

Nuclear Data Measurements, Analysis and Evaluation at the Oak Ridge National Laboratory in Support of Nuclear Criticality Safety

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This paper describes the activities in nuclear data measurement, nuclear data evaluation, and nuclear data testing at the Oak Ridge National Laboratory in support of the US Department of Energy Nuclear Criticality Safety Program. Data measurement's activities are performed at the Oak Ridge Electron Linear Accelerator (ORELA); data evaluations are done with the computer code SAMMY; and cross section processing is performed by the AMPX and/or NJOY code systems.

KEYWORD: *criticality safety, cross section evaluation, cross section, measurements, benchmark calculations*

I. Introduction

The Defense Nuclear Facility Safety Board (DNFSB) Recommendation 93-2 has motivated the US Department of Energy (DOE) to develop a comprehensive criticality safety program to maintain and to ensure the subcriticality of nuclear systems located throughout the DOE complex. To implement the response to the DNFSB 93-2 recommendation, a Nuclear Criticality Safety Program (NCSP) was created including the following tasks: Critical Experiments, Criticality Benchmarks, Training, Analytical Methods, and Nuclear Data. Elements of the Nuclear Data task consist of a variety of differential measurements performed at the Oak Ridge Electron Linear Accelerator (ORELA) at Oak Ridge National Laboratory (ORNL), data analysis and evaluation using the generalized least-squares fitting code SAMMY in the resolved, unresolved, and high energy ranges, cross section processing using the NJOY and/or AMPX codes, and the development and benchmark testing of a complete evaluation for inclusion into the Evaluated Nuclear Data Files (ENDF/B). The evaluated data are to be used in diverse criticality safety applications including the treatment of nuclear materials outside reactors, as for examples, the transportation and long-term storage of nuclear material. It has been noticed that unlike thermal and fast reactor systems there are criticality safety configurations for which the most important part of the neutron energy spectrum is in the intermediate energy region. Therefore, an accurate treatment of epithermal neutrons is needed in order to estimate the multiplication factors of critical experiments within the experimental uncertainties. In addition, criticality safety engineers recognize that many situations encountered in the DOE complex are characterized by neutron spectra in the intermediate energy region, as opposed to the high-energy region for fast reactors and fusion systems or the low energy region for thermal reactors. The Nuclear Data task of the NCSP has primarily focused on the intermediate energy region, so that upgrades to existing evaluated data will remove deficiencies in the current ENDF/B evaluations. The primary goal of the Nuclear Data Task is to utilize the experimental facility ORELA to obtain high-resolution data in the

intermediate energy region and to apply modern resonance formalisms of the code SAMMY to fit the data. In addition to the fitting of the experimental data, SAMMY also provides sensitivity and covariance information for subsequent use in criticality predictability applications in order to properly assess uncertainties in calculated results. The measured data are compiled and converted into the EXFOR format and submitted for inclusion in the Nuclear Data Bank at the Brookhaven National Laboratory. The evaluated data in the form of pointwise cross section and/or resonance parameters are converted into the ENDF/B library format and used in codes, such as NJOY and/or AMPX to generate continuous energy cross section libraries for calculations using Monte Carlo codes such as MCNP and/or VIM. Multi-group cross section libraries are generated for calculations with Monte Carlo code KENO and/or deterministic codes such as the one-dimensional discrete ordinates code XSDRNPM. After a large number of benchmark tests are performed, the new ENDF library is submitted to the Cross Section Evaluation Working Group (CSEWG) at Brookhaven National Laboratory for further tests, and a new release of the ENDF/B cross section library is made available to the user community.

II. Experimental Facility and Data Evaluation Code

1. ORELA

The ORELA experimental facility was built in the late 60's. ORELA is an electron accelerator which uses the time-of-flight technique for measuring high-resolution nuclear data in the energy range from few eV up to 50 MeV. Various kinds of data measurements for neutrons can be done at ORELA: transmission measurements (total cross sections), capture cross section, fission cross section, elastic cross section, and neutron production cross section. Since its start, ORELA has been used for data measurements in support of several reactor programs. Data measurements for application in thermal reactors design and operation and fusion reactors programs done at ORELA led to the ENDF/B-V library. In the 80's, the Integral Fast

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Reactor Program (IFR) under the leadership of Argonne National Laboratory, indicated the need for data measurements and evaluations for several actinides and structural materials. This program led to the ENDF/B-VI library. Several evaluations presently in the latest ENDF/B version were done based on data measurements carried out at the ORELA facility. A few of these measurements include ^{233}U , ^{235}U , ^{238}U , ^{239}Pu , ^{240}Pu , ^{241}Pu .

