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Zirconium Evaluations for ENDF/B-VII.2 for the Fast Region

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We have performed a new combined set of evaluations for $^{90-96}$Zr, including new resolved resonance parameterizations from Said Mughabghab for ^{90,91,92,94,96}Zr and fast region calculations made with EMPIRE-3.1. Because ^{90}Zr is a magic nucleus, ^{90}Zr is nearly spherical. Therefore, all even-even nuclei in the evaluation set were evaluated using EMPIRE's OPTMAN coupled channels code and a new soft-rotor optical model potential. This then led to improved (n,el) angular distributions, helping to resolve improper leakage in the older ENDF/B-VII.1β evaluation in KAPL proprietary, ZPR and TRIGA benchmarks. Another consequence of ^{90}Zr being a magic nucleus is that the level densities in both ^{90}Zr and ^{91}Zr are unusually low causing the (n,el) and (n,tot) cross sections to exhibit large fluctuations above the resolved resonance region. To accomodate these fluctuations, we performed a simultaneous constrained generalized least-square fit to (n,tot) for all isotopic and elemental Zr data in EXFOR, using EMPIRE's TOTRED scaling factor. TOTRED rescales reaction cross sections so that the optical model calculations are unaltered by the rescaling and the correct competition between channels is maintained. In this fit, all (n,tot) data in EXFOR was used for $E_{in} > 100$ keV, provided the target isotopic makeup could be correctly understood, including spectrum averaged data and data with broad energy resolution. As a result of our fitting procedure, we have full cross material and cross reaction covariance for all Zr isotopes and reactions.

I. INTRODUCTION

Because of its low (n,γ) cross section and corrosion resistance, zirconium is an important structural material and is used in many reactor applications:

- Nuclear fuel rod cladding
- Moderator (in form of ZrH)
- Inert material in fuel matrix

Despite zirconiums technological importance, its isotopes are notoriously difficult to evaluate. ^{90}Zr has a closed neutron shell so nearby nuclei have low level densities and the (n,el) and (n,tot) cross sections exhibit large fluctuations at relatively high energies. The evaluators of the ENDF/B-VI.8 evaluation remedied this by simply fitting experimental cross section data, but they ignored many of the outgoing particle distributions. Later, the ENDF/B-VII.0 evaluation was created using EMPIRE, providing the particle distributions, but ignoring the structure in (n,tot) and (n,el). In the ENDF/B-VII.1 evaluation, we attempted to capture the best of both evaluations by tuning EMPIRE calculations to data. Problems with (n,el) neutron angular distributions forced us to replace them with ones from JENDL-4.0. This inconsistency between cross section and outgoing particle distributions convinced us of the need to re-evaluate zirconium again. Here, we detail improvements to our zirconium evaluations for use in ENDF/B-VII.2.

II. THE RESONANCE REGION

For ENDF/B-VII.1, ^{90}Zr and ^{91}Zr resonances were re-evaluated giving these resonance integral and thermal cross sections. This is a summary of all of the new evaluations' resonances:

^{90}Zr: Revised for ENDF/B-VII.1 [1]

^{91}Zr: Revised for ENDF/B-VII.1 [1]

^{92}Zr: JENDL-4.0 [3]

^{93}Zr: WPEC-SG23 and Atlas of Neutron Resonances [2]

^{94}Zr: WPEC-SG23 and Atlas of Neutron Resonances [2]

^{95}Zr: JENDL-4.0 [3] (background must be corrected)

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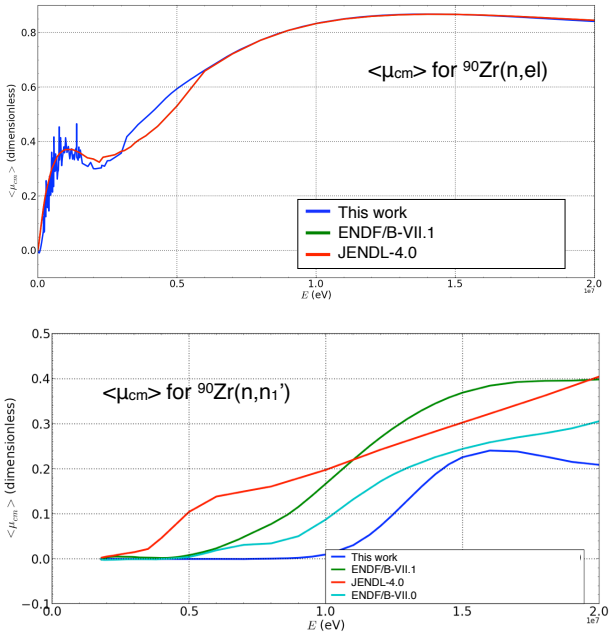

