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New Neutron Cross-Section Measurements at ORELA and Their Application in Nuclear Criticality Calculations

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Abstract. Many older neutron cross-section evaluations from libraries such as ENDF/B-VI or JENDL-3.2 show deficiencies in energy ranges that are important for criticality safety applications. 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 DOE Nuclear Criticality Safety Program, neutron cross-section measurements have been initiated at the Oak Ridge Electron Linear Accelerator (ORELA). 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 many nuclear criticality safety applications.

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INTRODUCTION

Design and analysis of any nuclear system—such as reactor core and fuel elements, storage of mixtures of nuclear waste with other materials, and burned fuel elements—require high-quality nuclear data. Appropriate nuclear data is necessary for waste transmutation, accelerator-driven systems, GEN-IV reactor design, and nuclear criticality calculations. All of the analysis codes for nuclear systems rely on the use of evaluated cross-section data from the libraries such as ENDF/B-VI or JENDL-3.2. However, the nuclear data found in these libraries show deficiencies. For example, evaluations for some nuclides do not cover energy ranges (1 eV to ~600 keV) that are important for criticality safety applications. Moreover, the description of some data in the neutron energy range above several tens of keV is crude.

There are many problems associated with existing nuclear data, such as the use of the incorrect pulse-height weighting functions, underestimation for neutron sensitivity backgrounds, poorly characterized samples, poor TOF resolution, and too restricted energy range. In addition, corrigenda were published after errors were discovered in the computer data reduction code (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 the 1.15-keV resonance in Fe measured with this technique compared with transmission measurements. Applying an experimentally determined weighting function, Corvi et al. [3] demonstrated that this discrepancy could be resolved. 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.

The neutron sensitivity of the experimental setup often was underestimated in previous neutron capture experiments. This 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. As a consequence many evaluations for nuclides having small neutron capture cross sections are erroneously large, as shown in Fig. 1 in the case of neutron capture on K. Although the neutron capture cross sections are small, they can be important absorbers in criticality calculations, and accurate cross-section data are essential.

Enriched samples are typically used in neutron capture experiments, but poorly characterized chemical composition of the samples resulted in large systematic errors in some measurements. The inventory form of the enriched isotope materials are in many cases oxides. They are known to be hygroscopic, and to absorb water fairly easily. Without treatments to remove this water content, moderation effects caused by the unsuspected hydrogen in the sample can lead to falsely large capture cross sections.

Some older cross-section measurements were performed with too coarse data binning because of the limited computer capacities. Consequently, the data sometimes have too few data points over the resonances for analysis programs, such as SAMMY [6], to calculate accurately corrections for effects such as Doppler broadening, self-shielding, and multiple scattering. In addition, many older experiments were run with an energy cutoff of around 3 keV, since at the time there was no interest in the resonance data below that cutoff. However, this missing energy range is sometimes of importance for current nuclear criticality calculations.

Any of these problems could be a possible cause of incorrect cross-section evaluations and hence result in unreliable nuclear criticality calculations. Thus, such problems are one of the prime motivations for performing new neutron cross-section measurements at ORELA.

EXPERIMENTAL APPARATUS AT ORELA

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 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 varying repetition rate between 1 and 1000 Hz and a burst width 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.

Capture Measurements

Neutron capture experiments were performed at ORELA on flight path 7 at a distance of 40 m from the neutron-producing target with a pair of deuterated benzene (C_6D_6) detectors using the pulse-height-weighting method. This apparatus [7] has been improved in several ways, over the past years. Firstly, the original C_6F_6 γ-ray detectors were replaced with C_6D_6 detectors, which have much lower neutron sensitivity. Secondly, much of structural material surrounding the sample and detectors was removed in order to reduce the neutron sensitivity even further. For example, the massive Al-sample changer and beam pipe were replaced the by a thin carbon

fiber tube. Additionally, the massive detector housings were replaced with reduced-mass detector mounts. Thirdly, the appropriate detector weighting function for each experiment is now calculated using the more sophisticated computer code EGS4. All structural materials within 30 cm of the detectors, including the sample, are incorporated into these calculations. More details about these improvements can be found in the papers by Koehler et al. [8, 9].