2. SAMMY

The computer code SAMMY¹⁾ has had several upgrades to provide new features such as the capability of performing nuclear data evaluation in the unresolved energy region and for very high energy regions. SAMMY was originally developed for the evaluation of neutron interaction cross section in the resolved-resonance region. SAMMY utilizes multilevel multi-channel R-matrix theory for its cross section representation and uses the Bayes' method (generalized least squares) for the fitting procedure. The Bayes' method allows evaluators to obtain data covariance and sensitivity parameter information which is important information for calculating uncertainties in integral benchmark evaluations. N. Larson, the author of the code SAMMY, will present a detailed description of new developments in the code SAMMY at this conference.

III. NCSP Activities of the Nuclear Data Group

To exemplify the work of the ORNL Nuclear Data Group we present some of the activities performed in response to the NCSP program:

1. Resonance Evaluation of the ^{233}U Cross Sections

A resolved resonance analysis of the ^{233}U cross sections up to 600 eV was performed to improve the results of criticality safety calculations for the Molten Salt Reactor Experiments (MSRE). A paper describing the resolved resonance evaluation using the code SAMMY is presented in this conference. An unresolved resonance evaluation for ^{233}U cross section was done with the computer code FITACS²⁾ from 600 eV up to 500 KeV. The code FITACS was incorporated into SAMMY which is now referred to as SAMMY/URR. SAMMY/URR is a statistical code that uses the Hauser-Feshbach formalism for the calculation of average total cross sections and partial cross sections. The parameters required for the calculations are the neutron strength-functions, the average level spacings and the average partial widths derived from the parameters in the resolved energy region or from independent optical model calculations. The experimental data used in the unresolved resonance analysis of the ^{233}U total cross sections were obtained from Guber *et al.*³⁾ neutron transmission measurements taken at ORELA 80-meter flight-path with a sample thickness of 0.0119 at/b (with ^{10}B filter in the beam) with the sample cooled to 11 K in. The Doppler broadening effect at 11 K are reduced by a

factor of two as compared to taking measurements at room temperature. The fission cross section measurements used in the evaluation were also from Guber *et al.*⁴⁾ The fission measurements were performed at an 80-m flight-path in the energy range from 10 eV to 700 KeV. Two capture measurements were used in the evaluation: the data of Weston *et al.*⁵⁾ obtained from simultaneous measurement of the fission and capture cross section performed at ORNL up to 2 keV, and a capture cross section measurement of Hopkins⁶⁾ made at LANL from 200 keV to 500 keV. The SAMMY/URR fits of these data are shown in Fig. 1. Also, shown in Fig. 1 is the ENDF/B-VI ^{233}U evaluation. The solid line is the SAMMY/URR evaluations, and the dashed line is the ENDF/B-VI evaluation. The top curve is the total cross section of Guber *et al.*, the middle curve is the fission cross section of Guber *et al.*, and the bottom curve is the capture cross section of Weston from 100 eV to 2 keV, and the capture cross section of Hopkins from 200 keV to 500 keV. This evaluation will be used in benchmark calculations for ^{233}U critical experiments with a neutron energy spectrum peaking in the energy range from 100 eV to 500 keV.

