Quantifying Nuclear Data Uncertainties in National Security Applications

I.C. Gauld, M.T. Pigni, S. Croft, A. Nicholson, M.L. Williams, V. Mozin, C.M. Mattoon, R. Vogt, and M.T. Swinhoe

***Abstract*— Nuclear data is fundamental to many of nuclear material measurements, for detector performance optimization, instrument calibration, and data reduction to unfold quantities of interest from measured signals. Due to limited experimental data for code validation, in nuclear safeguards and non-proliferation measurements frequently rely on models. These models can rely on nuclear data from theory or on data that has been extrapolated and has large uncertainties. Capabilities in uncertainty analysis are rapidly emerging that can provide code users with measures of the impact of uncertainties upon the reliability of calculations. This paper describes research initiated to develop uncertainty analysis capability for the nuclear data unique to national security applications. This work includes the development of covariance data (uncertainties and correlations) for passive emission processes. The tools necessary to apply these data to applications are currently non-existent. These components represent the basis of a comprehensive data uncertainty analysis capability. In addition to quantifying nuclear data uncertainty in applications, the tools are being developed to further identify and quantify the importance of the different nuclear data that contribute to uncertainty. These tools and data represent a cross-cutting R&D enabling capability that can be applied to objectively identify gaps and weaknesses in nuclear data to help prioritize future data investments that will have the greatest benefit for agency missions. This paper describes the components of uncertainty analysis developed for neutron based measurements and application of the tools to uncertainty analysis of a neutron multiplicity counter.**

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***Index Terms*— Nuclear data, uncertainty, covariance, national security, safeguards, nonproliferation.**

I. INTRODUCTION

NUCLEAR data provide fundamental information for modelling codes that simulate nuclear physics processes for material and radiation detection used in nuclear non-proliferation and nuclear safeguards. Application areas of nuclear forensics, radiation signature analysis, nuclear material verification for safeguards, reactor-based materials production detection, treaty verification, and nuclear test monitoring rely extensively on modelling to fill gaps where experimental data are unavailable and where measurements are not practical. Code calculations are widely employed in these applications for instrument design and performance assessment, optimization, calibration, and for interpretation of complex measured signals from measurements to unfold quantities of interest. However, nuclear data measurements have been prioritized to large extent by the nuclear criticality and reactor physics communities. Nuclear security modelling applications can easily depend on data that have limited or no experimental validation, leading to results that rely on low quality nuclear data containing gaps and inconsistencies. Since uncertainty analysis (inclusive of nuclear data) methods are not readily available for routine analyses, users are likely to be unaware when results are impacted by low fidelity or incomplete nuclear data.

Improving nuclear data is a goal common to many agencies that rely on code simulations to assess scenarios where direct measurement data for code validation are not available. In the US, recognition of the need for better nuclear data is evidenced by the Nuclear Data Needs Workshop [1] organized in 2015 by the Department of Energy Office of Science and the Office of Defence Nuclear Nonproliferation R&D to identify cross cutting data needs, and the subsequent formation of an interagency Nuclear Data Working Group [2] (NDWG) aimed at identifying high priority data needs and coordinating these needs with the data measurement community. These recent efforts reflect a consensus need for improved nuclear data in many application areas and better coordination of nuclear data measurement activities.

Several previous investigations have evaluated nuclear data needs for nuclear safeguards and nonproliferation applications. Schillebeeckx et al. [3] investigated nuclear data requirements for gamma- and neutron-based non-destructive assay methods of fissile material; Santi et al. [4] looked at uncertainties in the fundamental nuclear data that affect the precision of radiation transport codes in modelling the correlated neutron emissions from nuclear materials; Parker [5] has evaluated nuclear data needs focused on energies and intensities of gamma ray lines of interest in safeguards; and Sleaford [6] upgraded the Evaluated Nuclear Data File (ENDF) for neutron-induced prompt-gamma-ray spectrometry. These studies generally focused on assessment of a specific application and considered a subset of the nuclear data needs relevant to that application. Other studies, like that of Bahran et al. [7], performed a broad survey on data needs across various academic and research institutions. Bahran's review is comprehensive, with expert input from practitioners in safeguards, nuclear nonproliferation, forensics, and nuclear data communities. However, surveys are ultimately based on subjective experience, making any ranking of data priorities difficult. Funding agencies and end users need objective and quantitative information on existing data uncertainties to establish priorities for future data investments. While expert opinion is essential, intuition should not be the sole basis for prioritizing future data measurement programs.

This paper describes the development of covariance data for passive neutron emission processes, including specifically spontaneous fission and (α,n) emission where these data are currently unavailable, and application of nuclear data uncertainties and quantitative analysis tools for the systematic evaluation of complex systems that rely on a vast set of diverse nuclear data. These tools can be applied to assess total uncertainty in code calculations due to the data. However, the more ambitious goal of this work is to apply these tools to identify and quantify the individual contributions of nuclear data uncertainty to total model uncertainty. Such capability represents a basic cross-cutting R&D enabling capability that can help identify gaps and major weakness in nuclear data to prioritize data measurements that will have the greatest benefit for agency missions.

This work involved the collaboration of three U.S. National Laboratories: Oak Ridge National Laboratory (ORNL), Lawrence Livermore National Laboratory (LLNL), and Los Alamos National Laboratory (LANL), all with extensive experience in data development and uncertainty analysis. ORNL role was to quantify uncertainty in passive neutron source emission calculations from uncertainty in nuclear data, focusing on (α,n) reactions and spontaneous fission, and uncertainty in compositions and signatures during reactor-based material production. LLNL was responsible for evaluating uncertainties for neutron-induced cross sections as well as prompt fission parameters using nuclear data and covariances in ENDF/B-VII.1 [8]. The nuclear data uncertainties are applied to calculations using the MCNP code [9] by LANL to evaluate detector technologies. In this paper, the covariance data and uncertainty analysis tools are applied to evaluate nuclear data uncertainty importance for a LANL epithermal neutron multiplicity counter measuring PuO_2 items. This practical example problem exercises the uncertainty data for many of the prompt and delayed processes addressed in this research.

II. Approach

Uncertainties are inherent in nuclear data from the experimental measurements and data reduction and evaluation process. These data uncertainties can be used to estimate the accuracy of integral responses computed on the basis of nuclear material source characteristics and radiation detection models. This approach provides one of the few options for uncertainty analysis when direct experimental measurements are not available or possible to validate computer codes. This approach requires complete uncertainty information for all nuclear data that drive the major physics processes of the system being studied. These nuclear data uncertainties can be propagated through the model to estimate total uncertainty in calculated responses.

To this end, two different approaches are frequently used: (1) perturbation theory using solutions to the adjoint equations that describe the system, and (2) Monte Carlo uncertainty sampling. The former approach requires that adjoint equations be developed and added to the existing code and solved, whereas Monte Carlo sampling instead adjusts only the nuclear data used by the code within the uncertainty range of the data leaving the code unchanged. Adjoint solutions to continuous-energy transport codes have been implemented in MCNP [10] and SCALE [11], however these efforts have largely focused on uncertainty analysis for eigenvalue calculations for nuclear criticality safety. The application of generalized perturbation theory that can solve for other parameters such as reaction rates are not widely available, although recent progress in this area has been reported [12]. However, this approach cannot be used for general fixed-source problems common to non-proliferation and safeguards.

