Total Cross Section Measurements of Highly Enriched Isotopic Mo in the Resolved and Unresolved Energy Regions

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I. INTRODUCTION

High accuracy nuclear data is required in neutron transport calculations and contributes to the growing understanding of nuclear reaction theory. Molybdenum neutron cross section data in particular are important because molybdenum can exist in reactors as a high yield fission product or in alloyed form with applications in heat pipes, condenser tubes, and as an alternative advanced fuel [1,2].

High resolution neutron time-of-flight transmission measurements on highly enriched isotopic metallic samples of molybdenum were performed at the Rensselaer Polytechnic Institute Gaerttner Linear Accelerator (LINAC) Center in the energy range from 100 keV to 620 keV. Resolved resonance parameters can be found up to 2.1 keV for ^{95}Mo, 19.5 keV for ^{96}Mo, 52.6 keV for ^{98}Mo, and 26.1 keV for ^{100}Mo [3]. The high resolution data includes new resolved resonances beyond the current resolved resonance region. The characterization of these resonances can help extend the resolved resonance region in each molybdenum isotope.

At some point in energy, the average natural width of resonances becomes comparable to the experimental resolution width and only partially resolved structure is observed. Eventually, the level spacing between isolated resonances also becomes comparable to the average natural width of these resonances resulting in interferential structure as well. Even with perfect experimental resolution, the overlapping averaged resonances result in a pseudo-continuum. This transitional region is defined as the unresolved resonance region (URR). Non-statistical fluctuations in this region indicate the existence of partially resolved compound nucleus structure or intermediate structure described in terms of doorway states [4,5]. Several methods have been described [4,6-9] to determine the existence of intermediate structure based on fluctuations in the data.

The analysis in this paper is limited to the unresolved energy region treatment. The goal of this work is to extract average resonance parameters from the new transmission measurements in the energy range from 100 keV to 620 keV The new average parameters would improve the existing library database by representing the structure in the unresolved resonance region.

II. EXPERIMENTAL SETUP

The Gaerttner LINAC Center at Rensselaer Polytechnic Institute is home to an L-band (1300 MHz) traveling wave linear accelerator made up of a series of iris-loaded cavities spread over nine circular wave-guide accelerator sections. Neutrons are generated when short bursts of injected energetic electrons (≈50 MeV) are directed at a water-cooled and moderated tantalum target [10].

The high-resolution ^{100}Mo transmission measurements were taken with the newly developed Mid-Energy ^{6}Li-glass Neutron Detector Array (MELINDA) [11]. MELINDA employs four identical square-shaped modules each with a 0.5”-thick ^{6}Li-glass scintillator, two out-of-beam photomultiplier tubes coupled to fast electronics, and a low-mass, light-tight aluminum casing with inner reflective surfaces. The modular design allows operational reliability, functional versatility, relatively easy maintainability and lower overall life-cycle costs than a single all-in-one detector system. A detailed model of MELINDA is shown in Figure 1. The new detector is stationed at the 100-meter experimental flight station where flight tubes are positioned between the detector and the photoneutron target providing an evacuated pathway for the neutrons to travel.

Isotopically-enriched metallic molybdenum samples were prepared by Oak Ridge National Laboratory. The samples were stacked and mounted to a computer-controlled sample changer located at a ~13 m flight distance from the neutron-producing target. The atomic composition and number density of the stacked samples are shown in Table I.

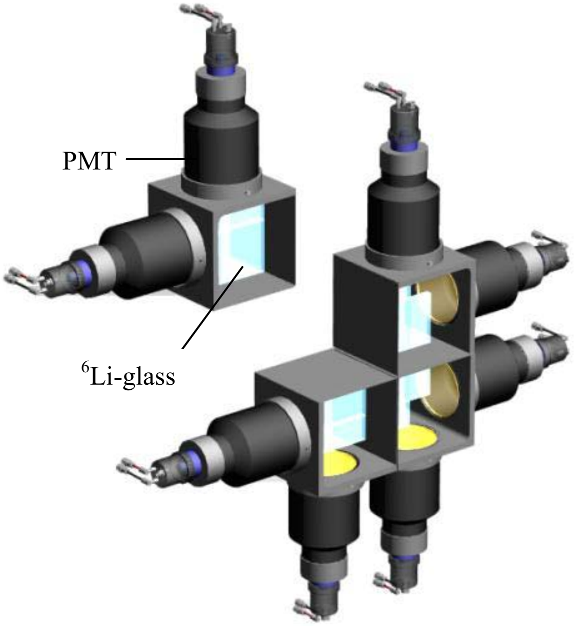

Fig. 1.Detailed 3D Computer Model of MELINDA.

