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Nuclear data uncertainty propagation for spectral reaction ratios

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ABSTRACT

This paper describes several approaches to propagating nuclear data uncertainty in integral capture and fission reaction rates, focusing on the spectral hardness indices of ^{193}Ir(n,n′)^{193m}Ir/^{191}Ir(n,g)^{192}Ir and ^{238}U(n,f)/^{235}U(n,f) in fast critical assemblies. Uncertainties in the nuclear data were propagated through the following methods: a sensitivity method (utilizing first order perturbation and covariance matrices), a BFMC method (random cross sections generated through covariance matrices), and Total Monte Carlo method (random cross sections generated from varying nuclear parameters). The uncertainty estimates from the three approaches are evaluated against the associated experimental uncertainties of a ZEUS/COMET critical assembly irradiation. The uncertainty estimates of the three approaches agree fairly well, and contribute a significant portion to the overall uncertainty of the spectral hardness indices.

1. Introduction

The propagation of nuclear data uncertainty in simulations is a current area of research for a variety of scenarios, from light water reactors to critical assemblies (Chadwick et al., 2005). As experimental measurements become more precise, including the impact of nuclear data when estimating the modeled uncertainty can be significant. This work evaluates a 2010 neutron irradiation experiment (IER-163) (Jackman, 2012) with observed discrepancies between simulated and measured integral responses. The purpose of this study is to evaluate whether the inclusion of nuclear data uncertainty helps explain those differences, and to determine the appropriate methodologies for the uncertainty calculations.

The IER-163 experiment irradiated a number of foils utilizing the Comet critical assembly machine, with this work specifically analyzing the ^{193}Ir(n,n′)^{193m}Ir/^{191}Ir(n,g)^{192}Ir and ^{238}U(n,f)/^{235}U(n,f) reaction ratios. The fission neutron spectra of critical assemblies have historically been defined by the hardness of the spectra, calibrated in terms of a ‘spectral index’. Originally the uranium fission ratio ^{238}U(n,f)/^{235}U(n,f) solely represented this index for fast critical assemblies, as the sensitivity range is approximately between 0.5 and 2 MeV shown in Fig. 1. Foil packets of highly enriched uranium (HEU) and depleted uranium (DU) could be placed at various locations (and under a variety of experimental conditions) in the assembly to evaluate the impact on neutron spectra though this “spectral index”. One problem with the fission ratio method was that the 0.5 MeV threshold did not probe the lower fission energy spectra; an additional spectral index was needed. Historic work in the 1960s demonstrated that the two naturally occurring iridium isotopics could serve in this role. Four reactions are dominant for iridium when considering critical assembly irradiations consisting of:

- ^{191}Ir(n,γ)^{192}Ir
- ^{191}Ir(n,2n)^{190}Ir
- ^{193}Ir(n,n′)^{193m}Ir
- ^{193}Ir(n,2n)^{192}Ir

While we are only interested in two of the reactions for the spectral index, the ^{193}Ir(n,2n)^{192}Ir reaction needs to be accounted for as a competing pathway to the ^{192}Ir measurement. For a measurement of ^{192}Ir that is evaluating the (n,γ) pathway, a correction is commonly applied relating the (n,2n) production of ^{190}Ir to the (n,2n) production of ^{192}Ir:

$$\frac{N_{191(n,\gamma)192}}{N^{o}_{191}} = \frac{N_{192}}{N^{o}_{191}} - \frac{N_{191(n,2n)190}}{N^{0}_{191}} \frac{\widehat{RR}_{193(n,2n)}}{\widehat{RR}_{191(n,2n)}}$$

Where the ratio of (n,2n) reaction rates, utilizing the ENDF/B-VII.1 library, is estimated to be 1.68 (insensitive to small perturbations is fast critical assemblies). The ratio can also be calculated specific for the application, and for the IER-163 irradiation was estimated to be 1.89. So with measurements of ^{190}Ir, ^{192}Ir, and ^{193m}Ir, we can estimate the spectral hardness with an approximate sensitive range from 0.08 MeV to 6.0 MeV (shown in Fig. 1).

Radiochemical data assessment can depend strongly on the use of these reaction rate ratios, and new measurements such as activation products in stainless steel (Keith, 2018) are commonly coupled with the spectral indices. By comparing measured reaction

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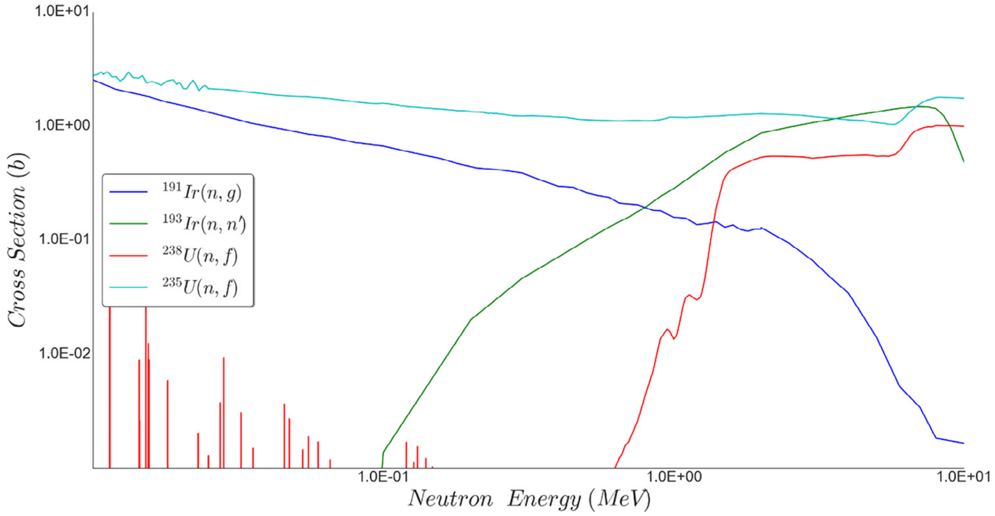