For problems involving relatively few data parameters of interest, the nuclear data can be adjusted manually within its uncertainty range to determine the impact on calculated results. However, for problems involving many thousands of nuclear data parameters, this approach is untenable and automated methods are needed. Data uncertainty propagation using Monte Carlo (statistical) uncertainty sampling techniques has been applied for continuous energy [13] and multigroup transport calculations [14] to assess a broad range of nuclear data uncertainty. However, this technique is computationally intensive as it involves adjusting the nuclear data according to the data uncertainties, and repeating the calculations many times to obtain statistical distributions for the results. This method provides total uncertainty in the calculated results due to all nuclear data that was adjusted in the problem. Correlation analysis of the results and nuclear data variations has been applied to identify the importance of dominant nuclear data uncertainties for the particular application. Such tools can be used to identify priority nuclear data measurement needs for specific applications.

The Monte Carlo approach was applied in this work using relatively new data analysis software called Kiwi [15]. Kiwi is designed as an interface between covariance data available in ENDF/B and large-scale data uncertainty studies and provides a targeted capability for handling the data complexities unique to this work. Kiwi can also be used within the LLNL Uncertainty Quantification Pipeline framework.

Uncertainties in nuclear data are usually represented as covariance matrices (uncertainties and correlations). However, covariances are a relatively new addition to nuclear data evaluations. When covariances are not available, retroactive methods have been successfully used to estimate missing covariance data using low-fidelity methods and physics models [16]. State-of-the-art neutron-induced reaction cross section evaluations have made significant advancement in expanding covariance data. Covariances in ENDF/B-VII.1 are now included for many neutron-induced neutron reactions for 190 of the 423 target materials, whereas release of ENDF/B-VII.0 contained almost no covariance information.

A comprehensive and systematic review of data uncertainties requires covariances describing all major nuclear processes in the model. This can involve a massive amount of nuclear data. For nondestructive assay measurements by neutron and gamma emission of fissile nuclear material, or irradiated nuclear material, the important nuclear data processes can include:

- Cross sections for neutron-induced reactions
- Neutron-induced prompt fission neutron production, spectrum and multiplicity
- Spontaneous fission prompt neutron production
- (α,n) reaction neutron production
- Prompt and delayed gamma ray emission
- Nuclear radioactive decay
- Fission product yields

Focus of our research is on neutron technologies, and therefore data related to gamma ray emission processes and detection has not yet been addressed. While nuclear data for many of these processes are in ENDF/B-VII.1, covariance information is only widely available for neutron reaction cross sections and prompt neutron production data. To provide a comprehensive assessment of data uncertainties for many of the data types important to non-proliferation detection, covariance data must first be generated for these other processes. To this end, our research has initiated development of methods to generate covariance data for missing processes, developed tools for the application of covariances in detection system models, and tests the covariances against available experimental data.

III. COVARIANCE DATA

Covariance data are the foundation of modern nuclear data uncertainty analysis. Covariances include the statistical and systematic components of uncertainties and correlations in the data that derive from the measurement method and models used in the nuclear data reduction procedures. Covariance data impact the uncertainty distributions of calculated quantities and output responses. Accurate assessment of the data uncertainties in complex systems requires accurate and complete covariance data. While there has been progress and recognition of the need for better and complete covariance data, large gaps remain. These gaps can be highlighted in nuclear security applications that frequently emphasize data of low importance to many nuclear safety applications and have not received adequate attention. Developing better and more complete covariance information has been identified as a priority topic by the US NDWG [2].

To address the gaps in covariance data needed for uncertainty analysis in this work, methods and tools for retroactive covariance data generation are applied using standard evaluation techniques and computer models that describe the physics of the nuclear processes. Fig. 1 shows the organization of this project by the national laboratories, nuclear data categories considered, and the computer models applied for covariance data generation and the application of covariance data to applications. The status of existing covariance data and data generation is described.

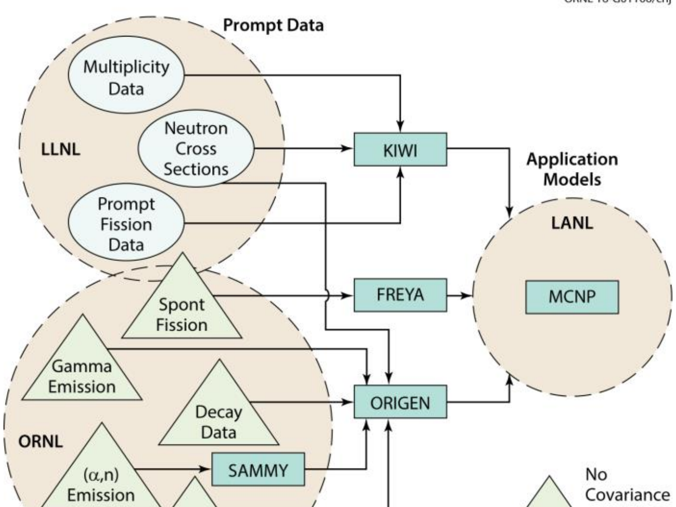

Fig. 1. Organizational roles, nuclear data types, and processing codes used for data uncertainty analysis applied in this research.

A. Neutron Cross section Data

Covariance data for neutron cross sections are advanced relative to most other nuclear data due to their importance in reactor analysis, nuclear criticality safety and stockpile stewardship. With the release of ENDF/B-VII.1 in 2011, covariance data for neutron reaction cross sections are available for 190 target materials, reflecting the increasing priority of uncertainty estimation in nuclear analyses. The neutron-induced reaction quantities in ENDF/B-VII.1 with covariance data include:

- total cross sections,
- elastic scattering cross sections,
- neutron capture (n,γ) cross sections,
- fission (n,f) cross sections,
- inelastic scattering (n,n′) cross sections,
- cross sections for (n,xn),
- charged-particle producing (n,p), (n,d), (n,t), (n,α) cross sections.

Not all neutron reactions and nuclear processes in ENDF/B-VII.1 have covariance data. The cross section covariance data applied in this work are adopted directly from ENDF/B-VII.1.

B. Prompt Neutron Production

The covariance data for prompt neutron generation via induced fission available with ENDF/B-VII.1 were used in this work, including the average neutron-induced fission number ($\bar{\nu}$) and prompt fission neutron spectrum (PFNS). These parameters play a significant role in neutron emission, particularly for items with large neutron multiplication factors. Even for highly irradiated nuclear fuel assemblies with typical discharge burnup and a relatively low effective multiplication factor (in water) of about 0.6, there are ≈ 2.5 prompt fission induced neutrons originating per passive neutron emitted. The PFNS has been identified as a large source of uncertainty in some criticality applications [17,18], and research is ongoing to quantify prompt fission neutron spectrum uncertainties in the high energy region.

The neutron multiplicity distribution $P(\nu)$, gives the distribution of the number of neutrons emitted by fission and is important to multiplicity counting. Currently, ENDF/B-VII.1 contains no fission neutron multiplicity data or uncertainties.

C. Passive Neutron Emission

Passive neutron emission by (α,n) reactions for nuclear materials in non-metal form and spontaneous fission are widely used for safeguards measurements and for many national security applications. Cross sections for alpha particle reactions are unavailable in ENDF/B-VII.1 but are available in the Japanese Evaluated Nuclear Data Library JENDL [19] and the TALYS-based evaluated nuclear data library TENDL [20], but without any covariances. Uncertainties in the nuclear data for these processes are incomplete and covariances are largely non-existent in any publications.