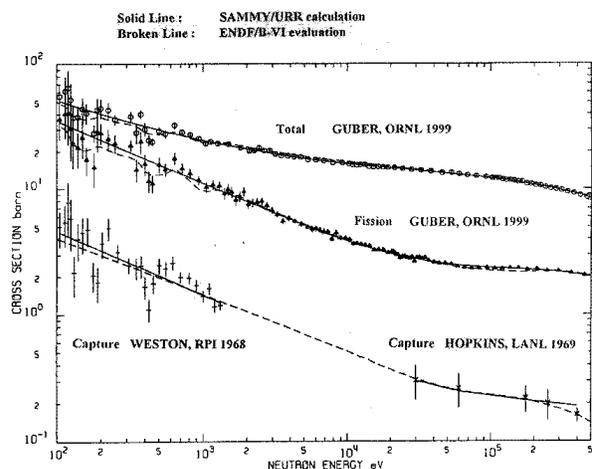


Fig. 1 SAMMY/URR fit of the total and fission cross sections of Guber, and capture cross section of Weston and Hopkins.

2. Resonance Evaluation of the Silicon Cross Section

The ENDF/B-VI Reich-Moore resonance evaluation of the three naturally-occurring isotopes of silicon, ^{28}Si , ^{29}Si , and ^{30}Si , were performed at ORNL based on the total cross sections of natural silicon and on the data for isotopic-enriched samples of silicon dioxide. When the evaluation were done, no capture cross section measurements were available. Therefore, the neutron capture widths in the ENDF/B-VI evaluation were obtained based on average values from the literature⁷⁾ and by using SAMMY to fit the total cross section. Because silicon is an important component in criticality safety applications, measurements of the silicon capture cross sections were done at ORNL by Guber *et al.*⁸⁾ using natural silicon samples for the energy range from 1 keV to 700 keV. The measured capture

data were used in the code SAMMY to re-evaluate the Reich-Moore resonance parameters for the three isotopes of silicon. The evaluation was initiated using the existing ENDF/B-VI evaluation. **Figure 2** shows a comparison of the ENDF/B-VI and ORNL capture cross sections in the energy region from few eV to 500 keV for ^{28}Si . The solid line is the ORNL evaluation and the dashed line is the existing ENDF evaluation. The capture cross section from the ORNL evaluation is consistently lower than the ENDF/B-VI evaluation.⁹⁾ Tests of the performance of the silicon evaluation in benchmark calculations using the MCNP code were done for five critical experiments. The MCNP pointwise cross sections were generated with the NJOY code. Similar calculations were done with the ENDF/B-VI silicon evaluation. The computed values of k_{eff} for the five critical experiments are given in **Table 1** for calculations using the silicon ENDF/B-VI evaluation and the ORNL evaluation.¹⁰⁾ The critical experiments are configurations with heterogeneous combinations of highly enriched uranium, silicone dioxide, and polyethylene. The measurements were done at the Institute for Physics and Power Engineering (IPPE), Obninsk, Russia. A benchmark model for these experiments by Tsiboulia *et al.* is documented in the Handbook of the International Criticality Safety Benchmark Evaluation Project (ICSBEP).¹¹⁾ In addition to the k_{eff} results, the average energy of neutrons causing fission (AEF), which is also shown in **Table 1**, which indicates the hardness of the neutron spectrum.

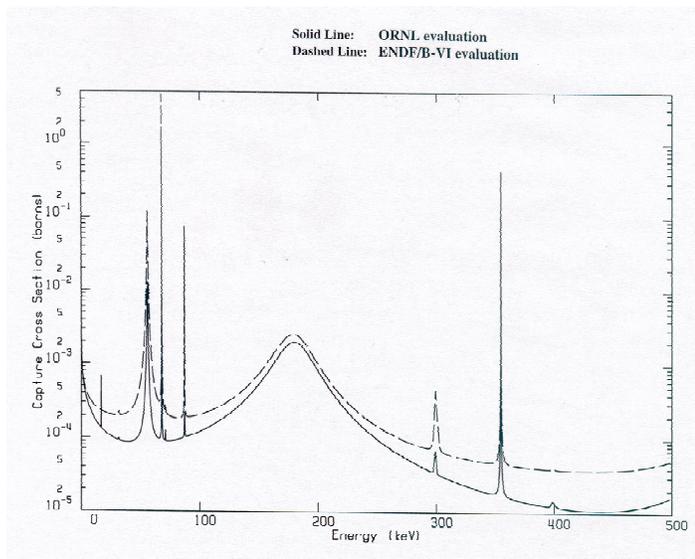


Fig. 2 Comparison of the ^{28}Si ORNL evaluation (solid line) with ENDF/B-VI evaluation (dashed line)

