Impact of Nuclear Data Uncertainties on Calculated Spent Fuel Nuclide Inventories and Advanced NDA Instrument Response

Jianwei Hu and **Ian C. Gauld**

Reactor and Nuclear System Division, Oak Ridge National Laboratory[1],
1 Bethel Valley Road, PO Box 2008, MS-6172, Oak Ridge, TN 37831-6172,
United States
E-mail: huj1@ornl.gov, gauldi@ornl.gov

Abstract:

The U.S. Department of Energy's Next Generation Safeguards Initiative Spent Fuel (NGSI-SF) project is nearing the final phase of developing several advanced nondestructive assay (NDA) instruments designed to measure spent nuclear fuel assemblies for the purpose of improving nuclear safeguards. Current efforts are focusing on calibrating several of these instruments with realistic spent fuel assemblies at two international spent fuel facilities. Modelling and simulation are expected to play important roles in predicting nuclide compositions, neutron and gamma source terms, and instrument responses in order to assist instrument calibrations. As part of NGSI-SF project, the purpose of this work is to assess the impacts of nuclear data uncertainties on the calculations of both spent fuel content and instrument responses.

Nuclear data is an essential part of many modern nuclear codes, including nuclear burnup codes and nuclear transport codes. Such codes are routinely used for analysis of spent fuel and NDA instruments. Hence, the uncertainties existing in the nuclear data used in these codes affect the accuracies of such analysis. In addition, nuclear data uncertainties represent the limiting (smallest) uncertainties that can be expected from the nuclear code predictions, and therefore define the highest attainable accuracy of the NDA instrument. This work studies the impacts of nuclear data uncertainties on calculated spent fuel nuclide inventories and the associated NDA instrument response. Recently developed methods within the SCALE code system are applied in this study. The Californium Interrogation with Prompt Neutron instrument was selected to illustrate the impact of these uncertainties on NDA instrument response.

Keywords: nuclear data; uncertainty; spent fuel safeguards; CIPN; NDA.

1. Introduction

The U.S. Department of Energy Next Generation Safeguards Initiative Spent Fuel (NGSI-SF) Project is nearing the final phase of developing several advanced nondestructive assay (NDA) instruments designed to measure spent nuclear fuel assemblies for the purpose of improving nuclear safeguards [1, 2]. As the project completes the initial R&D and instrument development phase, current efforts are focusing on instrument deployment and experimental measurements at the Swedish Central Interim Storage Facility for Spent Nuclear Fuel (Clab), operated by the Swedish Nuclear Fuel and Waste Management Company SKB, and at the Post Irradiation Experimental Facility at the Korea Atomic Energy Research Institute in the Republic of Korea (ROK).

The advanced NDA instruments must be accurately calibrated to enable measurement of the plutonium mass and other spent fuel attributes of interest to safeguards with high reliability. Advanced modelling and simulation codes, such as MCNPX [3] and SCALE [4], have been used extensively for instrument design, development, and calibration. Quantifying the uncertainties in these calculations is an essential task required for instrument calibration because these uncertainties will affect the NDA instrument performance prediction and limit the accuracy that can be attained. The uncertainties in calculated spent fuel content arise from various sources, such as irradiation history, burnup, irradiation conditions (e.g., exposure to burnable poisons), etc. These uncertainties are discussed in detail in a separate report [5]. The uncertainties in the underlying nuclear data used by the computer codes also affect the calculated nuclide concentrations in spent fuel and thus the calculated instrument responses for the spent fuel measurement; however, such impacts have not been previously studied under the NGSI program. Nuclear data uncertainties represent the limiting (smallest) uncertainties that can be expected from the code predictions, and therefore define the highest attainable accuracy of the instrument.

In this work, the impacts of nuclear data uncertainties on calculations of spent nuclear fuel content and associated NDA instrument responses are studied. Recently developed methods [6] within the SCALE code system are applied in this study. The Californium Interrogation with Prompt Neutron (CIPN) instrument [7] was selected to illustrate the impact of these uncertainties on instrument response. The study addresses only the uncertainties in the calculated nuclide concentrations of the spent fuel assembly; it does not include the impacts of nuclear data uncertainties on radiation transport calculations of the MCNPX detector model.