1) (α,n) reactions

Nuclear data describing charged-particle interactions on light nuclei are essential for calculating neutron emission, for instance, via (α,n) processes. Namely, neutron emission by (α,n) reactions can represent a significant neutron source in neutron-based measurements of uranium and plutonium in compound forms. In irradiated nuclear fuel achieving a moderate to high burn-up, the neutron source is typically dominated by ^{242,244}Cm spontaneous fission. However, in low-burn-up fuels such as those for material production, the (α,n) neutron processes can represent a large component of the total neutron source and are, therefore, important to neutron measurements involving irradiated materials.

The probability of an α-particle undergoing an (α,n) reaction with a target nuclide of atom density N_i, normalized to the total atom density of the material, N, (i.e., atom fraction) is given by

$$p_i(E_\alpha) = \frac{N_i}{N} \int_{E_\alpha}^{0} \sigma_i(E)/\epsilon(E)\, dE$$

where E_α is the maximum (emitted) energy of the α particle, $\sigma_i(E)$ is the (α,n) cross section (cm^2), and $\epsilon(E)$ is the stopping power cross section (eV·cm^2) which is approximated using the Bragg-Kleeman relationship for compounds and mixtures. An important observation from this equation is that both the cross section and the stopping power data contribute to the uncertainty in the neutron emission rate (although the stopping power is not strictly nuclear data since it is dominated by electronic stopping). Therefore, uncertainty contributions were evaluated for both data terms.

Covariance data development for ^{17,18}O(α,n) reactions

Although nuclear data describing charged particle interactions on light nuclei are essential for calculating neutron emission, e.g., via (α,n) processes, there are no (α,n) cross sections or covariance data in ENDF/B-VII.1. Passive neutron calculations performed using MCNP do not include (α,n) sources. These must be calculated externally and input as a fixed source to the code. The SOURCES 4C code [21] is widely used for this purpose, and more recently similar routines have been used in the MISC package [22]. The SOURCES algorithms and data have also been integrated as the neutron source module for the ORIGEN code [23] to provide analysis of complex time-dependent sources arising from material irradiation and decay. However, no source uncertainty estimates are available with these codes.

To address this gap, uncertainties in the data for neutron production by ^{17,18}O(α,n) reactions were evaluated. The impact of the uncertainty related to both ^{17,18}O(α,n) cross sections and stopping power cross sections of oxygen and uranium propagated to neutron source generation is estimated using the ORIGEN code in a typical uranium oxide fuel type. Details on the methodology are described in reference [24] and, here, we limit the discussion to give a concise overview of the work. In this regard, we developed a methodology to evaluate charged-particle-induced cross section data and generate related covariance information with the R-matrix code SAMMY [25].

SAMMY evaluation methodology

The SAMMY code is a modern tool for calculating reaction cross sections mainly used for nuclear data evaluations in the neutron resolved resonance region. The most recent version of SAMMY has built-in capabilities that also allow the code to evaluate cross sections for other incident particles including charged particles. The SAMMY code incorporates selected R-matrix approximations coupled to the Bayesian method to fit experimental data and ultimately to generate a set of resonance parameters with a related parameter covariance matrix. For this task, we based our results on the Reich-Moore formalism, which approximates the expression for elastic and reaction cross sections better than other single- and multi-level variants of R-matrix theory.

To show the feasibility of the method, thin-target ^{17,18}O(α,n) cross sections measured by Bair [26,27] were used to estimate a set of resonance parameters and a related covariance matrix. In the analyzed energy range from 1.0 to 5.1 MeV, as the result of the Bayesian update procedure used to update the preliminary set of parameters by taking into account the experimental data sets, we obtain both the evaluated cross sections, uncertainties, and the covariance matrix. On average, cross section uncertainties estimated for ^{17}O and ^{18}O were 5.6% and 5.8%. The experimental data and evaluated cross sections are shown in Fig. 2.

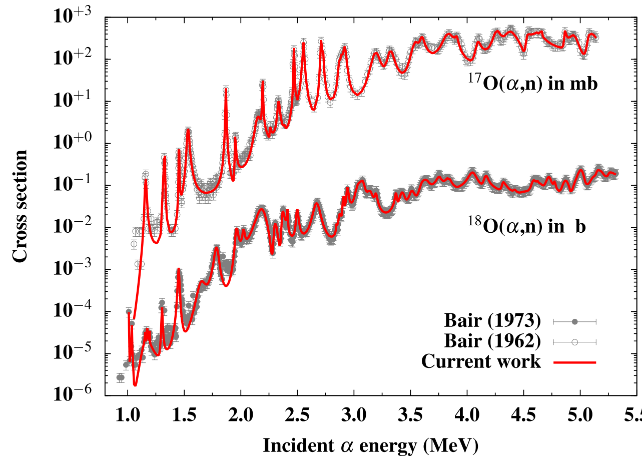

Fig. 2. Bair's experimental data for ^{17,18}O(α,n) cross sections and the cross section evaluations performed in this work using the SAMMY code.

An important outcome of the SAMMY evaluation is the generation of a covariance matrix. The correlation matrix corresponding to the ^{18}O(α,n) cross sections is shown in Fig. 3.

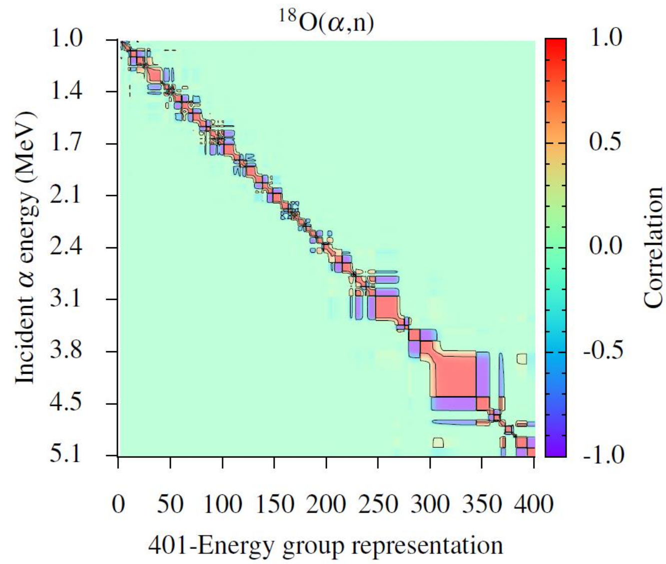

Fig. 3. Correlation matrix of ^{18}O(α,n) cross sections in 400-energy group representation in the range 1.0 – 5.1 MeV.

Stopping power cross sections

The stopping power cross sections used in SOURCES4C code are based on data from Ziegler for Z < 93 and Northcliffe and Schilling for Z of 93 to 105. These data are fit to an energy-dependent analytic function computed on the basis of a few parameters. In order to quantify the contribution of the stopping power cross sections to the uncertainty of the output neutron source intensity, stopping power cross sections based on ASTAR data [28] for oxygen and uranium were fit to the analytic function.

The results of the fitting procedure using the ASTAR data for oxygen and uranium are shown in Fig. 4, and the values of the related coefficients and uncertainty information were obtained using an implementation of the nonlinear least-squares Marquardt-Levenberg algorithm. This procedure generated a covariance matrix that could be used for uncertainty analysis using Eq. (1) as implemented in the SOURCES and ORIGEN codes.