As presented in **Table 1**, the results using the ENDF/B-VI and ORNL evaluations for silicon are quite similar with exception of the BFS-79/5 benchmark. The first four results in **Table 1** correspond to experiments for which the neutron spectra are predominantly in the thermal region as indicated by the values of AEF. However, for the BFS-79/5 benchmark for which the neutron spectrum is in the epithermal energy region the results with the ORNL evaluation are better relative to the ENDF/B-VI results. This result indicates that the ORNL evaluation

Table 1 Comparison of k_{eff} for critical experiments with calculations using ENDF/B-VI and ORNL silicon evaluations

| Experiment | AEF (eV) | k_{eff} | |
|------------|----------|---------------------|---------------------|
| | | ENDF/B-VI | ORNL |
| BFS-79/1 | 49.7 | 1.0024 ± 0.0004 | 1.0030 ± 0.0004 |
| BFS-79/2 | 14.7 | 1.0123 ± 0.0004 | 1.0127 ± 0.0004 |
| BFS-79/3 | 2.81 | 1.0121 ± 0.0004 | 1.0117 ± 0.0004 |
| BFS-79/4 | 185.0 | 1.0044 ± 0.0004 | 1.0043 ± 0.0004 |
| BFS-79/5 | 4710.0 | 0.9975 ± 0.0004 | 0.9997 ± 0.0004 |

improves criticality safety calculations in the intermediate energy region for silicon.

3. Resonance Evaluation of the Chlorine Cross Section

Chlorine is an important material in criticality safety applications where chlorides are present in significant amounts. An example is the polyvinyl chloride pipe (PVC) tubes in which 57 % of chlorine by weight percent is present. To improve the cross section data for chlorine in the ENDF/B library data, measurements were performed at ORELA and an evaluation of ^{35}Cl , ^{37}Cl , and $^{\text{NAT}}\text{Cl}$ neutron cross sections in the resolved resonance region is underway. The evaluation is also being performed using the multilevel Reich-Moore R-matrix resonance formalism of the code SAMMY. **Table 2** gives selected information about the data sets used in the analysis. The evaluation for chlorine include recent high-resolution capture and transmission measurements for $^{\text{NAT}}\text{Cl}$ performed by Guber, *et al.*¹²⁾ at ORELA and total cross section data for $^{\text{NAT}}\text{Cl}$ done by Cierjacks *et al.*¹³⁾ Singh *et al.*¹⁴⁾ and Brugger *et al.*¹⁵⁾ Since the $^{35}\text{Cl}(n, p)^{35}\text{S}$ reaction yields a significant contribution to the total cross section from thermal energies up to ~ 10 keV, the (n, p) cross section data of Koehler¹⁶⁾ were fit to obtain proton widths for several unbound resonances as well as parameters for a negative energy resonance to fit the low energy data. The code SAMMY has been updated to enable it to calculate charged particle penetrabilities for the proton exit channel. The evaluation provides resonance energies and widths for 384 resonances in the range $0.2 < E_n < 1200$ keV for which both Doppler and resolution broadening effects are incorporated. Two negative-energy resonances are included to account for bound levels and several high-energy resonances are included to account for the effect of resonances above 1200 keV, including s- and p-wave. **Figure 3** and **Figure 4** illustrate how well the ORNL evaluation represents chlorine cross sections in the energy region up to 1200 keV. **Figure 3** shows a

comparison of the total cross section generated with the ORNL evaluation (solid line) and the evaluation in the ENDF/B-VI library (dashed line) with the experimental data of Guber *et al* (crosses) taken at ORELA in the energy region of few electron volts up to 100 keV. As can be seen, the present Reich-Moore cross section representation for chlorine using the ORNL evaluation is much better than the ENDF/B-VI evaluation. This representation should be extremely useful for criticality safety applications. In order to properly treat charged particles in an exit channel an algorithm to calculate charged particle penetrabilities and shifts was incorporated in the SAMMY code. Resonance parameters for the (n,p) channel in the ^{35}Cl isotope were determined by a consistent SAMMY analysis of the (n,p) cross sections. A sequential analysis of the total cross section and the (n,p) cross section was made with SAMMY to determine the Γ_{np} widths. **Figure 4** shows a comparison of the Reich-Moore representation of the ^{35}Cl (n,p) experimental cross section (solid line) and the experimental data are of Koehler (crosses) in the energy region of few electron volts up to 10 keV. The resonance parameter representation of the (n,p) cross section, as can be seen from **Fig. 4**, is excellent. The present ENDF format does not allow for a resonance parameter representation of charge particle cross sections. Work is underway to define a new ENDF format that will allow this data to be included.