2. Uncertainties in nuclear data

Burnup codes are routinely used to calculate nuclide concentrations in spent fuel. These calculations require simulation of both neutron transport and nuclear depletion and decay. There are three main types of nuclear data involved in burnup calculations: 1) neutron cross sections (e.g., fission and absorption cross sections); 2) fission product yields (e.g., fission product generation due to the fission of an actinide); and 3) decay data (e.g., half-lives, branching ratios). Uncertainties exist in all nuclear data; for example, uncertainties exist in the cross-section values, measured half-lives, and branching ratios. In addition, many of the data are correlated, and accurate representations of these data correlations (covariance files) are necessary for rigorous uncertainty analysis.

The majority of the research effort in uncertainty analysis has been directed at expanding the covariance data for nuclear cross sections. The most recent release of the Evaluated Nuclear Data Files, ENDF/B-VII.1 [8], provides extensive data on cross-section uncertainties (covariance data evaluations) for 190 isotopes that are particularly important in nuclear technology applications. The previous release, ENDF/B-VII.0 [9], contained neutron cross-section covariances for only 26 materials, of which 14 were considered a complete representation of the reaction energy range and major reaction channels. The expansion of neutron cross-section covariance data represents one of the major advances in the latest nuclear data library. The neutron cross-section covariance data used in this work were developed prior to the release of ENDF/B-VII.1, and are distributed with the SCALE code system. Selected covariance evaluations were taken from the pre-release of ENDF.B-VII.1, while most of the data were taken from ENDF/B-VII.0, ENDF/B-VI, JENDL, and additional low-fidelity data for more than 300 nuclides developed by U.S. national laboratories under a DOE project for nuclear criticality safety [10]. Cross-section covariances for a total of 401 materials were available.

ENDF/B-VII and other international evaluated nuclear data files currently do not include covariance information for fission product yields, which are highly correlated. The evaluations contain uncertainties for the direct and cumulative fission yields, but not the correlations necessary to apply the data for fission product uncertainty analysis. To support uncertainty analysis for fission products, correlation matrices for direct fission yields have recently been developed by Oak Ridge National Laboratory (ORNL) [6] using the nuclear data and uncertainties in the ENDF/B-VII.0 evaluations, developed by England and Rider [11], and these covariance files have been implemented for use in SCALE. The decay data are generally correlated to a lesser degree, and the uncertainties for decay data are available through ENDF/B-VII. The covariance files are utilized by SCALE for the uncertainty analyses.

3. Uncertainty analysis methods

A newly developed uncertainty analysis tool within SCALE, named Sampler [6], was applied to the burnup calculations used to support NGSI spent fuel analysis in this work. Sampler generates perturbed nuclear data libraries that have been adjusted by Monte Carlo (stochastic) sampling of the data in a manner that is consistent with the uncertainties and correlations in the data. This stochastic sampling of the correlated nuclear data uncertainties is performed using the XSUSA code [12] developed by GRS in Germany. Sampler can be applied to any SCALE sequence (e.g., burnup and criticality calculations). Sampler repeatedly calls the SCALE sequence to perform the calculation, each time using a different set of perturbed nuclear data libraries, and then post-processes the results to obtain the distribution and statistical parameters on the calculated quantities. Figure 1 shows the flowchart of Sampler.

The TRITON module within SCALE is widely used to perform burnup calculations, and is used within the NGSI-SF project to generate the reference spent fuel inventories for the spent fuel assemblies being measured at the Clab facility in Sweden and the assemblies measured in ROK. For each set of the perturbed data libraries, an individual SCALE/TRITON calculation was executed and the responses (e.g., nuclide concentrations in this case) due to the different data libraries were obtained. The variance in the responses attributed to the nuclear data uncertainties can thus be assessed. Sampler will post-process the response distributions to compute statistical parameters (e.g., standard deviation of the concentration of a particular nuclide). Sampler can also perform perturbations to modelling parameters of a system to assess the impacts of uncertainties in material densities, temperatures, dimensions, etc.

SCALE/TRITON couples the two-dimensional deterministic neutron transport code NEWT, which was used in this work, or the three-dimensional Monte Carlo KENO code for the neutron transport calculation, with the ORIGEN code for nuclide depletion and decay calculations. Therefore, uncertainties in the neutron cross sections (used in both the neutron transport and depletion calculation), fission product yields, and nuclear decay data are all included in the total uncertainty analysis.