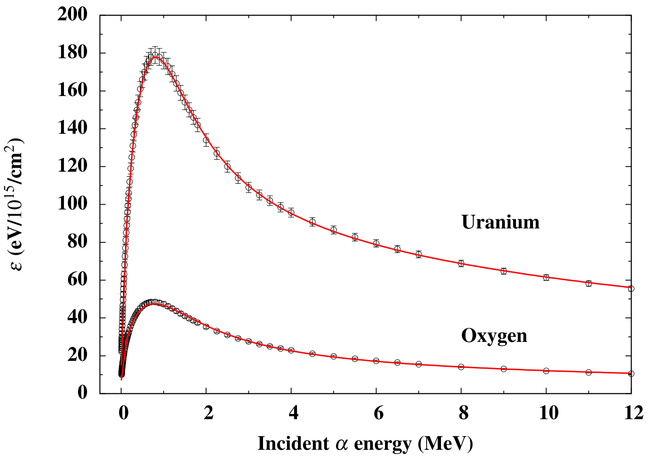

Fig. 4. Stopping cross sections from ASTAR and results from fitting to a few-parameter analytic function used to develop covariances.

2) Spontaneous fission

Covariance information on *spontaneous* fission yields, energy distributions, and neutron multiplicity is very limited. Covariances for spontaneous fission in ENDF/B-VII.1 are only available for ^{252}Cf. No uncertainties are given for prompt or delayed spontaneous fission $\bar{\nu}$ values for any uranium or plutonium isotopes. Consequently, a retroactive approach to covariance data generation for the spontaneous fission neutron spectrum was investigated using a fission event modelling code.

FREYA fission simulation code

FREYA [29] is an event-by-event Monte Carlo fission modelling code developed at Lawrence Livermore National Laboratory (LLNL). FREYA was used in the current work to generate covariances for spontaneous fission based on the parameters constraints in the fission model. Nuclear data uncertainties in the fission process can be large, and there is very little experimental data available for code validation, and essentially no covariance data for uncertainty analysis. The FREYA code can help fill gaps in data with modelling and can produce correlations and covariances based on model input parameters to enable uncertainty sampling. FREYA is being implemented for release in MCNP6 as a prompt and spontaneous fission model; however, there are no plans currently to include ranges of fission parameter values (uncertainties) and therefore no uncertainty estimates are provided. Evaluating parameter uncertainties is a goal of present research.

This work applied FREYA to generate covariances for the fission parameters based on correlations in physics model and conservation laws. An important element of this work is to constrain individual parameter uncertainties to be consistent with experimental observations. In this approach, options will be implemented in FREYA to vary the parameters, and therefore the spectra, to include uncertainty contributions rather than the current practice of using a fixed set of fit parameters.

Application of FREYA to ^{240}Pu spontaneous fission

Figures 5 and 6 illustrate the influence of one of the model input parameters, the excitation energy (x), on the energy neutron spectrum and average neutron energy for ^{240}Pu spontaneous fission. The development of fission model parameters in FREYA have focused on neutron induced fission in U and Pu materials ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{238}Pu, ^{240}Pu, ^{242}Pu. There is much less experience in spontaneous fission that is important to irradiated U and Pu materials (e.g., ^{242}Cm and ^{244}Cm) and unirradiated materials (e.g., ^{240}Pu).

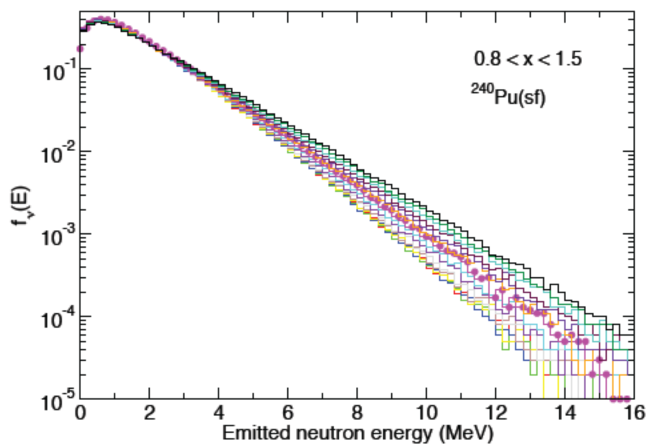

Fig. 5. FREYA-generated spontaneous fission spectra for variations in the excitation energy parameter, x.

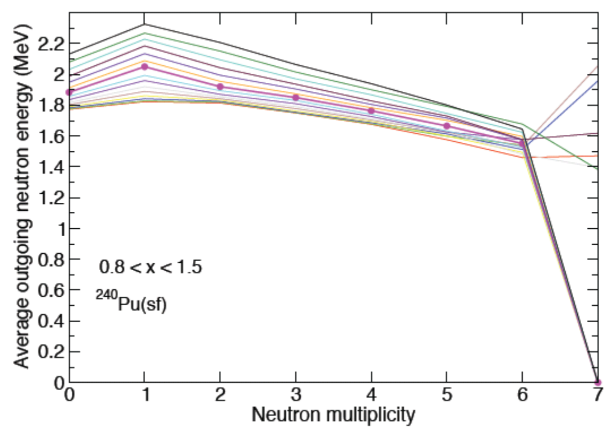

Fig. 6. FREYA-generated average outgoing neutron energy for variations in the excitation energy parameter, x.

The FREYA code was used to generate covariance data for the P(ν) distribution and prompt fission neutron spectrum (PFNS) of spontaneous fission of ^{240}Pu. The Total Kinetic Energy (TKE) and related uncertainties measured by Schillebeeck [30] were used to generate 100 input TKE files. The output responses from FREYA, namely, P(ν) distribution and PFNS , were collected to compute their mean values that were compared to measurements. Calculations show that the distribution of the number of neutrons (Figure 7) is rather poor compared to data of Boldeman [31] and an evaluation performed by Croft in this work, and that the multiplicity distribution model needs to be improved, or other model parameters need to be considered. Comparison of calculations to Alexandrova's experimental data [32] above 3 MeV (Figure 8) shows good agreement for PFNS calculations. Also shown in this figure is the default Watt fission spectrum obtained using the Watt fission parameter currently used in MCNP. The FREYA calculation is significantly improved compared to the Watt spectrum.

An important outcome of the FREYA model for this work is the covariance matrix and related correlation matrix needed for uncertainty analysis. The correlation matrix of the neutron multiplicity distribution (Figure 9) shows strong positive and negative correlations. The correlation matrix of the prompt neutron energy distribution (Figure 10) shows a pattern of both negative and positive correlations around the peak of the distribution suggesting a competition effect. The correlation matrix of the PFNS is mostly uncorrelated for energies above 10 MeV. Strong positive correlations are seen at the peak of the spectrum (about 1 MeV). For energies above 2 MeV strong negative correlations are also visible.

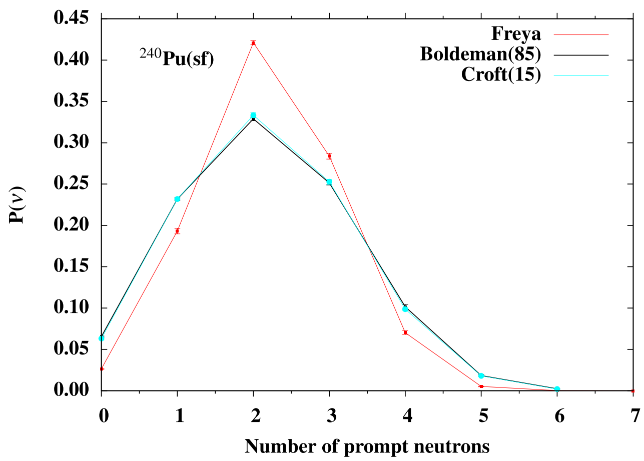

Fig. 7. FREYA-generated neutron multiplicity for ^{240}Pu spontaneous fission based on the excitation energy parameter, x, uncertainty of Schillebeeck [30] compared to other evaluations.