Table 2 Natural Chlorine and ^{35}Cl Data Sets for the Chlorine Evaluation

| Data | Authors | Facility | Energy (keV) |
|-----------------------|------------------------|-----------------------------------|--------------|
| Total | Guber <i>et al</i> | ORELA 79.822 m | 0.02 - 1250 |
| Total | Singh <i>et al</i> | Columbia Cyclotron 202.05 m | 0.02 - 400 |
| Total | Brugger <i>et al</i> | MTR fast Chopper 45 m | 0.03 - 15 |
| Total | Cierjacks <i>et al</i> | KFK Cyclotron 57.54 m | 500 - 1250 |
| Total | Newson <i>et al</i> | Duke Van der Graaff | 7 - 194 |
| Capture | Guber <i>et al</i> | ORELA 40 m | 600 |
| $^{35}\text{Cl}(n,p)$ | Koehler | LANSCE | 0.01 - 35 |

IV. Summary and Conclusion

Nuclear data measurements, evaluation, and testing for criticality safety applications are performed at ORNL as part of the US/DOE nuclear criticality safety program. The needs for

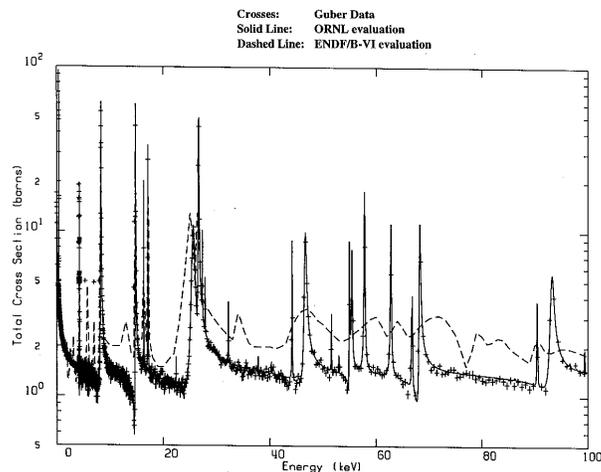


Fig. 3 Comparison of the natural chlorine total cross section for ORNL evaluation (solid line), ENDF/B-VI (dashed line) and measurements of Guber *et al.* (crosses)

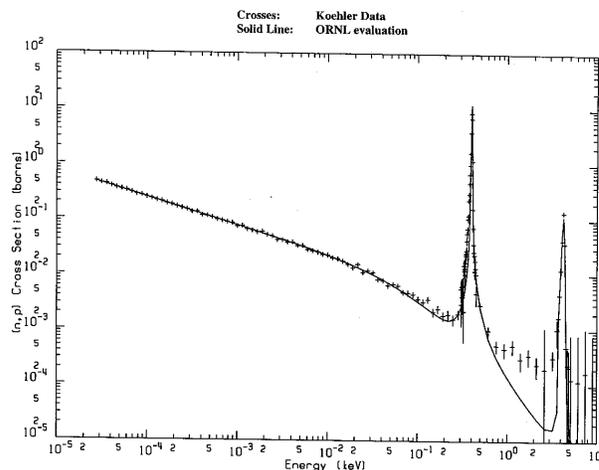


Fig. 4 Comparison of the (n,p) cross section for ^{35}Cl for ORNL evaluation (solid line) and measurements of Koehler (crosses).

new evaluations for criticality safety applications are identified by members of the ORNL Nuclear Data group by means of nuclear data priority list. Data measurements are done at the time-of-flight machine, ORELA, and evaluated using the Bayesian computer code SAMMY. This paper provides an overview of the present nuclear data measurements and evaluation work performed at ORNL.

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