The neutron flux was measured with a 0.5-mm-thick ^{6}Li-glass scintillator at a distance of 39.695 m from the neutron target. The final 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 [10].

Over the past few years, several neutron capture cross-section experiments have been performed with this improved setup. Many of these measurements were on nuclides of interest for nuclear criticality because they often occur in mixtures with fissile materials. Most of these nuclides have small (n,γ) cross sections. However, because they are present in large amounts in these mixtures they can play important roles as neutron absorbers. Samples measured included extremely high purity aluminum (0.01520 atom/b and 0.04573 atom/b), natural silicon (0.07831 atom/b), a natural LiCl sample (0.09812 atom/b), a Teflon (for fluorine) sample (0.05086 atom/b), and a natural K_2CO_3 sample (0.0088791 atom/b), as well as an enriched ^{41}KCl sample.

Transmission Measurements

Usually neutron capture experiments are performed with a fairly thick sample; therefore the corrections for self-shielding and multiple scattering can be sizeable. In order to apply corrections for these experimental effects, we made corresponding total cross-section measurements. These data also can be useful in making isotopic assignments as well as for

observing some resonances not visible in the (n,γ) data. For the total cross-section measurements on Al, we used two samples with different thickness (0.0189 atom/b and 0.1513 atom/b). These were mounted in a computer-controlled sample changer positioned at about 10 m from the neutron target in the beam of ORELA. For the chlorine transmission measurement, a natural CCl_4 (thickness for Cl 0.2075 atom/b) sample and a corresponding C compensator were mounted in the sample changer. The potassium transmission measurement was carried out using two metallic samples (0.013367 and 0.10517 atom/b). A presample collimator limited the beam size to about 2.54 cm at the sample position and allowed only neutrons from the water moderator part of the neutron source to be seen by the detector. As a neutron detector we used an 11.1-cm-diameter, 1.25-cm-thick ^{6}Li-glass scintillator positioned in the beam at the 80-m flight station at a distance of 79.815 m from the neutron source.

EXPERIMENTAL RESULTS

We find significant differences between our new capture and transmission experiments compared with the evaluated nuclear data files. Overall, our new neutron capture cross sections for Al, Cl, F, K, and Si are smaller than previous results. In many previous cases, capture widths were severely overestimated and resonances were missed due to large backgrounds. Additionally, the new total cross-section measurements reveal previously misassigned resonances and extend the resolved resonance region to much higher energies. The data were then analyzed using the computer code SAMMY. As a consistency check of these resulting evaluated cross sections, criticality benchmark calculations were performed. As an example, the Oak Ridge National Laboratory (ORNL) evaluation for Si shows serious discrepancies from capture cross sections found in the nuclear data library ENDF/B-VI. We observed new

resonances in ^{28}Si; found a misassigned resonance in ^{28}Si, which is actually in ^{30}Si; and conclusively showed that a previously reported resonance in ^{30}Si at 2.235 keV does not exist. Similar results were obtained in the cases of Al and Cl; the previous neutron capture and total cross sections were too large in the ENDF/B-VI and JENDL3.2 evaluations. Additionally, the resolved energy range is greatly expanded in our evaluations, which were analyzed up to 700 keV and 1.2 MeV for Al and Cl, respectively. The result of the total cross section for ^{35}Cl is shown in Fig. 2.

CONCLUSIONS

To support the Nuclear Criticality Safety Program, we performed new neutron total and capture measurements at ORELA over broad energy ranges. In all analyzed and evaluated cases, we found serious discrepancies with respect to existing evaluations and we were able to extend the resolved resonance region to much higher energies. Over the past ten years, the results of our new neutron capture cross-section measurements at ORELA for samples having large scattering cross sections are generally smaller than the previous results. Therefore, many of the older evaluations for samples with small capture cross sections are of questionable accuracy, especially if the applied corrections for neutron sensitivity were sizeable.