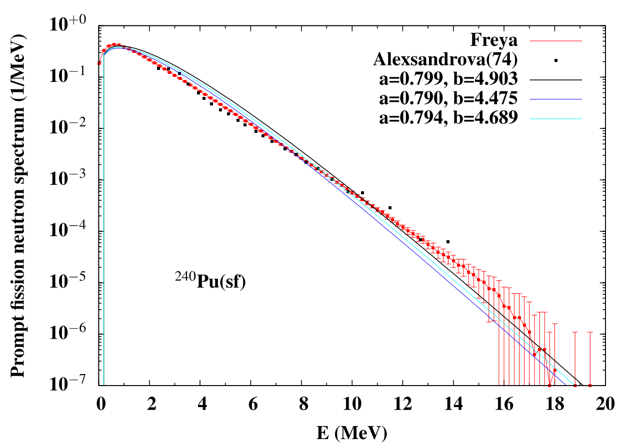

Fig. 8. FREYA-generated neutron energy spectrum for ^{240}Pu spontaneous fission based on the excitation energy parameter, x, uncertainty of Schillebeeck [30]. Also shown is the default Watt fission spectrum based on spectrum parameters used in MCNP. [Note: remove the Watts spectrum uncertainty lines]

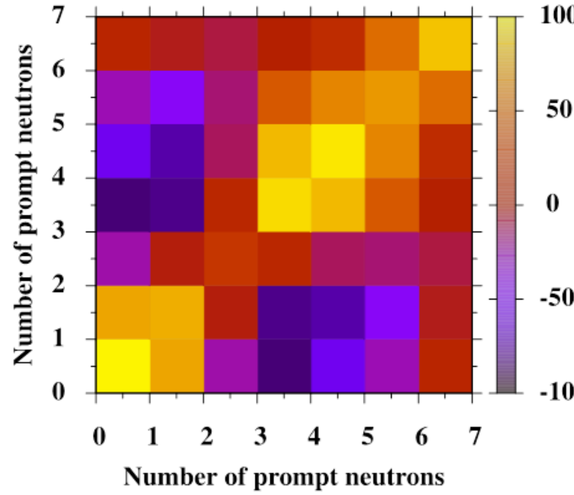

Fig. 9. Correlation matrix (%) for the ^{240}Pu spontaneous fission neutron multiplicity based on results of the FREYA calculations.

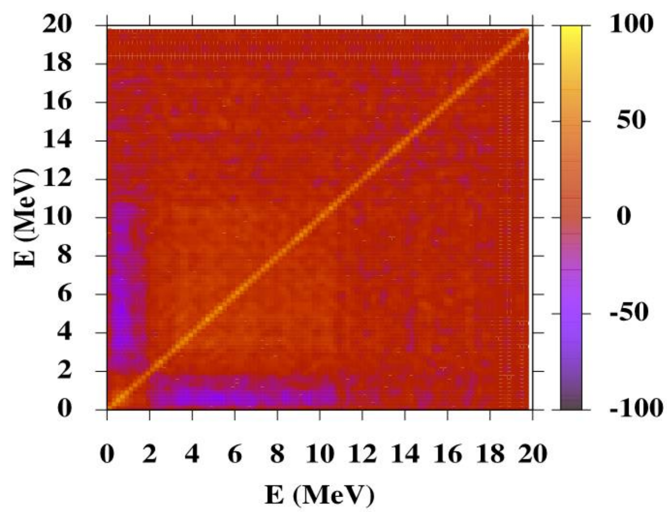

Fig. 10. Correlation matrix (%) for the ^{240}Pu spontaneous fission neutron energy spectrum based on results of the FREYA calculations.

D. Nuclear Decay Data

Nuclear decay modes, half-lives, and decay branches are considered to be relatively well known compared to other nuclear data. Uncertainties are available for most nuclides in the ENDF/B-VII.1 decay files; however covariance data are not available and these data are therefore usually considered to be uncorrelated in uncertainty analysis. Nuclear decay data for spontaneous fission half-lives and branching fractions, decay alpha particle emission energies and intensities that contribute to passive neutron emission uncertainty are of particular interest in this work.

E. Fission Product Yields

Fission product yields are important to the evaluation of signatures during reactor irradiation of nuclear fuel, for some active neutron interrogation measurement techniques, and nuclear fallout analysis. The yields describe the distributions of fission products resulting from neutron-induced (and spontaneous) fission that depend on both the fission target material and incident neutron energy causing fission. ENDF/B-VII.1 data are based primarily on the 1992 evaluations of England and Rider.

Uncertainty values are given for the fission yields; however no covariance data were published with the evaluations. The direct fission yields are highly correlated in charge (Z) and mass (A), and isomeric state (I) and consideration of these correlations is essential for accurate uncertainty analysis. Consequently, retroactive approaches have been applied to develop yield covariance data, but results are generally sensitive to assumptions used in data reconstruction. These approaches are performed without access to all information and models used in the original evaluations, which can lead to possible inconsistencies between yield values and the covariance data.

Fission yield covariance data have been generated previously [33] using a Bayesian approach that is constrained by the cumulative yields, the decay chains, and summation constants. These covariances have been applied for fission product uncertainty analysis using the ORIGEN burnup code In addition to generating covariances, some modification of the direct fission yields was required to preserve the cumulative yields and the end of decay chains that are generally based on experimental measurements. These changes were required due to differences in the decay data in ENDF/B-VII.1 and the older decay data used in the evaluations by England and Rider. There is consensus on the need to revitalize capability to measure and evaluate fission product yields for many applications, and improving fission yield data is identified as a priority topic by the US NDWG.

IV. Covariance Applications with Kiwi

Libraries for uncertainty analysis of neutron transport using MCNP were generated using the Kiwi interface [15]. A Monte Carlo sampling technique implemented in the Kiwi code is used to generate sets of randomly sampled nuclear data libraries. Each randomly sampled library, or realization, represents a new set of cross sections varied within the range of data uncertainty attributed to the measurements.

Kiwi applies principal component analysis to covariance matrices from the ENDF/B library to quantify the related uncertainty and correlations for all available reaction channels. ENDF/B-VII.1 covariance data are first pre-processed onto a common set of energy bins, and then random library realizations were generated. Given a covariance matrix M, Kiwi extracts a set of eigenvalues λ and eigenvectors Λ such that the most important eigenvectors correspond to the largest eigenvalues. After obtaining the eigenvalues and eigenvectors, Kiwi draws a random vector V from a Gaussian distribution, by default, with σ = 1.0, and constructs a random realization vector R such that

$$R_i = \sum_j \eta_j \Lambda_{j,i}$$

where

$$\eta_j = \sqrt{\lambda_j}\,\Lambda_j$$

and the summation is carried out for each realization, j. Vector R is a linear combination of eigenvectors, where each eigenvector is weighted by η_j. The principal eigenvectors (with large λ_j) receive larger weights on average. Small negative eigenvalues arise from round-off error and are discarded prior to constructing vector η, to avoid complex realizations. A new realization is constructed from data in the original ENDF/B-VII.1 library L and vector R by multiplication: $L\,(1 + R)$.

For data with large relative uncertainty (30% or higher), Kiwi sampling may produce unphysical negative values. These can be dealt with either by truncating negative values to 0, or by treating the uncertainty distribution as log-normal rather than Gaussian. In that case, the final realization is constructed by $L\,e^R$.

