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New neutron-induced cross-section measurements for improved nuclear data

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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 nuclear reactor and criticality safety applications. These deficiencies may occur in the resonance region and high energy region. Consequently, these evaluated data may not be adequate for radiation transport calculations where effects such as self-shielding, multiple scattering, or Doppler broadening are important. To support the U.S. Nuclear Criticality Safety Program, a series of new measurements has been undertaken in response to deficiencies identified in nuclear data libraries. New data and evaluations including covariances are required for several materials found in mixtures with uranium. For this purpose neutron capture and total cross section measurements have been performed for ^{58,60}Ni, natural chromium, and ^{53}Cr.

1. Introduction

High-quality nuclear data are essential for the design and analysis nuclear systems, such as reactor cores layout, fuel elements, and stored mixtures of nuclear waste with other materials, as well as burned fuel elements. Relevant nuclear data are also necessary for waste transmutation, accelerator-driven systems, and GEN-IV reactor design. Analysis codes for nuclear systems rely on the use of evaluated cross-section data from libraries such as ENDF/B-VII, JEFF3.1, or JENDL-3.3. Concerns about data deficiencies in some existing cross-section evaluations for nuclear criticality calculations have been the prime motivator for new cross-section measurements at the Oak Ridge Electron Linear Accelerator (ORELA). Many older neutron cross-section evaluations show signs of 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, some evaluated data may not be adequate for nuclear criticality calculations where effects such as self-shielding, multiple scattering, and Doppler broadening are important. Furthermore, many evaluations for nuclides in the nuclear data libraries are missing covariance data, which are increasingly required by the more sophisticated codes to analyze and simulate nuclear systems.

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Covariance data for a cross-section data set in the libraries can only be created by a reevaluation of the data, and sometimes this is impossible since the data used in the analysis and evaluations are no longer available. This shortfall can be overcome only by new experimental data included in a new evaluation.

New neutron capture cross-section measurements for nuclides with small neutron capture cross sections have revealed that these data are erroneously large because the neutron sensitivity of old measurements was underestimated. Although their neutron capture cross sections are small, these nuclides can be important absorbers in many criticality safety calculations, and consequently 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, total, and capture cross sections in the energy range from 1 eV to ~600 keV, which is the important range for nuclear criticality safety applications.

2. Cross section measurements at ORELA

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 variable repetition rate (1 to 1000 Hz) and burst width (2 to 30 ns). At maximum power, the average neutron flux is 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 layout of the ORELA facility is shown in Fig. 1. 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 total, capture, fission, elastic, and neutron production cross sections, as well as neutron-induced charge particle reactions.

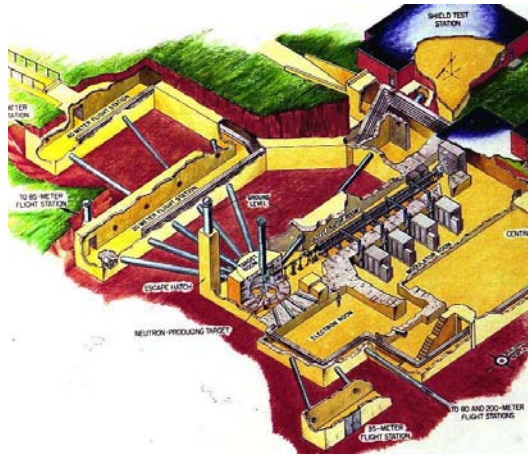

Fig. 1. Layout of ORELA.

During the last decade, ORELA operation was unreliable due to accelerator vacuum problems and electron-gun problems. Therefore ORELA has recently undergone some refurbishments in order to establish more reliable operation. The vacuum level for steady operation was successfully reestablished after two leaks were identified and sealed and the guard vacuum system overhauled. By redesigning the electron gun, a better shielding of the ceramic insulating shell against high-voltage arcs was achieved. As a result there have been no electron-gun failures due to this cause since the redesign. Since fall 2006, about 2000 hours of beam time for experiments has been run at ORELA.

2.1. Capture measurements

Neutron capture experiments for ^{58,60}Ni were performed at the 40-m flight station using flight path 7 (Figs. 2 and 3). A recently constructed capture measurement apparatus at flight path 6 in the 40 m flight station was used for the natural Cr(n,γ) experiment (Fig. 4). In both cases the pulse-height-weighting method, using a pair of deuterated benzene (C_6D_6) detectors, was employed.

