EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Measurement of ^{94,95,96}Mo(n,γ) relevant to Astrophysics and Nuclear Technology

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Abstract We propose neutron capture cross section measurements at the n_TOF facility to improve the data accuracies for the stable ^{94}Mo, ^{95}Mo, and ^{96}Mo isotopes, which are relevant for nuclear astrophysics and nuclear technology and presently known with large uncertainties. Measurements will be carried out at EAR1 and EAR2 under similar conditions as previous neutron capture cross section measurements successfully performed at n_TOF, using an optimized detection setup consisting of specifically designed detectors for accurate (n,γ) cross section determination. Isotopically highly-enriched samples will be used, thus reducing the background introduced by other molybdenum isotopes and allowing for a better isotope assignment of the resonances. These measurements are part of an ongoing nuclear data initiative within the framework of the EU Horizon-2020 SANDA project.

Requested protons: 8×10^{18} protons on target
Experimental Area: EAR1 and EAR2

1 Introduction and scientific motivation

Molybdenum is a very favourable element to study nucleosynthesis because of the variety of processes involved in its origin. 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). Neutron capture cross sections are key nuclear physics quantities for understanding and modelling these processes.

As an example, studies to interpret traces of s-process pollution 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 ^{94}Mo and ^{95}Mo [3].

An additional example is provided by the recent spectroscopic observations of evolved low mass stars [4], showing in their photosphere both Nb and Tc. The enhanced Nb, produced through the mass-exchange processes characterizing binary systems known as *Ba-stars* (see e.g. [5]), can be an effective seed for producing neutron-rich Mo isotopes more efficiently than so far imagined, starting from ^{94}Mo (following the decay of ^{94}Nb).

Neutron captures on ^{94,95,96}Mo have been identified as strong key reactions impacting the s-process in AGB stars and a reduction in uncertainty in the cross section is highly desirable as pointed out in a recent investigation by Cescutti *et al.* (2018) [6]. While neutron captures on ^{94,96}Mo are mainly important in the thermal pulses of the AGB star, ^{95,96}Mo(n,γ) appear as important reactions in the interpulse period in models covering a wide range of possible stellar conditions.

The mentioned complex blend of processes affecting Mo isotopes is therefore bound to reveal more subtleties, whose disentangling cannot be done without a superior quality in neutron capture cross sections data.

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 [7, 8, 9]. An accurate ^{95}Mo(n,γ) reaction cross section is required for criticality safety studies that are based on a burnup 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). Therefore, reliable nuclear data for Mo are relevant for safety assessments at the back-end of the nuclear fuel cycle [10].

While nuclear data of several Mo isotopes are important as just outlined, we have limited the present proposal to the three isotopes ^{94}Mo, ^{95}Mo and ^{96}Mo. This choice has been made considering, in addition to their relevance in the fields mentioned above, other constrains such as the uncertainties of the experimental information already known, the availability of sample material, and the number of protons required to reach a target uncertainty on the measurements.

2 State-of-the-art

Experimental data for neutron induced capture reactions on Mo isotopes that are reported in the literature suffer from large uncertainties. This is reflected in the status of the latest evaluated and compiled data libraries, as illustrated in Figure 1. This figure plots the relative uncertainties as a function of neutron energy for the cross sections of the ^{94}Mo(n,γ), ^{95}Mo(n,γ) and ^{96}Mo(n,γ) reactions which are recommended in the ENDF/B-VIII.0 library [11], for the cross sections at thermal energy compiled by Mughabghab and for the maxwelllian-averaged capture cross section (MACS) at 30 keV in the KADONIS database [12]. In the thermal energy region, the relative uncertainties of the cross sections