4. Impact of nuclear data uncertainties on nuclide concentrations

A simplified assembly model of a typical 15×15 PWR design with 16 guide tubes and 1 central instrument tube was developed for this work, and it is shown in Figure 2. All the fuel rods were modelled using a single fuel material mixture (uniform composition). In reality, the fuel content will vary from rod to rod, but for the purposes of this study, uniform rod compositions were used because the focus here is to establish minimum uncertainties due to the nuclear data. The fuel has an initial ^{235}U enrichment of 4.5 wt% and was irradiated to 45 GWd/tU and cooled for 5 years.

A total of 120 separate burnup calculations were performed, with each calculation using a different set of perturbed cross section, fission yield, and decay libraries. By examining the distribution of nuclide concentrations from these calculations, the standard deviation for each nuclide was obtained. Figure 3 shows relative uncertainty in calculated ^{239}Pu content caused by nuclear data uncertainties. As shown, the uncertainty of ^{239}Pu increases with burnup and reaches 1.3% at 45 GWd/tU due to the accumulation of nuclear data uncertainties at higher burnups. Figure 4 shows the distribution of ^{239}Pu content after the 5-year cooling time for all 120 samples, indicating that most of the predicted ^{239}Pu content is within the range of 27 to 28 mol per tonne U (tU) (~0.6% in heavy metal concentration). The mean value and relative standard deviation of the distribution is 27.42 mol/tU ± 1.3%. This value presents the expected uncertainty in the calculated result due to the nuclear data alone. Uncertainties for any other nuclide or calculated quantities can be obtained in a similar manner. The distribution of the results will approach a normal distribution as the number of samples increases.

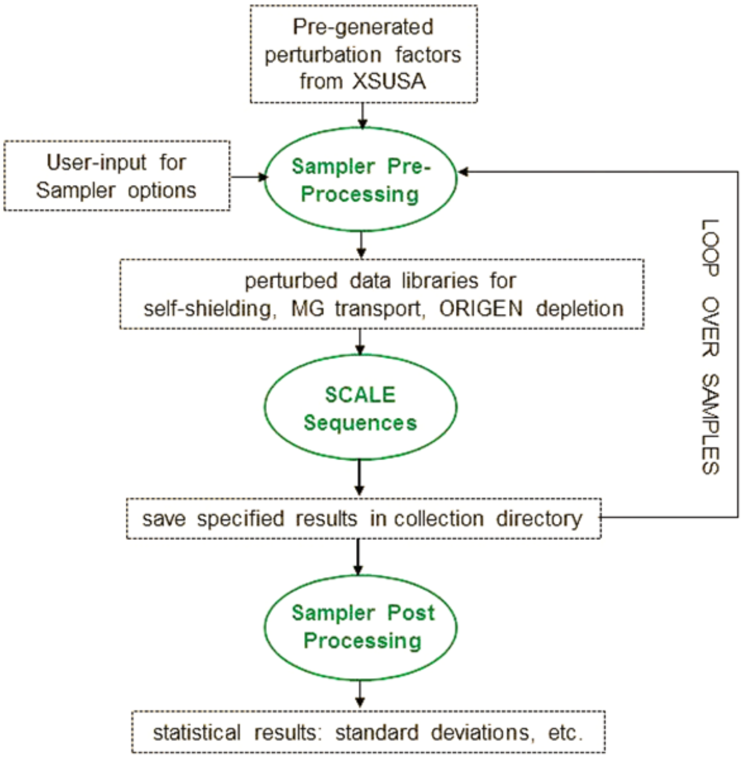

Figure 1. Sampler flowchart [6].

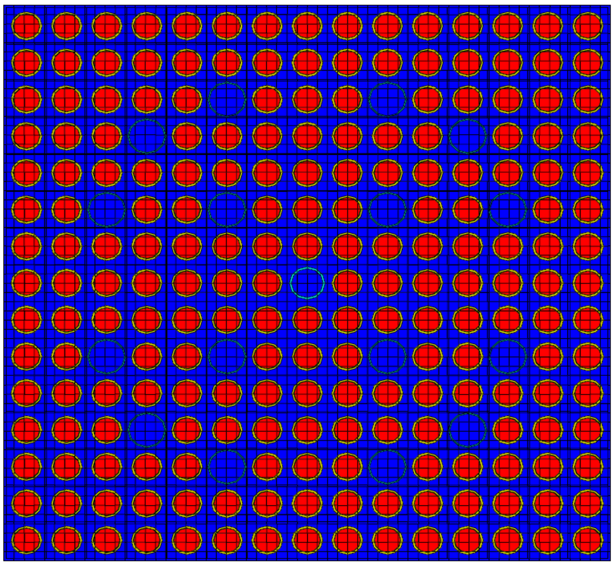

Figure 2. The simplified 15×15 PWR spent fuel assembly as modeled in TRITON.

