NEW IMPROVED NUCLEAR DATA FOR NUCLEAR CRITICALITY AND SAFETY*

K. Guber and L. Leal
Oak Ridge National Laboratory
P.O. Box 2008
Oak Ridge, TN 37831, USA
guberkh@ornl.gov and leallc@ornl.gov

C. Lampoudis, S. Kopecky, P. Schillebeeckx, F. Emiliani, R. Wynants and P. Siegler
Nuclear Physics Unit, EC-JRC-IRMM
Retieseweg 111, 2440 Geel, Belgium
Christos.Lampoudis@ec.europa.eu, Stefan.Kopecky@ec.europa.eu, Peter.Schillebeeckx@ec.europa.eu, Federica.Emiliani@ec.europa.eu, Ruud.Wynants@ec.europa.eu and Peter.Siegler@ec.europa.eu

ABSTRACT

The Geel Electron Linear Accelerator (GELINA) was used to measure neutron total and capture cross sections of ^{182,183,184,186}W and ^{63,65}Cu in the energy range from 100 eV to ~200 keV using the time-of-flight method. GELINA is the only high-power white neutron source with excellent timing resolution and ideally suited for these experiments. Concerns about the use of existing cross-section data in nuclear criticality calculations using Monte Carlo codes and benchmarks were a prime motivator for the new cross-section measurements.

Key Words: Neutron Cross Section, Time of Flight, Nuclear Data

1 INTRODUCTION

To support the Nuclear Criticality Safety Program, neutron cross-section measurements were initiated using GELINA at the EC-JRC-IRMM. Concerns about data deficiencies in some existing cross-section evaluations from libraries such as ENDF/B, JEFF, or JENDL for nuclear criticality calculations were the prime motivator for new cross-section measurements. Over the past years many troubles with existing nuclear data have emerged, such as problems related to proper normalization, neutron sensitivity backgrounds, poorly characterized samples, and use of improper pulse-height weighting functions. These deficiencies may occur in the resolved- and unresolved-resonance region and may lead to erroneous nuclear criticality calculations. An example is the use of the evaluated neutron cross-section data for tungsten in nuclear criticality safety calculations, which exhibit discrepancies in benchmark calculations and show the need for reliable covariance data. We measured the neutron total and capture cross sections of ^{182,183,184,186}W and ^{63,65}Cu in the neutron energy range from 100 eV to several hundred keV. This will help to improve the representation of the cross sections since most of the available evaluated data rely only on old measurements. Usually these measurements were done with poor experimental resolution or only over a very limited energy range, which is insufficient for the current application.

Additionally, evaluations in the nuclear data libraries are missing, in most cases, covariance data. These data are increasingly prerequisite for the more sophisticated codes to analyze and simulate nuclear systems. New covariance data for the libraries can be created only by a reevaluation of the cross-section data; however, sometimes this is impossible because the data used

* Notice: This manuscript has been authored by UT-Battelle, LLC, under contract DE-AC05-00OR22725 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

in the analysis and evaluations are no longer available. In such cases, this shortfall can be overcome only by new experimental data included in a new evaluation. In addition, new experiments can give the full experimental covariances.

2 EXPERIMENTS AT GELINA

The experiments were performed at the time-of-flight (TOF) facility GELINA [1]. Neutrons are produced by photo-nuclear and -fission reactions from a rotating uranium target after producing Bremsstrahlung from a pulsed electron beam. The depleted uranium target is mercury cooled with two beryllium canned water containers for neutron moderation. With the help of a special bunching magnet, the electron beam of the GELINA is compressed to a 1 ns pulse, with maximum electron energy of 150 MeV and a repletion rate of up to 800 Hz. The combination of a short pulse, small neutron production target, and long flight paths (up to 400 m) leads to an excellent TOF, and thus neutron energy resolution, which facilitates resolving individual resonances in the neutron cross sections. Since GELINA is a white neutron source—that is, the neutron spectrum ranges from thermal up to several MeV—the cross section in the resolved and unresolved resonance region could be determined in a few experiments.

