

MEASUREMENT, EVALUATION AND VALIDATION OF MOLYBDENUM CROSS SECTIONS

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ABSTRACT

Available nuclear data for molybdenum included in the nuclear data libraries are not of sufficient quality for reactor physics or criticality safety issues and indeed information about uncertainties and covariance is either missing or leaves much to be desired. Therefore, IRSN and JAEA performed experimental measurements on molybdenum at the J-PARC (Japan Proton Accelerator Research Complex) facility in Japan. The aim was to measure capture cross section and transmission of natural molybdenum at the ANNRI (Accurate Neutron-Nucleus Reaction measurement Instrument) in the MLF (Material Life and science Facility) of J-PARC. The measurements were performed on metallic natural molybdenum samples with various thicknesses. A NaI detector, placed at a flight-path length of about 28 m, was used for capture measurements and a Li-glass detector (flight-path length of about 28.7 m) for transmission measurements. Following the data reduction process, the measured data are being analyzed and evaluated to produce more accurate cross sections and associated uncertainties.

KEYWORDS: differential experiments, molybdenum, capture, transmission, J-PARC

1. INTRODUCTION

A precise knowledge of neutron cross sections is of a great importance to accurately calculate reaction rates and detailed neutron flux distributions in many nuclear applications. Reducing uncertainties in the neutron cross section data can result in an enhanced safety of present and future nuclear systems.

Molybdenum isotopes are mainly encountered in irradiated fuel as fission products or in molybdenum alloys in research or naval reactors. In the nuclear fuel cycle, ⁹⁵Mo is also taken into account in criticality safety studies considering burn-up credit for transportation casks or irradiated fuel storage. ⁹⁵Mo is one of the 15 main absorbing fission products in Light Water Reactors (LWR) irradiated fuel assemblies. Besides, in reprocessing plants, during the dissolution process, some UPu-MoZr deposits could appear in specific equipments. Thus, accurate nuclear data of molybdenum isotopes in a wide energy range are of great importance. As an example, molybdenum isotopes are among the greatest contributors to the global nuclear data uncertainty on calculated k_{eff} in UPu-MoZr deposits configurations.

The available nuclear data for molybdenum included in the nuclear data libraries are not of sufficient quality for reactor physics or criticality safety issues and indeed information about uncertainties and

covariance are either missing or leaves much to be desired. Therefore, IRSN and JAEA performed experimental measurements on molybdenum at the J-PARC (Japan Proton Accelerator Research Complex) facility in Japan. The aim was to measure the capture cross section and transmission of natural molybdenum.

2. MEASUREMENTS AT THE J-PARC FACILITY

In 2018, IRSN submitted a proposal to the J-PARC system to perform experiments with molybdenum samples and was granted 7 days of neutron beam. Molybdenum capture cross sections and transmission measurements were performed on five metallic natural molybdenum samples of various thicknesses. Additional measurements were performed to determine the background and normalization factors.

2.1. J-PARC facility

J-PARC is a proton accelerator facility operated by JAEA and KEK in Tokai-Mura. The measurements were carried out using the ANNRI (Accurate Neutron-Nucleus Reaction measurement Instrument) in the MLF (Material Life and science Facility for the purpose of using neutrons to investigate material properties) of J-PARC [1].

Neutrons were produced via spallation reactions induced by a 3-GeV pulsed proton beam impinging on a mercury target of MLF. The accelerator was operated in single-bunch mode at a repetition rate of 25 Hz. The proton beam power on the spallation target was around 500 kW. The number of incident protons was recorded and used for normalization of each measurement.

3 types of detectors are available (see Figure 1) at the ANNRI, allowing for transmission and capture cross sections measurements:

- a 4π germanium spectrometer,
- a NaI spectrometer,
- a Li-glass detector.

