

Results of Time-of-Flight Transmission Measurements for ^{nat}Mo at a 50 m Station of GELINA

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Abstract. Transmission measurements on natural molybdenum samples have been performed at the time-of-flight facility GELINA to validate neutron resonance parameters for Mo isotopes. The measurements were carried out at the 50 m transmission station using a Li-glass scintillator with the accelerator operating at 400 Hz. This report provides the experimental details required to deliver the data to the EXFOR data library which is maintained by the International Network of Nuclear Reaction Data Centres (NRDC). The experimental conditions and data reduction procedures are described. In addition, the full covariance information based on the AGS concept is given such that nuclear reaction model parameters together with their covariances can be derived in a least-squares adjustment to the data.

1 Introduction

To study the resonance structure of neutron induced reaction cross sections, neutron spectroscopic measurements are required, which determine with a high accuracy the energy of the neutron that interacts with the material under investigation. To cover a broad energy-range such measurements are best carried out with a pulsed white neutron source, which is optimized for time-of-flight (TOF) measurements [1].

The TOF-facility GELINA [2][3] has been designed and built for high-resolution cross section measurements in the resonance region. It is a multi-user facility, providing a white neutron source with a neutron energy range from 10 meV to 20 MeV. The GELINA facility can host up to 10 experiments at measurement stations located between 10 m and 400 m from the neutron production target. The electron linear accelerator provides a pulsed electron beam with a maximum energy of 150 MeV, a maximum peak current of 10 A and a repetition rate ranging from 50 Hz to 800 Hz. A compression magnet reduces the width of the electron pulses to about 2 ns [4]. The electron beam hits a mercury-

cooled uranium target producing Bremsstrahlung and subsequently neutrons via photonuclear reactions [5]. Two water-filled beryllium containers mounted above and below the neutron production target are used to moderate the neutrons. By applying different neutron beam collimation conditions, experiments can use either a fast or a moderated neutron spectrum. The neutron production rate is monitored by BF_3 proportional counters which are mounted in the ceiling of the target hall. The output of the monitors is used to normalize the time-of-flight spectra to the same neutron intensity. The measurement stations are equipped with air conditioning systems that maintain a constant temperature to reduce electronic drifts in the detection chains due to temperature changes.

This report describes the transmission measurements carried out at GELINA with ^{nat}Mo metallic samples. To reduce bias effects due to e.g. dead time and background, the measurement and data reduction procedures recommended in Ref. [1] have been followed. The report provides the information required for extracting resonance parameters by using the resonance shape analysis code REFIT [6]. In the description of the data the recommendations resulting from a consultant's meeting organized by the Nuclear Data Section of the IAEA (NDS/IAEA) have been followed [7].

2 Experimental conditions

The transmission experiments were performed at the 50 m measurement station of flight path 4 with the accelerator operating at 400 Hz. The moderated neutron spectrum was used. A shadow bar made of Cu and Pb was placed close to the uranium target to reduce the intensity of the γ -ray flash and the fast neutron component. The flight path forms an angle of 9° with the direction normal to the face of the moderator viewing the flight path. The samples and detector were placed in an acclimatized room to keep them at a constant temperature of 20 °C.

The partially thermalized neutrons scattered from the moderators were collimated into the flight path through an evacuated aluminum pipe of 50 cm diameter with annular collimators, consisting of borated wax, copper and lead. A set of Pb, Ni and Cu annular collimators was used to reduce the neutron beam to a diameter of 45 mm at the sample position. Additional lithium and B₄C collimators were installed to absorb neutrons that are scattered by the collimators. A ¹⁰B overlap filter with an areal density of about 0.08 at/b was placed close to the neutron target to minimize the contribution of slow neutrons coming from previous accelerator bursts. The impact of the γ -ray flash in the neutron detector was reduced by a 16 mm thick Pb filter.

The samples were placed in an automatic sample changer at a distance of approximately 24 m from the neutron source. Close to the sample position Na, Co and W black resonance filters were mounted in independent and automatic filter changers to determine the background contribution at 2850 eV, 132 eV and 18 eV, respectively, and to obtain its time dependence. The Co filter was permanently in the beam to continuously monitor the background level and to account for the impact of the sample or other filters placed in the beam [1]. The neutron beam passing through the sample and filters was further collimated and detected by a 6.35 mm x 151.6 mm diameter NE912 Li-glass scintillator. The scintillator was connected through a boron-free quartz window to a 127 mm EMI 9823 KQB photomultiplier (PMT), which was placed outside the neutron beam perpendicular to its axis. The detector was placed at about 47 m from the neutron target and the diameter of the neutron beam at the detector position was about 90 mm.

