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# Estimated <sup>55</sup>Mn and <sup>90</sup>Zr cross section covariances in the fast neutron energy region

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We completed estimates of neutron cross section covariances for <sup>55</sup>Mn and <sup>90</sup>Zr, from keV range to 25 MeV, considering the most important reaction channels, total, elastic, inelastic, capture, and (n,2n). The nuclear reaction model code EMPIRE was used to calculate sensitivity to model parameters by perturbation of parameters that define the optical model potential, nuclear level densities and strength of the pre-equilibrium emission. The sensitivity analysis was performed with the set of parameters which reproduces the ENDF/B-VII.0 cross sections. The experimental data were analyzed and both statistical and systematic uncertainties were extracted from almost 30 selected experiments. Then, the Bayesian code KALMAN was used to combine the sensitivity analysis and the experiments to obtain the evaluated covariance matrices.

# I. INTRODUCTION

Neutron cross section covariances are highly demanded by applications, probably the most prominent being the Global Nuclear Energy Partnership (GNEP) and the U.S. Nuclear Criticality Safety Program (NCSP). In GNEP, improved nuclear concepts are being considered with fuel and reactor characteristics that are well outside the design envelope of existing and prior systems. Therefore, a wide effort in advanced simulations must be preceded with the adequate adjustment of the recently released ENDF/B-VII.0 library [1]. Nuclear data covariances (uncertainties and correlations) are essential for such adjustment. NCSP is developing computational tools to enhance criticality safety predictive capabilities. For testing these tools an extensive amount of covariance data is needed, giving rise to the recent "low-fidelity project" [2].

This project was charged to provide a rough set of covariances covering all relevant reaction channels for all 393 materials in the ENDF/B-VII.0 library, emphasizing completeness rather than precision. In addition, NCSP needs high-quality covariances for specific materials, such as <sup>55</sup>Mn and <sup>90</sup>Zr. This need was partly met by the new ORNL evaluation of <sup>55</sup>Mn in the resonance region [3], including also <sup>55</sup>Mn(n, $\gamma$ ) dosimetry reaction for which covariance re-evaluation is required [4].

The present work is addressing covariances for <sup>55</sup>Mn and <sup>90</sup>Zr in the fast neutron region. Although the low-fidelity project was useful starting point, we made an important step forward by including almost 30 sets of experimental data.

The paper is organized as follows. In Section II, we describe the methodology used to produce the cross section covariances, while Section III and IV discuss, respectively, the results and the conclusions.

## **II. METHODOLOGY**

Our methodology is based on the nuclear reaction model code EMPIRE [5], Bayesian code KALMAN [6] and due in-

clusion of experimental data, see adjacent paper for more details [7]. The EMPIRE code system incorporates an extensive set of nuclear reaction models capable of describing all relevant reaction mechanisms, coupled to the up-to-date library of input model parameters [8] and providing reasonable overall description of nuclear observables even if default parametrization is used. EMPIRE was used to calculate neutron cross sections and sensitivity matrices. Then, these sensitivity matrices were used as prior by KALMAN in order to incorporate, one by one, experimental data including their statistical and systematic uncertainties.

We emphasize that our goal is to produce covariance estimates, not to re-evaluate cross sections. Therefore, our modeling and parametrization aims to reproduce ENDF/B-VII.0 somewhat approximately, just giving us enough confidence in covariance estimates.

#### A. Reaction Models and Parameters

Four nuclear reaction models were adopted that should sufficiently well describe the physics of nuclear reactions at neutron energies from 10 keV to 25 MeV for both <sup>55</sup>Mn and <sup>90</sup>Zr. The spherical optical model, in case of <sup>90</sup>Zr, and the coupled channels formalism, in case of <sup>55</sup>Mn, take care of the total cross sections and neutron scattering. The Hauser-Feshbach statistical model describes the bulk of particle emission, and the exciton pre-equilibrium model describes major features of fast particle emission at higher incident energies.

