New Neutron Cross-Section Measurements at ORELA for Improved Nuclear Data Calculations

K. H. Guber, L. C. Leal, R. O. Sayer, P. E. Koehler, T. E. Valentine, H. Derrien, and J. A. Harvey

Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN 37831-6354, USA

Abstract. 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. 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 the Oak Ridge Electron Linear Accelerator (ORELA). ORELA is the only high-power white neutron source with excellent time resolution still operating in the United States. It is ideally suited to measure fission, neutron total, and capture cross sections in the energy range from 1 eV to ~600 keV, which is important for many nuclear criticality safety applications.

INTRODUCTION

Concerns about data deficiencies in some existing cross-section evaluations from libraries such as ENDF/B-VI or JENDL-3.2 for nuclear criticality calculations have been a prime motivator for new cross-section measurements at the Oak Ridge Electron Linear Accelerator (ORELA). Many older neutron cross-section evaluations exhibit deficiencies or do not cover energy ranges that are important for criticality safety applications. Moreover, many older evaluations were derived from measurements made with poor time-of-flight (TOF) resolution, and the description of some data in the neutron energy range above several tens of keV is crude. 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. Furthermore, many evaluations for nuclides having small neutron capture cross sections are erroneously large because the neutron sensitivity of the old measurements was underestimated. 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. Over the last three decades, many neutron-induced cross-section measurements have been performed at ORELA. It is the only operating high-power white neutron source in the United States with excellent time resolution in the energy range from thermal to about 1 MeV. ORELA is ideally suited to measure 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.

NUCLEAR DATA

High-quality nuclear data are essential for the design and analysis of any nuclear system, such as reactor core and fuel elements and storage of mixtures of nuclear waste with other materials, as well as burned fuel elements. Pertinent nuclear data are also necessary for waste transmutation, accelerator-driven systems, and GEN-IV reactor design. All of the analysis codes for nuclear systems rely on the use of evaluated cross-section data from libraries such as ENDF/B-VI or JENDL-3.2.

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A wide variety of problems with existing nuclear data have come to light over the past few years. These include problems related to improper pulse-height weighting functions, neutron sensitivity backgrounds, poorly characterized samples, poor TOF resolution, and restricted energy range. Also, after errors were discovered in the computer data reduction code, corrigenda were published (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]).

After a 20% discrepancy was found in the neutron width of 1.15-keV resonance in Fe measured with C_6D_6, compared to transmission measurements, the validity of the calculated pulse-height weighting function used in the neutron capture experiments was questioned. Corvi et al. [3] showed that using an experimentally determined weighting function could overcome this problem. On the other hand, using the Monte Carlo code EGS4 [4], Perey et al. [5] showed that a careful calculation of the weighting function could also resolve this problem.

As shown in Fig. 1, the neutron sensitivity of the experimental setup often was underestimated in previous (n,γ) measurements. This neutron sensitivity background is caused by neutrons scattered from the sample and captured in the detector or surroundings within the time corresponding to the width of the resonance. Also, it led in some cases to incorrectly large capture kernels in the current ENDF/B-VI evaluation.

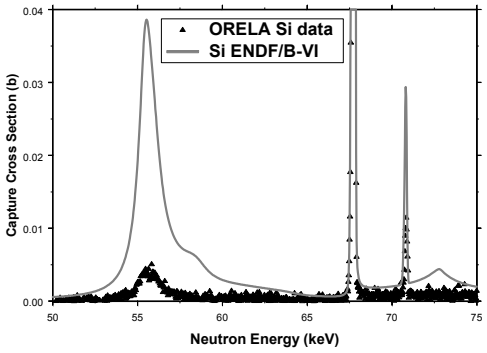

FIGURE 1. The large neutron sensitivity of older measurements led to many erroneously large resonance areas in current evaluations. The triangles represent the new ORELA experimental data, and the solid line is the calculated cross section including all experimental effects using the ENDF/B-VI evaluation (which is based on the older measurement) for Si.

Even though, whenever it was necessary, enriched samples were used in many experiments, the sometimes poorly characterized chemical composition of the samples resulted in large systematic errors in some measurements. This could especially be the case when oxide samples were used that were not correctly treated. Some oxides, which are in many cases the inventory form of the enriched isotope material, are known to be hygroscopic and therefore to pick up water fairly easy. If no precautions were taken, the rising water content in the sample could lead (via moderation effects) to falsely large results.