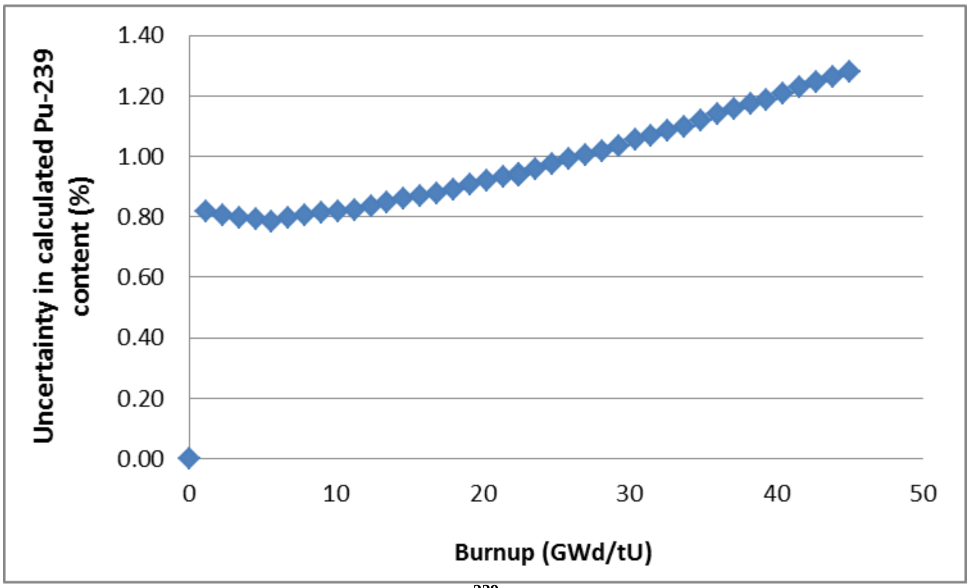

Figure 3. Uncertainty in calculated ^{239}Pu content as a function of burnup.

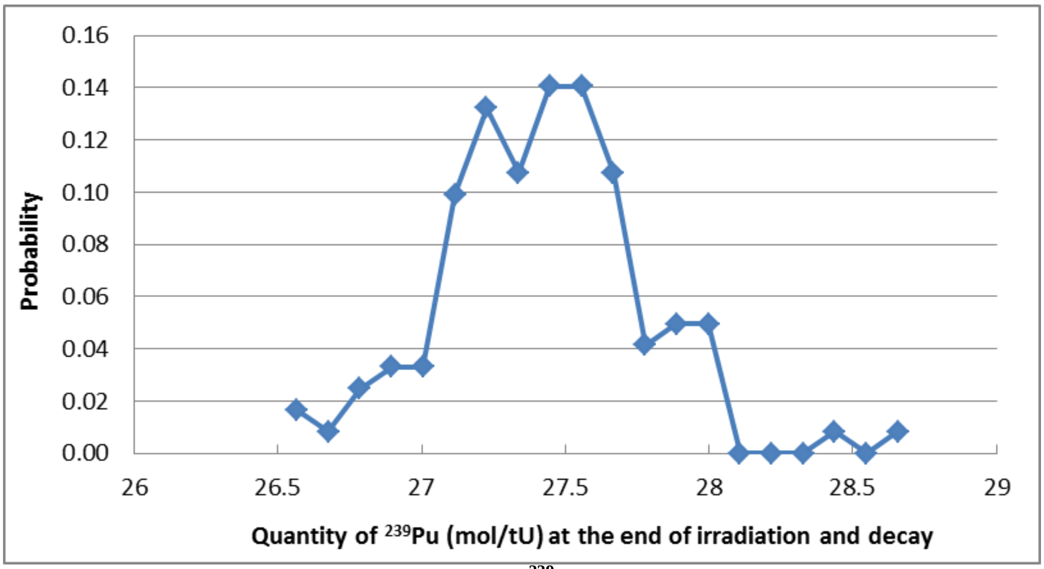

Figure 4. Distribution of calculated ^{239}Pu mass results for 120 samples.

