

Neutron Cross-Section Measurements at ORELA for Improved Nuclear Data and their Application

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ABSTRACT

To support the Nuclear Criticality Safety Program, the Oak Ridge Electron Linear Accelerator (ORELA) has been used to measure the total and capture neutron cross sections of several nuclides in the energy range from 100 eV to ~600 keV. Concerns about the use of existing cross-section data in nuclear criticality calculations have been a prime motivator for the new cross-section measurements. Our new capture cross sections of aluminum, silicon, chlorine, fluorine, and potassium in the energy range 100 eV to 600 keV are substantially different from the cross sections in evaluated nuclear data files of ENDF/B-VI and JENDL-3.2.

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INTRODUCTION

Concerns about the use of existing cross-section data in the nuclear criticality calculations using Monte Carlo codes and benchmarks have been a prime motivator for the new cross-section measurements. Most of the older neutron-induced cross-section data used in libraries such as ENDF/B-VI or JENDL-3.2 show deficiencies or do not cover the neutron energy range which is important for a wide variety of applications such as systems that include Al, Si, Cl, F, K, and ^{235}U . For example, many of the older measurements suffer from poor time-of-flight (TOF) resolution. Moreover, the description of some data in the neutron energy range above several tens of keV is crude. As a result, these data may not be sufficient for applying certain corrections, such as self-shielding, multiple scattering, or Doppler broadening of individual resonances. This impacts not only the resolved resonance region but also the unresolved region and could lead to problems in the correct processing of data from data libraries.

The experiments were performed using the Oak Ridge Electron Linear Accelerator (ORELA) to measure the total and capture neutron cross sections in the energy range from 100 eV to ~600 keV. ORELA, the only high-power white neutron source with excellent time resolution that is still operating in the United States, is ideally suited for these experiments. In

many cases, the capture cross sections found in these experiments are significantly smaller than those in most recent evaluations.

MEASUREMENTS

For the neutron capture measurements, we used rectangular samples (2.54×5.08 cm) of high-purity aluminum (0.01520 a/b and 0.04573 a/b), a high-purity natural silicon sample (0.07831 a/b), a natural LiCl sample (0.09812 a/b), a Teflon sample (0.05086 a/b) for fluorine, and a natural K_2CO_3 sample (0.0088791 a/b). The capture samples and C_6D_6 gamma-ray detectors were situated at a distance of 40.12 m from the neutron production target on FP-7 at ORELA. This capture system⁽¹⁾ has been reengineered to minimize structural material surrounding the sample and detectors in order to reduce the background due to sample-scattered neutrons (neutron sensitivity). In addition, the less-neutron-sensitive C_6D_6 scintillators replaced the previously used C_6F_6 scintillators and the computer code EGS4⁽²⁾ was used to calculate the detector weighting function. A 0.5-mm-thick 6Li glass scintillator served as the neutron flux monitor. Pulse-height weighting was employed with the C_6D_6 detectors, and normalization of the capture efficiency was carried out in a separate measurement using the “black resonance” technique⁽³⁾ by means of the 4.9-eV resonance from a 0.00508-cm-thick gold sample.

To apply all sample-related corrections in an analysis and evaluation of the neutron capture cross-section, total cross section data are indispensable. Therefore, we made corresponding transmission measurements when needed. For the Al transmission measurements, we used two high-purity samples (0.0189 a/b and 0.1513 a/b), which were mounted in the sample changer positioned at about 10 m from the neutron production target in the beam of ORELA. The chlorine transmission measurement was performed using a natural

CCl₄ (thickness for Cl 0.2075 a/b) sample. For the potassium transmission, we mounted two metallic samples (0.013367 a/b and 0.10517 a/b) in a sealed brass holder in the sample changer at 10 m. A presample 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. As a neutron detector, we used an 11.1-cm-diameter, 1.25-cm-thick ⁶Li-glass scintillator, which was 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. During the experiment the samples were cycled periodically through the neutron beam. Additional measurements with corresponding “compensators” in the open beam and with a thick polyethylene sample were used to determine the γ -ray background from the neutron source.

RESULTS

Comparisons of our recent results for Al, Cl, F, K, and Si (Figure 1) with the evaluated nuclear data obtained from the ENDF/B-VI or JENDL-3.2 nuclear data libraries reveals significant differences in the neutron capture cross sections. In many cases the capture widths were severely overestimated or resonances were missed due to large backgrounds.

These discrepancies arose for primarily two reasons; firstly, 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. For all structural materials within 30 cm of the detectors, including the sample to be investigated, the electron and γ -ray interaction are to be included in the input of the EGS4 code.⁽⁴⁾ 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. Secondly, the underestimated neutron sensitivity of the experimental setup led to an overestimation of the capture cross-section. In order to reduce the neutron sensitivity, the experimental setup was redesigned (i.e., the surrounding structural material was reduced by removing the massive Al-sample changer and replacing the beam pipe with a thin carbon fiber tube). Also, the massive detector housings were removed and the two C_6F_6 scintillation detectors were replaced by less-neutron-sensitive C_6D_6 detectors with reduced-mass detector mounts. More details about these improvements can be found in the papers by Koehler et al.^(5, 6), 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.