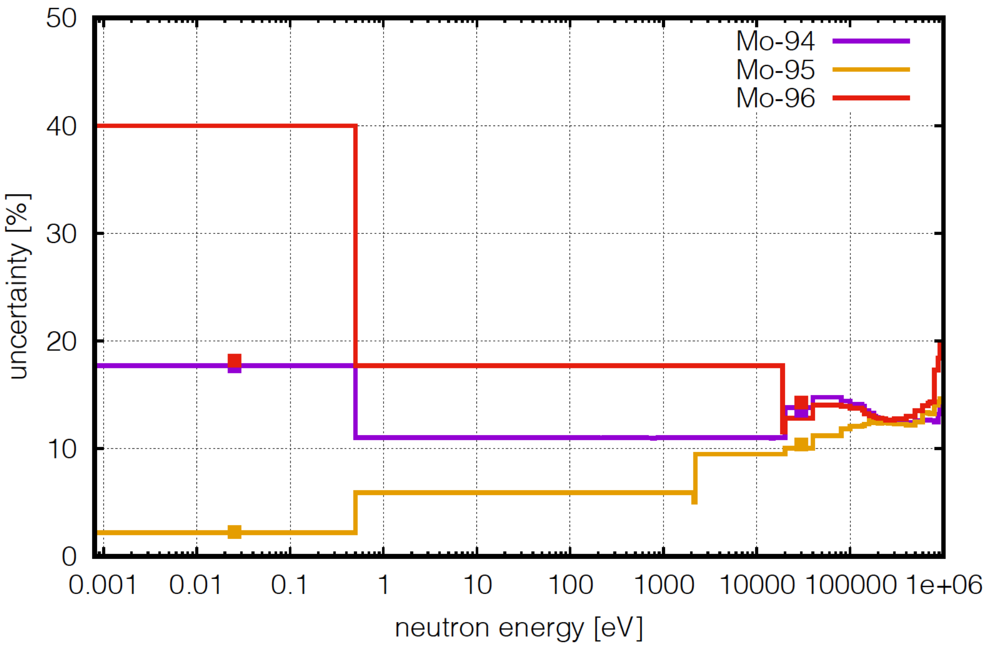

Figure 1: Relative uncertainties of the cross sections for the ^{94}Mo(n,γ), ^{95}Mo(n,γ) and ^{96}Mo(n,γ) reactions that are recommended in the ENDF/B-VIII.0 library (full line). The uncertainties for the cross sections at thermal energy compiled by [13] and at 30 keV in the KADONIS database [12], are represented by a dot.

for the ^{94}Mo(n,γ), ^{95}Mo(n,γ) and ^{96}Mo(n,γ) reactions in ENDF/B-VIII.0 [11] are 18%, 3%, and 40% respectively, while from 0.5 eV to 2 keV, the relative uncertainties are 12%, 7%, and 18%. Above 2 keV, the relative uncertainties are in the order of 10 – 20%.
Cross sections data for the ^{94}Mo(n,γ), ^{95}Mo(n,γ) and ^{96}Mo(n,γ) reactions are mainly available based on results of measurements by Weigmann and Schmid [14], Musgrove *et al.* [15] and Leinweber *et al.* [16].
Weigmann and Schmid performed TOF measurements at the GELINA facility of the JRC Geel (BE), applying the total energy detection principle using a Moxon-Rae detector and natural molybdenum samples. Capture yields were derived in the energy region between 10 eV and 25 keV. Capture kernels for resonances below 1.5 keV were derived from an area analysis. For energies between 1 keV and 25 keV an average capture cross section for natural Mo was derived.

Musgrove *et al.* performed TOF measurements at a 40 m station of the ORELA facility (US) using a pair of fluorocarbon liquid scintillators and enriched samples. They derived capture yields for neutron energies ranging between 3 keV and 90 keV. A shape analysis was performed to derive capture kernels for ^{92}Mo and $^{94-98}$Mo, for neutron energies below 50 keV, 20 keV, 4 keV, 20 keV, 3.5 keV and 31 keV, respectively. The capture kernels derived from these data are in reasonable agreement with those derived by Weigmann and Schmid. Musgrove *et al.* noted the impact of the neutron widths that were adopted to derive capture kernels and the lack of proper transmission data for a reliable analysis of the capture yield data in particular for an analysis of overlapping resonances. The capture yields were used to derive average capture cross sections for energies between 1 keV and 100 keV.

The capture data of Musgrove *et al.* were used in Ref. [17] and [18] to evaluate MACS. The MACS at 30 keV reported in [17] and [18] differ by about 30%. In addition, in the data reduction and analysis no correction for the neutron sensitivity of the detection system was applied. Hence, the results of the capture data of both Weigmann and Schmid and Musgrove *et al.* might suffer from bias effects due to the impact of the neutron sensitivity of the detection system. Unfortunately, no numerical capture yields are available to verify this effect.

More recently, Leinweber *et al.* derived capture yields and transmissions from TOF measurements at the RPI facility (US) for neutron energies between 10 eV and 2 keV using natural samples. The capture detection system was a total absorption detector based on an array of NaI scintillator detectors. Capture kernels for neutron energies below 2 keV were derived from a resonance shape analysis of the data. To avoid corrections for the neutron sensitivity of the detection system, the analysis of the capture data was limited to neutron energies below 600 eV.