Figure 5 shows the relative standard deviation of the major actinides based on the 120 samples. As shown, the relative standard deviations caused by the uncertainties in nuclear data are generally within 2% for most actinides, and they vary from one nuclide to another because their production paths are different. The standard deviations for ^{239}Pu, ^{240}Pu, and ^{241}Pu, the three major plutonium isotopes, are 1.3%, 1.4%, and 1.0%, respectively. Because ^{244}Cm is a dominant passive neutron source in spent fuel, the large relative standard deviation of ^{244}Cm (6.5%) may have significant impacts on the responses of the NDA instruments that measure passive neutrons. The isotopes ^{235}U, ^{239}Pu, and ^{241}Pu are the primary fissile nuclides in spent fuel, and ^{240}Pu and ^{241}Am are the primary neutron absorbers. These nuclides have a significant impact on the neutron multiplication factor in spent fuel and thus on NDA neutron signals.

Figure 6 shows the relative standard deviation for several important fission products. Compared to the actinides, the fission products uncertainties are much larger because the uncertainties in fission yield data are generally greater than those in neutron cross sections. It is important to note that the fission product yield covariance file used in this study is known to overestimate the fission product uncertainties. Therefore, the results shown here represent conservative estimates of uncertainties in the simulations. As shown, for most fission products, the standard deviation varies from 5% to 10%. The standard deviations for ^{137}Cs and ^{134}Cs are 2.3% and 5.1%, respectively (^{137}Cs and ^{134}Cs are two of the primary photon source nuclides). Many of these nuclides (e.g., ^{133}Cs, ^{143}Nd, ^{149}Sm, ^{154}Eu) are major neutron absorbers in spent fuel, and they have a significant impact on NDA instrument neutron signals.

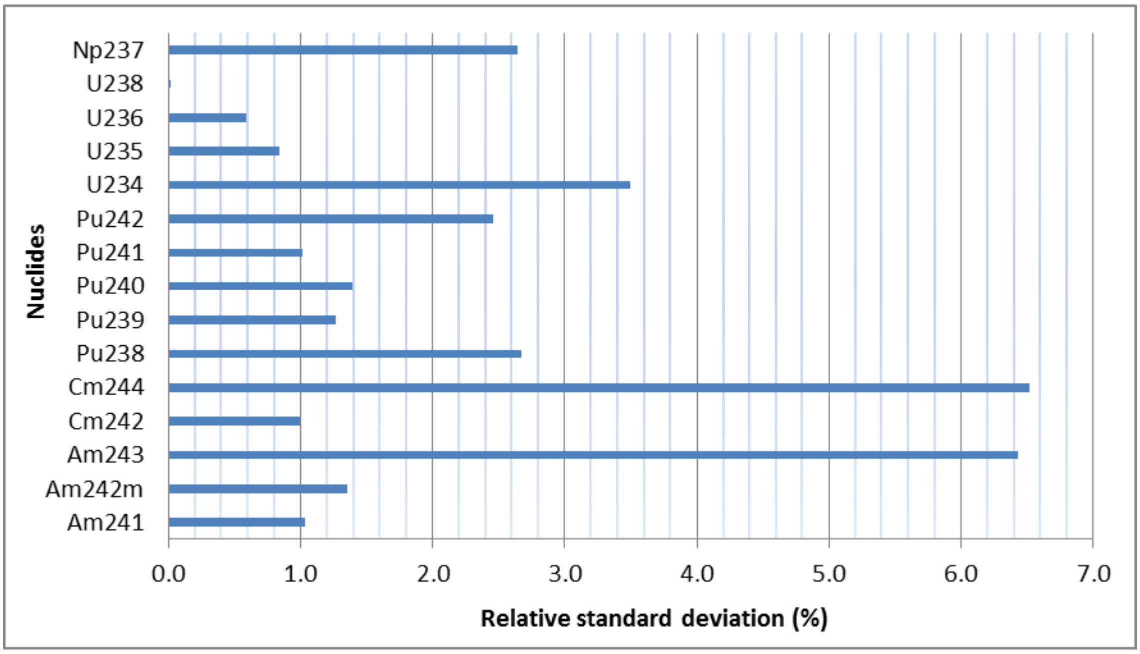

Figure 5. Relative standard deviation of major actinides due to nuclear data uncertainties.