Because of computer storage systems limitations, many of the existing cross-section measurements were performed with data bins too coarse. As a consequence, the data sets sometimes have too few data points over the resonance for the analysis programs to calculate accurately the corrections of experimental effects such as Doppler broadening, self-shielding, and multiple scattering. In addition, many of the older experiments were run with an energy cut-off of around 3 keV, since there was no interest in the resonance data below that cut-off. It was found that this missing energy range is sometimes of importance for nuclear criticality calculations.

The current apparatus at ORELA has been designed to overcome all of these problems.

EXPERIMENTAL SET UP AT ORELA

ORELA is the only high-power white neutron source with excellent time resolution in the keV neutron energy range still operating in the United States. ORELA consists of a 180-MeV electron linear accelerator, neutron-producing target, underground and evacuated flight tubes, sophisticated detectors, and data acquisition systems. It is a highly flexible accelerator with a varying repetition rate between 1 and 1000 Hz and a burst between 2 and 30 ns. This leads to an average neutron flux of 10^{14} neutrons per second. 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 is used for measuring neutron-induced cross-section data in the energy range from a few eV up to 50 MeV, such as transmission measurements (total cross sections), capture cross sections, fission cross sections, elastic cross sections, and neutron production cross sections.

Capture Measurements

The neutron capture experiments were performed at the 40-m flight station of ORELA utilizing flight path 7. The employed experimental technique was the pulse-height-weighting method using a pair of deuterated benzene (C_6D_6) detectors. Compared to the old ORELA setup [6], the capture system has been improved in several ways: (i) The amount of structural material surrounding the sample and detectors in order to reduce the background due to sample-scattered neutrons (neutron sensitivity) was minimized. This was achieved by removing the massive Al-sample changer and replacing the beam pipe with a thin carbon fiber tube. In addition, the massive detector housings were removed and replaced with reduced-mass detector mounts. (ii) The more neutron-sensitive C_6F_6 γ-ray detectors were replaced with C_6D_6, which has much lower neutron sensitivity. More details about these improvements can be found in the papers by Koehler et al. [7,8] in which the impact of the neutron sensitivity was impressively demonstrated in a high-resolution TOF measurement for ^{88}Sr. For the two prominent resonances at 289 and 325 keV with neutron widths $g\Gamma_n$ =24,932 and 22,082 eV, respectively, a reduction of capture widths by an average factor of five was reported. (iii) In addition to replacing the old data acquisition system, the more sophisticated computer code EGS4 is used to calculate the appropriate detector weighting function for each experiment. All structural materials within 30 cm of the detectors, including the sample, are incorporated into these calculations. The code was used to calculate the response functions of the detector for various monoenergetic γ-rays. The resulting pulse-height spectra were then broadened using a resolution function. The final weighting function was calculated from these broadened spectra using a least-squares fitting code.

With this new setup, we performed several neutron capture cross-section experiments over the last few years, mainly on isotopes that have small (n,γ) cross sections and are of interest for nuclear criticality. Samples measured include (i) extremely high-purity aluminum (0.01520 atom/b and 0.04573 atom/b), (ii) a high-purity natural silicon sample (0.07831 atom/b), (iii) a natural LiCl sample (0.09812 atom/b), (iv) a Teflon (for fluorine) sample (0.05086 atom/b), and (v) a natural K_2CO_3 sample (0.0088791 atom/b), as well as an enriched ^{41}KCl sample.

A 1.27-cm-thick Pb filter was used to reduce the γ-ray background from the neutron production target, and a 0.48-g/cm^2 ^{10}B filter served to eliminate pulse overlap by absorbing low-energy neutrons. Otherwise, these neutrons would be interpreted from the data-acquisition system as higher-energy neutrons from the next neutron pulse. Normalization of the capture efficiency was carried out in a separate measurement using the "saturated resonance" technique by means of the 4.9-eV resonance from a gold sample [9]. A 0.5-mm-thick ^{6}Li-glass scintillator at a distance of 39.695 m from the neutron target was used to monitor the neutron flux.