To determine the resonance parameters, we performed transmission and neutron capture experiments at GELINA with high TOF resolution. The samples used in these experiments were enriched isotopic metallic disks on loan from the Oak Ridge National Laboratory (ORNL).

2.1 Neutron Capture Experiments

The neutron capture experiments were carried out at flight path 14 using the 60 m flight stations. With a set of four deuterated benzene (C_6D_6) detectors [2], gamma rays were recorded after the neutron was captured in the sample, which was placed 58.567 m from the neutron production target in the beam. For each capture event the TOF and gamma-ray energy was recorded; for the latter the pulse-height weighting method was applied [2]. The Monte Carlo code MCNP [3] was used to calculate the appropriate weighting function for each sample. In this computation all structural materials surrounding the detectors and sample were included. With a ^{10}B-loaded ionization chamber, located 80 cm upstream of the sample, the neutron flux was determined. The samples used for the capture experiments were metallic disks isotopically enriched in ^{182,183,184,186}W and ^{63,65}Cu. Since tungsten is very brittle, some of the disks were broken, but the complete set of pieces could be placed in a thin-walled aluminum can to form a complete disk. Details of tungsten disks are compiled in Table I; by combining different disks a sample of the desired thicknesses could be achieved.

Table I. Parameters for the enriched tungsten disks

Isotope	Label	Weight (g)	Diameter (cm)	Thickness (cm)
^{182}W (93.8%)	1	47.6457	6.6	0.129
	2	45.4032	6.6	0.129
	3	47.1538	6.6	0.129
	4	48.3836	6.6	0.129
^{183}W (80.9%)	4	45.645	7.1	0.130
	1+5	93.6633	7.1	0.285
^{184}W (94.5%)	1	45.2854	7.061	0.115
	4	45.6467	6.94	0.113
	5	44.7428	6.786	0.114
	6	44.9424	6.835	0.1145
	7	43.4668	6.82	0.111
^{186}W (96.9%)	2	45.4761	6.952	0.109
	3	45.8759	6.917	0.110
	4	44.7659	6.92	0.109