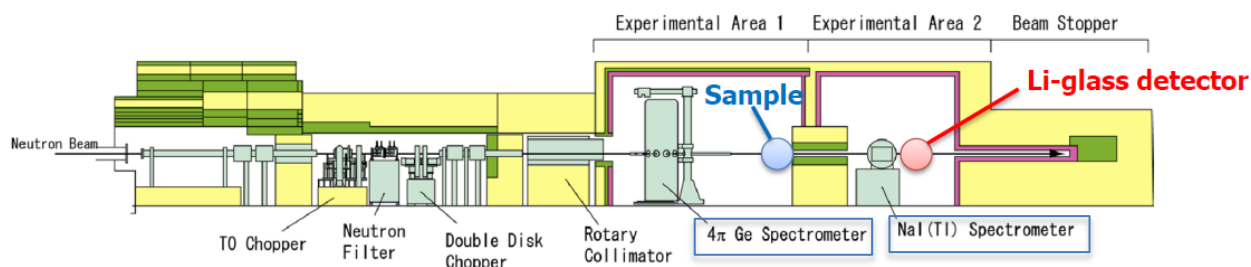


Figure 1. ANNRI experimental device

(The sample position shown corresponds to transmission measurements using Li-Glass detector)

2.2. Capture cross-section measurements

A NaI detector (flight-path length of about 28 m) was used for capture measurements (see Figure 2).



Figure 2. NaI detector in ANNRI (left) and 0.1 mm Mo sample in the FEP film (right)

Three samples of various thicknesses were used during the measurements: 0.1 mm, 0.5 mm and 2 mm made by the stacking of four 0.5 mm samples. Caution was taken to ensure that there is no air gap between the 4 samples and reproducibility experiment was performed. The samples weight was measured using a micro analytical balance AND BM 22 type, which precision is 0.001 mg. Table I gives the characteristics of the Mo samples used for capture measurements.

Table I. Characteristics of the Mo samples for capture measurements

Thickness (mm)	Dimensions (mm x mm)	Weight (mg)
0.1	4.9 x 4.4 ^a	21.025
0.5	5 x 5 ^c	128.023
2 ^b	5 x 5 ^c	541.747

- a.* Dimensions were measured using a micrometer, which precision is 0.05 mm
b. The sample was made by stacking four 0.5 mm thick samples.
c. Cutting specifications

Additional reference measurements were also performed on lead, gold and boron in order to determine the normalization factors for the capture cross section. The lead sample is a Pb-208 sample, which is 5 mm in diameter and 159.7 mg in weight. The gold reference sample is a 6 mm diameter Au-197 sample with a thickness of 10 μm .

Blank measurements, meaning only with the FEP film used to cover the samples, were also performed to determine the background of capture measurements.

2.3. Transmission measurements

Transmission measurements were performed on a thin metallic sample (0.5 mm) and a thick one (5 mm) using a Li-glass detector (flight-path length of 28.71 m). Table II gives the characteristics of the Mo samples used for transmission measurements. Figure 1 shows the sample and Li-glass detectors position. Figure 3 shows the Li-glass detectors and the 5 mm thick Mo sample installed on the experimental device used for the transmission measurements.

Table II. Characteristics of the Mo samples for transmission measurements

Thickness (mm)	Bin width (μs)	Surface density (g/cm^2)	Sample thickness (atoms/barn)
0.5	1	0.538	3.37664E-03
5		5.091	3.19526E-02
0.5	0.1	0.538	3.37664E-03
5		5.091	3.19526E-02



Figure 3. Li-glass detectors (left) and 5 mm thick Mo sample (right) installed on the experimental device used for the transmission measurements

Additional measurements were performed using Co, Mn, In, Ag resonance filters. Blank measurement (without sample) was also performed to check the Background.

3. MEASUREMENTS ANALYSIS AND EVALUATION

Natural molybdenum on which the transmission measurements were performed is comprised of seven major isotopes, amongst which two are particularly important for criticality safety, namely, ^{95}Mo and ^{96}Mo since they have strong resonances in the low energy range (see Figure 4).