Transmission Measurements

High-resolution transmission experiments for determining the total cross section are not only indispensable for an evaluation but also a necessity for the analysis of neutron capture cross sections, in order to apply all the corrections for the experimental effects. Because capture experiments cannot be performed with an infinitely thin sample (in fact, sometimes the samples are quite thick), the corrections for self-shielding and multiple scattering can be sizeable. Therefore, we made corresponding total cross-section measurements when needed. In addition, some resonances with small radiation widths are not visible in the (n,γ) data and vice versa. For the Al transmission measurements, the two samples (0.0189 atom/b and 0.1513 atom/b) were mounted in the sample changer positioned at about 10 m from the neutron target in the beam of ORELA. For the chlorine transmission measurement, we used a natural CCl_4 (thickness for Cl 0.2075 atom/b) sample and a corresponding C compensator in the open beam. For the potassium transmission, we used two metallic samples (0.013367 and 0.10517 atom/b) mounted in a sealed brass holder. A pre-sample collimation limited the beam size 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}Li-glass scintillator positioned in the beam at 79.815 m from the neutron source. The scintillator was viewed on edge by two 12.7-cm-diameter photomultipliers that were placed outside the neutron beam to decrease backgrounds. To reduce systematic uncertainties, the samples and their compensators or corresponding empty containers were cycled periodically through the neutron beam; and the neutron flux was recorded for each sample and cycle. Additional measurements with a thick polyethylene sample were used to determine the gamma-ray background from the neutron source.

EXPERIMENTAL RESULTS

The results of our new capture and transmission experiments show significant differences compared to the evaluated nuclear data for Al, Cl, F, K, and Si obtained from the ENDF/B-VI or JENDL-3.2 nuclear data libraries. In general, all of our new neutron capture cross sections are smaller than previous results. Our new capture data show that in many previous cases, capture widths were severely overestimated and resonances were missed as a result of large backgrounds. Our new total cross-section measurements reveal previously misassigned resonances and extend the resolved resonance region to much higher energies. An example of the data is given in Fig. 2, where we plotted the transmission data for natural metallic potassium and the corresponding capture data from our K_2CO_3 sample.

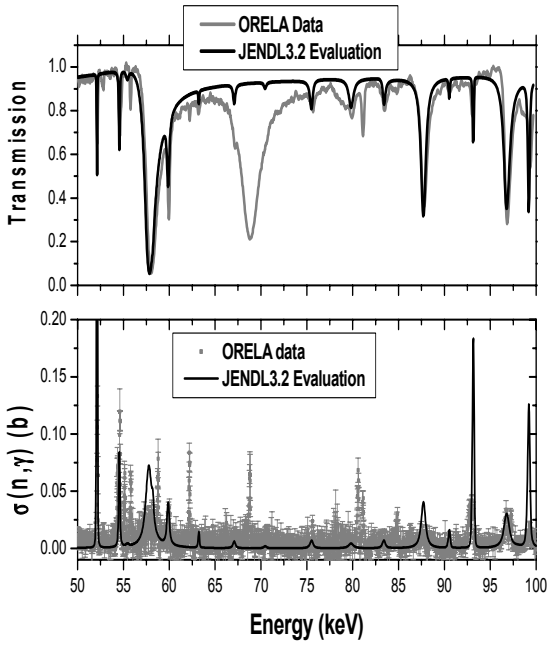

FIGURE 2. Transmission and capture of natural potassium compared to the transmission and capture calculated using JENDL3.2 parameters.

The observed discrepancies between our data and the evaluated data from the nuclear data libraries mainly have two reasons; first, the use of improper weighting functions resulted in mismatched detector response functions. In the new experiments, the more sophisticated computer code EGS4 was used for the correct determination of the weighting function. Secondly, underestimated neutron sensitivity of the experimental setups led to previous capture cross sections that were too large. In addition, better characterized samples, superior TOF-resolution, and well-understood experimental setups and backgrounds helped to produce more reliable cross-section data in the present case.