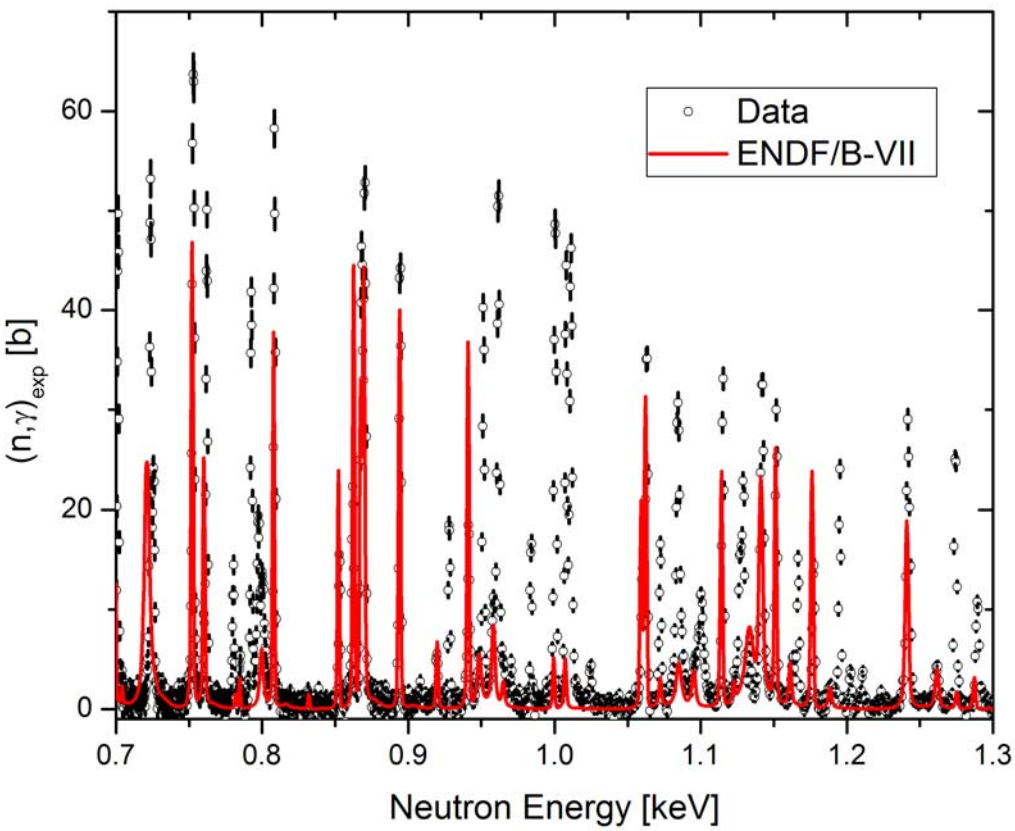

Figure 1. Neutron capture for ^{183}W compared to ENDF/B-VII evaluation. Many resonances for ^{183}W are missing in the evaluation or have erroneous parameters.

The enriched copper disks have a diameter of 8 cm and were used without any canning. From the sample dimension we calculated 0.00862354 at/b for ^{63}Cu and 0.009269207 at/b for ^{65}Cu.

GELINA was operated at 800 Hz repetition rate with 1 ns pulse width. The measurements included additional runs using an empty sample holder, the aluminium canning, and a ^{208}Pb sample in the beam in order to determine the level of background originating from the sample holder and sample scattered neutrons. Using black resonance filters in the neutron beam supplementary background measurements were performed and a ^{10}B slab served as a frame overlap filter.

To determine the normalization factor for the capture yield data obtained for flight path 14, several runs using natural iron samples were performed. The resonance at 1.15 keV in ^{56}Fe was used as a standard for this procedure. In this case the well-known resonance has $\Gamma_n << \Gamma_\gamma$, and if $n\sigma << 1$, the capture is mostly determined by Γ_n. Thus a transmission measurement can provide the normalization. Additionally runs with a combination of natural iron and tungsten disks were performed. Silver samples were used for the 50 Hz runs at flight path 5, and by applying the saturated resonance technique [4] the normalization factor was obtained.

As an example of our neutron capture data, we show in Fig. 1 the experimental neutron capture cross section for ^{183}W, compared to the cross -section calculation using the resonance parameters for the ENDF/B-VII evaluation. This calculation included all experimental effects.

2.2 Neutron Transmission Experiments

To obtain a complete set of resonance parameters, additional neutron transmission experiments were performed using the same samples as for the capture experiments. A set of good neutron widths is not only essential for the description of the cross section of the resolved resonance region,

but is also used to calculate the appropriate correction factors for the capture yield using the analysis program. In addition, some resonances with small radiation widths are not visible in the neutron capture data and vice versa. In this way a complete set of resonance parameters can be obtained for the resolved resonance region.

The measurements were performed simultaneously with the capture experiments using the 50 m fight station of flight path 4. At a distance of 49.33 m, a 0.635-cm-thick ^{6}Li-loaded glass scintillator, viewed by one photomultiplier placed outside the neutron beam, was used to detect the sample-transmitted neutrons. The neutron beam was collimated to 4.5 cm diameter at the sample position, which was mounted in the sample changer located at the 30 m flight station. With the computer-controlled sample changer, the samples were cycled in and out of the beam, as well as the appropriate open beam. The background as a function of TOF was determined by the "black resonance" technique [5]. This method relies on the use of certain samples with strong resonances to scatter all neutrons out of the beam (i.e., the transmission is zero).