The DANCE array was used by Sheets *et al.* [19] to study the properties of γ-rays emitted after ^{94,95}Mo(n,γ) reactions in the resonance region. No capture yields were derived. However, their results include spin and parity assignment of resonances for neutron interactions with ^{94}Mo and ^{95}Mo, which are essential input data to perform a resonance shape analysis of experimental capture yields or transmissions.

The resolved resonance parameters and average capture cross sections in the unresolved region for Mo isotopes in the main data libraries are primarily based on the compilation of Mughabghab [13]. The lack of proper evaluated data is primarily due to the limited number of well documented and accurate experimental transmissions and capture yields. To improve the status of evaluated data libraries for Mo isotopes and in particular to improve the quality of the recommended capture cross sections, a collaborative effort has been planned as part of the SANDA project supported within the EU Horizon 2020 framework programme [20]. It includes capture measurements at n_TOF and transmission measurements at the GELINA facility of the JRC-Geel using isotopically enriched Mo metallic samples.

3 Proposed experimental setup

We propose to perform time-of-flight measurements to determine the cross section for capture reactions on ^{94}Mo, ^{95}Mo and ^{96}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. This set-up was optimised by the n_TOF collaboration [21] to minimise its neutron sensitivity. In addition, the procedures to derive accurate weighting functions and capture yields are well defined [22].

The measurements are proposed at both EAR1 and EAR2 measuring stations in order to accurately estimate the neutron capture cross-section for neutron energies between thermal (25.3 meV) and 100 keV. From previous similar experiments at n_TOF, it was demonstrated that at EAR2 the cross section at near thermal energy can be estimated more accurately because of the absence of sizeable changes in the neutron beam profile due to the gravitational effects, as is the case in EAR1 [23]. On the other hand the excellent energy resolution of EAR1 enables the extraction of accurate resonance parameters in the epithermal region and above.

The samples will be metallic discs with a 30 mm diameter, produced from three batches of material enriched in ^{94}Mo, ^{95}Mo and ^{96}Mo to about 98.9 wt%, 94.3 and 95.7%, respectively. Five samples will be produced. For each isotope a sample with a total mass of about 2 g will be available for capture measurements. For ^{95}Mo and ^{96}Mo two disks of different masses will be produced. The partition in a thin and a thick sample is done to optimize the sample thickness for the transmission as well as capture measurements. The main characteristics of the isotopically enriched samples are specified in Table 1. In addition, metallic discs of natural molybdenum will be used. Results of transmission and capture measurements using these samples will be used to assign resonances to the correct Mo isotope and ensure consistency between the data for the individual isotopes and cross section data for natural Mo. As for past measurements, samples of ^{197}Au, ^{208}Pb and ^{nat}C with similar geometrical properties will be used for additional normalization and background measurements.

	^{94}Mo	^{95}Mo		^{96}Mo	
Enrichment [%]	98.90	> 94.3		> 95.7	
Mass [g]	2.0	0.1	1.9	0.2	1.8
Area density [at/b]	1.8×10^{-3}	9.0×10^{-5}	1.7×10^{-3}	1.7×10^{-4}	1.6×10^{-3}

Table 1: Mass and areal density of the three enriched molybdenum samples.

4 Beam-time request

Expected count rates were calculated to estimate the number of protons required to derive capture kernels and average capture cross sections with an uncertainty of the level of a few percent.

Calculations were performed for the samples specified in Table 1 using the cross sections recommended in ENDF/B-VIII.0 and assuming a 20% detection efficiency for the C_6D_6

