Zirconium Nuclear Data Campaign: Measurement of ^{90}Zr (n, γ) cross section

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Abstract. The isotopes of Zr with $A = [90, 91, 92, 94]$ make up more than 97% of naturally occurring Zr and are important to many nuclear applications such as nuclear reactors. One of the attractive qualities of naturally occurring Zr isotopes is that they have a low σ_γ/σ_t ratio at most neutron energies. Thus, they improve the neutron economy in reactors by preferentially scattering neutrons rather than absorbing them. This same quality also presents a challenge to measuring the capture cross section, σ_γ, of Zr isotopes. The ENDF/B-VIII.0 library has a relative uncertainty of approximately 10–20% for incident neutron energies < 0.1 MeV and an uncertainty greater than 20% for energies > 0.1 MeV for the majority of natural Zr isotopes. This motivated the Nuclear Criticality Safety Program to embark on a campaign to accurately measure and evaluate these Zr isotopes. In this work, we demonstrate energy-dependent neutron capture cross section measurements for the first enriched sample to be measured: ^{90}Zr.

1 Introduction

Isotopes of Zr are important to many applications—notably, reactors that contain cladding alloys with significant fractions of natural Zr and reactors that employ ZrH_x as moderators. In addition to these isotopes' importance for current nuclear applications, the capture cross sections of Zr are also important for fundamental science, such as modeling stellar nucleosynthesis [1, 2]. The natural composition of Zr includes five major isotopes: ^{90}Zr, ^{91}Zr,^{92}Zr, ^{94}Zr, and ^{96}Zr. The atom percent abundance is given in Table 1. Because of the distribution of isotopic abundance in naturally occurring Zr, measurements on natural samples are largely incapable of identifying which nuclear resonances belong to which isotope. To accurately model nuclear cross sections and associated uncertainties for isotopes of Zr, isotopically enriched samples must be fabricated, measured, and evaluated. A campaign of measurements of isotopically enriched Zr samples is being carried out at the Geel Electron Linear Accelerator (GELINA) facility of the Joint Research Center – Geel (JRC-Geel) of the European Union. Because isotopic enrichment is a costly process, long-term activation of any of the samples must be avoided, as it would hinder future radiation-sensitive measurements. An activation analysis based on the measurement being presented herein is described in a report by Brown et al. [3]. After the activation analysis verified that the Zr samples would not be activated by the proposed measurements, the ^{90}Zr sample was fabricated at Oak Ridge National Laboratory (ORNL) and shipped to GELINA. The GELINA measurements of the ^{90}Zr (n, γ) cross sections are presented in this document.

Table 1. The natural abundance of Zr (from Baum et al. [4]) is not dominated by any single isotope. The largest, ^{90}Zr, accounts for only half of natural Zr.

Isotope	Atom %
^{90}Zr	51.45
^{91}Zr	11.22
^{92}Zr	17.15
^{94}Zr	17.38
^{96}Zr	2.80

2 Current status for ^{90}Zr

The ^{90}Zr nucleus has a remarkably low neutron capture cross section because of a closed neutron shell (a magic number of neutrons is 50) and the fact that it is an even–even nucleus (even neutron and proton numbers). This makes it desirable from a neutron economic point of view (fewer neutrons are lost to capture), but it also makes the capture cross section difficult to measure, as counting statistics from the neutron capture reaction are typically poor. The most updated evaluations of ^{90}Zr come from the

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ENDF/B-VIII.0, JEFF-3.3, and JENDL-4.0 libraries. The capture cross sections for each of these libraries are very similar, though there are discrepancies between them regarding the neutron energy at which resonances become unresolved. The capture cross sections from each of these three libraries are plotted in Fig. 1, along with their associated uncertainties. The uncertainties have greater deviation when compared library to library. JEFF-3.3 places higher confidence in the cross section values at almost all energies compared to ENDF/B-VIII.0 and JENDL-4.0. ENDF/B-VIII.0 and JENDL-4.0 have almost identical variances reported in the evaluations. All three libraries report large uncertainty in the capture cross section, which motivates new measurements and evaluations that could reduce uncertainties.