The TOF of the detected neutron was derived from the time difference between the stop signal T_s , obtained from the anode pulse of the PMT, and the start signal T_0 , given at each electron burst. This time difference was processed with a multi-hit fast time coder with a 1 ns time resolution. The TOF and the pulse height of each detected event were recorded in list mode using a multi-parameter data acquisition system developed at the JRC Geel [8]. Each measurement was subdivided in different cycles. Only cycles for which

the ratio between the total counts in the transmission detector and in the neutron monitor deviated by less than 1 % were selected. The dead time of the detection chain $t_d = 3305$ (10) ns was derived from a spectrum of the time-interval between successive events. The uncertainties due to dead time corrections in the region of interest are very small and can be neglected.

The measurements were performed with natural Mo metallic samples of 2 mm and 5.4 mm thicknesses. The first sample consists of a disk of 2 mm thickness and a diameter of 80 mm with reference number NP2013-09-01. The second sample was obtained by stacking six square foils of about 65 mm length and between 0.53 and 0.65 mm thickness to the first sample. The main characteristics of the disk and the foils are reported in Table 1. The areal density of the samples was derived from a measurement of the weight and the area with an uncertainty better than 0.1 %. The mass was determined by substitution weighing with a microbalance from Mettler Toledo. The area was determined by an optical surface inspection with a microscope system from Mitutoyo [9]. The areal density for the stack of all the samples amounts up to 3.5348 (35) x 10^{-2} at/b. It is worth to mention the presence of tungsten contamination in the squared foils which amount for 3.5×10^{-4} of the total of atoms in the thick sample.

Table 1 Characteristics of the samples used for the transmission measurements. Each areal density $n_{\rm d}$ was calculated by using the experimentally determined mass and area.

ID	Thick/mm	Mass/g	Area/mm2	Areal Density (at/b)
09-01	2	114.320 (10)	5077 (5)	1.4136 (15) x 10 ⁻²
02-01	0.65	26.959 (10)	4208 (4)	0.4020 (4) x 10 ⁻²
02-02	0.53	21.292 (10)	4106 (4)	0.3254 (4) x 10 ⁻²
02-03	0.65	26.262 (10)	4118 (4)	0.4003 (4) x 10 ⁻²
02-04	0.53	20.989 (10)	4055 (4)	0.3248 (4) x 10 ⁻²
02-05	0.53	21.762 (10)	4102 (4)	0.3329 (4) x 10 ⁻²
02-06	0.53	19.738 (10)	3686 (4)	0.3360 (4) x 10 ⁻²

3 Data reduction

The AGS code [10][11], developed at the JRC Geel, was used to derive the experimental transmission from the TOF-spectra. The code is based on a compact formalism to propagate all uncertainties starting from uncorrelated uncertainties due to counting statistics.

3.1 Experimental transmission

The experimental transmission T_{exp} as a function of TOF was obtained from the ratio of a sample-in measurement C_{in} and a sample-out measurement C_{out} , both corrected for their background contributions B_{in} and B_{out} , respectively:

$$T_{exp} = N \frac{C_{in} - KB_{in}}{C_{out} - KB_{out}}.$$
(3.1)

The TOF spectra, C_{in} and C_{out} , were corrected for losses due to the dead time in the detector and electronics chain. All spectra were normalized to the same TOF-bin width structure and neutron beam intensity. The latter was derived from the response of the BF₃ beam monitors. To avoid systematic effects due to slow variations of both the beam intensity and detector efficiency as a function of time, data were taken by alternating sample-in and sample-out measurements in cycles of about 600 seconds. Such a procedure reduces the uncertainty on the normalization to the beam intensity to less than 0.25 %. This uncertainty was evaluated from the ratios of counts in the ⁶Li transmission detector and in the flux monitors. To account for this uncertainty the factor N= 1.0000 (25) was introduced in Eq. (3.1). The background as a function of TOF was

approximated by an analytic expression applying the black resonance technique [1]. The factor K = 1.00 (3) in Eq. (3.1) was introduced to account for systematic effects due to the background model. Its uncertainty was derived from a statistical analysis of the difference between the observed black resonance dips and the estimated background [12].