The experimental apparatus at flight path 7 has been improved in several ways compared to the old ORELA setup (Macklin and Allen, 1971). The amount of structural material surrounding the sample and detectors was minimized in order to reduce the background due to sample-scattered neutrons (neutron sensitivity). The old apparatus was improved 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. 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. (1996, 2000) in which the impact of the much reduced neutron sensitivity was demonstrated in a high-resolution TOF measurement for ^{88}Sr. For two prominent

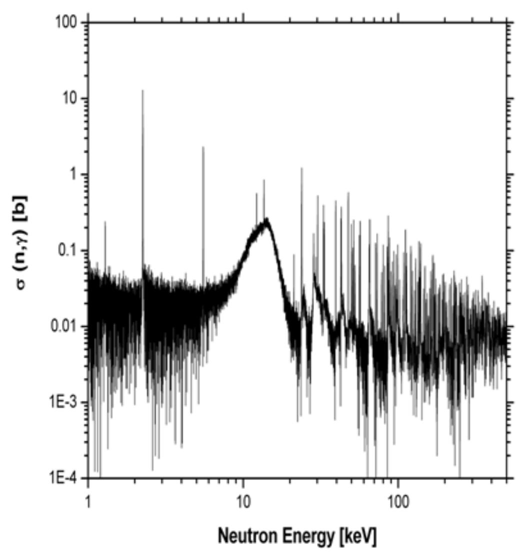

Fig. 2. Neutron capture cross section for ^{60}Ni, measured using the apparatus on flight path 7.

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. In addition to these improvements, the old data acquisition system was replaced, and the more sophisticated computer code EGS4 was used to calculate the pulse-height weighting functions for each experiment. All structural materials within 30 cm of the detectors, including the sample, were 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 Bayesian least-squares fitting code.

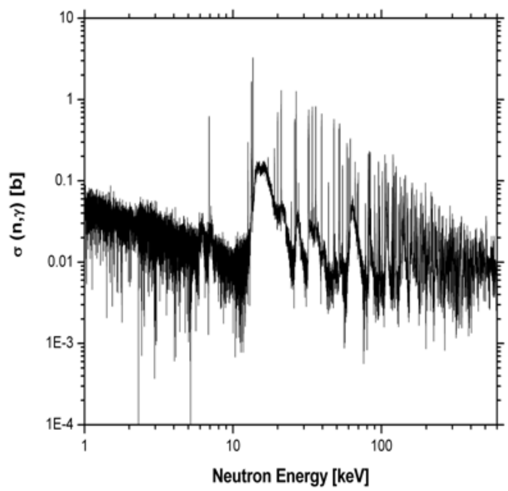

Fig. 3 . Neutron capture cross section for ^{58}Ni, measured on flight path 7.

This knowledge acquired over the last years was used for the construction of the capture apparatus on flight path 6 in the 40 m flight stations. This capture system was built with emphasis on reducing mass around the sample and detectors. Instead of the Monte Carlo code EGS4, the code MNCP is now used for weighting function calculations for both flight path 6 and 7.

Neutron capture cross-section experiments for nickel isotopes were performed using enriched metallic samples. For ^{58}Ni, the sample was a 2.54-cm by 5.08-cm disk with an enrichment of 99.9% and a thickness of 0.03635 atom/barn. The 99.62% enriched ^{60}Ni sample had the same dimensions except for a thickness of 0.03598 atom/barn. The samples were placed at a distance of 40.12 m from the neutron target, and gamma rays following neutron capture were detected by two C_6D_6-detetors. For the natural chromium experiment, a 2.54-cm-diameter disk was used in flight path 6 with a thickness of 0.64cm. The chromium disk was placed at a distance of 38.412m from the neutron target in the beam. For ^{53}Cr, an enriched sample of chromium oxide was used on flight path 7. It was combined by two 2.54- by 2.54-cm disks with a thickness of 0.0122 atom/barn.

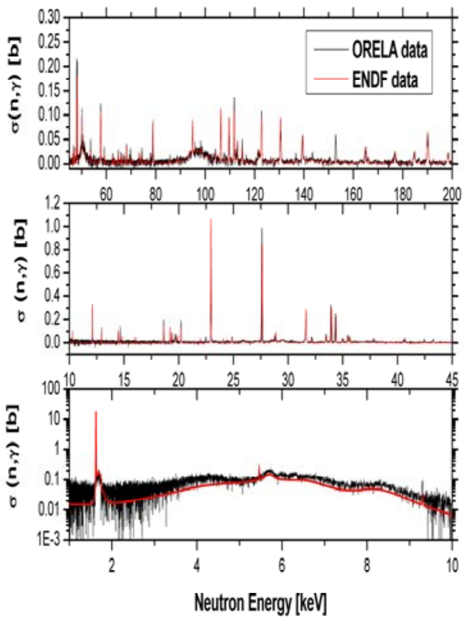

Fig. 4. Neutron capture for natural chromium, measured on flight path 6 compared to the ENDF evaluation.

