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Neutron Cross-Section Measurements at ORELA

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Abstract—The Oak Ridge Electron Linear Accelerator (ORELA) is the only high-power white neutron source with excellent time resolution still operating in the United States and is ideally suited to measure fission, neutron total, and capture cross sections. For many nuclear criticality safety applications in the important neutron energy range from 1 eV to ~600 keV, many of the neutron cross sections from data libraries such as ENDF/B-VI or JENDL-3.2 exhibit large deficiencies. These deficiencies may occur in the resolved and unresolved-resonance regions. Consequently, these evaluated data may not be adequate for nuclear criticality calculations where effects such as self-shielding, multiple scattering, or Doppler broadening are important. To support the Nuclear Criticality Predictability Program, neutron cross-section measurements have been initiated at ORELA.

INTRODUCTION

For the last several years, concerns about existing nuclear data have been the prime motivator for new cross-section measurements at the Oak Ridge Electron Linear Accelerator (ORELA). Many older neutron cross-section evaluations from libraries such as ENDF/B-VI or JENDL-3.2 exhibit deficiencies or do not cover energy ranges that are important for criticality safety applications. Because many of the older evaluations were derived from measurements made with poor time-of-flight (TOF) resolution, the description of some data in the neutron energy range above several tens of keV is crude. Deficiencies may occur in the resolved- and unresolved-resonance regions. Therefore, some of these evaluated data may not be adequate for criticality calculations where effects such as self-shielding, multiple scattering, or Doppler broadening are important. Furthermore, many evaluations for nuclides having small neutron capture cross sections are erroneously large. Although their neutron capture cross sections are small, these nuclides can be important absorbers in many criticality calculations, and accurate cross-section data are essential. Of the several neutron sources in the United States, ORELA is the only operating high-power white neutron source with excellent time resolution in the energy range from thermal to about 1 MeV. Therefore, ORELA is ideally suited for measuring fission, neutron total, and capture cross sections in the energy range from 1 eV to ~600 keV, which is important for nuclear criticality safety applications. In fact, over 180 isotopes found in the evaluated libraries originated from ORELA cross-section measurements.

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WHITE NEUTRON SOURCES IN THE UNITED STATES

There are several white neutron sources still operating in the United States. Among them are the Los Alamos Neutron Scattering Science Center (LANSCE) facility at the Los Alamos National Laboratory (LANL) with the Lujan center and the Weapons Neutron Research Facility (WNR), the Intense Pulsed Neutron Source (IPNS) at the Argonne National Laboratory (ANL), the Gaertner Linear Accelerator at Rensselaer Polytechnic Institute (RPI), and ORELA. LANSCE and IPNS are spallation neutron sources driven by proton accelerators, whereas the RPI linac and ORELA are electron-accelerator-driven neutron sources. Each facility has its own characteristics, depending on the design of the accelerator and the neutron production target. For example, the Lujan center and IPNS are optimized for a high neutron flux in the thermal neutron energy range, which requires rather large moderators and broad pulses of the primary protons. On the other hand, the electron-linac-driven sources use smaller neutron target moderator assemblies and have shorter, variable-width pulses. Two important parameters define the quality of a neutron source and are used for classification. First, there is the neutron flux and spectrum, which should be suitable for the desired cross section measurement of the isotope of interest. Second, there is the neutron energy resolution, which defines how well individual resonances in the cross section can be distinguished from one another. High resolution can be of major importance, even if only the unresolved or average cross-section region is of primary interest, because average cross sections for applications can most reliably be determined from average resonance parameters resulting from the analysis of high-resolution measurements.

One useful figure of merit for comparing facilities is the neutron flux divided by the square of the resolution as a function of neutron energy. Relevant facility parameters are compiled in Table 1. In Figure 1, the figure of merit for three facilities at given flight path lengths and primary beam characteristics are plotted for comparison.

Table 1. U.S. white neutron source parameters for the intermediate energy range

Facility parameters	ORELA	LANSCE/Lujan	IPNS	RPI
Sources	e ⁻ linac	p spallation	p spallation	e ⁻ linac
Particle E (MeV)	140	800	450	>60
Flight path (m)	10–200	7–55	~6–20	10–250
Pulse width (ns)	2–30	125	40	15–5000
Max power (kW)	50	100	6.3	>10
Rep rate (Hz)	1–1000	20	30	1–500
Intrinsic resolution (ns/m)	0.01	3.9	2.0	0.06
Neutrons/s	1×10^{14}	1.2×10^{16}	8.1×10^{14}	4×10^{13}