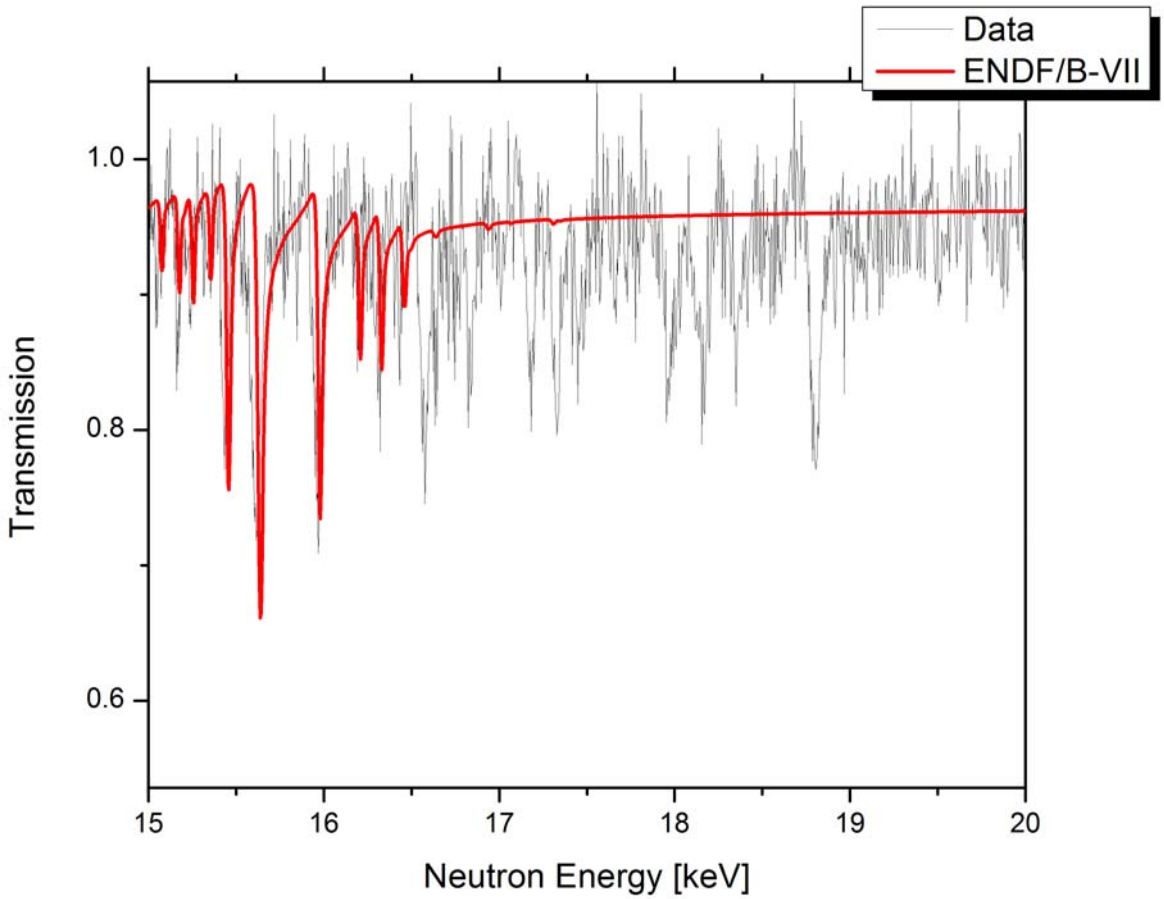

Figure 2. Neutron transmission data for ^{184}W compared to ENDF/B-VII evaluation.

The calculated transmission data from ENDF/B-VII resonance parameters for ^{184}W+n is compared in Fig. 2 to the experimental data. The ENDF/B-VII evaluation has no resonance parameters above 16 keV. But because the resolution of our transmission data is good, many more resonances are visible.

2.3 Data Reduction

For the neutron capture measurements at the 60 m flight station, both capture and flux data were recorded in cycles and stored in list mode simultaneously. During the first stage of the data

reduction, the data were corrected for gain shifts and checked for stability using scalers and counts for different TOF windows of the spectra. Afterwards, the software code AGS (Analysis of Geel Spectra) [6] is used to transform the recorded count rates into observables such as transmission data and reaction yield, including corrections for backgrounds. In addition, AGS performs a full uncertainty propagation starting from counting statistics, which results in a full covariance matrix for the measured observable.

Applying the normalization factor from the iron runs, the tungsten and copper neutron capture yields can be normalized and compared to the latest evaluations. The result is shown in Fig. 1 for the ^{183}W experimental data and the calculated cross section using resonance parameters. This computation was performed using the analysis code SAMMY [7] including experimental effects such as Doppler and resolution broadening, self-shielding, and multiple scattering. As can be seen, the newly measured capture data cannot be described by the evaluation. Many resonances are missing or have incorrect parameters.