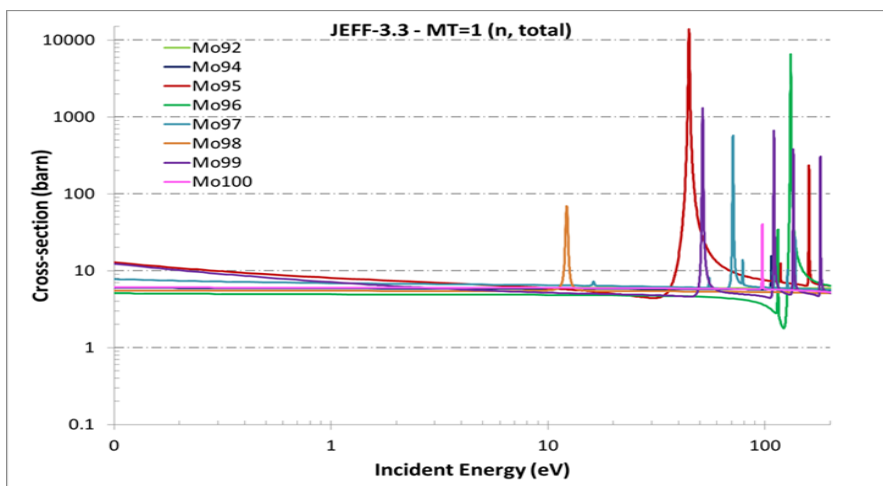


Figure 4. Mo isotopes total cross sections.

The abundances of molybdenum stable isotopes and associated thermal cross sections and resonance integral are reported in Table III. The target spin is 0^+ for ^{92}Mo , ^{94}Mo , ^{96}Mo , ^{98}Mo , ^{100}Mo and $5/2^+$ for

⁹⁵Mo, ⁹⁷Mo. An inspection of the evaluations included in the ENDF (JEFF-3.3 evaluation and others) show that resonance parameters for ⁹⁵Mo are available from 10⁻⁵ eV to 2145 eV with 23 resonances corresponding to $l = 0$ (*s-wave*) and 34 resonances to $l = 1$ (*p-wave*). However, in the JEFF-3.3 evaluation [2], as well as ENDF, the correspondence between the spin groups and the channel spin is not correctly addressed for these odd isotopes. Work has been done to correct this issue and assign physically accepted spin sequences.

Table III. Abundances of molybdenum isotopes and associated thermal cross sections and resonance integrals.

Isotope	Abundance (atom %)	Thermal Cross Section (barns)*	Resonance Integral (barns)*
⁹² Mo	14.84	0.08 ± 0.02	0.83
⁹⁴ Mo	9.25	0.34 ± 0.02	1.12
⁹⁵ Mo	15.92	13.4 ± 0.3	118 ± 7
⁹⁶ Mo	16.68	0.5 ± 0.3	17 ± 3
⁹⁷ Mo	9.55	2.2 ± 0.2	14.4 ± 3.0
⁹⁸ Mo	24.13	0.130 ± 0.006	6.7 ± 0.3
¹⁰⁰ Mo	9.63	0.199 ± 0.002	3.76 ± 0.15

* Mughabghab

3.1. Channel spin for the compound nucleus n+target for ⁹⁵Mo and ⁹⁷Mo

The channel spin s in the entrance channel is determined by a combination of the neutron spin ($1/2$) and the target spin I . Hence, according to the quantum spin addition rule the entrance channel spin can be $s=1/2 - I$ and $s=1/2 + I$. It is easy to see that for target spin I greater than 0 there will always be two possible channel spin. The resonance total angular momentum j is obtained by combination of the spin channel s and orbital angular momentum l . The isotopes of molybdenum ⁹⁵Mo and ⁹⁷Mo have spin of $5/2$. Existing evaluations for these isotopes (ENDF, JEFF, JENDL, etc) do not consider separation of the two spin channels for $l > 0$. Consequently, interference effects in the scattering cross section (total cross section) are not properly accounted for. The evaluation presented in this work takes into account the spin channel issue in the fitting procedure by the SAMMY code [3]. It is of great importance since it can reflect the interferences in the entrance channels.

The quantum numbers j , l , and s are provided in Table IV for ⁹⁵Mo and ⁹⁷Mo. The neutron spin and parity is $1/2+$.

Table IV. Channel spins for molybdenum isotopes.