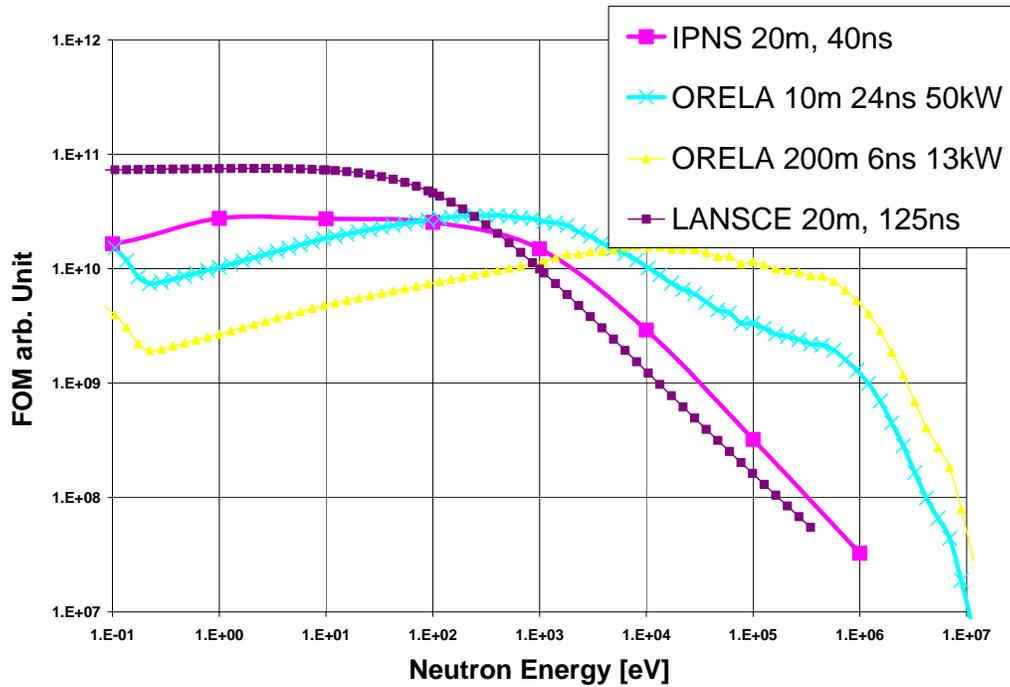


Figure 1. Comparison of figures of merit (neutron flux divided by the square of the resolution as a function of neutron energy) for the different neutron sources in the United States. In the case of LANSCE, the neutron flux was taken for the flight path of the Detector for Advanced Neutron Capture Experiments.

NUCLEAR DATA

Design and analysis codes for nuclear systems rely on evaluated cross sections and covariances from nuclear data libraries. These libraries, in turn, are built from cross-section measurements. Therefore, high-quality nuclear cross-section measurements are required for efficiency and safety analyses of new GEN-IV reactor designs; nuclear waste storage, transportation, and transmutation; and accelerator-driven subcritical systems.

Problems with existing nuclear data have emerged over the past few years, such as improper pulse-height weighting functions, neutron sensitivity backgrounds, poorly characterized samples, poor TOF resolution, and restricted energy ranges. Furthermore, corrigenda were published after discovering errors in the computer data reduction code (the correction factors ranged from 0.7480 to 1.1131 for 46 nuclides from ^{24}Mg to ^{232}Th [1] and from 0.9507 to 1.208 for 47 nuclides from ^{23}Na to ^{206}Pb [2]).

The validity of the calculated pulse-height weighting function used in the neutron capture experiments was questioned after a 20% discrepancy was found in the neutron width of 1.15-keV resonance in ^{56}Fe . Corvi *et al.* [3] overcame this problem by using an experimentally determined weighting function. On the other hand, using the Monte Carlo code EGS4 [4], Perey *et al.* [5] showed that a careful calculation of the weighting function also could resolve this problem.

The neutron sensitivity background appears to have been incorrectly accounted for in some of the old neutron capture data. This background is caused by neutrons scattered from the sample and subsequently captured in the detector or surroundings within the time corresponding to the width of the resonance. It has led to erroneously large capture areas in the old data for resonances having large neutron widths, as shown in Figure 2. In those cases it has led to incorrectly large capture kernels in the current ENDF/B-VI evaluations. The new neutron capture apparatus at ORELA has been improved in many ways compared to the old apparatus with the result that this neutron sensitivity background has been reduced to the point where it is no longer a problem.