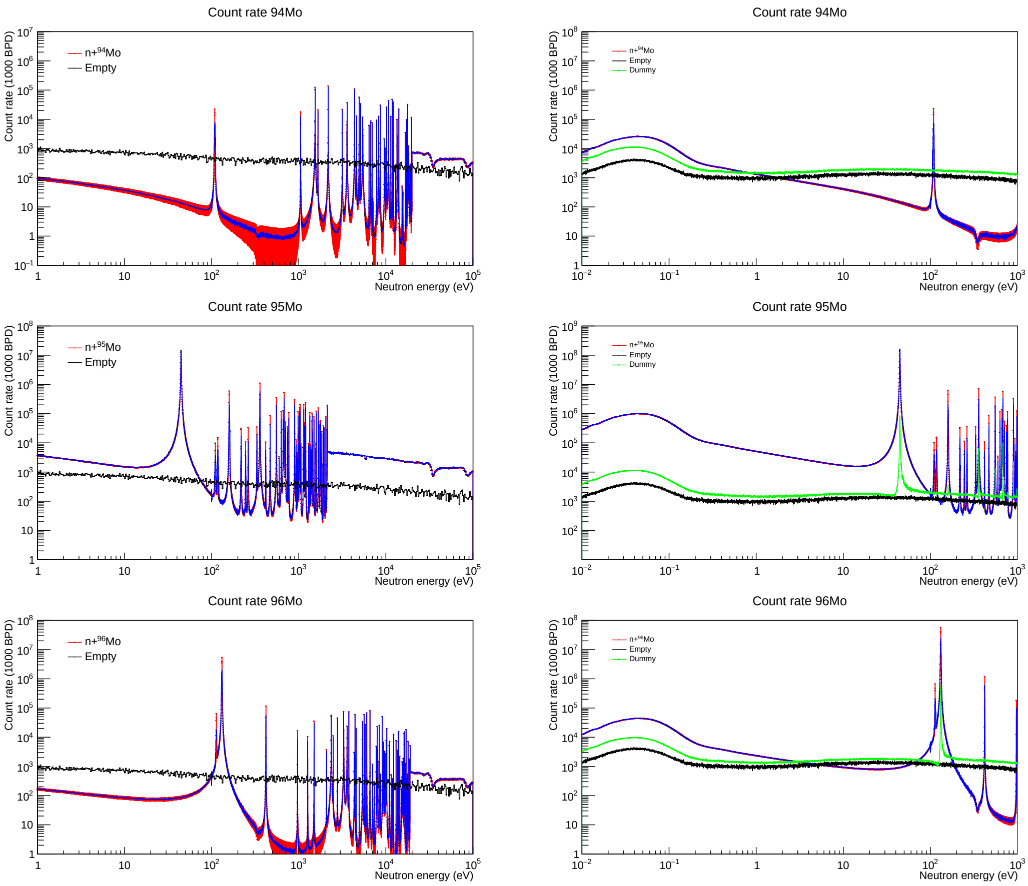

Figure 2: Expected count rate at EAR1 (left column) and EAR2 (right column) for ^{94}Mo (top), ^{95}Mo(center) and ^{96}Mo (bottom) samples. The count rates (blue) with uncertainties due to counting statistics (red) are shown compared to the n_TOF background (black) and a dummy sample (green, see text for details), at a resolution of 1000 bins per neutron energy decade. A total of 2×10^{18} and 0.5×10^{18} protons were considered in the calculations for each sample for EAR1 and EAR2, respectively.

array, typical for the experimental setup considered. The expected count rates, with 1000 bins per energy decade, are displayed in Figure 4 for the proposed measurements at EAR1 (left panels) and for EAR2 (right panels). They were obtained considering 2×10^{18} protons in EAR1 and 0.5×10^{18} in EAR2, respectively.
As already mentioned, the energy range between 10 eV and 100 keV can be better studied in EAR1 because of the better energy resolution than in EAR2. This energy range covers completely the resolved resonance region of the three molybdenum isotopes. In EAR2 the calculation was restricted in the region of major interest, that is between 10 meV and 1 keV, although we cannot exclude at the moment that the EAR2 data in the unresolved region can be used for the extraction of the cross section. This possibility will be explored, once that the background in the region above 1 keV will be extracted from the experimental data.
In the calculation of Figure 4, the estimation of the background is based on previous capture measurements. In particular, for EAR1 it is based on a capture measurement with an empty sample (therefore named Empty). For EAR2, we added an estimation of the background contribution from scattered neutrons from the molybdenum samples (labelled as Dummy, in the Figure). The requested beam time for the normalization procedure and the study of the background is equal to 5×10^{17} additional protons on target.

5 Conclusion

In conclusion, we request a total of 8×10^{18} protons (6×10^{18} for EAR1 and 2×10^{18} for EAR2, respectively) on target to carry out the proposed measurement and improve the accuracy of molybdenum nuclear data. The capture data from n_TOF, together with the transmission data from GELINA will be included in future evaluations, as proposed in the H2020 SANDA project.
Summary of requested protons: 8×10^{18}.

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