TABLE I: *Prior* optical-model parameter uncertainties (in %): r-radius, a - diffuseness, V - real depth, W - imaginary depth. The subscripts v, s, and w, respectively, denote real volume, real surface, and imaginary surface. The superscripts,  $tg \equiv n + \frac{A}{Z}$  and  $np \equiv p + \frac{A+1}{Z-1}$ , identify nucleon-nucleus interaction.

$\Delta r_s^{tg}$	$\Delta r_v^{tg}$	$\Delta r_w^{tg}$	$\Delta V_v^{tg}$	$\Delta W_s^{tg}$
3-5	3	3	3	3-3
$\Delta W_v^{tg}$	$\Delta a_s^{tg}$	$\Delta a_v^{tg}$	$\Delta V_v^{np}$	$\Delta W^{np}_s$
5	5	5	5	5

The parametrization was taken from RIPL-3 [8]. For <sup>55</sup>Mn

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we used optical model parameters of Koning-Delaroche [9] and for <sup>90</sup>Zr the dispersive potential used by us earlier [1]. Parameter uncertainties were those used in Ref. [2]. The optical model parameters, for which uncertainties (3% or 5%) were considered, are listed in Tab. I. The list of 8 parameters relevant for the Hauser-Feshbach and the exciton model plus a parameter taking into account the deformation of <sup>55</sup>Mn, is shown in Table II. The uncertainties given in Tabs. I, II represent the *prior* information on the model parameters required as a starting point in the Bayesian update procedure.

TABLE II: *Prior* parameter uncertainties (in %) used for the Hauser-Feshbach and exciton models:  $\tilde{a}$  - total level density,  $\tilde{g}$  - singleparticle level density,  $f_{\gamma}$  - gamma-ray strength functions, and mfp nucleon mean-free path; Def - deformation in the DWBA. The superscripts refer to  $cn \equiv$  compound,  $tg \equiv$  target,  $n2n \equiv$  (n,2n) residue,  $np \equiv$  (n,p) residue.

$\Delta \tilde{a}^{cn}$	$\Delta \tilde{a}^{tg}$	$\Delta \tilde{a}^{n2n}$	$\Delta \tilde{a}^{np}$	$\Delta \tilde{g}^{np}$	$\Delta \tilde{g}^{tg}$	$\Delta f_{\gamma}$	$\Delta m f p$	$\Delta \mathrm{Def}$
15	15	15	15	15	15	10-15	25	35

#### B. Sensitivities and Bayesian Update

Matrix elements  $s_{i,j}$  of the sensitivity matrix **S** were calculated as

$$s_{i,j} = \frac{\partial \sigma(E_i, \mathbf{p})}{\partial p_j}, \qquad (1)$$

where  $\sigma$  is the cross section,  $E_i$  is the energy and **p** is the vector of model parameters including  $p_j$ . The partial derivatives were computed numerically, by varying the parameters as defined by the uncertainties given in Tabs. I and II.

The Bayesian update procedure was used to update prior results by taking into account new data. We used the code KALMAN which is based on the iterative generalized least-squares approach. Applying the Bayesian equations is straightforward, an update being a simple algebraic operation,

$$\mathbf{p}_{n+1} = \mathbf{p}_n + \mathbf{P}_n \mathbf{S}^{\mathsf{T}} \mathbf{Q}_{n+1} (\boldsymbol{\sigma}_{n+1}^{\exp} - \boldsymbol{\sigma}(\mathbf{p}_n))$$

$$\mathbf{P}_{n+1} = \mathbf{P}_n - \mathbf{P}_n \mathbf{S}^{\mathsf{T}} \mathbf{Q}_{n+1} \mathbf{S} \mathbf{P}_n .$$
(2)