To reduce the γ-ray background from the neutron production target, the neutron capture experiments were run with a 1.27-cm-thick lead filter in the beam. A 0.48-g/cm^2 ^{10}B filter served as overlap filter by absorbing the low-energy neutrons from the pulse. 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 (Macklin, 1979). A 0.5-mm-thick ^6Li-glass scintillator placed in the beam approximately 1 m in front of the capture sample was used to monitor the neutron flux.

2.2. 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 corrections for 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.

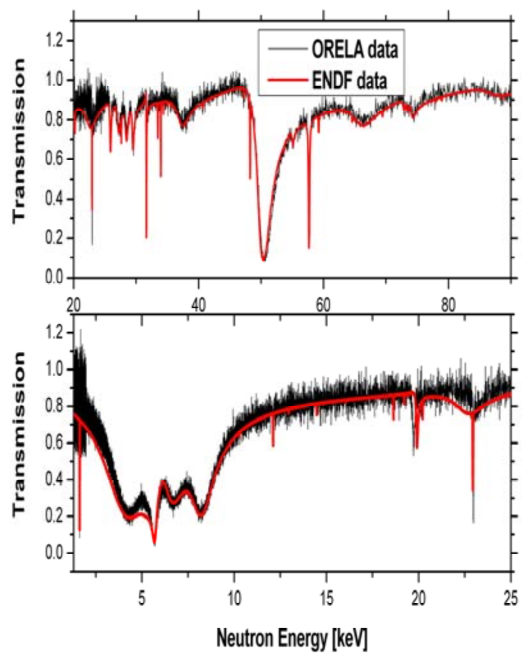

Fig. 5. Transmission of natural Cr in the energy range from 1 to100 keV. SAMMY was used to calculate the transmission using the ENDF/B-VII resonance parameters.

Since high-resolution transmission data for the nickel isotopes are available from previous ORELA experiments (Perey, 1983, 1993), we made transmission measurements only for chromium in the low neutron energy range where no previously high-quality data are available (Fig. 5). For the natural chromium transmission experiment, we used two metallic disks which were 0.0531 and 0.02627 atom/barn thick. These samples were mounted on a computer-controlled sample changer at a distance of 10 m from the neutron target. A pre-sample collimation limited the beam size to about a 2.54-cm diameter 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 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. To determine the overlap from low-energy neutron with the subsequent pulse, low-repetition-rate runs with a cadmium filter were made. Measurements with a thick bismuth sample were used for background checks.

3. Results

The results of our new capture and transmission experiments show differences compared to the evaluated nuclear data for ^{58}Ni, ^{60}Ni, and natural chromium obtained from the ENDF/B-VII or JENDL-3.3 nuclear data libraries. In Fig. 4 we show the neutron capture cross section of natural chromium in the neutron energy range from 1 to 200 keV compared to the cross section calculated using the most recent resonance parameter set from the ENDF/B-VII library. SAMMY (Larson, 2006) was used to calculate the neutron capture cross section including all experimental effects. We were able to resolve resonance up to 500 keV with sufficient statistics. Together with the high-resolution transmission data (compare to Fig. 5), a good data set is now available for a new evaluation. The neutron capture cross section for the nickel isotopes 58 and 60 are shown in Figs. 2 and 3. In these data sets, resonances can be resolved up to 600 keV with sufficient statistics.

In general, all of our new and recent 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. Results from our new total cross-section measurements will help in the analysis of our new capture cross-section measurements.

The observed discrepancies between our data and evaluations from 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 MCNP was used for the correct determination of the weighting function. Secondly, underestimated neutron sensitivity of the experimental apparatus led to previous capture cross sections that were too large. In addition, the better characterized samples, superior TOF resolution, and well-understood experimental apparatus and backgrounds helped to produce more reliable cross-section data in the present case.

The neutron total and capture cross-section data will be analyzed using the computer code SAMMY. This analyzing program applies all of the necessary corrections for 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 are available and suitable, this evaluation will include other existing experimental data sets. The final result will then be checked for consistencies using criticality benchmark calculations.

4. Conclusion

To support the Nuclear Criticality Safety Program, we performed new neutron total and capture measurements at ORELA over broad energy ranges. The obtained results will be analyzed using the multilevel R-matrix code SAMMY. With these new high-resolution data it should be possible to extend the resolved resonance region to much higher energies than the existing evaluations and improve the cross sections in the resolved and unresolved neutron energy range. 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 for Al, Si, ^{88}Sr, F, Cl, K, ^{41}K, Cr, and ^{58,60}Ni at ORELA have shown the tendency to be smaller than the data found in the nuclear data libraries, such as ENDF/B-VII, JEFF3.1, or JENDL-3.3. These are all nuclides with rather small capture cross sections and mostly scatter neutrons. 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.

5. Acknowledgement

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