Fig. 1. ENDF/B-V11.1 evaluated pointwise cross-section data for ^{191}Ir(n,γ), ^{193}Ir(n,n′), ^{235}U(n,f), and ^{238}U(n,f) reactions.

rates in critical assemblies to calculated results (radiation transport simulations), a validation of these spectral indices under a variety of energy-dependent spectra can be provided. The intention of the IER-163 experiment was to establish the basis for future measurements at critical assemblies, such as activation products and fission-product chain yields (Jackman, 2012), by measuring the well-known spectral ratios. Due to experimental constraints, the foil irradiation occurred at the core/reflector interface (not measured in past experiments). The core consisted of cylindrical highly enriched uranium (HEU) metal plates with a metallic copper reflector, resulting in a high positional sensitivity for the reaction ratios considered (Favorite, 2012). It has been reported that cross-section validation can be unreliable when the assembly neutron spectra is not established (Chadwick, 2007), as is the case for the IER-163 measurements. However, we can estimate the influence that the neutron spectra uncertainty (radiation transport) has on the reaction ratios and ideally help explain the discrepancies between measured and calculated results.

In this study, three methods were chosen to propagate nuclear data uncertainty to the integral ratios. One method utilized perturbation theory to compute sensitivity coefficients as a function of isotope and energy (Rising, 2015). This can be combined with the available covariance matrices to obtain the uncertainty. Another set of approaches uses Monte Carlo sampling of nuclear data distributions to produce the uncertainty estimates. One method builds the distribution based on the evaluated covariance matrices, referred to as Brute Force Monte Carlo (BFMC) (Rising, 2015). The other method evaluated in this work combines the Unified Monte Carlo (UMC) method (Capote and Smith, 2008) and the Total Monte Carlo (TMC) method (Koning and Rochman, 2008). The UMC + TMC method (Rochman, 2011, 2016) builds the nuclear data distribution directly from distributions in differential data and theoretical model parameters. One objective of this work was to determine applicability of perturbation relative to Monte Carlo methods in areas of high sensitivity (core/reflector interface).

2. Methods/results

The uranium fission ratio ($^{238}U_{n,f}/^{235}U_{n,f}$) and the iridium activation ratio (^{193m}Ir/^{192}Ir) underpin multiple radiochemical assessments and codes (Lee, 2016). However, minimal systematic effort has been made to evaluate the influence of neutron spectra uncertainties on these particular reaction ratios. While other foils were irradiated during the experiment (Au, Pu, etc.), the measurement capability is well established (particularly with the uranium fission ratio) for the spectral indices described here. This ideally allows the evaluation of nuclear data uncertainty, while minimizing the influence of measurement uncertainty on the analysis. The first step in the analysis was to evaluate the experimental uncertainty through gamma/beta measurements, cumulative fission chain yields, and foil masses. Since the goal was to evaluate if nuclear data propagation helped explain the discrepancies (between calculated and experimental), it was important to quantify any potential uncertainty/bias associated with the measurements.

After the experimental measurements were characterized, the uncertainties associated with both the uranium (core) and copper (reflector) cross sections could be propagated for the IER-163 critical assembly calculations. The spectral indices calculations were performed through MCNP, a Monte Carlo based radiation transport approach, utilizing ACE files (a compact version of the Evaluated Nuclear Data File format) containing the nuclear data. Due to the differences in the uncertainty propagation methodologies (see BFMC and TMC sections), the use of two nuclear data libraries were needed. The random ACE files required for the BFMC approach were obtained through the ENDF/B-VII.1 library (Chadwick, 2011). The covariance files of the ENDF/B-VII.1 library were processed into 187 groups, and the established ENDF/B-VII.1 ACE file was “randomly drawn” (based on the 187-group covariance matrix) to produce the random ACE files (Koning, et al., 2017). The TMC approach, on the contrary, utilized random nuclear data by weighting randomly sampled input parameters in the TALYS code system which can then be processed into ACE files (Koning and Rochman, 2012). The random ACE files used in the TMC approach is then inherently based on the TENDL nuclear data library. The isotopics considered for the uncertainty propagation were based on sensitivity calculations performed for the IER-163 experiment (spectral ratio responses). The isotopics responsible for the largest sensitivities in the calculations were found to be ^{235}U, ^{238}U, ^{63}Cu, and ^{65}Cu. Based on the results, the random ACE files for uncertainty propagation were obtained for those select isotopics. The cross-isotope correlation for the iridium ratio was not found, therefore nuclear data uncertainty is only propagated through the prompt fission neutron spectra and transport within the assembly.

3. IER-163 irradiation

The computational model was modified from the fifth Zeus experiment (HEU-MET-FAST-073) in the International Handbook