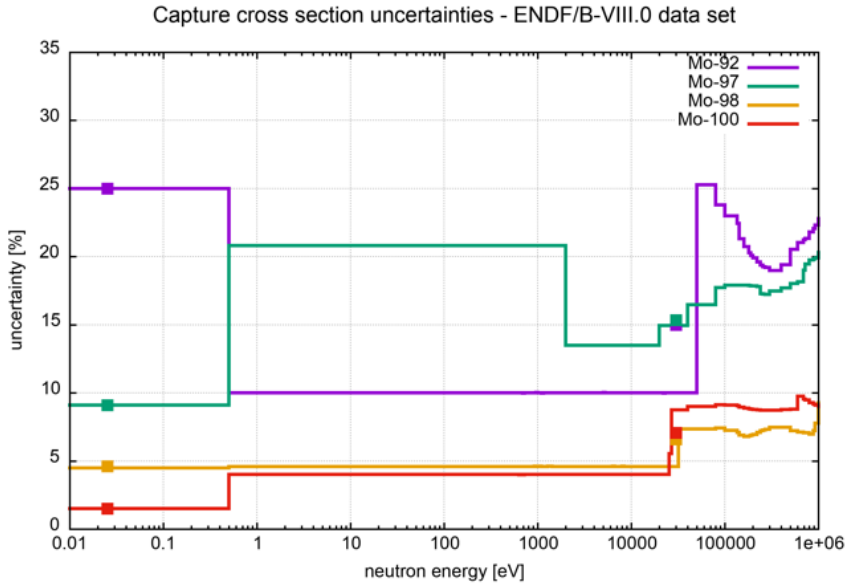

Figure 1: Relative uncertainties of the cross sections for the ^{92}Mo(nγ), ^{97}Mo(nγ), ^{98}Mo(nγ), and ^{100}Mo(nγ) reactions recommended in the ENDF/B-VIII.0 library. The uncertainties for the cross sections at thermal compiled by [10] and at 30 keV in the KADoNIS database [11] are represented by a dot.

the cross section near thermal energies can be estimated with higher accuracy at EAR2 because of the high thermal neutron flux available (thermal neutrons are suppressed, on purpose, in the beam line leading to EAR1 with the use of borated water moderation). On the other hand, the excellent energy resolution of EAR1 allows an accurate determination of resonance parameters in the epithermal region and above. The samples will be metallic disks of 20 mm diameter with an isotopical enrichment $> 95\%$ for the four isotopes. For each isotope a sample with a total mass of about 2 g will be produced. In addition, metallic disks of natural molybdenum will be used. As for past measurements, ancillary measurements with ^{197}Au, ^{208}Pb, and ^{nat}C with similar geometrical properties will be performed for normalization and background estimation. In addition, the MACS of ^{98}Mo could also be obtained with an activation measurement at the NEAR station, using techniques already tested at n_TOF [22]. This activation measurement could be performed parasitically, without increasing the number of requested protons.
The preliminary analysis on the neutron capture and transmission measurements on ^{94,95,96}Mo proposed in INTC-P-569 [8] and performed using the same detection setups, shows some discrepancies with the data in the latest evaluations and the possibility to extend the resolved resonance region for these isotopes and improve the accuracy on the cross section [23, 24]. This supports the appropriateness of the experimental technique for these measurements.

4 Beam time request

The expected count rates were calculated to estimate the number of protons required to obtain capture kernel with an uncertainty of the level of few percent. Calculations were

performed assuming, for each sample, a metallic disk of 20 mm diameter with a mass of 2 g. For the count rate estimation, the capture cross section recommended in ENDF/B-VIII.0 was used. The effect of the resolution function was included in the calculation using the SAMMY code [25]. The expected count rates, with 1000 Bins Per energy Decade (BPD), are shown in Fig. 2,3. For the measurements at EAR1 (Fig. 2,3, left panel), 2.0×10^{18}, 1.0×10^{18}, 1.7×10^{18}, and 1.7×10^{18} protons were considered for ^{92}Mo, ^{97}Mo, ^{98}Mo, and ^{100}Mo, respectively. For the measurements in EAR2 (Fig. 2,3, right panel), 6×10^{17}, 4×10^{17}, 5×10^{17}, and 5×10^{17} protons were considered for the four samples. The number of protons for each sample was chosen to obtain an uncertainty due to counting statistics below 3% in the resonance region and in the unresolved resonance region considering a binning of 1000BPD and 100BPD, respectively. In Fig. 2,3 is also reported an estimation of the background obtained in previous capture measurements with the same detection setup proposed in here. The background for both areas was estimated from measurements with an empty sample in the beam (therefore labeled Empty in the figure). The requested beam time for the normalization procedure and the study of the background is equal to 1.1×10^{18} and 5×10^{17} for EAR1 and EAR2 respectively. An overview of the requested protons is shown in Tab.1. Given the considerate amount of proton beam required to reach the required statistical uncertainties for the four isotopes, the actual measurements can be performed over a period of several year runs (e.g., one or two isotope/year).