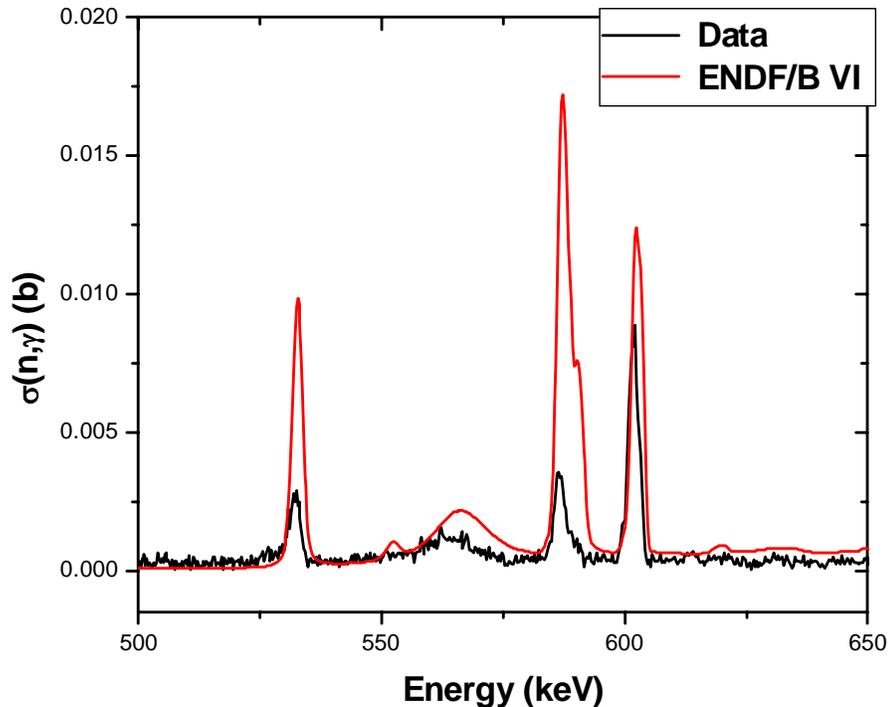


Figure 2. The large neutron sensitivity of older measurements led to many erroneously large resonance areas in current evaluations. The black curve represents the new ORELA experimental data. The red curve is the calculated cross section including all experimental effects using the ENDF/B-VI evaluation (which is based on the older measurement) for Si.

Although efforts were made to use highly (isotopically) enriched and chemically pure samples, it appears that sometimes problems with a sample's chemical composition led to large systematic errors in some measurements. For example, some samples were available only as oxides, which can be hygroscopic and therefore pick up water fairly easily. A substantial water content in the sample can lead (via moderation effects) to erroneously large cross sections.

Because of computer storage system limitations, many of the older cross-section measurements were performed with data bins that were too coarse. As a consequence, those data sets sometimes have too few data points over narrow resonances to accurately calculate corrections for experimental effects such

as Doppler broadening, self-shielding, and multiple scattering. In addition, many of the older experiments were run with a low-energy cut-off of around 3 keV. However, this missing energy range can be important for current nuclear criticality calculations. Hence, new measurements are needed in these cases.

EXPERIMENTAL SETUP AT ORELA

ORELA consists of a 180-MeV electron linear accelerator, a neutron-producing target, underground and evacuated flight tubes, sophisticated detectors, and data acquisition systems. The accelerator is highly flexible because of its variable repetition rate (1–1000 Hz) and burst width (2–30 ns). At full power, the average neutron flux is 10^{14} neutrons/s. Simultaneous measurements are possible at 18 detector stations on 10 separate flight paths at distances between 9 and 200 m from the neutron source. The TOF technique has been used in the energy range from thermal up to 50 MeV to measure neutron total, capture, fission, elastic, inelastic, (n,α) , and γ -ray and neutron production cross sections.

Capture Measurements

Neutron capture experiments at ORELA are usually performed at the 40-m flight station, on flight path 7. A pair of deuterated benzene (C_6D_6) detectors is used to detect the capture γ -rays; the pulse-height-weighting method is applied. Over the last couple of years, the system has been improved in many ways compared to the old ORELA apparatus [6]: First, most of the structural material surrounding the sample and detectors was reduced to decrease the background from sample-scattered neutrons (neutron sensitivity background). This was accomplished by replacing the massive Al sample changer and beam pipe with a thin carbon fiber tube. The steel detector housings were replaced with reduced-mass detector mounts. Second, the C_6F_6 scintillator was replaced with a C_6D_6 scintillator, which has much lower neutron sensitivity. More details about these improvements can be found in the papers by Koehler *et al.* [7, 8]. For two resonances in ^{88}Sr , at 289 and 325 keV with neutron widths $g\Gamma_n=24,932$ and 22,082 eV, respectively, the measured capture widths were an average factor of five smaller than reported from measurements using the old system (after a correction for neutron sensitivity already had been applied to the old data). Third, calculation of the detector weighting functions has been improved by using the Monte Carlo code EGS4 and by including the sample and all structural materials within 30 cm of the detectors in the calculations.