The time-of-flight (t) of a neutron creating a signal in the neutron detector was determined by the time difference between the start signal (T_0) and the stop signal (T_s):

$$t = (T_s - T_0) + t_0, (3.2)$$

with t_0 a time-offset which was determined by a measurement of the γ -ray flash. The flight path distance L = 47.670 (8) m, i.e. the distance between the centre of the moderator viewing the flight path and the front face of the detector, was derived previously from results of transmission measurements using uranium standard references [13].

3.2 Background correction

The background as a function of TOF for the measurement with the ¹⁰B overlap filter was parameterized by an analytical function:

$$B(t) = b_0 + b_1 e^{-\lambda_1 t} + b_2 e^{-\lambda_2 t} + b_3 e^{-\lambda_3 (t+\tau_0)}.$$
(3.3)

It consists of a time independent and three time-dependent exponential terms. The time independent component b_0 is related to the ambient radiation and background contributions that lost any time correlation. The first time-dependent component is due to 2.2 MeV γ -rays resulting from neutron capture in hydrogen present in the moderator, the second exponential term is due to neutrons scattered inside the detector station and neutrons scattered at other flight paths, and the last one originates from slow neutrons coming from previous accelerator pulses. The decay constants λ_1 and λ_2 were derived from transmission data measured with additional black resonance filters, while λ_3 was obtained by extrapolating the TOF spectra behavior at large times, i.e. for $t > 1 \times 10^6$ ns. The parameter τ_0 is related to the operating frequency of accelerator ($\tau_0 = 2.5$ ms for 400 Hz). This extrapolation applied to each measurement also provides the amplitudes b_0 and b_3 . The time dependence of the first and the second time-dependent background components was studied by including short cycles with Na, Co and W filters in the beam. The b_1/b_2 ratio obtained in these short cycles was used for adjusting the amplitudes b_1 and b₂ together with the information of the black resonance dip of the permanent Co filter. Example of a dead-time corrected and normalized sample-in spectrum with the background contributions from Eq. (3.3) are shown in Figure 1.

ID	b ₀ /10 ⁻⁹	b1/10 ⁻⁷	λ1/10 ⁻⁵ ns ⁻¹	b ₂ /10 ⁻⁸	λ2/10 ⁻⁶ ns ⁻¹	b ₃ /10 ⁻⁷	λ ₃ /10 ⁻⁶ ns ⁻¹
Bin	7.10	4.55	2.40	4.16	1.35	4.78	2.30
B_{out}	7.47	5.08	2.40	4.67	1.35	5.28	2.30

Table 2. Parameters for the analytical expressions of the background correction for the sample-in and sample-out measurements for the natural molybdenum sample of 2 mm thick.

Table 3. Parameters for the analytical expressions of the background correction for the sample-in and sample-out measurements for the natural molybdenum sample of 5 mm thick.

ID	b ₀ /10 ⁻⁹	b ₁ /10 ⁻⁷	λ ₁ /10 ⁻⁵ ns ⁻¹	b ₂ /10 ⁻⁸	λ ₂ /10 ⁻⁶ ns ⁻¹	b ₁ /10 ⁻⁷	λ ₃ /10 ⁻⁶ ns ⁻¹
Bin	7.59	4.54	2.40	3.93	1.35	4.51	2.30
Bout	8.53	5.62	2.40	5.13	1.35	5.17	2.30

Figure 1. TOF spectrum with the 2 mm thick ^{nat}Mo sample and the Co filter in the beam at 400 Hz together with the total background (B_{in}) and its different components as given in Eq. (3.3).



4 Results

The AGS code [10][11] was used to derive the experimental transmission and propagate both the correlated and uncorrelated uncertainties. The code is based on a compact formalism to propagate all uncertainties starting from uncorrelated uncertainties due to counting statistics. It stores the full covariance information after each operation in a concise, vectorized way. The AGS formalism results in a substantial reduction of data storage volume and provides a convenient structure to verify the various sources of uncertainties through each step of the data reduction process. The concept is recommended by the NDS/IAEA [7] to prepare the experimental observables, including their full covariance information, for storage into the EXFOR data library [14][15].

The experimental transmission resulting from the measurement with the 2 mm thick ^{nat}Mo sample is shown in Figure 2. The experimental transmission is compared with the transmission calculated with the resonance parameters in the JEFF-3.3 In order to obtain the theoretical transmission, the following expression is implemented in the REFIT code:

$$T_{M}(t) = \int R(t, E) e^{-n\sigma_{tot}(E)} dE.$$
(4.1)

The response function of the TOF spectrometer, R(t,E), represents the probability that a neutron with energy E is detected with a time-of-flight t. The response function can be considered as the convolution of the duration of the accelerator burst, the time resolution

of the detection system and the neutron transport in the neutron target and in the detector [1][16]. The residuals in the figure indicate that the evaluated resonance parameters can be improved.