Isotope ($I=5/2+$)	l	$s (I/2+I)$	j^π
⁹⁵ Mo, ⁹⁷ Mo	0	2	2+
		3	3+
	1	2	1-
		2	2-
		3	
		2	3-
		3	
		3	
		3	4-

3.2. SAMMY fit of the experimental data

In the sequential fit of the experimental data at each step of the evaluation process an updated resonance parameter and covariance is generated, which are, subsequently feedback into the analysis of the next experimental data in the sequence. This process is repeated several times until a good fit of the experimental data is achieved. An important figure-of-merit that indicates a good fitting of the experimental data is the χ^2 provided at the end of each SAMMY run. The resulting set of resonance parameters is expected to reproduce well the experimental data within the quoted experimental uncertainty.

Using the transmission measurements performed at the J-PARC facility for natural molybdenum, IRSN began a SAMMY fit of the 0.5-mm thickness sample in the energy range 0-600 eV. The covariance matrix of cross sections was produced as an output. The resonance parameter and covariance were used for fitting the experimental data corresponding to the 5-mm thickness sample. The results of the SAMMY fitting of the experimental data are shown in Figure 5. The data resolution above 350 eV is not good enough to obtain a good fit of the experimental data.

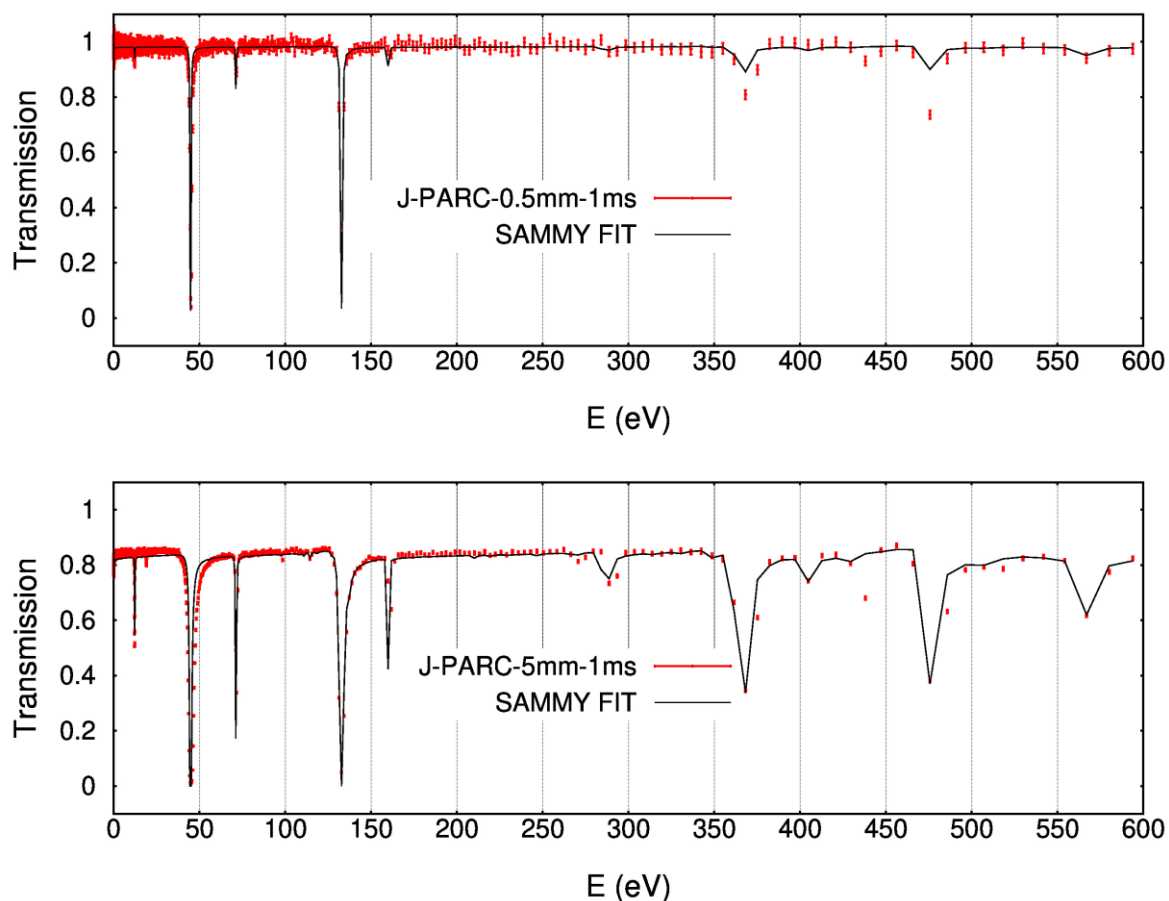


Figure 5. Fit of the J-PARC measurement values for 0.5-mm (top) thick sample and reconstruction of resonance for 5-mm (bottom) thick sample.

3.3. Comparison of thermal cross section values

Comparisons of the thermal capture and scattering cross sections (0.0253 eV) between SAMMY results, calculated with resonance parameters derived in this work, based on the J-PARC measurements and reference values from Mughabghab [4] were done for ^{95}Mo , ^{96}Mo and ^{97}Mo isotopes. They are reported in Table V. Good agreement is observed between the calculated value and the reference one, giving quite good confidence in the produced fit.

Table V. Comparison of thermal cross sections at 0.0253 eV.

Isotope	SAMMY		Mughabghab	
	σ_γ (barns)	σ_s (barns)	σ_γ (barns)	σ_s (barns)
^{95}Mo	13.635	6.2112	13.4 ± 0.3	6.49 ± 0.37
^{96}Mo	0.439	5.06575	0.5 ± 0.2	4.85 ± 0.09
^{97}Mo	2.55741	6.64238	2.2 ± 0.2	6.61 ± 0.14

4. FUTURE WORK AND PERSPECTIVES

Capture cross sections measurements were also performed at the J-PARC facility and the data reduction process is in progress. A SAMMY fitting will be performed on these measurements in order to provide more accurate capture data for molybdenum isotopes.

Besides, additional measurements on ^{95}Mo , ^{96}Mo , and ^{97}Mo enriched samples are already envisioned for the next years.