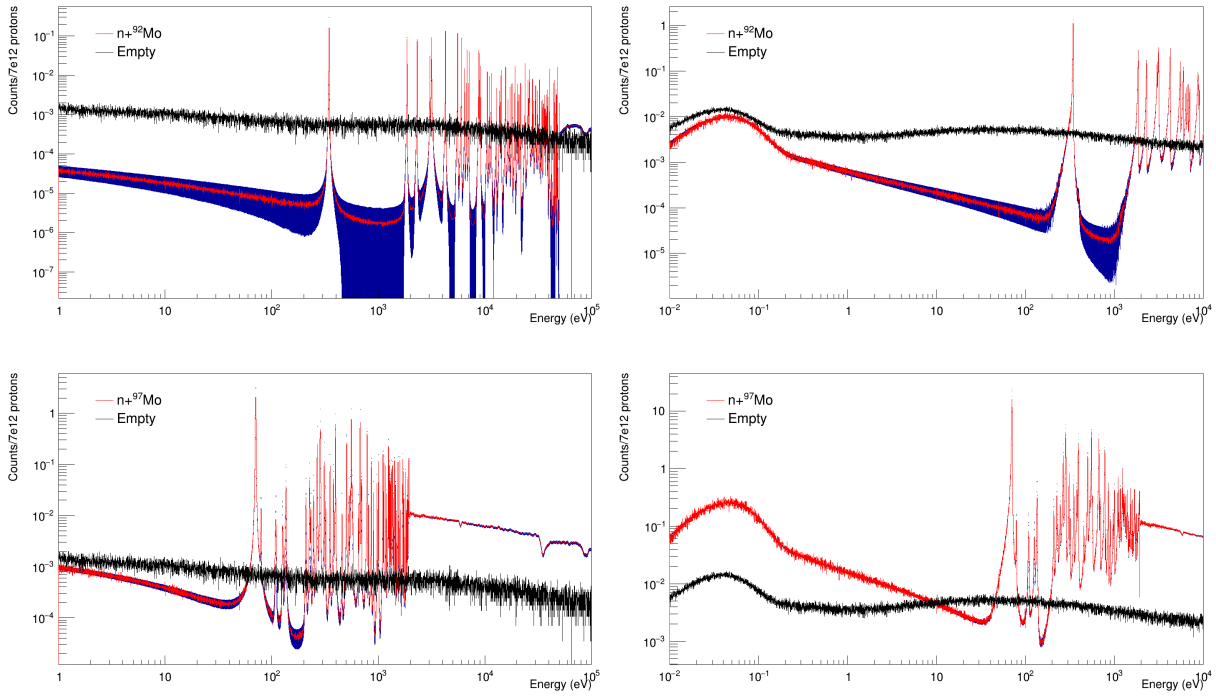

Figure 2: Expected count rate at EAR1 (left panel) and EAR2 (right panel) for ^{92}Mo (top) and ^{97}Mo (bottom). The count rates (red) with uncertainties due to counting statistics (blue) are shown compared to the n_TOF background (black), at a resolution of 1000 bins per energy decade. A total of 2.0×10^{18} and 1.0×10^{18} were considered in EAR1 for ^{92}Mo and ^{97}Mo, respectively. For the calculation of EAR2, 6×10^{17} and 4×10^{17} protons were considered for ^{92}Mo and ^{97}Mo, respectively.