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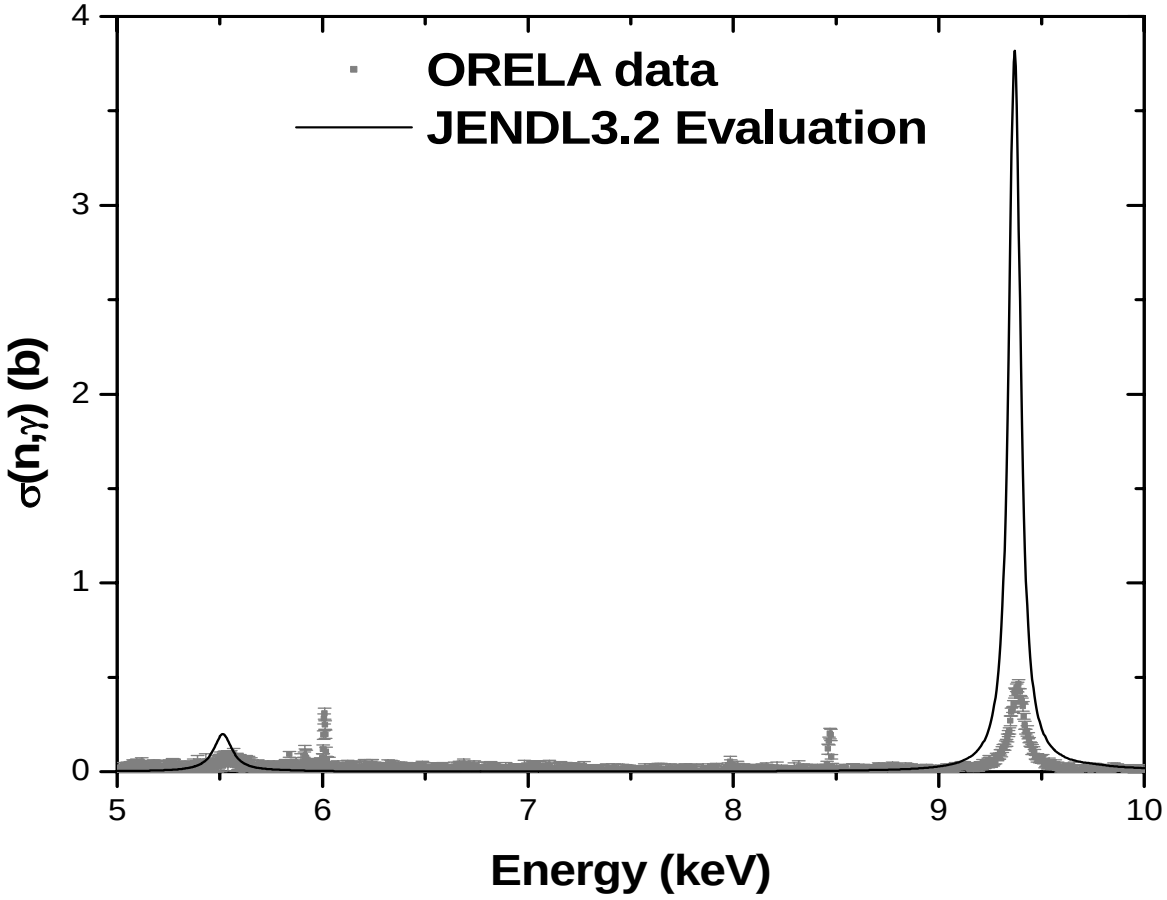

FIGURE 1: The large and underestimated neutron sensitivity of older measurements has led to many erroneously large neutron capture resonance areas in current evaluations. The symbols represent the new ORELA experimental data. The solid curve is the calculated cross section, including experimental effects, using the JENDL3.2 evaluation (which is based on the older measurement) for natural K.

