ORNL Neutron Cross Section Measurements of ^{90}Zr

Klaus Guber, * Jesse Brown, * Carlos Paradela†, Stefan Kopecky†, Jan Heyse†, Peter Schillebeeckx†

**Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA, guberkh@ornl.gov*

†Nuclear Physics Unit, EC-JRC Geel, Retieseweg 111, 2440 Geel, Belgium

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INTRODUCTION

Nuclear criticality modeling and simulations rely on the quality of the existing evaluated nuclear data libraries such as Evaluated Nuclear Data File (ENDF)/B, the Joint Evaluated Fission and Fusion (JEFF) nuclear data library, or the Japanese Evaluated Nuclear Data Library (JENDL). In some cases, the cross-section evaluations of those libraries were found to be deficient in describing criticality benchmarks accurately. More than two decades ago, the US Nuclear Criticality Safety Program (NCSP) established a Nuclear Data (ND) task which encompassed experiments and evaluations. In response to this, the Oak Ridge National Laboratory (ORNL) formed a Nuclear Criticality and Data group which performed ND experiments, data analysis, and evaluations to produce ENDF files for the ND libraries as identified in the NCSP Five-Year Plan. Before being submitted to the ENDF library, files were processed and tested for performance by running benchmark calculations. This procedure was centralized in the ORNL group and is now often referred to as the *ND pipeline*.

NCSP collaborates with the Joint Research Center (JRC) of the European Commission in Geel, Belgium, to perform high-resolution neutron-induced cross section measurements at the Geel Linear Accelerator (GELINA). The objective is to address emerging ND problems in criticality calculations. Difficulties with ND include insufficient neutron energy range, missing covariances, and previously unrecognized inaccuracies with experiments.

New neutron total and capture cross sections of ^{90}Zr in the neutron energy range from 100 eV to several hundred keV were recently performed. These measured data will be used, together with existing high-resolution transmission data from a metallic ^{90}Zr sample, to improve representation of the cross sections.

EXPERIMENTS

The neutron time-of-flight (TOF) experiments were performed at the GELINA facility [1]. Currently, GELINA is the only remaining TOF facility with excellent neutron energy resolution over a broad range, which is essential for measuring the cross sections in the resolved resonance range (RRR). The GELINA accelerates electrons to a maximum energy of 150 MeV, and with the help of a bunching magnet, the electron pulse is compressed to a 2 ns pulse. This pulsed electron beam is stopped by a rotating depleted uranium target, and the induced Bremsstrahlung radiation produces neutrons via photonuclear and photofission reactions. Two beryllium canned water containers mounted above and below the mercury-cooled uranium target aid as neutron moderators. The combination of the long flight path (up to 400 m) and the short burst result in a TOF resolution which is unsurpassed. This enables high-resolution experiments in the RRR that are essential for obtaining good average resonance parameters.

The experiments were performed using a metallic disk of enriched ^{90}Zr that was fabricated by the ORNL Isotopes Division (97.65% ^{90}Zr, 0.93% ^{91}Zr, 0.7% ^{92}Zr, 0.56% ^{94}Zr, and 0.16% ^{96}Zr). The disk's diameter was 5.08 cm, its measured average thickness was 0.124 cm, and it weighed 15.398 g. During fabrication, the disk became warped and slightly non-uniform. However, a sample having the least number of imperfections possible is desirable for good transmission experiments. Because good transmission data [2] from previous Oak Ridge Electron Linear Accelerator (ORELA) experiments are available, it was decided to accept the disk for capture experiments, which was the main goal of this effort.

Transmission Experiments

Neutron capture cross section experiments are typically performed using a rather thick sample, so corrections were applied by the data analysis programs. The corrections for self-shielding and multiple scattering can be quite sizeable, requiring reliable neutron widths as input to the data analysis programs. Without this information, erroneous capture cross section data can result, as shown by Koehler et al. [3]. Neutron transmission experiments were performed using the ^{90}Zr sample placed along Flight Path 4 with the neutron detector located 47.46 m from the neutron production target. Even though the sample used was not very uniform, the transmission data that were obtained can be used to test the correction applied by the analysis program. Because good transmission data are available from previous ORELA experiments using 80 m and 200 m flight paths, reliable neutron width data are available from those experiments for comparison with the new data.

To determine the time-dependent and -independent backgrounds for the transmission experiments, the so-called black resonance filter technique [4] was used. This involved inserting material (e.g., W, Co, Na and S) into the neutron beam, which is thick enough to remove all neutrons at certain neutron energies from the beam. Energy points are fitted to define the shape of the background. Typical uncorrected transmission spectra for the sample, open beam, and corresponding backgrounds are shown in Fig.1.