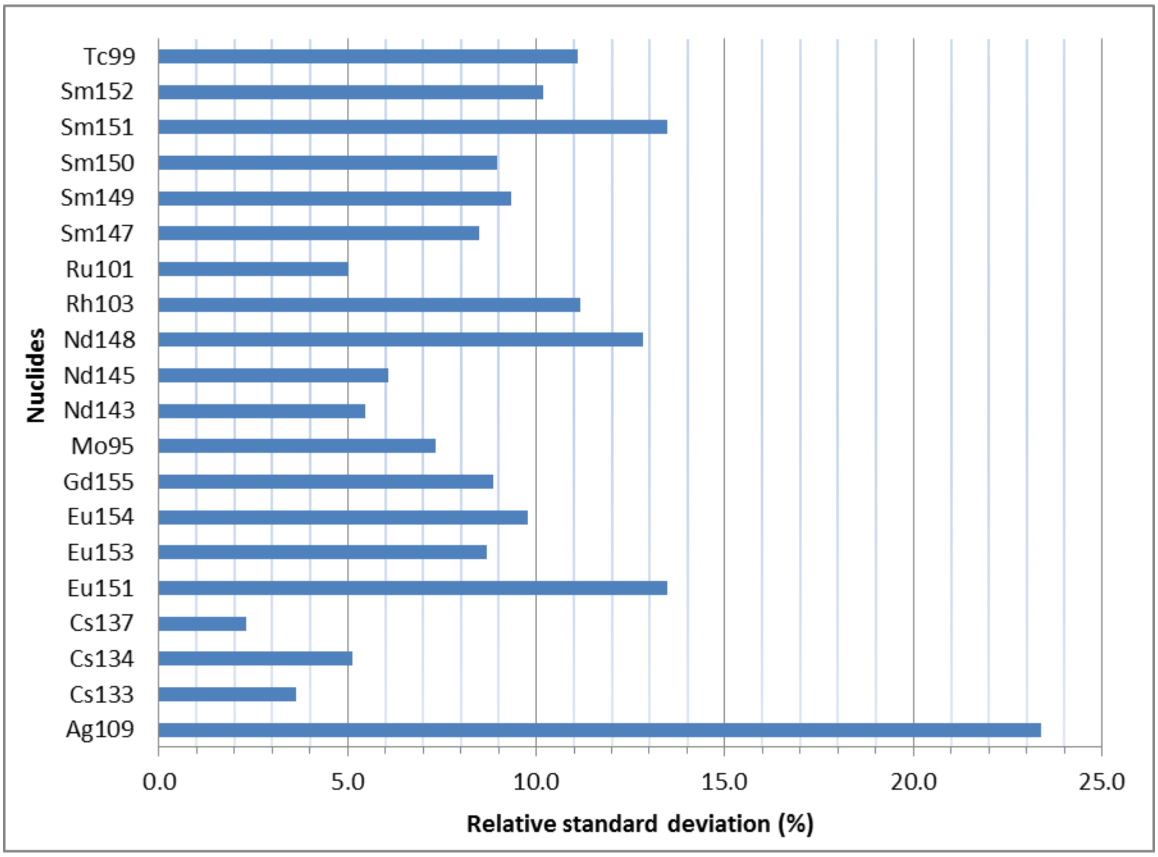

Figure 6. Relative standard deviation of important fission products due to nuclear data uncertainties.

5. Impacts on NDA instrument responses

While the impact of nuclear data uncertainties on the spent nuclear fuel nuclide contents is important to this analysis, ultimately for nuclear safeguards purpose it is the net effect of the nuclide uncertainties on the instrument response that is required. CIPN is one of the advanced NDA instruments developed under the NGSI-SF project that is being used in field tests [2]. CIPN was selected to evaluate the impact of uncertainties for this study because its detection capability extends across the entire fuel assembly.