Over the last few years, we used the new setup to perform several neutron capture cross-section experiments on elements with small capture cross sections (Al, Cl, F, Si, and K) that are of interest for nuclear criticality. A 1.27-cm-thick Pb filter was employed to reduce the γ -ray background from the neutron production target; and pulse overlap neutrons were eliminated with a 0.48-g/cm^2 ^{10}B filter. Absolute cross sections were determined by using the saturated resonance technique, employing the 4.9-eV resonance in gold [9]. A 0.5-mm-thick 6Li -glass scintillator, placed 42.1 cm upstream of the sample position, was used to measure the energy dependence of the neutron flux.

Transmission Measurements

High-resolution transmission experiments for determining the total cross sections are not only indispensable for evaluations but are also necessary for calculating self-shielding and multiple-scattering corrections for neutron capture cross-section measurements. Given the fact that capture experiments cannot be performed with an infinitesimally thin sample (in fact, sometimes the samples are quite thick), these corrections can be sizeable. Consequently, we made corresponding total cross-section measurements when needed. Also, transmission measurements sometimes can be more sensitive to certain resonances than (n,γ) measurements. The neutron beam was collimated to about 2.54 cm on the samples and allowed only neutrons from the water moderator part of the neutron source to be used. The neutron detector was an 11.1-cm-diameter, 1.25-cm-thick ${}^6\text{Li}$ -glass scintillator positioned in the beam at a distance of 79.815 m from the neutron source. For background reduction, the scintillator was viewed edge-on by two 12.7-cm-diameter photomultipliers that were placed outside the neutron beam. To reduce systematic uncertainties, we cycled the samples and their compensators or corresponding empty containers periodically through the neutron beam; the neutron flux was recorded for each sample and cycle. Additional measurements with a thick polyethylene sample were used to determine the γ -ray background from the neutron source.

EXPERIMENTAL RESULTS

We found significant differences between our new capture and transmission data and the evaluations for Al, Cl, F, K, and Si obtained from the ENDF/B-VI or JENDL-3.2 nuclear data libraries. For these elements with small capture cross sections, our new results are even smaller than the previous ones. It is evident from our new data that in many previous cases, the capture widths were severely overestimated and resonances were missed as a result of large backgrounds. The new high-resolution total cross-section measurements reveal previously misassigned resonances and enabled us to extend the resolved resonance region to much higher energies. An example of the data is given in Figure 3, where we plotted the transmission data for natural metallic potassium and the corresponding capture data from our K_2CO_3 sample.

The discrepancies observed between our data and the evaluated data from the nuclear data libraries have two main causes. First, the use of improper weighting functions resulted in mismatched detector response functions. Second, underestimated neutron sensitivity of the experimental setups resulted in previous capture cross sections that were too large. Together with the better characterized samples, the superior TOF resolution, and the well-understood experimental setups and backgrounds, the new experimental data enable us to produce more reliable resonance parameters.

The neutron total and capture cross-section data were analyzed with SAMMY [10]. This code made the necessary corrections for the experimental effects, such as Doppler and resolution broadening, self-shielding, and multiple-scattering effects. The new resonance parameters were then used as a starting point for an evaluation. In this evaluation other existing experimental data sets were included whenever they were available and suitable. The new cross-section data set was then used for criticality benchmark calculations and was checked for inconsistencies. As an example, the ORNL evaluation for Si shows large discrepancies from capture cross sections found in the ENDF/B-VI nuclear data library. We observed two resonances for ${}^{28}\text{Si}$ that had not been previously reported. In addition, we determined that one resonance previously assigned to ${}^{28}\text{Si}$ is actually in ${}^{30}\text{Si}$. Furthermore, a reported resonance in ${}^{30}\text{Si}$ at 2.235 keV was not visible in our new capture data and was not in transmission measurements of an enriched ${}^{30}\text{Si}$ sample. The result of this evaluation [11] is shown in Figure 4, where the unbroadened capture cross sections are plotted. For the correct capture of Si, the direct capture component has to be added to the resonant capture of the ORNL evaluation.

In the case of Cl we found similar results [12]. The neutron capture cross section is too large in the ENDF/B-VI evaluation, and the resolved energy range is very limited compared with our data (Figure 5).

