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Neutron Cross-Section Measurements on Structural Materials at ORELA

K. H. Guber,* P. E. Koehler, D. Wiarda and J. A. Harvey
Oak Ridge National Laboratory, Oak Ridge, TN 37831-6356, USA

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Neutron capture experiments, using isotopically enriched and natural samples of chromium and titanium, were performed on flight paths 6 and 7 at the 40 m flight station of ORELA. The experimental data were acquired using a pair of deuterated benzene detectors employing the now well-established pulse-height-weighting technique. These data were complemented by new total cross-section measurements where no useful previous data were available.

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I. INTRODUCTION

Most analysis and simulation codes rely on evaluated cross-section data from libraries such as ENDF/B-VII, JEFF3.1, or JENDL-3.3. These codes require high-quality nuclear data for the design of nuclear systems. Similar nuclear data are also needed for nuclear astrophysics. 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. For example, 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 was 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. 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 because the data used in the analysis and evaluations are no longer available. In such cases, this shortfall can be overcome only by new experimental data included in a new evaluation.

*E-mail: guberkh@ornl.gov

II. EXPERIMENTS

ORELA, a high-power white neutron source with excellent time resolution in the keV neutron energy range, was used to perform the reported neutron cross-section measurements. It consists of a 180 MeV electron linear accelerator; a neutron-producing, water-cooled Ta target; underground and evacuated flight tubes; sophisticated detectors; and data acquisition systems. ORELA is a highly flexible accelerator with a varying repetition rate between 1 and 1000 Hz and a neutron burst width between 2 and 30 ns. Average neutron production of up to 10^{14} neutrons per second is obtainable. 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 data in the energy range of a few eV up to 50 MeV, such as total, capture, fission, elastic, and neutron production cross sections.

1. Neutron Capture Experiments

The neutron capture experiments were performed at the 40 m flight station of ORELA using flight paths 6 and 7. The employed experimental technique was the pulse-height-weighting method using, for each flight path, a pair of deuterated benzene (C_6D_6) detectors.

Compared to the old ORELA setup [1], the capture system at flight path 7 has been improved in several ways. First, the amount of structural material surrounding the sample and detectors was minimized to reduce the background due to sample-scattered neutrons (neutron sensitivity). 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. Second,

the more neutron-sensitive $C_6F_6\gamma$-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.* [2,3], in which the impact of the much reduced the neutron sensitivity was impressively demonstrated in a high-resolution TOF measurement for ^{88}Sr. Finally, more accurate pulse-height weighting functions were calculated using the computer code MCNP. 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 least-squares fitting code.

The neutron capture apparatus on flight path 6 incorporates these same improvements. With this setup we measured the neutron capture for natural Cr by means of a 2.54 cm diameter disk that was 0.0188 atom/b thick. The ^{53}Cr(n,γ) cross section was measured at flight path 7 using a 2.54 $\times$ 5.08 cm isotopically enriched sample with 0.0137 atom/b thickness. For the natural Ti sample, several 2.54 $\times$ 5.08 cm rectangles were combined to form a sample that was fully illuminated by the neutron beam at flight path 7. In this way a sample thickness of 0.03519 atom/b was achieved for the natural Ti. The enriched ^{48}Ti sample was made out of titanium oxide placed in an Al can to prevent the sample form absorbing moisture. From the dimensions a thickness of 0.009138 atom/b was calculated. To normalize the obtained neutron capture cross sections, the saturated resonance method using ^{197}Au was applied [4].

2. Transmission Experiments

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 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, corresponding total cross-section measurements were made when needed. In addition, some resonances with small radiation widths are not visible in the neutron capture data and vice versa. A presample collimation limited the beam size to a diameter of 2.54 cm at 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 periodically cycled 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 γ-ray background from the neutron source.

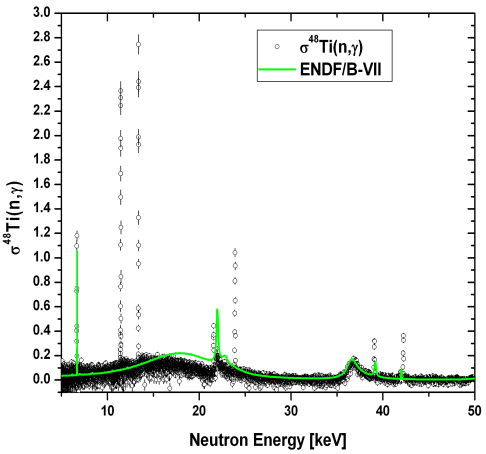

Fig. 1. (Color online) Neutron capture on ^{48}Ti oxide compared to ENDF/B-VII evaluation parameters.

New transmission experiments were conducted because good-quality data were not always available. For the Cr isotopes, high-resolution data were available, but with low neutron energy cut off at about 40 keV. So, new transmission experiments were necessary to cover the whole resolved resonance range. These experiments were performed with natural Cr thicknesses of 0.05310 and 0.0262696 atom/b. For samples having ^{53}Cr, we used a 0.017984 atom/b sample of enriched chromium oxide. The natural Ti and ^{48}Ti oxide had thicknesses of 0.05297 and 0.028186 atom/b, respectively.