The format in which the numerical data will be stored in the EXFOR data library is illustrated in the Appendix. The data include the full covariance information based on the AGS concept. The total uncertainty and the uncertainty due to uncorrelated components are reported, together with the contributions due to the normalization and background subtraction. Applying the AGS concept the covariance matrix V of the experimental transmission can be calculated by:

$$V = U_u + S(\eta)S(\eta)^T, \qquad (4.2)$$

where U_u is a diagonal matrix containing the contribution of all uncorrelated uncertainty components. The matrix S contains the contribution of the components $\eta = \{N, K\}$ creating correlated components. The uncertainty due to the dead time correction can be neglected. The experimental details, which are required to perform a resonance analysis on the data, are summarized in the Appendix.

Acknowledgements

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Appendix

A. SUMMARY OF EXPERIMENTAL DETAILS

A. 1 Experiment description (ID 1)

1.	Main Reference		[a]
2.	Facility	GELINA	[b]
3.	Neutron production		
	Neutron production beam	Electron	
	Nominal average beam energy	100 MeV	
	Nominal average current	50 μ A	
	Repetition rate (pulses per second)	400 Hz	
	Pulse width	2 ns	
	Primary neutron production target	Mercury cooled depleted uranium	
	Target nominal neutron production intensity	3.4 x10 ¹³ s ⁻¹	
4.	Moderator		
	Primary neutron source position in moderator	Above and below uranium target	
	Moderator material	2 water filled Be-containers around U- target	
	Moderator dimensions (internal)	2 x (14.6 cm x 21 cm x 3.9 cm)	
	Density (moderator material)	1 g/cm ³	
	Temperature (K)	Room temperature	
	Moderator-room decoupler (Cd, B,)	None	
5.	Other experimental details		
	Measurement type	Transmission	
	Method (total energy, total absorption,)	Good transmission geometry	[c][d]
	Flight Path length (m) (moderator centre-detector front face) Flight path direction	L = 47.670 (8) m 9° with respect to normal of the moderator face viewing the flight path	
	Neutron beam dimensions at sample position Neutron beam profile	45 mm in diameter	
	Overlap suppression	¹⁰ B overlap filter	
	Other fixed beam filters	Co. Pb	
6.	Detector		-
	Туре	Scintillator	
	Material	Li-glass	
	Surface Dimensions	152.4 mm diameter	
	Thickness (cm)	6.35 mm	
	Detector(s) position relative to neutron	In the beam	
	beam		
	Detector(s) solid angle	-	
_			
7.	lype (metal, powder, liquid, crystal)		
		$1.4126(15) \times 10^{-2}$ - 5/5	
		1.4130 (15) X 10 - at/D	
	Sample mass (g)	114.320 (10) g	
	Geometrical snape (cylinder, sphere,)	Cymder	

	Surface dimension	5077 (5) mm ²	
	Nominal thickness (mm)	2 mm	
	Containment description	None	
Da	ta Reduction Procedure		[d][e]
8.	Dead time correction	Done (< factor 1.05)	
	Back ground subtraction	Black resonance technique	
	Flux determination (reference reaction,)	-	
	Normalization	1.0000 (25)	
	Detector efficiency	-	
	Self-shielding	-	
	Time-of-flight binning	Zone length bin width 16384 1 ns 10240 2 ns 8192 8 ns 6144 16 ns 6144 32 ns 4096 64 ns 4096 128 ns 1024 512 ns 1024 1024 ns	
•	Response function		
9.		Normal distribution, FWHM = 2 ns	
	larget / moderator assembly	Numerical distribution from MC	[f][g]
	Detector	Analytical function defined in REFIT manual	[h]

A. 2 Experiment description (ID 2)

1.	Main Reference		[a]
2.	Facility	GELINA	[b]
3.	Neutron production		
	Neutron production beam	Electron	
	Nominal average beam energy	100 MeV	
	Nominal average current	50 μ A	
	Repetition rate (pulses per second)	400 Hz	
	Pulse width	2 ns	
	Primary neutron production target	Mercury cooled depleted uranium	
	Target nominal neutron production intensity	3.4 x10 ¹³ s ⁻¹	
4.	Moderator		
	Primary neutron source position in moderator	Above and below uranium target	
	Moderator material	2 water filled Be-containers around U- target	
	Moderator dimensions (internal)	2 x (14.6 cm x 21 cm x 3.9 cm)	
	Density (moderator material)	1 g/cm ³	
	Temperature (K)	Room temperature	
	Moderator-room decoupler (Cd, B,)	None	
5.	Other experimental details		
			1