The neutron total and capture cross-section data were analyzed with the computer code SAMMY,⁽⁷⁾ applying all the corrections for the experimental effects. The resonance parameters obtained are the basis for an evaluation of the cross section performed by the ORNL Nuclear Data Group. 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 two nuclear data libraries ENDF/B-VI and JENDL-3.2.⁽⁸⁾

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 an overall small capture cross-section are

questionable, or at least much more uncertain, especially if the applied corrections for neutron sensitivity were sizeable.

ACKNOWLEDGEMENTS

ORNL is managed by UT-Battelle, LLC, for the U.S. Department of Energy under contract no. DE-AC05-00OR22725. The work that is presented in this paper was sponsored by the U.S. Department of Energy (DOE) Nuclear Criticality Safety Program.

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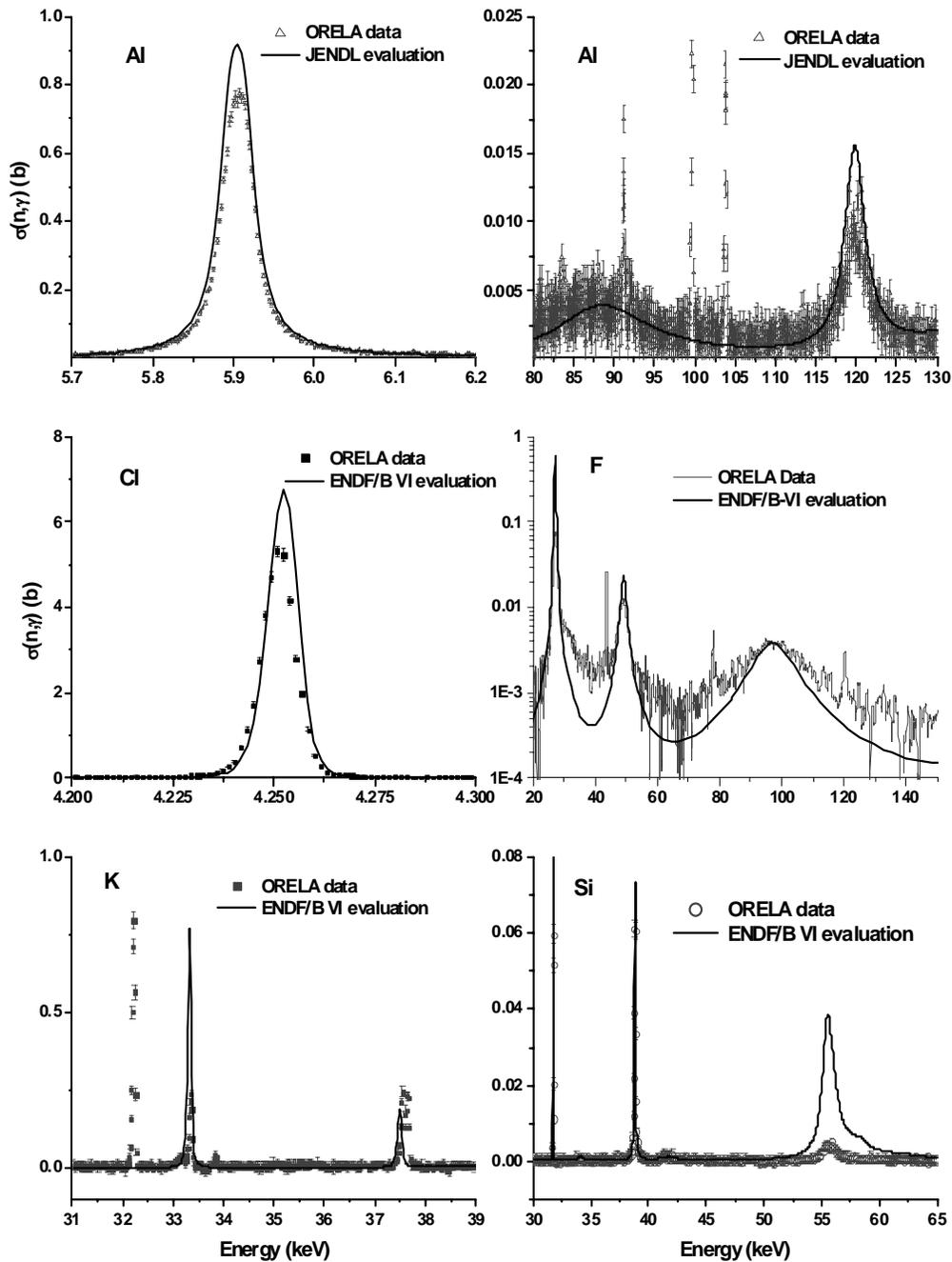


Fig. 1. Selected examples of the measured neutron capture cross sections at ORELA for Al, Cl, F, K, and Si compared with the most recent evaluations from JENDL and ENDF/B VI. Due to the underestimated neutron sensitivity in the older measurements, the cross sections were seriously overestimated.