III. RESULTS

The results of our capture and transmission experiments show conflicting differences compared to the evaluated nuclear data files for all isotopes measured. These differences can be seen in Figs. 1 through 5, in which we show examples of the transmission and neutron capture cross-section data of natural Ti, Cr, ^{53}Cr, and ^{48}Ti. These data are compared to the cross sections or transmission calculated using the most recent resonance parameter set from the ENDF/B-VII library. SAMMY [5] was used to calculate the neutron capture cross section and transmission, including all experimental effects. In most cases, resonances up to 600 keV were resolved 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

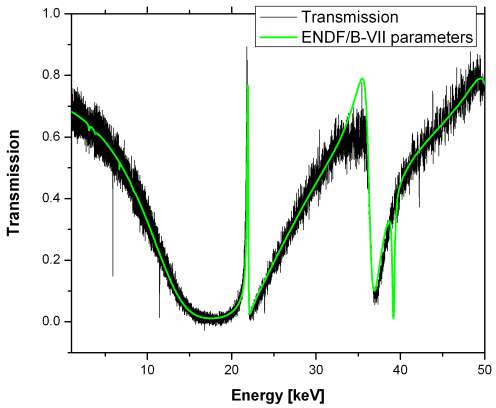

Fig. 2. (Color online) Transmission data for ^{48}Ti oxide compared to parameters from the ENDF/B-VII evaluations.

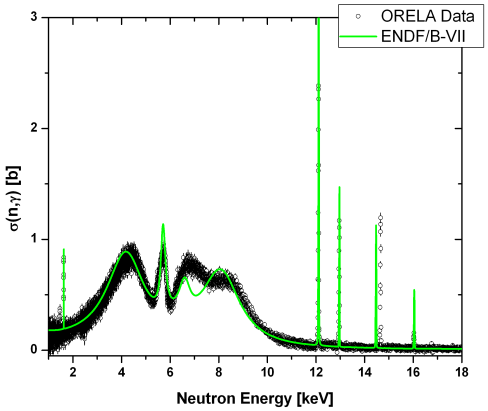

Fig. 4. (Color online) Neutron capture for ^{53}Cr oxide compared to ENDF/B-VII evaluation parameters.

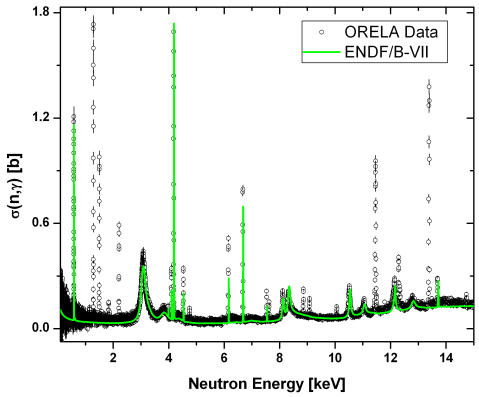

Fig. 3. (Color online) Neutron capture on natural Ti compared to ENDF/B-VII evaluation parameters.

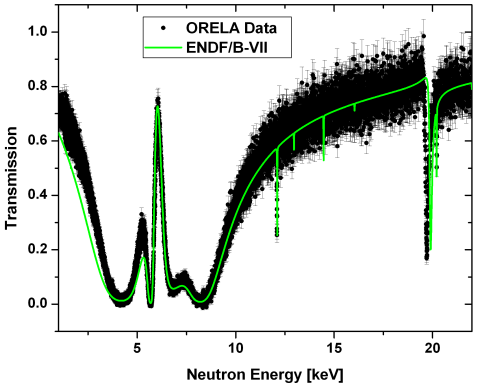

Fig. 5. (Color online) Transmission for ^{53}Cr oxide compared to ENDF/B-VII evaluation parameters.

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 the older experimental data and evaluations from nuclear data libraries mainly had two causes. 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. Second, underestimated neutron sensitivity of the experimental apparatus led to previous capture cross sections that sometimes were too large for resonances having very large neutron widths. In addition, 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.

New neutron total and capture cross-section evaluations will be made using the computer code SAMMY. This R-matrix analysis program applies all the necessary corrections for experimental effects, such as Doppler and resolution broadening, self-shielding, and multiple scattering to the data. Resonance parameters obtained from the SAMMY fit to the experimental data will serve as the basis for new cross-section evaluations. When these parameters are available and suitable, evaluations will include other existing experimental data sets. The result of the Cr cross section evaluation will be presented in a contribution at this conference [6]. The final result will then be checked for consistencies using criticality benchmark calculations.

IV. CONCLUSION

To support the U.S. Nuclear Criticality Safety Program, new neutron total and capture measurements at ORELA were performed over broad energy ranges. The new Ti and Cr neutron cross-section measurements complement a series of previous ORELA experiments for structural materials such as Mn and Ni. The newly ob-

tained neutron cross-section data for the Cr and Ti isotopes show serious discrepancies compared to the most recent evaluations from JENDL and ENDF, and the resolved resonance regions could be extended to much higher neutron energies. The data, including all available experimental data, will be analyzed with the multilevel R-matrix code SAMMY (see for example Ref. 6). With these new data sets it will be possible to produce new, more accurate evaluations and consequently improve the nuclear criticality safety calculations.

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