6 Detector	
6. Detector	
Notorial Li class	
Material Li-glass	
Surface Dimensions 152.4 mm diameter	
Enickness (cm) 6.35 mm	
beam	
Detector(s) solid angle -	
Sample	
7. Type (metal, powder, liquid, crystal) Metal	
Chemical composition ^{nat} Mo (100 at %)	
Sample composition (at/b) $3.5348 (35) \times 10^{-2} at/b$	
Temperature 20 °C	
Sample mass (g) 251.322 (25) g	
Geometrical shape (cylinder, sphere,) Cylinder and square foils	
Surface dimension	
Nominal thickness (mm) 5.4 mm	
Containment description None	
Data Reduction Procedure [d][e]
8. Dead time correction Done (< factor 1.05)	
Back ground subtraction Black resonance technique	
Flux determination (reference reaction,)	
Normalization 1.0000 (25)	
Detector efficiency -	
Self-shielding -	
Time-of-flight binning Zone length bin width 16384 1 ns 10240 2 ns 8192 8 ns 6144 16 ns 6144 32 ns 4096 64 ns 4096 128 ns 1024 512 ns 1024 1024 nc	
1024 1024 115	
Response function Named distribution Entities	
Response function Normal distribution, FWHM = 2 ns 9. Initial pulse Normal distribution, FWHM = 2 ns	
Response function Normal distribution, FWHM = 2 ns 9. Initial pulse Normal distribution from MC Target / moderator assembly Simulations	

B. Data format

Column	Content	Unit	Comment				
1	Energy	eV	Relativistic relation using a fixed flight path length				
			(L = 47.670 m)				
2	tı	ns	Low bound				
3	th	ns	High bound				
4	T _{exp}		Transmission				
5	Total Uncertainty						
6	Uncorrelated		Uncorrelated uncertainty due to counting				
	uncertainty		statistics				
7	AGS-vector (K)		Background model uncertainty (uk/K=3 %)				
8	AGS-vector (N)		Normalization $(u_N/N = 0.25 \%)$				

Comments from the authors:

- The AGS concept was used to derive the experimental transmission

$$T_{exp} = N \frac{C_{in} - KB_{in}}{C_{out} - KB_{out}},$$

and to propagate the uncertainties, both the uncorrelated due to counting statistics and the uncertainty due to the normalization and the background contributions.

- The quoted uncertainties are standard uncertainties at 1 standard deviation

B.1 DATA (ID 1)

E/ a)/	t / nc	t/nc	т	21		AGS	
L/ EV	u// 115	th / IIS	l exp	u_t	u_u	К	Ν
300128.776	6292	6293	0.906756	0.008662	0.008360	-0.000011	0.002267
300033.361	6293	6294	0.900847	0.008455	0.008161	-0.000012	0.002252
682.74245	131896	131904	0.595702	0.016976	0.016898	-0.000648	0.000149
682.65964	131904	131912	0.551043	0.016021	0.015945	-0.000714	0.000138
3.00519	1987584	1988608	0.962126	0.141936	0.127987	-0.061313	0.002405
3.00209	1988608	1989632	0.941141	0.126881	0.115073	-0.053400	0.002353

	t. / no	t. / pc	т			AGS	
E/ ev	u/ ns	th / ns	l exp	u_t	u_u	K	Ν
300128.776	6292	6293	0.755008	0.009839	0.009656	-0.000030	0.001888
300033.361	6293	6294	0.755007	0.009648	0.009462	-0.000029	0.001888
682.74245	131896	131904	0.258394	0.013611	0.013541	-0.0012199	0.000646
682.65964	131904	131912	0.251201	0.013195	0.013126	-0.0011967	0.000628
3.00519	1987584	1988608	0.805276	0.171171	0.128389	-0.113189	0.002013
3.00209	1988608	1989632	0.640788	0.173776	0.114051	-0.131103	0.001602
 682.74245 682.65964 3.00519 3.00209	 131896 131904 1987584 1988608	 131904 131912 1988608 1989632	 0.258394 0.251201 0.805276 0.640788	 0.013611 0.013195 0.171171 0.173776	 0.013541 0.013126 0.128389 0.114051	 -0.0012199 -0.0011967 -0.113189 -0.131103	 0.000646 0.000628 0.002013 0.001603

B.2 DATA (ID 2)

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