FIG. 1. Average forward scattering cosine ($\mu = \cos\theta$) in the center of mass frame for neutrons emitted in the (n,el) and (n,n$'_1$) reactions from ^{90}Zr.

^{96}Zr: Modified from ENDF/B-VII.1: scattering radius and negative energy resonance modified to better fit thermal neutron scattering data

III. MODELING THE FAST REGION

For our new evaluations we used EMPIRE-3.1 (Rivoli) [4]. This latest release of the EMPIRE code system includes several major improvements that impact our evaluation:

- New version of Coupled Channel code ECIS-2006
- Coupled Channel code OPTMAN for soft-rotor calculations, which is used to modeling even-even spherical nuclei
- Parity and spin dependent level density improvements, including a new Enhanced Generalized Superfluid Model (EGSM) level densities
- The inclusion of anisotropic angular distributions from inelastic excitations

This last improvement leads to the non-trivial (n,n$'_1$) $\langle\mu\rangle$ seen in Fig. 1. In this figure, one can also see the impact of cross section fluctuations on the elastic $\langle\mu\rangle$ as a result of the fitting procedure discussed in the next section. In addition to these improvements, there were many bug fixes which improve the overall predictability and reliability of the EMPIRE system.

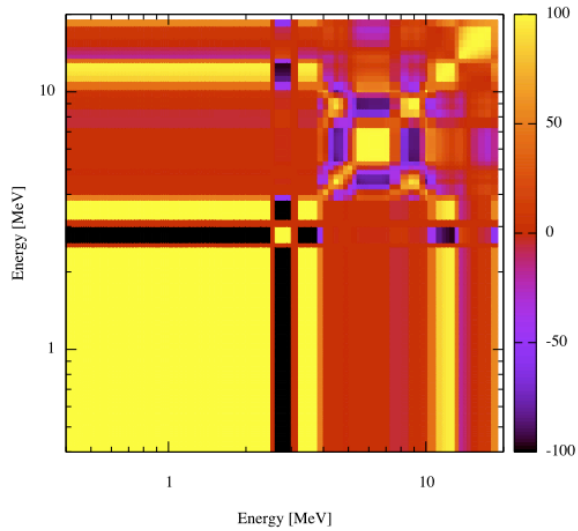

FIG. 2. Typical covariance matrix computed with the KALMAN approach. Here we show the ^{96}Zr(n,γ) covariance.

Fortunately, the changes to EMPIRE were small enough that we could reuse the EMPIRE input files used for the ENDF/B-VII.1 evaluation which include parameter tunings performed by H.I. Kim. This provides a solid prior for use in the fits described in the next section.

IV. FITTING THE CROSS SECTIONS

In order to both generate covariance data for our evaluations and to encapsulate the fluctuations in the (n,tot) and (n,el) reactions, we are resorting to a two-step fitting procedure for energies > 100 keV. In the first step, we use the KALMAN code to fit EMPIRE model parameters to the isotopic cross section data. A sample covariance from this procedure is shown in Fig. 2. Table I summarizes the database of EXFOR sets used in both this and the next fitting steps.

To include the ^{nat}Zr, we use a standalone Bayesian update code which uses the covariances created with the KALMAN procedure as a prior. Using the ^{nat}Zr(n,tot) data this way induces cross-material covariance. None of the ^{nat}Zr (n,γ), (n,el) or (n,2n) data is high enough quality to justify use in the fit so they are used to validate our results (as are the copious angular distribution data). Our update code generates modified EMPIRE inputs with rescaled reaction cross sections. The resulting fits build the fluctuations in (n,tot) into the compound cross section, creating fluctuations in (n,tot), (n,γ) and (n,el) (through the influence of the compound elastic channel). See Fig. **??**. This procedure also has the positive effect of the building fluctuations into $\langle\mu\rangle$ since typical cross sections are a sum of direct and compound contributions: $\langle\mu\rangle = \frac{1}{\sigma}\int d\mu\mu(d\sigma/d\mu) = \int d\mu\mu(d\sigma_{dir}/d\mu)/(\sigma_{dir} + \sigma_{CN})$.