Kiwi generates an ENDF-6 formatted nuclear data library for each realization. Processed with NJOY-2012, these sampled libraries are then processed into the ACE format to be used by MCNP, or they can be processing into other code formats. An example illustrating the variations in ten random realizations of the ^{239}Pu (n,f) cross section is shown in Fig. 11, as the vector R, the change in the cross section relative to the ENDF/B-VII.1 values.

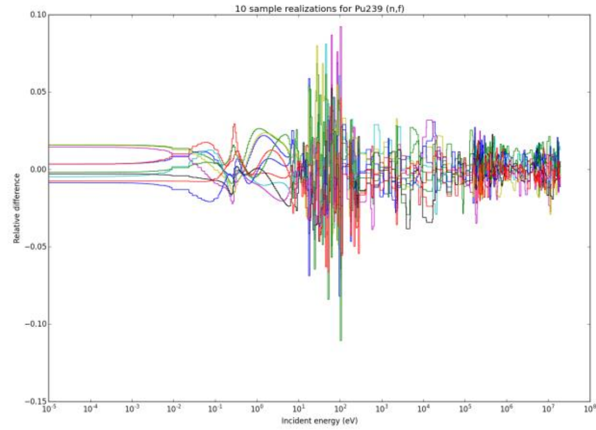

Fig. 11. Vector R for ten sample realizations of the ^{239}Pu (n,f) cross sections based on ENDF/B-VII.1 covariance data.

By performing separate MCNP calculations using the perturbed cross section data sets, distributions in the results can be obtained and assessed for impact due to the data uncertainty. However, identifying individual data contributions to the total uncertainty is needed to rank and prioritize data needs. This can be done by limiting the uncertainty sampling to individual data components. However, this is untenable for compilations like ENDF/B-VII.1 due to the massive amount of data that must be sampled individually.

To obtain information on individual data contributions, data analysis is being performed to correlate the MCNP results and the data variations to identify the data components with the highest correlations or anticorrelations. The number of input data parameters is large, so the task of searching for significant correlations is automated using Python scripts in a data analysis framework used with Kiwi.

V. Analysis of Reactor Materials Production

Another potentially important class of problems involves material production by reactor irradiation. Nondestructive assay or nuclear forensics measurements of material or effluents produced by reactor irradiation can provide information that can be used to verify declared materials during routine operations and also detect undeclared activities or diverted material. This category of problems involves the additional complexity of uncertainties in the material compositions due to modelling of material generation and depletion by neutron transmutation. Unlike most MCNP analyses that involve static material compositions, analysis of uncertainties in irradiated materials must address time-dependent transmutation and time-dependent nuclear data importances. The transmutation processes involve neutron reaction cross section data, decay data and fission product formation by fission (yields). Current uncertainty analysis tools generally do not address transmutation or decay processes.

Direct perturbation of the nuclear data is possible if there are a small number of input data of interest, i.e., as in the case of spontaneous fission and (α,n) neutron sources. However, the general transmutation equation contains about 50,000 different data parameters; therefore a massive computational effort is required to compute sensitivities for all data by perturbing each individual parameter.

A more efficient approach for computing data sensitivities is to use perturbation theory for the transmutation equations [34]. Perturbation theory requires solving the adjoint transmutation equation given by,

$$A^T(\alpha)N^*(t) = -\frac{dN^*(t)}{dt} \qquad t \in (T_f, 0)$$

and $$N^*(t = T_f) \to R$$

where A^T is the transpose of the transition matrix A containing nuclear transition data elements α (neutron cross sections, nuclear decay data and branching fractions, and fission product yields), $N^*(t)$ is the adjoint solution of the nuclide concentrations at time t, T_f is the time of interest after irradiation, and R is the response of interest.

The adjoint equation is very similar to the forward transmutation equation except the time derivative is negative and the transmutation matrix is transposed. With the negative derivative, the adjoint is effectively solved backwards in time as a final-value problem, where the final condition is equal to the response vector. After performing the forward and adjoint transmutation calculations, sensitivity coefficients are computed for an arbitrary data parameter α using the expression,

$$S_\alpha = \frac{\alpha}{R}\int_0^{T_f} N_R^*(t)\frac{\partial A(\alpha(t))}{\partial \alpha} N^T(t)dt$$

Therefore, one forward and one adjoint solution can provide sensitivities for all data in the ~50,000 element transmutation matrix.

Recently a new solution algorithm based on the Chebyshev Rational Approximation Method was implemented in ORIGEN which provides the capability to perform adjoint transmutation calculations. This capability is publicly available ORIGEN code released in SCALE version 6.2 [35]. As an illustrative example of a transmutation problem, forward and adjoint ORIGEN calculations were performed for typical commercial fuel to identify the dominant nuclear data components important to the production of ^{238}Pu. The sensitivity coefficents for all nuclear data are obtained and ranked. The dominant data elements are listed in Table I.

Table I

NUCLEAR DATA SENSITIVITY COEFFICIENTS FOR ^{238}PU PRODUCTION FROM URANIUM TRANSMUTATION

Parent nuclide	Product nuclide	Data category	Data type (α)	S_α
^{235}U	^{236}U	Cross section	(n,γ)	0.5928
^{236}U	^{237}U	Cross section	(n,γ)	0.5877
^{237}Np	^{238}Np	Cross section	(n,γ)	0.6136
^{241}Pu	^{241}Am	Decay data	Half life	0.2132
^{238}U	^{239}U	Cross section	(n,γ)	0.2155
^{242}Cm	^{238}Pu	Decay data	Branching fraction	0.2182
^{241}Pu	^{241}Am	Decay data	Branching fraction	0.2176
^{242}Am	^{242}Cm	Decay data	Branching fraction	0.2175
^{239}Pu	^{240}Pu	Cross section	(n,γ)	0.1740
^{238}U	^{237}U	Cross section	(n,2n)	0.1576
^{238}Pu	^{239}Pu	Cross section	(n,γ)	-0.1475

These quantify the dominant nuclear data for ^{238}Pu production. In this example, the final ^{238}Pu concentration is most sensitive to the neutron capture cross sections of ^{237}Np, ^{235}U, and ^{236}U, which indicates that the major buildup chain for ^{238}Pu in this system, and starts with ^{235}U through the chain:

$$^{235}\text{U (n,}\gamma) \to {}^{236}\text{U (n,}\gamma) \to {}^{237}\text{U } (\beta^-) \to {}^{237}\text{Np (n,}\gamma) \to {}^{238}\text{Np } (\beta^-) \to {}^{238}\text{Pu}$$

High sensitivities for decay branching-fractions of ^{242}Cm alpha decay, ^{241}Pu, and ^{242}Am indicate that ^{238}Pu buildup from the chain beginning with ^{238}U also plays a significant role.

As currently implemented, this capability provides sensitivities of all nuclear data to the calculated concentration of any nuclide or a response due to aggregate nuclides. The sensitivities give the importance of the data to the calculated result, but currently does not reflect the uncertainty of the data. The next challenge in this development is to integrate the covariances for these data to calculate not only sensitivities, but the uncertainty contributions for all nuclear data.