For the transmission measurements, the data for valid events were recorded using equal measuring time for sample in and out. The data of selected cycles were summed to histograms after scalers and consistency checks. The obtained histograms for sample in and sample out are normalized to the same neutron monitor counts. The AGS code was used to produce from the histograms transmission data with a full covariance matrix, and background corrections were made using the black resonance filtered runs.

3 RESULTS AND DATA ANALYSIS

The results of our new capture and transmission experiments show differences compared to the evaluated nuclear data for tungsten and copper obtained from the ENDF/B-VII or JENDL 3.3 nuclear data libraries. This is demonstrated exemplarily in Figs. 1 and 2. We were able to resolve resonances with sufficient statistics to much higher neutron energies compared to the evaluations. In general, all of our new and recent neutron capture cross sections exhibit serious discrepancies. Our new capture data show that in many previous cases, capture widths for the tungsten isotopes were over- and underestimated and resonances were missed. Results from our new total cross-section measurements are a valuable addition and will help in the analysis of our new capture cross-section measurements.

There are mainly two reasons for the observed discrepancies: First, previous capture experiments used an improper weighting function; this resulted in mismatched detector response functions. In the new experiments, the computer code MCNP was used for the correct determination of the weighting function. Second, underestimated backgrounds might have led to previous capture cross sections that were too large. In addition, the better characterized metallic samples, superior TOF resolution, and well-understood experimental apparatus and backgrounds helped to produce more reliable cross-section data in the present case.

The high resolution data obtained with the new experiments at GELINA will allow us to extend the resonance analysis for the tungsten and copper isotopes to much higher neutron energies. The resonance parameters obtained are the basis for an evaluation of the cross sections calculated by the Nuclear Data and Criticality Safety Group of the Reactor and Nuclear Systems Division at ORNL. For a sound data evaluation it is necessary to include as many data sets as possible in the analysis, preferably from experiments at different facilities. For this, the EXFOR database is searched for usable data—that is, data with sufficient resolution and information on the experiment and, if possible, data from the ORELA transmission data library. In the case of the copper isotopes, four ORELA transmission data sets were obtained for the analysis. As an example, the ORNL transmission data for ^{63}Cu are shown in Fig. 3; this high-resolution copper transmission data will enable us to extend the resonance analysis far beyond the current cutoff of the ENDF/B-VII parameters.