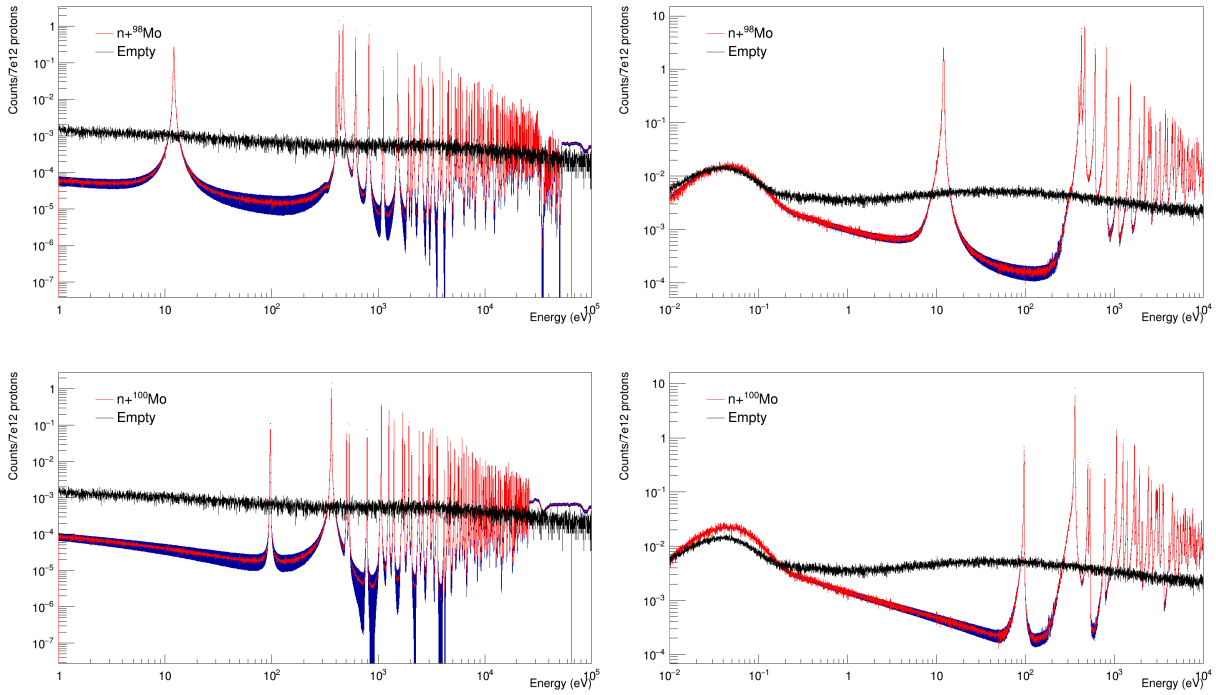

Figure 3: Expected count rate at EAR1 (left panel) and EAR2 (right panel) for ^{98}Mo (top) and ^{100}Mo (bottom). The count rates (red) with uncertainties due to counting statistics (blue) are shown compared to the n_TOF background (black), at a resolution of 1000 bins per energy decade. A total of 1.7×10^{18} and 5×10^{17} protons were considered for each sample for EAR1 and EAR2, respectively.

Sample	EAR1 (C_6D_6)	EAR2 (sTED)
^{92}Mo	2.0×10^{18}	6×10^{17}
^{97}Mo	1.0×10^{18}	4×10^{17}
^{98}Mo	1.7×10^{18}	5×10^{17}
^{100}Mo	1.7×10^{18}	5×10^{17}
Normalization (Au)	2×10^{17}	1×10^{17}
Background (Empty/Dummy, C, Pb)	9×10^{17}	4×10^{17}
Total	7.5×10^{18}	2.5×10^{18}

Table 1: Requested number of protons for ^{92}Mo, ^{97}Mo, ^{98}Mo, and ^{100}Mo in EAR1 and EAR2. The requested number of protons for ancillary normalization and background estimation measurements are also reported.

5 Conclusion

The proposed measurement is to be carried out in both EAR1 and EAR2. We request a total of 7.5×10^{18} protons in EAR1 and 2.5×10^{18} in EAR2, respectively. The objective of the measurements is to improve the accuracy of molybdenum nuclear data for all naturally occurring isotopes. Given the considerate amount of proton beam required to reach the required statistical uncertainties for the four isotopes, the actual measurements can be performed over a period of several year runs.

Summary of requested protons: 7.5×10^{18} in EAR1 and 2.5×10^{18} in EAR2

References

[1] T. Stephan, R. Trappitsch, A. M. Davis, M. J. Pellin, D. Rost, M. R. Savina, R. Yokochi, N. Liu, *CHILI – the Chicago Instrument for Laser Ionization – a new tool for isotope measurements in cosmochemistry*, International Journal of Mass Spectrometry **407** (2016) 1–15.

[2] N. Liu, T. Stephan, S. Cristallo, R. Gallino, P. Boehnke, L. R. Nittler, C. M. O. Alexander, A. M. Davis, R. Trappitsch, M. J. Pellin, I. Dillmann, *Presolar Silicon Carbide Grains of Types Y and Z: Their Molybdenum Isotopic Compositions and Stellar Origins*, The Astrophysical Journal **881** (1) (2019) 28.

[3] B. Cheng, Y. Kim, P. Chou, Nucl. Eng. Tech. **48** (2016) 16–25.

[4] X. Iltis, H. Palancher, J. Allenou, F. Vanni, B. Stepnik, A. Leenaers, S. V. D. Berghe, D. Keiser, I. Glagolenko, EPJ Nuclear Sci. Technol. **4** (2018) 49.

[5] M. Gilbert, L. Packer, T. Stainer, Nuclear Fusion **60** (2020) 106022.