VI. MULTIPLICITY COUNTER APPLICATION MODEL

The Epithermal Neutron Multiplicity Counter (ENMC) [36], a state-of-the-art neutron detector for fissile material measurements, was selected as an example case to demonstrate application of the covariance data and uncertainty tools developed for this work, and to demonstrate the mechanics of integrating the large set of nuclear data in a real-world problem. Neutron coincidence and multiplicity counting is routinely used to measure plutonium materials and is used for nonproliferation applications and arms control measurements. The technique uses the time structure of the detected neutrons to distinguish spontaneous fission, (α,n) production, and induced fission contributions to the total counting rate to determine Pu mass of the measured item [37].

The case of ENMC measurement of plutonium oxide is analysed to evaluate total uncertainty and uncertainty contributions from nuclear data. While performance of this system is well understood and has relatively small uncertainties, it was selected because it is widely encountered, the counter is used at LANL, and this case exercises a broad range of data addressed in this work, including:

- plutonium and oxygen neutron reaction cross sections,
- prompt neutron-induced fission data,
- ^{240}Pu spontaneous fission neutron spectrum,
- P(ν) for ^{240}Pu spontaneous fission,
- absolute intensity of spontaneous fission neutrons,
- intensity of neutrons emitted from (α,n) reactions, and
- neutron spectrum from (α,n) reactions.

Demonstration using the ENMC was desired before applying the tools to more complex problems.

A. *The ENMC Detector*

The ENMC detector consists of 121 high pressure (10 atm) ^{3}He tubes dispersed in a polyethylene matrix. It has a detection efficiency of around 65% and a die-away time of ~19 μs. The detector configuration is shown in Fig. 8. MCNP6 was used to calculate the probabilities of single counts, double counts (correlated pairs) and triples (correlated sets of three counts).

Pu items with different masses (1 g, 700 g and 5.8 kg PuO_2) were analysed to assess the impact of level of subcritical neutron multiplication. Two different isotopic compositions (weapons grade and non-weapons or reactor grade) were also simulated to investigate the impact of variations in the spontaneous fission and (α,n) neutron emission reactions.

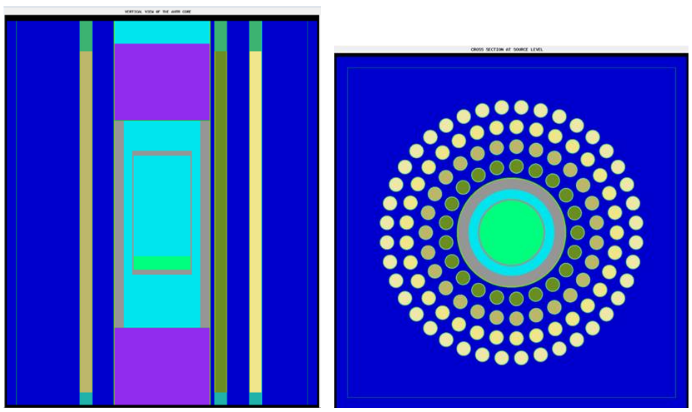

Fig. 8. Vertical and horizontal cross section views of the ENMC geometry, showing the central measurement cavity and PuO_2 material (1) and 121 ^{3}He tubes in four rings (2).

B. *ENMC Uncertainty Analysis*

There are three different analysis methods widely used for plutonium mass determination based on the measured count rates: (1) passive calibration curve, (2) known-alpha or multiplication corrected method, and (3) multiplicity analysis. The passive calibration curve method uses a calibration curve of doubles versus ^{240}Pu effective mass established with reference standards. The known-alpha method is similar but the doubles rate is corrected using the doubles to singles ratio to estimate the effect of neutron multiplication. Neutron multiplicity uses all three measured quantities linked through point model equations to give values for ^{240}Pu effective mass, alpha value that denotes the (α,n) to spontaneous fission neutrons created in the item, and neutron multiplication. Each method applies measured counts differently and emphasizes different nuclear data. Therefore, the methods are expected to exhibit different uncertainties depending on their sensitivity to the different nuclear data.

The Kiwi code was used to generate 100 perturbed nuclear data libraries based on ENDF/B-VII.1 data and covariances. Covariances for prompt fission $\bar{\nu}$ and PFNS are also applied by Kiwi when generating the libraries. The nuclear data that were varied for this study included cross sections and outgoing fission neutron energy and multiplicity for ^{236}Pu, ^{237}Pu, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{244}Pu, ^{246}Pu, and ^{16}O. The perturbed libraries were then used in the MCNP model to assess the effect of these uncertainties on the neutron transport and multiplication inside the PuO_2 items. Uncertainties attributed to the detector materials were not addressed.

A sensitivity study was made to determine the importance of the multiplicity distribution, $P(\nu)$, of the dominant spontaneous fission neutron source, ^{240}Pu. The reference distribution was perturbed to increase and decrease $P(\nu)$ by 1 standard deviation. This gave three $P(\nu)$ distributions that were used to determine the effect of multiplicity uncertainty on the measurements.

ENDF/B-VII.1 does not contain covariance data for spontaneous fission (except for ^{252}Cf) and neutron yield and energy spectrum uncertainties are therefore not included in the Kiwi analyses. The approach adopted to address spontaneous fission neutrons was to apply the FREYA model and experimental data to estimate covariances for spontaneous fission of ^{240}Pu. These covariances were then sampled and input to the MCNP6 model as a fixed neutron source. The effect of uncertainties in the total neutron emission rate and energy spectrum were analysed separately from other effects. [Expand this section]

The dominant (α,n) neutron sources in the ENMC model is from alpha particles emitted by ^{239}Pu, ^{240}Pu and ^{241}Am. Uncertainties in the neutron emission rate were estimated using the covariances developed for the ^{17,18}O(α,n) cross sections and stopping powers described in this paper. Uncertainties in the decay data, alpha particle intensity and energy, were not considered. The covariances were sampled to generate 1000 realizations of the data libraries used by the ORIGEN code applying the SOURCES neutron source method. The resulting distribution of the neutron emission rate (n/s) calculated by ORIGEN has a relative standard deviation of about 1.2%. Breakdown of these values indicates that 0.4% of the effect arises from the cross section and the rest is attributed to the stopping power data.

The (α,n) neutron spectrum is also calculated by SOURCES using branching fractions for product nuclide level formation. Many of the branching levels are calculated using the GNASH code and no uncertainty is available for this data. For our work, we applied a low-fidelity approach that compared the calculated spectra and measured spectra from different experiments. These data included thick target measurements of Jacobs and Liskien [38] using mono-energetic incident alpha particle energies, and measured spectra for plutonium oxide by Herold [39] and Anderson [40]. The (α,n) spectrum depends on the Pu isotopic concentrations present as their alpha particle energies are unique. For typical plutonium compositions, an incident alpha particle energy of 5.5 MeV would be appropriate, but for low ^{238}Pu and ^{241}Am compositions, an average alpha particle energy of 5.0 MeV is more representative. Both the 5.5 MeV and 5.0 MeV Jacobs and Liskien spectra were evaluated, although the differences were found to be small when applied in the ENMC model. The different energy spectra are compared in Fig. 9.