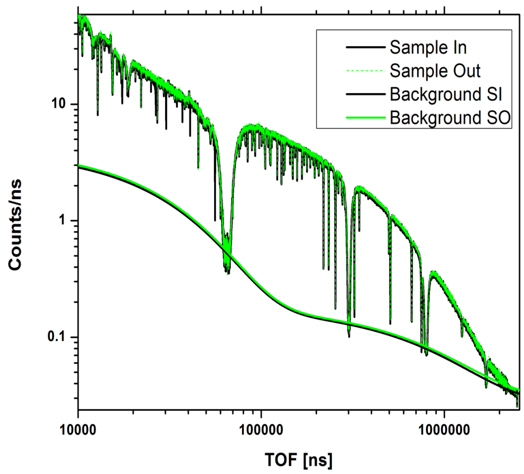

Fig. 1. Background determination using the back resonance technique for the ^{90}Zr transmission experiment

A ^{6}Li-glass detector with a 0.635 cm thickness was used to detect the sample transmitted neutrons. Mounting the photomultiplier on the bottom of the scintillation glass and a pre-sample collimation ensured that neutrons only hit the ^{6}Li-glass scintillator. The samples and their corresponding empty containers were cycled in dependence of the accelerator pick-up signal through the neutron beam. This helped reduce systematic uncertainties. As normalization, the total neutron count in the target hall for each sample and cycle was recorded.

The acquired list mode data for transmission were sorted into TOF spectra which were processed by the Analysis of Geel Spectra (AGS) software [5], which transforms recorded count rates into observables such as transmission. Using resonance parameters from the ENDF/B-VIII library, the theoretical transmission for ^{90}Zr sample was computed using the data analysis program SAMMY [6], including all experimental effects. The same computation was performed for the old ORELA 80 and 200 m flight path data, including the details of the ORELA experiments. This was possible because the authors had access to the experiment logbooks.

The transmission spectra for ^{90}Zr from the GELINA and ORELA experiments compared to the computed theoretical transmissions are shown in Figs. 2 and 3.

Neutron Capture Experiments

The neutron capture experiments for ^{90}Zr were performed at the 60 m flight station on Flight Path 14 using four C_6D_6 detectors. The detectors were mounted at an angle of 125° to the neutron beam and registered the gamma rays after neutron capture in the sample. The pulse-height weighting method was applied to the recorded data using a weighting function. This function was determined by simulating the experimental set-up, including the sample using the Monte Carlo N-Particle (MCNP) code. The zirconium sample was mounted in the well-collimated beam on a low mass sample holder at a distance of 58.56 m from the neutron production target. In parallel, the neutron flux was recorded by a ^{10}B ionization chamber located in front of the sample. The obtained capture data were normalized by means of the well-known 1.15 keV resonance in ^{56}Fe using a natural Fe sample. An empty frame was used to measure the open beam, and by measuring a ^{208}Pb sample, the effect of neutrons scattered by ^{90}Zr and subsequently captured by the surroundings of the detector could be approximated. For resonances with large scattering-to-capture ratios, this effect can be non-negligible and may lead to false capture signals if capture in the surrounding occurs within the width of the resonance.

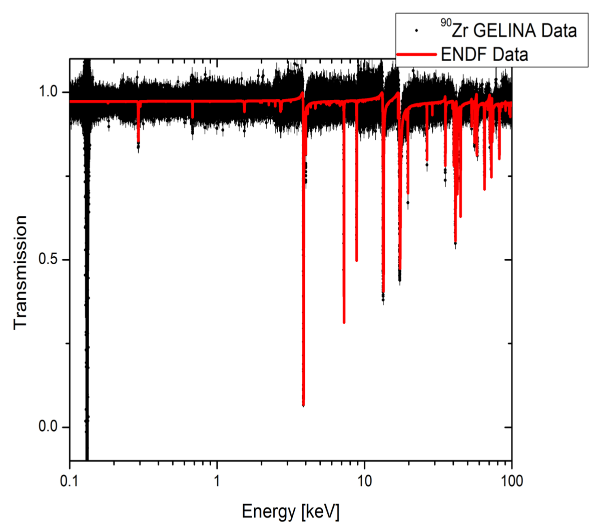

Fig. 2. ^{90}Zr transmission data from the GELINA experiment compared to the computed transmission from ENDF/B-VIII parameters.

The experiments were run with a set of black resonance filters to control the backgrounds. Because GELINA operated at 400 Hz, a boron slab in the beam served as a neutron overlap filter to take out all slow energy neutrons which would be interpreted by the data acquisition system as high-energy neutrons of the next neutron burst. The neutron capture data (TOF and pulse height) were stored in list mode on disk. The data were then weighted with the weighting function and sorted into the TOF spectra, which were processed by the Analysis of Geel Spectra (AGS) software code [4]. AGS applies the necessary corrections and transforms the recorded count rates into capture yield. The data reduction for the ^{90}Zr data is ongoing at this writing.

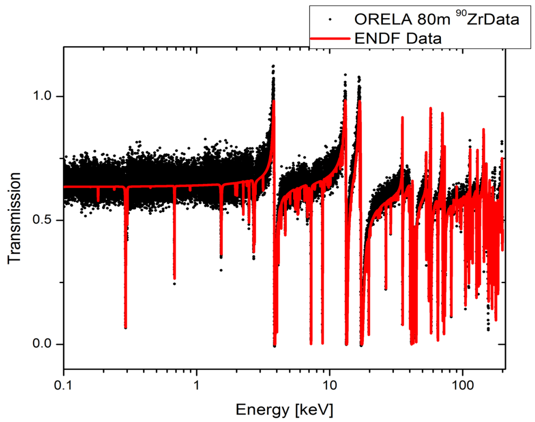

Fig. 3. ^{90}Zr transmission data from the 80 m ORELA experiment compared to the computed transmission from ENDF/B-VIII parameters.