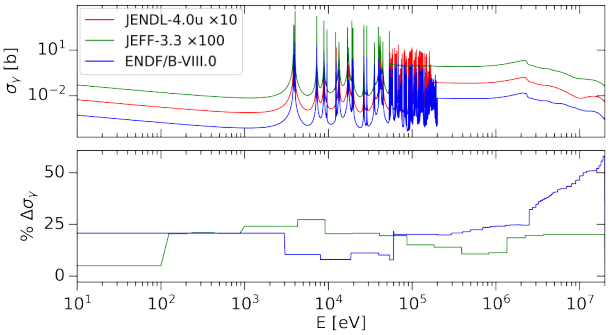

Figure 1. Major evaluation libraries report very similar capture cross sections, with differences in the energy regions of the resolved and unresolved resonance regions. The JEFF-3.3 library reports the lowest uncertainty in capture cross section.

High-accuracy, energy-dependent measurements of isotopically enriched samples of ^{90}Zr are relatively rare, though they do exist. Measurements containing data in the energy region of interest and available before this work were those of Tagliente et al. [1] and Bartolome et al. [5] for ^{90}Zr(n,γ) as well as de L. Musgrove et al. [6] for ^{90}Zr(n,tot). These measurements are discussed further in the following subsections.

2.1 Capture

The most recent high-resolution capture yield measurement was performed by Tagliente et al. at the nTOF facility. The measured sample consisted of isotopically enriched (97.7%) zirconium oxide 0.003531 at./b (1.3 mm) thick inside an Al can, placed along a flight path 185.2 m from the pulsed neutron source. A much older measurement is by Bartolome et al. at the RPI linear accelerator (Linac) with a 97.65% isotopically enriched, 0.0109 at./b thick, zirconium oxide sample in an Al can, placed along a flight path of 25.5 m from the pulsed neutron source. Both datasets are shown for energies 1-100 keV in Fig. 2. Unfortunately, the Bartolome measurement does not include any uncertainties on the data, and the energy alignment is contradictory to evaluated standards; the original, raw Bartolome data are unlikely to be recovered.

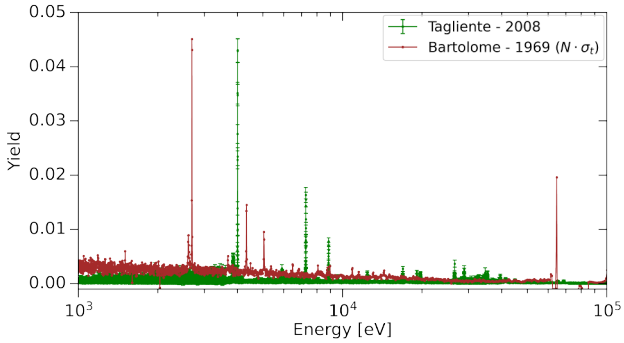

Figure 2. Two measurements met the criteria for new resonance analysis: those of Tagliente et al. and Barolome et al. The Bartolome data do not include any uncertainty information and have resonance energies discrepant with all related data and will likely not be used in future evaluations.

2.2 Transmission

The highest quality transmission measurements available were performed using the Oak Ridge Electron Linear Accelerator (ORELA) at ORNL by de L. Musgrove et al. There are two separate measurements at different flight paths of 80 m and 200 m with a metallic 0.0827 at./b thick Zr sample enriched to 97% ^{90}Zr. With the 80 m data, the count rate is increased at a loss of resolution, and at 200 m, the resolution is improved at the cost of count rate; the combination of the two measurements provides as much useful information as possible. These measurements are shown from 1 keV to 1 MeV in Fig. 3. A measurement by Good et al. [7] might prove useful to future evaluations, but with limited uncertainty information, resolution, and energy range.