Using the computer code SAMMY [10], the neutron total and capture cross-section data were analyzed. SAMMY applied all of the necessary corrections for the experimental effects, such as Doppler and resolution broadening, self-shielding, and multiple-scattering effects to the data. The resonance parameters obtained are the basis for an evaluation of the cross sections calculated by the Nuclear Data Group of the Nuclear Science and Technology Division of the Oak Ridge National Laboratory (ORNL). When they were available and suitable, this evaluation included other existing experimental data sets. The final result was then checked for consistencies using criticality benchmark calculations. As an example, the ORNL evaluation for Si shows serious discrepancies from capture cross sections found in the nuclear data library ENDF/B-VI. We observed two resonances for ^{28}Si that had not been previously reported. In addition, we determined that one resonance previously assigned to ^{28}Si is actually in ^{30}Si. Furthermore, a reported resonance in ^{30}Si at 2.235 keV was not visible in our new capture data nor in transmission measurements using an enriched ^{30}Si sample. The result of this evaluation [11] is shown in Fig. 3, where the unbroadened capture cross sections

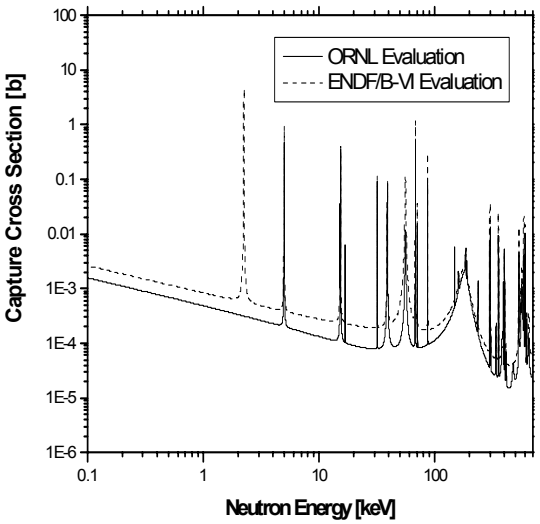

FIGURE 3. Evaluations of natural Si from ENDF/B-VI (dashed line) compared to the ORNL evaluation (solid line). Note for the correct ORNL neutron capture, the contribution of the direct capture calculation has to be added to the resonant capture from the ORNL evaluation.

are plotted. Note for the correct capture of Si, the direct capture (DC) component has to be added to the resonant capture of the ORNL evaluation. The average capture cross sections are calculated and compiled in Table 1.

In the case of Al we find similar results. The neutron capture cross section is too large in the ENDF/B-VI and JENDL3.2 evaluations; and the resolved energy range is very limited compared to our data, which was analyzed up to 700 keV. This is shown in Fig. 4.

TABLE 1. The calculated average neutron-capture cross sections for natural Si.

Energy Range (keV)	DC (mb)	ORNL Resonant Capture (mb)	Sum DC + Resonant Capture (mb)	ENDF/B-VI (mb)
1 - 250	0.31	0.978	1.29	2.172
250 - 450	0.50	0.212	0.71	0.532
450 - 700	0.64	0.326	0.96	0.875

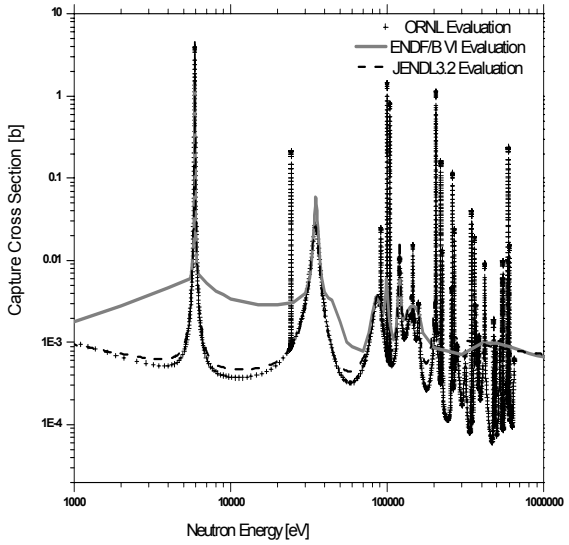

FIGURE 4. Evaluations of Al from ENDF/B-VI (grey curve) and JENDL3.2 (dashed curve) compared to the ORNL evaluation (cross).

CONCLUSIONS

To support the Nuclear Criticality Safety Program, we performed new neutron total and capture measurements at ORELA over broad energy ranges. The obtained results were then analyzed using the multilevel R-matrix code SAMMY. 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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