Here,  $\mathbf{p}_n$  is the vector of model parameters,  $\mathbf{P}_n$  is their covariance matrix and  $\boldsymbol{\sigma}_{n+1}^{\text{exp}}$  is the new experimental data set. The updated (posterior) values are denoted by the superscript n+1. The matrix  $\mathbf{Q}_{n+1}$  is defined as an inverse of the covariance matrix  $\mathbf{C}_n$  and the experimental covariance matrix  $\mathbf{C}_{n+1}^{\text{exp}}$ 

$$\mathbf{Q}_{n+1} = (\mathbf{C}_n + \mathbf{C}_{n+1}^{\exp})^{-1}.$$
 (3)

Then, the updated (posterior) cross section covariance matrix is obtained by the well known "sandwich" equation

$$\mathbf{C}_{n+1} = \mathbf{SP}_{n+1}\mathbf{S}^{\mathrm{I}} \,. \tag{4}$$

The experimental data were analyzed and both statistical and systematic uncertainties were extracted for selected experiments. The covariance matrix of the  $n^{\text{th}}$ -experiment is

$$\mathbf{C}_n^{\exp} = \mathbf{U}_n + \mathbf{W}_n \,, \tag{5}$$

where  $U_n$  and  $W_n$  are the covariance matrices of the statistical and systematic uncertainties, respectively. In the explicit notation and omitting the subscript n, the matrix elements are given by

$$c_{i,j}^{\exp} = \begin{cases} u_{i,j} + w_{i,j} & i = j \\ w_{i,j} & i \neq j \end{cases},$$
 (6)

where the off-diagonal terms are obtained assuming that the systematic uncertainties are fully correlated.

The quality and consistency of the evaluated cross sections can be assessed by scalar quantity

$$\chi^2 = \sum_{n} (\boldsymbol{\sigma}_{n+1}^{\exp} - \boldsymbol{\sigma}(\mathbf{x}_n))^{\mathrm{T}} (\mathbf{C}_{n+1}^{\exp})^{-1} (\boldsymbol{\sigma}_{n+1}^{\exp} - \boldsymbol{\sigma}(\mathbf{x}_n)).$$
(7)

High value of  $\chi^2$  per one degree of freedom suggests that the obtained uncertainties are under-estimated and it is fairly common practice to use this factor to rescale these uncertainties to get their final values.

## III. RESULTS AND DISCUSSION

We calculated neutron cross sections and their covariance matrices for  ${}^{55}$ Mn and  ${}^{90}$ Zr at 63 incident energies between 1 keV and 25 MeV, considering the five reaction channels, total, elastic, inelastic, (n,2n), and capture. We used data from 22 experiments for  ${}^{55}$ Mn and 7 experiments for  ${}^{90}$ Zr. First, we discuss  ${}^{55}$ Mn and focus on energies above the ORNL evaluation [3], that is, above 122 keV.

Fig. 1 compares our cross sections with ENDF/B-VII.0 and three sets of experimental data [10–12] found to be the basis of the ENDF/B-VII.0 evaluation. Due to the necessity of retaining validated ENDF/B-VII.0 cross sections, our estimation of covariances exclusively depends on these selected experiments. The optical model predicts a smooth, averaged behavior of cross sections and cannot reproduce fluctuating values extending as high as 4 MeV and adopted by the ENDF/B-VII.0. Accordingly, below 4 MeV we adopted the uncertainties deduced from the experiments. Since related experimental information was limited, we estimated these uncertainties conservatively as 5%. At higher energies, our uncertainties are based on KALMAN and take into account careful measurement by Cierjacks *et al.* [10].

In Fig. 2,  $^{55}$ Mn(n,n') reaction is shown. Our cross sections are in reasonable agreement with the ENDF/B-VII.0 evaluation. Relative uncertainties are fairly large at the threshold region, while in the energy range of about 0.7-10 MeV they drop to about 15-30%. As expected, the uncertainties rise at higher energies where cross sections become small.

Cross sections for <sup>55</sup>Mn(n,2n), obtained with EMPIRE-KALMAN using the experimental data of Refs. [13–24], appear to agree well with ENDF/B-VII.0 as shown in Fig. 3.



FIG. 1: Reaction <sup>55</sup>Mn(n,tot). *Prior, posterior*, and ENDF/B-VII.0 cross sections are compared with experimental data [10–12]. Relative uncertainties are in red (point-wise representation).