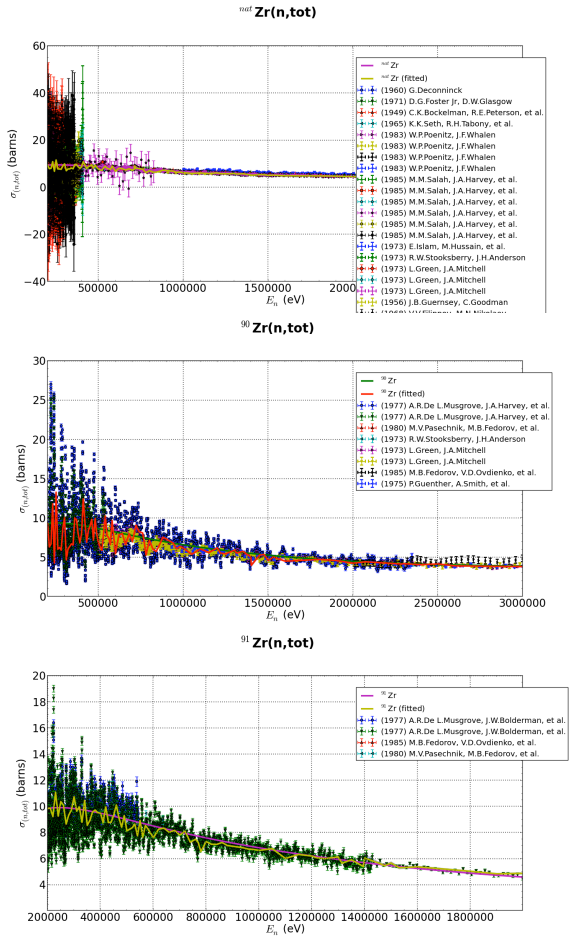

FIG. 3. Fitted (n,tot) cross sections for ^{nat}Zr, ^{90}Zr and ^{91}Zr.

	^{nat}Zr	^{90}Zr	^{91}Zr	^{92}Zr	^{94}Zr	^{96}Zr
(n,tot)	59966*	17408$^+$	22472$^+$	695$^+$	84$^+$	5917**
	(37 sets)	(9 sets)	(4 sets)	(6 sets)	(4 sets)	
(n,el)	65	24	2	21	1	
	(16 sets)	(9 sets)	(6 sets)			
(n,γ)	8308	4955	4960	4911	95	18
	(14 sets)	(7 sets)	(9 sets)	(6 sets)	(3 sets)	(15 sets)
(nn$'_1$)					109	104
					(3 sets)	
(n,2n)	19	288	2			46
	(3 sets)	(54 sets)	(2 sets)			(14 sets)
(n,p)		274	76	79	57	1
		(37 sets)	(22 sets)	(27 sets)	(26 sets)	

TABLE I. Summary table of our experimental cross section database: each cell contains the number of data points and data sets (if more than one set). ^{93}Zr and ^{95}Zr are not included here: there is no data for ^{95}Zr and ^{93}Zr has 16 points for (n,γ) in 3 sets. Several sets were removed as noted: * EXFOR #40883003 V.V. Filippov, EXFOR #30134003 E. Islam, *et al.* (bad normalization) and EXFOR #22560002 G. Deconninck (no cross section uncertainty was given); $^+$EXFOR #40935 M.B. Fedorov, *et al.* (poor target characterization); and **EXFOR #13754002 A.R. De L. Musgrove, J.A. Harvey, W.M. Good (target contaminated with ZrO_2).

V. CONCLUSIONS

A lot of work remains to finalize this evaluation, including retuning of prior parameters in preparation for redoing the KALMAN fits, redoing the ^{nat}Zr(n,tot) fits and making fixes to the resonance parameters. In addition, we have not yet begun testing in the ZPR, TRIGA and KAPL benchmarks which were essential to the testing in the ENDF/B-VII.1 evaluations [1]. We anticipate finalizing the evaluations in the summer of 2013.

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