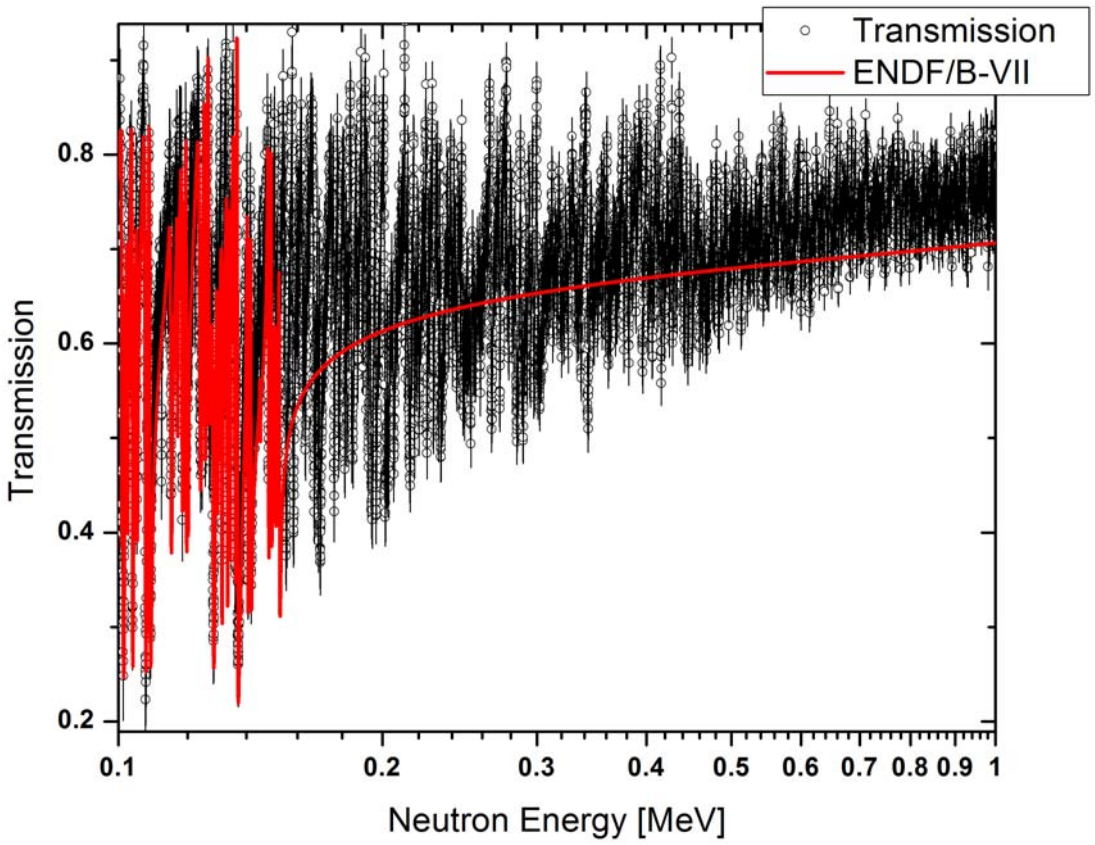

Figure 3. ORELA ^{63}Cu transmission compared to ENDF/B-VII evaluation.

Using the R-matrix code SAMMY, we analyzed the neutron total and capture cross-section data. SAMMY is capable of applying to the data all the necessary corrections for the experimental effects, such as Doppler and resolution broadening, self-shielding, and multiple-scattering effects. The data are fitted sequentially using the fitting results with covariance data from previous fits to different data. Results from the preliminary analysis for ^{184}W are shown in Fig. 4.

The final result of the evaluation will be checked for inconsistencies using criticality benchmark calculations.

4 CONCLUSIONS

To support the Nuclear Criticality Safety Program of the U.S. Department of Energy, we performed at GELINA high-resolution neutron total and capture cross section for the ^{182,183,184,186}W and ^{63,65}Cu isotopes. Since the measurements could be expanded over a broader neutron energy range than the previous evaluation, many more resonances could be resolved. The obtained results are the basis for a new evaluation that will result in a more precise description of the neutron induced cross section.