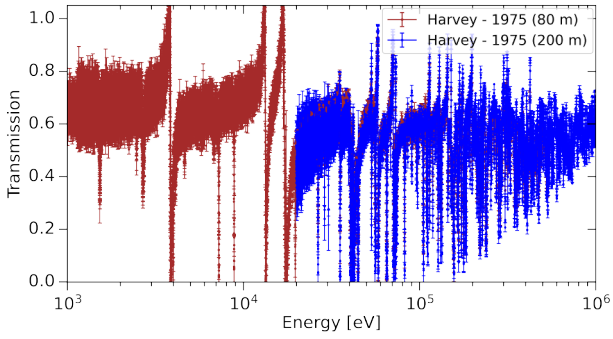

Figure 3. Transmission measurements by Harvey et al. (published by de L. Musgrove et al.) provide very high–resolution data with an extensive energy range and multiple experiments with different flight paths.

3 New measurements

Since ^{90}Zr has no appreciable chemical reactions to air, a bare metallic sample was chosen for the current measurement. Compared to zirconium oxide samples, the metallic

samples have a greater density of Zr nuclei, more consistent sample composition (e.g., pressed oxide powders risk having hidden voids), and avoid confounding effects from neutron interactions with oxygen. The cylindrical sample was 0.005086±0.04% (1.2 mm) thick, and had a radius of ~25 mm. The sample was measured at a flight path (FP) length of ~60 m, using four C_6D_6 detectors on FP14 of the GELINA facility. The associated transmission measurement used the same sample on FP4 at 47 m. During both measurements the beam was pulsed at a rate of 400 Hz and pulse widths were approximately 2 ns at full-width-half-max. Sample temperature was constant at approximately 293 K.

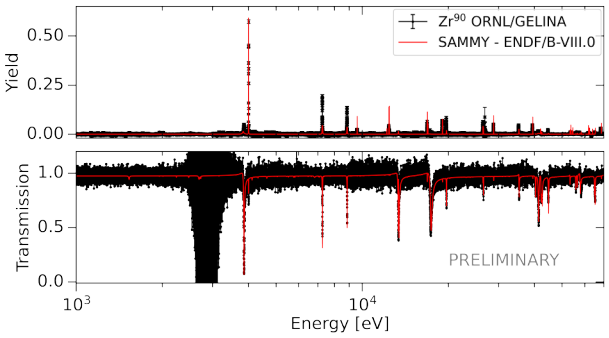

Figure 4. Preliminary analysis of ^{90}Zr capture yield measured at GELINA suggests that the new measurements will lead to lower uncertainties in evaluated cross sections. Note that these are preliminary mean values, and uncertainties and are subject to change. Plotted below the capture yield is the corresponding transmission measurement at GELINA of the same sample; this will help validate the capture analysis.

Preliminary analysis of the data, shown in Figure 4, suggests that the new measurement will provide high-resolution resonance information up to approximately 100 keV. The average statistical uncertainty of capture yield at resonances near 10 keV are about 6–7%, which could allow updates to resonance evaluations including these data to reduce the overall uncertainty in capture yield. With better resonance region data, it is also possible to validate quantum numbers of each resonance and potentially improve angular distributions of resonant scattering. The transmission measurement of the same sample at GELINA can be used to validate the capture analysis.

4 Conclusions

Isotopes of Zr and thorough evaluation of their cross sections are important to a wide range of nuclear applications. In this work, we present the first of four high-resolution time-of-flight measurements of isotopically enriched isotopes of Zr: that of ^{90}Zr. New measurements in this work will be combined with historical measurements and the planned measurements of ^{91}Zr, ^{92}Zr, and ^{94}Zr to produce a new comprehensive evaluation of Zr. Ultimately, this leads to increasing the accuracy of neutronics models and reducing the uncertainty in those models resulting from nuclear data.

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