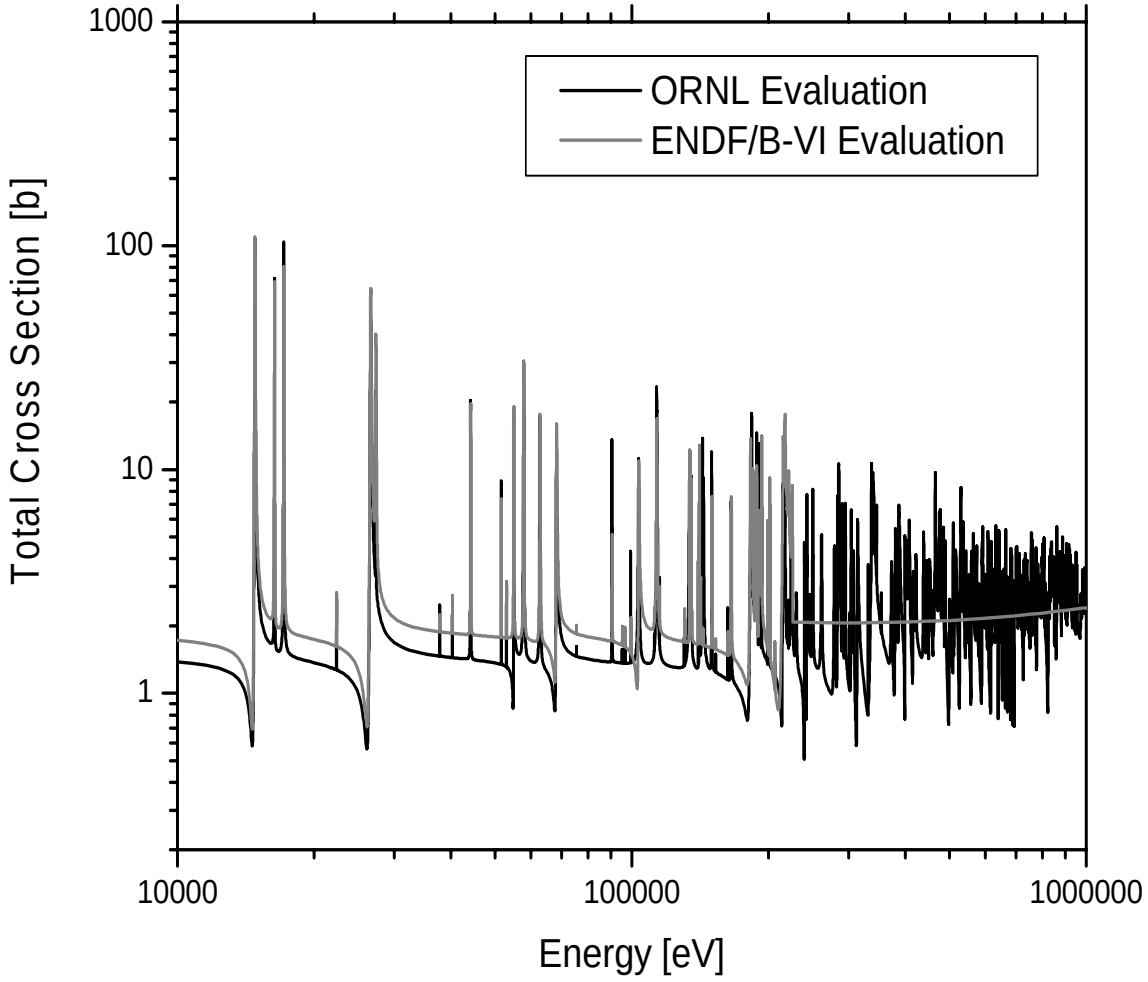

FIGURE 2: Evaluations of ^{35}Cl from ENDF/B-VI (grey curve) compared with the ORNL evaluation (black curve).