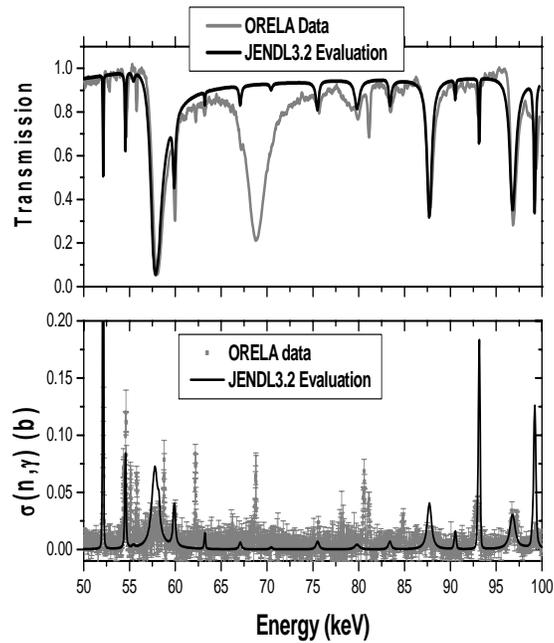


Figure 3. Transmission and capture of natural potassium compared with the transmission and capture calculated from JENDL3.2 parameters. The fact that the strong resonance at 68.8 keV in transmission is not reported seem to be puzzling and could be very well a misprint in JENDL.

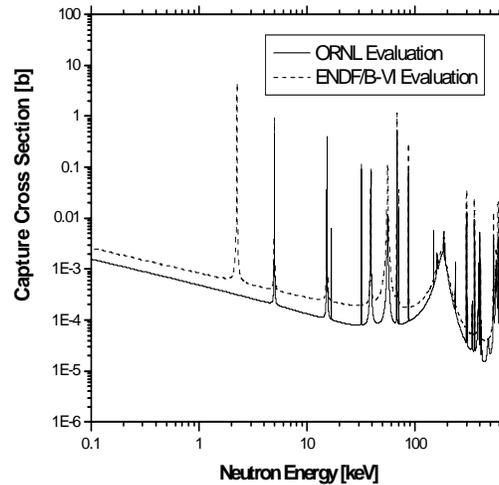


Figure 4. Evaluations of natural Si from ENDF/B-VI (dashed curve) compared with the ORNL evaluation (solid curve). To obtain the correct ORNL neutron capture, the contribution of the direct capture calculation must be added to the resonant capture from the ORNL evaluation.

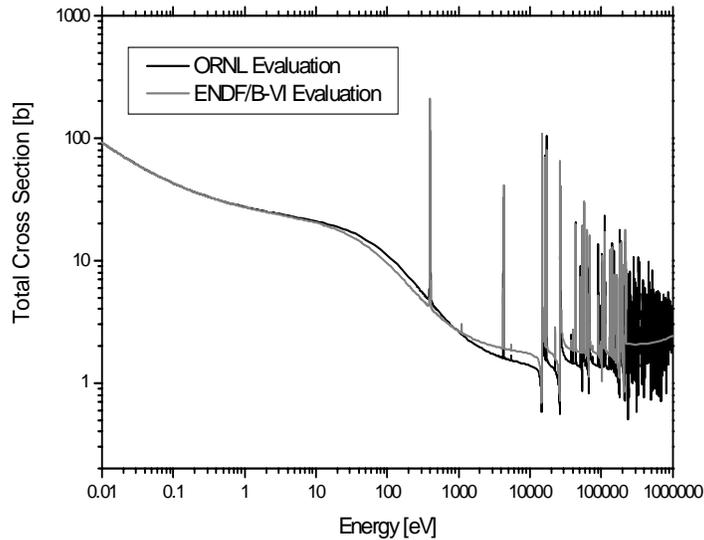


Figure 5. Evaluations of Cl from ENDF/B-VI (grey curve) compared with the ORNL evaluation (black curve).

CONCLUSIONS

To support the Nuclear Criticality Safety Program, we performed new neutron total and capture measurements at ORELA over broad energy ranges. We then used the SAMMY multilevel R-matrix code. In all analyzed and evaluated cases, we were able to extend the resolved resonance region to much higher energies than the existing evaluations. These new evaluations should lead to much more reliable nuclear criticality calculations.

We would like to emphasize one particular finding. Over the past ten years, the results of our new neutron capture cross-section measurements at ORELA for samples with large scattering cross sections have shown the tendency to be smaller than the data found in the nuclear data libraries. Therefore, many of the older measurements for samples with small capture cross sections are questionable, or at least much more uncertain, especially if the applied corrections for neutron sensitivity were sizeable.

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