[6] *Feasibility of Producing Molybdenum-99 on a Small Scale Using Fission of Low Enriched Uranium or Neutron Activation of Natural Molybdenum*, no. 478 in Technical Reports Series, 2015.

[7] P. Harper, K. A. Lathrop, F. Jiminez, R. Fink, A. Gottschalk, Radiology **85** (1) (1965) 101–109.

[8] C. Massimi, et al., *Measurement of ^{94,95,96}Mo(n,γ) relevant to Astrophysics and Nuclear Technology* CERN-INTC-2020-052, INTC-P-569 (2020).

[9] D. Brown, et al., Nucl. Data Sheets **148** (2018) 1–142.

[10] S. Mughabghab, *Atlas of neutron resonances*, 2006.

[11] I. Dillmann, T. Szücs, R. Plag, Z. Fülöp, F. Käppeler, A. Mengoni, T. Rauscher, Nuclear Data Sheets **120** (2014) 171–174.

[12] H. Weigmann, G. Rohr, J. Winter, in: Proceedings of the Third Conference on Neutron Cross Sections and Technology, Conf-710301, National Technical Information Center, Springfield, Virginia, Vol. 2, 1971, p. 749.

[13] A. De L. Musgrove, B. Allen, J. Boldeman, R. Macklin, Nucl. Phys. A **270** (1976) 108–140.

[14] O. A. Wasson, G. G. Slaughter, Phys. Rev. C **8** (1973) 297–314.

[15] R. E. Chrien, G. W. Cole, G. G. Slaughter, J. A. Harvey, Phys. Rev. C **13** (1976) 578–594.

[16] H. Weigmann, S. Raman, J. A. Harvey, R. L. Macklin, G. G. Slaughter, Phys. Rev. C **20** (1979) 115–127.

[17] O. A. Wasson, B. J. Allen, R. R. Winters, R. L. Macklin, J. A. Harvey, Phys. Rev. C **7** (1973) 1532.

[18] C. L. Walker, M. Krtička, B. Baramsai, F. Bečvář, T. A. Bredeweg, A. Chyzh, R. C. Haight, M. Jandel, J. Kroll, G. E. Mitchell, J. M. O'Donnell, R. S. Rundberg, J. L. Ullmann, S. Valenta, J. B. Wilhelmy, Phys. Rev. C **92** (2015) 014324.

[19] P. Mastinu, R. Baccomi, E. Berthoumieux, D. Cano-Ott, F. Gramegna, C. Guerrero, C. Massimi, P. Milazzo, F.Mingrone, J. Praena, G. Prete, A. García, n_TOF-PUB-2013-002 .

[20] V. Alcayne, et al., *A segmented total energy detector (sTED) for (n,γ) cross section measurements at n_TOF EAR2*, ND2022 Conference, EPJ Web of Conferences (2022).

[21] U. Abbondanno, et al., *New experimental validation of the pulse height weighting technique for capture cross-section measurements*, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment **521** (2) (2004) 454–467.

[22] E. Stamati, A. Manna, et al., *Neutron capture cross section measurements by the activation method at the n_TOF NEAR Station*, Tech. rep., CERN, CERN-INTC-2020-052, INTC-P-569 (2020).

[23] R. Mucciola, et al., *Neutron capture and total cross-section measurements on ^{94,95,96}Mo at n_TOF and GELINA*, EPJ Web of Conf. **284** (2023) 01031.

[24] *Neutron capture and total cross-section measurements on ^{94,95,96}Mo at n_TOF and GELINA, presented by R. Mucciola at* , Sesto Incontro Nazionale di Fisica Nucleare (2023).

[25] N. M. Larson, ORNL/TM-9179/R8 ENDF-364/R2 (2008).

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
C6D6, SIMON, sTED	☒ To be used without any modification ☐ To be modified

HAZARDS GENERATED BY THE EXPERIMENT

Additional hazard from flexible or transported equipment to the CERN site:

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Pressure	☐	[pressure] [bar], [volume][l]
	Vacuum	☐	
	Machine tools	☐	
	Mechanical energy (moving parts)	☐	
	Hot/Cold surfaces	☐	
Cryogenic Safety	Cryogenic fluid	☐	[fluid] [m3]
Electrical Safety	Electrical equipment and installations	☐	[voltage] [V], [current] [A]
	High Voltage equipment	☒	C6D6: 1500V; sTED: 800V
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 Safety	Laser	☐	[laser], [class]
	UV light	☐	
	Magnetic field	☐	[magnetic field] [T]
Workplace	Excessive noise	☐	
	Working outside normal working hours	☐	
	Working at height (climbing platforms, etc.)	☐	
	Outdoor activities	☐	
Fire Safety	Ignition sources	☐	
	Combustible Materials	☐	
	Hot Work (e.g. welding, grinding)	☐	
Other hazards			