TABLE I. Measured Sample Enrichment and Number Density.

Isotope	Enrichment %	atoms / barn
^{95}Mo	96.5 ± 0.1	0.03998 ± 0.00006
^{96}Mo	96.8± 0.1*	0.05653 ± 0.00006
^{98}Mo	95.83 ± 0.03	0.03847 ± 0.00004
^{100}Mo	97.8 ± 0.15	0.05440 ± 0.00013

*Estimated value

The axial water-moderated photoneutron target [12] was used in the high resolution transmission measurements utilizing a 12 ns electron burst width from the linear accelerator. All measurements were performed with a fixed high-Z filter in the beam (0.5” Pb or 1” ^{238}U) and a ^{10}B-enriched boron disc to minimize overlap neutrons between LINAC pulses. Separate background measurements were performed by cycling different materials with strong black resonances into the beam. The dominant time-dependent γ-ray background component (mainly a result of thermal neutron capture in the water moderator) was determined by placing several thicknesses of polyethylene in the beam and extrapolating the gamma-background to zero-thickness polyethylene. Black notch filters of Na, Al, Mg, S, Li, and Be were used to determine the time-dependent neutron background at specific energies across the measured range.

III. ANALYSIS

Transmission data reduction was performed using the internal processing codes RPIXDR, MONCHK, BACK, FIT and TRANS [13]. RPIXDR produces dead time-corrected, run-summed data files that can be grouped and displayed in counts per second. Statistical checks and correlations between beam monitor data and time-of-flight transmission data were performed with MONCHK. For background corrections, a function was fitted through the black notch filter background points. The experimental transmission and its associated error were determined by TRANS. The total cross section is related to the experimental transmission by,

$$T(E) = \exp[-N\sigma_t(E)] \qquad (1)$$

N = number density of sample (atoms/barn)
T = probability of a neutron of energy E to pass through the sample without interaction, i.e. the transmission.
σ_t= total microscopic neutron cross section

The final cross section data were compared to four reference experimental datasets [7,14-16] on which the latest Evaluated Nuclear Data File (ENDF/B-VII.1) total cross section is based in this energy range [17]. The resolution of the present ^{100}Mo transmission experiment is significantly better than the resolution of the previous four measurements [7,14-16]. Direct comparisons to previous isotopic measurements with broader resolution or elemental measurements with comparable resolution [18] are difficult. The ability to observe resonance structure in the unresolved region depends on both the level density of the isotopes (affected by the purity of the sample), sample thickness, statistical accuracy of the measurement, and the maximum energy resolution of the experiment. A snippet of the present high-resolution data is shown in Figure 2 showing partially-resolved structure as compared to previous data and the latest evaluation.

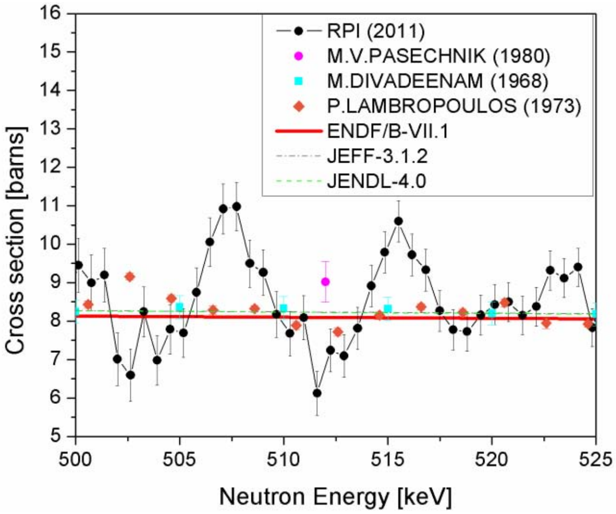

Fig. 2.^{100}Mo data compared to current evaluations.

Generally, the presence of unresolved resonance structure in transmission data can produce an effective measured cross section that is lower than the true theoretical average cross section. This “transmission enhancement” effect results when the energy bins of the measured data are larger than the widths of the underlying structure. This is typically a result of manual transmission averaging, resolution energy broadening or transition into the region where the level spacing between resonances becomes comparable to the average natural widths. This phenomenon is best described by considering the mathematical relationship between the true average cross section and average transmission [19].

$$\langle\sigma_t\rangle = \frac{1}{n}ln\langle e^{-n\sigma_t}\rangle + \frac{1}{n}ln\left(1+\frac{n^2}{2}var\sigma_t - \cdots\right) \qquad (2)$$

The first term is what is often reported as the total cross section [19]. The second term represents a correction for resonance effects containing the cross section variance and higher moments of its distribution quantifying the contribution of underlying resonances to the average cross section [20]. There are several examples of different methods for quantifying and applying a correction for the transmission enhancement based on information obtained from the resolved resonance region [19-22]. Ultimately, the observable partially-resolved structure in the current high-resolution cross-section data, when averaged, can account for the transmission enhancement correction and provides average values that are closer to the theoretical average. This correction was estimated to be on the order of 1% for this dataset and was not applied to the experimental data.