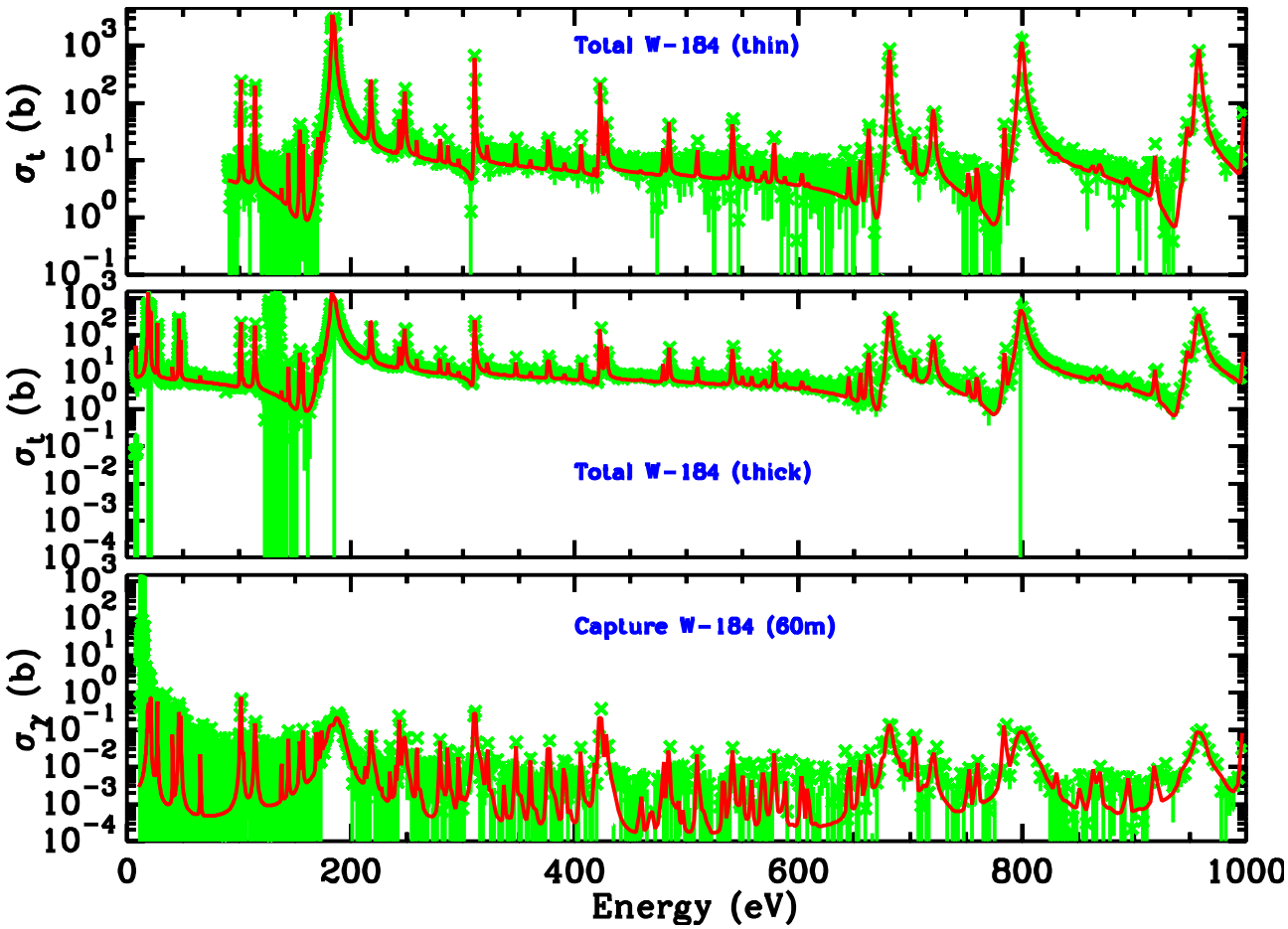

Figure 4. Preliminary SAMMY analysis for the ^{184}W total and capture cross-section data.

5 ACKNOWLEDGMENTS

ORNL is managed by UT-Battelle, LLC, for the U.S. Department of Energy under Contract No. DE-AC05-00OR22725. The U.S. Department of Energy Nuclear Criticality Safety Program sponsored the work presented in this paper.

6 REFERENCES

1. M. Flaska et al., *Nuclear Instruments and Methods A*, **531**, 394 (2004).
2. A. Borella et al., "The use of C_6D_6 detectors for neutron induced capture cross-section measurements in the resonance region," *Nuclear Instruments and Methods in Physics Research A*, **577**, 626–640 (2007).
3. J. F. Briesmeister, Ed., "MCNP A General Monte Carlo N-Particle Transport Code Version 4C," LA-13709-M, Los Alamos National Laboratory (March 2000).
4. R. L. Macklin et al., "Absolute neutron capture yield calibration," *Nuclear Instruments and Methods in Physics Research A*, **164**, 213 (1979).
5. D. B. Syme, "The black and white filter method for background determination in neutron time-of-flight spectrometry," *Nuclear Instruments and Methods in Physics Research*, **198**, 357–364 (1982).
6. C. Bastian et al., "AGS, A Computer Code for Uncertainty Propagation in Time-of-Flight Cross Section Data," *Physor 2006*, Vancouver, Canada (2006).
7. N. M. Larson, *Updated Users Guide for SAMMY: R-matrix Fits to Neutron Data Using Bayes Equations*, ORNL/TM-9179/R8, Oak Ridge National Laboratory, Oak Ridge, Tenn. (2008).