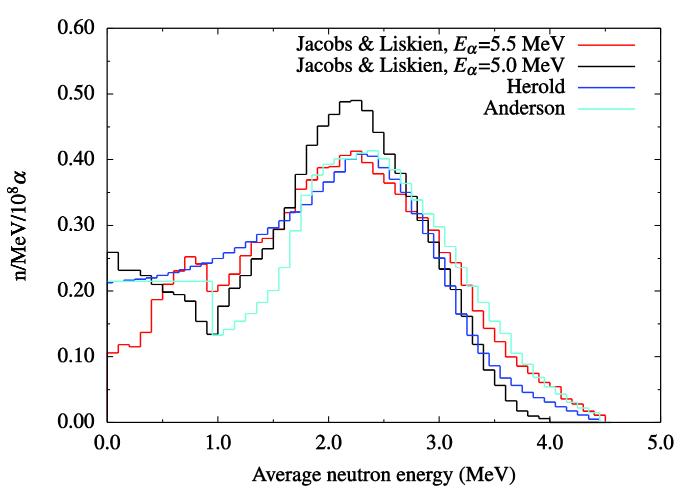

Fig. 9. Neutron energy spectra from SOURCES 4C calculations and measurements of Jabobs and Liskien, Herold, and Anderson.

C. Results

Neutron transport calculations were performed for the MCNP6 model of the ENMC with different plutonium isotopic compositions and item masses. The nuclear data for each of the data categories discussed in this paper were varied independently to quantify the impact on the measured Pu mass based on the count rates as derived using the three different data reduction methods.

The results of the ENMC uncertainty analysis are summarized in Table II for the case of the 700 gram PuO_2 item and a non-weapon reactor grade Pu isotopic vector. The table breaks down the uncertainty contributions from all neutron cross section and the PFNS, and contributions from passive neutron emission. For the non-multiplication-corrected plutonium mass the standard deviation of the derived mass is 0.6% and for multiplicity calculated mass is 0.09%. This indicates that the passive calibration curve (or "non-multiplication-corrected") analysis technique is the most sensitive to uncertainties in the nuclear data. These results illustrate that the nuclear data uncertainties are not only specific to the measurement system, but also the data reduction methods.

The impact of uncertainties in the neutron cross sections and prompt fission data, as applied by Kiwi in the ACE libraries, include contributions from many materials and reaction processes. To identify the dominant contributor to uncertainty, a critical step in identifying data weaknesses, LLNL further analysed the data by correlating MCNP results with variations in each of the individual data values. Initial studies indicated that the dominant effect on the measured Pu mass was due to the PFNS, which masked many other lower-importance data contributions. Therefore, MCNP uncertainty calculations were performed with and without the PFNS variations.

Further correlation analysis was performed for each reaction type and energy group to help isolate the specific nuclear data and energy regions contributing to these uncertainties. In particular, the neutron singles, gated doubles, and gated triples are highly sensitive to changes in the ^{239}Pu PFNS (and to a lesser extent to the ^{240}Pu PFNS). Softening of the PFNS (i.e., higher probability of neutrons at lower energies) is observed to be positively correlated with number of detected neutrons, while hardening of the spectrum is anti-correlated with counts. The largest correlation in the ^{239}Pu PFNS occurs in the energy region near the peak of the spectrum. Other important contributions include ^{239}Pu (n,γ) and various reactions on other Pu isotopes.

Comparing different item masses and isotopic compositions reveals interesting differences. The sensitivity to ^{240}Pu (n,γ) decreases for the weapons grade sample compared to reactor grade sample. Both the large and small mass configurations are less sensitive to the ^{239}Pu PFNS compared to the standard configuration. The only significant sensitivities to ^{16}O appear for the small mass case (1g Pu), where ^{16}O(n,γ) is anti-correlated with multiplication-corrected Pu mass. No single nuclear data quantity dominates the sensitivity for the small mass simulations.

The largest uncertainty contribution to the reactor grade Pu case is ^{240}Pu(n,γ) cross section in the energy region around 600 keV.

The results in Table II indicate that uncertainties in passive neutron emission data are major contributors to measurement uncertainty and can exceed the uncertainties in ENDF/B-VII.1 data that are typically addressed in uncertainty studies. The passive neutron emission rates only contribute significantly to the uncertainty on the multiplication-corrected plutonium mass, but this is a widely used technique. This level of uncertainty is comparable with the observed measurement uncertainty on good quality measurements and clean items. The uncertainty of the multiplicity distribution and energy spectrum of ^{240}Pu spontaneous fission contribute almost equally to the uncertainty of all three techniques, but at a level less than the observed measurement error on all but the most careful measurements. The uncertainty on the (α,n) spectrum only contributes to the multiplication-corrected plutonium mass at a moderate level.

TABLE II

SUMMARY OF NUCLEAR DATA UNCERTAINTIES ON THE PU MASS MEASUREMENTS USING THE ENMC DETECTOR

Data type	Multiplication-corrected	Non-multiplication-corrected	Multiplicity calculated
700 gram PuO_2 item			
Neutron cross sections	0.04%	0.61%	0.09%
PFNS			
SF neutron intensity	0.64%	1.02%	1.09%
(α,n) neutron intensity	0.62%	0.10%	0.00%
P(v) for ^{240}Pu	0.16%	0.44%	0.38%
SF energy spectrum ^{240}Pu	0.18%	0.40%	0.50%
Energy spectrum (α,n)	0.27%	0.04%	0%

VII. CONCLUSIONS

Uncertainty in computational studies introduced by nuclear data represents an important component of modelling uncertainty, particularly for applications in safeguards and nuclear non-proliferation which can frequently rely on data that is developed from theory and has a limited experimental basis.

This work describes an analysis of nuclear data uncertainties related primarily to passive neutron measurement of nuclear items as frequently encountered in nuclear safeguards and treaty verification. These applications are found to depend on nuclear data that extends beyond what is currently available in ENDF/B-VII.1 and codes such as MCNP. In cases where data exist in ENDF/B-VII.1 they are found in some cases to differ from the consensus data values used by the safeguards community. When covariances are available in ENDF/B-VII.1, they have been used directly in this work for uncertainty analysis. However, uncertainties are not reported for all nuclear data and in many cases the associated covariance data needed for uncertainty analysis is not provided. In these cases, methods for retroactive covariance data development have been explored in our work by re-evaluating experimental data and using physics modelling codes.

Analysis of the nuclear data uncertainty for national security applications required a diverse and somewhat eclectic tool set. This is a direct consequence of the diverse nature of the nuclear data that describes the different physics phenomena. While this work does not provide a unified framework for uncertainty analysis, these covariance files and tools provide the basic building blocks of a modern computational data uncertainty capability.

Application of the tools and covariances to the analysis of the ENMC detector illustrates several important findings:

1) Uncertainties attributed to spontaneous fission and (α,n) neutron source intensity and neutron energy spectrum are important and can exceed contributions from data considered in ENDF/B-VII.1. Uncertainties for these data are not included in most evaluations and methods to treat these uncertainties are not available in current tools.
2) The importance of nuclear data uncertainties are highly dependent on the measurement system and the characteristics of the nuclear material being measured.
3) Uncertainty can depend not only on the detector system and material, but also on data reduction methods used to analyze the measurements.

Quantitative assessment of data uncertainties can provide important information to guide future nuclear data measurements by funding agencies and the measurement community by informing where data uncertainties have the greatest impact on applications. However, it is insufficient to define nuclear data prior data needs for broad applications such as safeguards, nuclear forensics, nonproliferation detection, and other national security studies. To address the question of nuclear data needs, it is critical to define the specific problem: the detection technology, design, observable signatures, material configuration, and data reduction methods.

This work describes an initial venture into generating the covariance data and the application tools to address a broad application space. While these tools are currently not integrated for general systems analysis, they do represent the basic components of such a toolset. Integrating elements of this work into codes such as MCNP, already widely used for nonproliferation studies, represents one avenue for integration of methods and data. This work demonstrates the potential utility of these tools for total uncertainty analysis and to support quantitative nuclear data needs assessments for specific application scenarios.

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