RESULTS AND OUTLOOK

Several observations can be made from the comparison of the three transmission data sets with the computed transmission from SAMMY calculations using the ENDF/B-VIII resonance parameters. In the 200 m flight path data from ORELA, resonances are well visible above 1 MeV, which is a factor of three more compared to previous experiments. Above 140 keV, serious discrepancies between the ORELA data and the ENDF/B-VIII library resonance parameters are found compared to the data shown in Fig. 4.

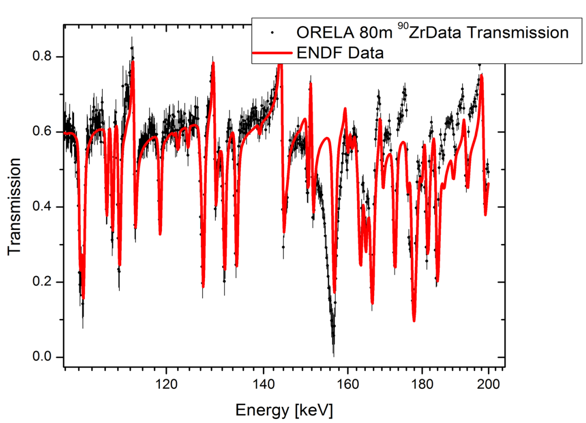

Fig. 4. ^{90}Zr transmission data above 100 keV from the ORELA experiment compared to the computed transmission from ENDF/b-VIII parameters. Serious differences are seen at high neutron energies.

The ORELA data were measured using a thicker sample (0.0845 at/b of Zr) compared to the new experiments that used a 0.00508 at/b sample for Zr. The thinner sample data combined with the better resolution of the GELINA data help to exclude the reported doublet in the ENDF library, as demonstrated in Fig. 5.

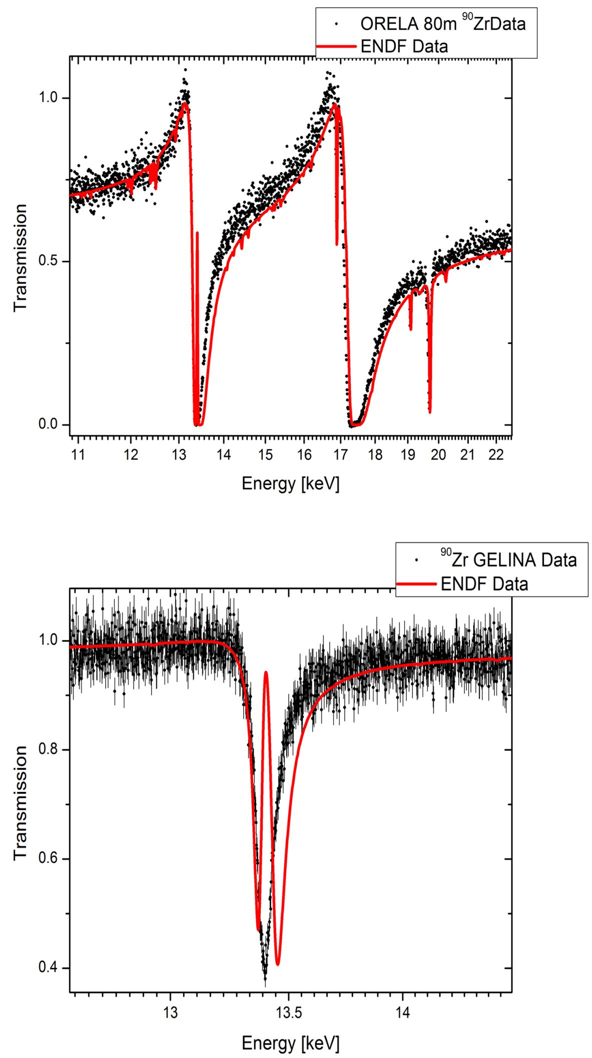

Fig. 5. ^{90}Zr transmission from the ORELA experiment (top) and GELINA data (bottom) compared to the transmission calculated from ENDF/B-VIII parameters. The experiments excluded the reported doublet.

With the high-resolution transmission data from ORELA and GELINA, the resolved resonance region for ^{90}Zr can be extended to a new analysis and evaluation. Furthermore, the transmission data obtained with thin and thick samples will serve as a guide for spin assignments of the resonances during the analysis. The previously obtained data for natural Zr will be a necessary addition to the existing data sets to allow for a new evaluation and to serve as a good sanity check for the data acquired with separated isotopes. These data will also eventually be supplemented by new neutron capture data of the remaining separated isotopes.

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