IV. RESULTS & CONCLUSIONS

The measured cross section in the URR can contain non-statistical fluctuations due to partially-resolved overlapping resonance structure or intermediate structure that cannot be treated with the Breit-Wigner formulism. Instead, accurate fits to the total cross section were obtained using the Bayesian Hauser-Feshbach (with width fluctuations) statistical model code FITACS [23] which is currently incorporated into the SAMMY code [24]. This option was chosen based on its comprehensive treatment of the data, immediate availability and ability to produce average resonance parameters with covariance matrices. Average parameters were obtained by fitting the calculated theoretical cross section to the experimental cross section by solving the Bayes’ equation relative to the variable parameters. The fitted quantities obtained from the SAMMY analysis were the neutron strength functions for s-wave and p-wave resonances, the level spacing *D*, and the “effective” or “potential scattering radius” [25], R’. The results were compared to values from the resolved resonance region obtained from the Atlas of Neutron Resonances [3] and were in agreement as shown in Table II (within 2σ of the experimental uncertainty of previous data for S_1 and R’). There is a substantial difference in S_0 compared to the Atlas value. ^{100}Mo cross-section data from Duke University was recently re-evaluated by Mughabghab in the unresolved resonance region which shows a smaller S_0 value of 0.58 [26]. It is worth noting that there is a peak in the p-wave strength function at around A=100 and a minimum in the s-wave strength function (based on the optical model)[3]. Further comparisons to quantitative values in the fast energy region obtained from the optical model should also be explored.

TABLE II. Comparison of ^{100}Mo Fitted Quantities.

	Atlas [3]	SAMMY
R'	6.9 ± 0.2 eV	6.16 ± 0.04
S_0	0.8 ± 0.22	0.31 ± 0.08
S_1	5.14 ± 0.71	6.00 ± 0.07
D_0	617 ± 60	Not Varied
D_1	236 ± 21	212.7

The final fits obtained from SAMMY were evaluated by how well they reproduced the experimental cross section and how they compared to the latest total cross section evaluated libraries. The fit is shown in Figure 3 and its error bars are smaller than the line width. The SAMMY fit has closer agreement to the JEFF and JENDL evaluations and is higher than the ENDF/B-VII.1 evaluation over most of the energy range. The JEFF and JENDL evaluations are nearly identical in the unresolved resonance region. The latest Evaluated Nuclear Data File (ENDF/B-VII.1) for ^{100}Mo is based on a spline fit [17] of four lower resolution experimental data sets [7,14-16].

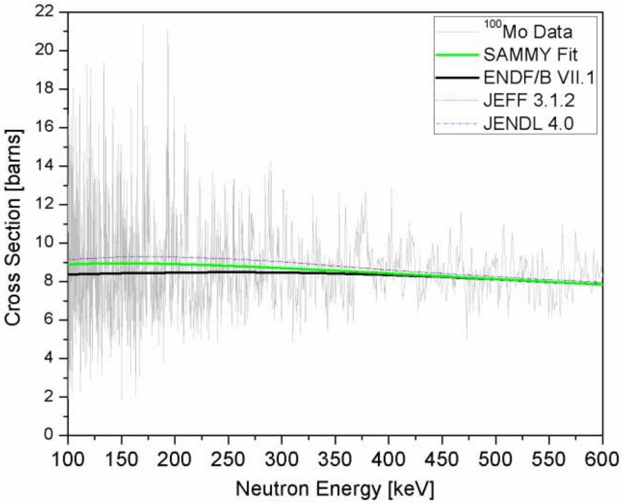

Fig. 3.SAMMY fit compared to data and evaluations. Error bars on the SAMMY fit are within the thickness of the line.

The observed difference between the new fit and ENDF was expected based on the newly resolved structure that provides an average cross section that is closer to the theoretical average value. The fit to the high-resolution data shows that the latest ENDF/B-VII.1 ^{100}Mo evaluation could be underestimating the cross section in the unresolved resonance region (as much as 5% at 150 keV).

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