FIG. 2: Reaction <sup>55</sup>Mn(n,inl). *Prior, posterior*, and ENDF/B-VII.0 cross sections are compared with experimental data. Relative uncertainties are in red.

Relative uncertainties exhibit expected U-shape, starting with large values at the threshold region of  $\sim 10$  MeV, at energies  $\leq 22$  MeV being essentially flat. At higher energies, in the absence of experimental data, the uncertainties again increase.

Fig. 4 displays <sup>55</sup>Mn radiative capture cross sections and their uncertainties. Similar to (n,tot) reaction, below 1 MeV the ENDF/B-VII.0 adopted fluctuating cross sections following the experiment by Garg *et al.* [25]. Consequently, we adopted Garg's experimental uncertainties. At higher energies EMPIRE-KALMAN method was adopted. Relative uncertainties are lower than 10% in the energy range of 0.1-15 MeV, followed by expected sharp increase at higher energies.

We proceed with the discussion of <sup>90</sup>Zr reactions showing first <sup>90</sup>Zr(n,tot) and <sup>90</sup>Zr(n,el) in Figs. 5 and 6. Total as well as elastic cross sections compare well with ENDF/B-VII.0 and



FIG. 3: Reaction  $^{55}$ Mn(n,2n). *Prior, posterior*, and ENDF/B-VII.0 cross sections are compared with experimental data [13–24]. Relative uncertainties are in red.



FIG. 4: Reaction  ${}^{55}$ Mn(n, $\gamma$ ). *Prior, posterior*, and ENDF/B-VII.0 cross sections are compared with experimental data. Relative uncertainties are in red.

experimental data. Except for the low energy region, the uncertainties are fairly flat around 2.5%. In contrast, uncertainties for (n,inl) are much larger throughout the whole energy range (Fig. 7) since no experimental data were used. Generally, uncertainties should be low whenever a wealth of experimental data is used in the evaluation.

Finally, in Fig. 8 the <sup>90</sup>Zr(n,2n) cross sections obtained with EMPIRE-KALMAN method are shown. Compared are *prior*, *posterior*, and ENDF/B-VII.0 cross sections with experimental data [30–34] included in our evaluation showing good agreement with both ENDF/B-VII.0 and data. Relative cross section uncertainties exhibit expected U-shape.



FIG. 5: Reaction <sup>90</sup>Zr(n,tot). *Prior*, *posterior*, and ENDF/B-VII.0 cross sections are compared with experimental data [26–29]. Relative uncertainties are shown in red.



FIG. 6: Reaction <sup>90</sup>Zr(n,el). *Prior*, *posterior*, and ENDF/B-VII.0 cross sections are compared with experimental data. Relative uncertainties are shown in red.

## **IV. CONCLUSIONS**

We produced estimates of neutron cross section covariances for  ${}^{55}$ Mn and  ${}^{90}$ Zr in the fast neutron energy region. This work was primarily motivated by the needs of the U.S. Nuclear Criticality Safety Program, though the results are of interest for other applications such as GNEP and dosimetry. Our results are based on the EMPIRE-KALMAN approach using statistical and systematic uncertainties taken from almost 30 selected experiments.

Our covariances should be considered as being of intermediate quality. For high-fidelity results one should perform complete re-evaluation of cross sections simultaneously with covariances, and preceded with detailed analysis of all ex-



FIG. 7: Reaction <sup>90</sup>Zr(n,inl). Shown are *prior*, *posterior*, and ENDF/B-VII.0 cross sections. Relative uncertainties are in red.



FIG. 8: Reaction  ${}^{90}$ Zr(n,2n). *Prior, posterior*, and ENDF/B-VII.0 cross sections are compared with experimental data [30–34]. Relative uncertainties are shown in red.

perimental data. <sup>55</sup>Mn represents additional challenge due to many data available, including high resolution measurements that exhibit strong fluctuations up to a few MeV.

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