The SAMMY fitting procedure has currently been applied to the J-PARC measured data in the 0-600 eV energy range. Future works will consist of extending the evaluation to higher energies covering at least the range of resonance parameters up to 2145 eV. It is expected that this range could be extended beyond this energy to account for resonances identified in the measured data.

The production of covariance data for transmission and capture cross sections for molybdenum is essential for criticality safety applications. Indeed, this data is used in the GLLSM methodology to establish the prior uncertainty due to nuclear data in criticality safety applications, which is the result of the propagation in terms of Δk_{eff} of cross sections uncertainties from the covariance matrix. Molybdenum isotopes covariances will be established using these new J-PARC measurements, which will surely help criticality practitioners with establishment of the bias applied on their industrial configurations using tools such as MACSENS [5].

Finally, the impact of the new molybdenum isotopes evaluation on benchmark calculation will be studied, on a basis of about 30 critical experiments issued from the ICSBEP handbook [6] and French proprietary programs [7] involving natural molybdenum in various neutron energy spectra. Moreover, new integral experiments involving molybdenum in thermal spectra are being designed to be performed at SPRF/CX at the SANDIA National Laboratories [8]. These future experiments will also be an opportunity to validate the new molybdenum nuclear data produced in the frame of this collaborative work between JAEA and IRSN.

5. CONCLUSIONS

New differential experiments were performed at J-PARC facility on natural molybdenum samples of various thicknesses in the frame of a JAEA-IRSN collaboration. The transmission measurements were analyzed and the SAMMY fitting procedure was already applied on the 0-600 eV energy range. On-going work is focused on the capture experimental data analysis and on the extension of the evaluation to higher energies covering at least the range of resonance parameters up to 2145 eV. Resonance parameter covariances for the evaluated isotopes will also be derived. The impact of the new molybdenum isotopes evaluation on benchmark calculation will be studied using integral experiments involving natural molybdenum in various neutron energy spectra.

This collaborative work between IRSN and JAEA will help to address issues concerned to criticality safety and reactor physics applications thru more accurate data and uncertainties.

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