CIPN is a relatively low-cost and lightweight instrument that resembles a Fork detector, except that CIPN has an active interrogation source (^{252}Cf). CIPN shows promising capability for determining fissile content and detecting diversion of fuel rods in spent nuclear fuel assemblies [7]. Figure 7 shows the cross-sectional views of the CIPN instrument at two axial levels: Z = -3 cm and Z = 3 cm (the center of the assembly is set at Z = 0). As shown, there are four fission chambers in the instrument to detect neutrons and two ion chambers to detect photons. CIPN can operate in both passive and active modes. In the passive mode, the californium source is not present, and the neutrons and photons emitted from the spent fuel assembly itself are measured. In the active mode, the californium source is placed in proximity to the assembly. The neutrons emitted from the californium source will induce fissions in the fuel, and these fission neutrons will add to the neutron signal in addition to the passive neutrons. The difference in neutron counts between the active and passive mode, or the net neutron count, is related to the neutron multiplication factor of the assembly and thus the fissile content [7]. (For photon counts, the active mode is similar to the passive mode because addition of the active neutron source does not appreciably impact the photon counts.) The net neutron counts are mainly driven by the external neutron source (californium) and the multiplication factor, which is primarily determined by the combined effect of several fissile nuclides and neutron-absorber nuclides. In addition to the passive gamma signal, both the passive and active neutron signals have been studied in this work.

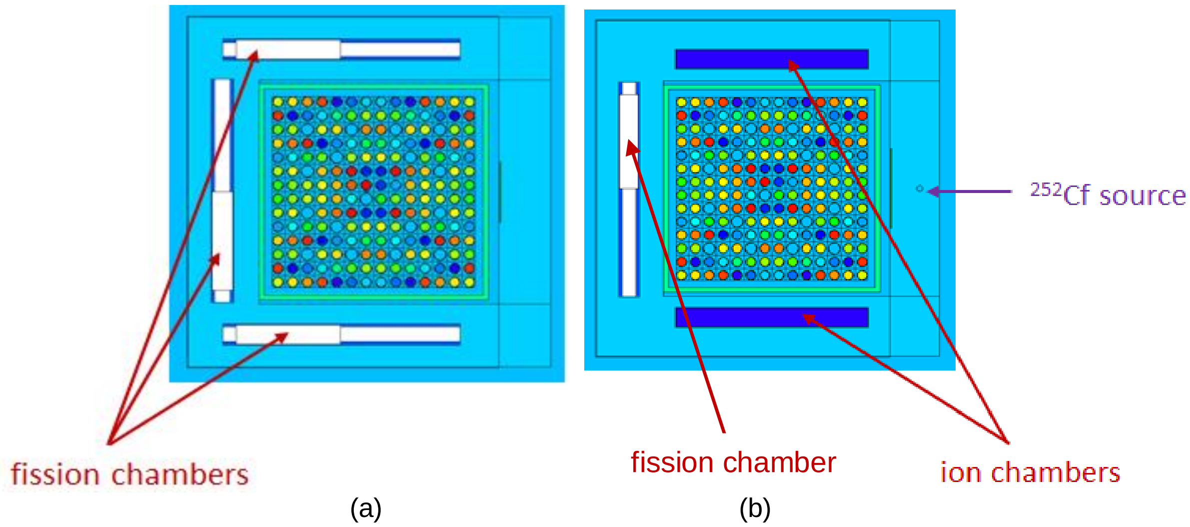

Figure 7. Cross-sectional views of the CIPN instrument at two axial levels: (a) Z = -3 cm; (b) Z = 3 cm.

Given the high computational demand of MCNPX simulation, only 20 detector simulation calculations were performed for this study. These 20 sets, as a subset of the 120 samples, of assembly nuclide concentrations based on the perturbed nuclear data libraries were applied in the MCNPX model used to simulate uncertainties in the CIPN count rates. These assembly nuclide concentrations can also be applied to test any other NDA instruments using different MCNPX models. Figure 8 shows the relative percent difference between the passive gamma count rates for each of the 20 perturbed cases from that of the reference case (in which the nuclear data were not perturbed). For the relatively long cooling time (5 years) used, cesium isotopes, especially ^{137}Cs, are the main gamma sources. As shown, the uncertainties in nuclear data introduce an average uncertainty in the CIPN passive gamma count rates of 2.0% (relative standard deviation), a value that is similar to the uncertainty in ^{137}Cs shown in Figure 6. Figure 9 shows the uncertainty in the passive neutron count rate, dominated by ^{244}Cm. The average uncertainty in the CIPN passive neutron count rates is 6.4%, which is similar to that of ^{244}Cm, as shown in Figure 5. The nuclear data uncertainties have a larger impact on passive neutron count rates than gamma count rates, because ^{244}Cm is more sensitive to nuclear data uncertainties than ^{137}Cs.

The net neutron count rate can be obtained by subtracting the passive count rate from the active count rate. Figure 10 shows the percent difference of the net neutron count rate of the samples from that of the reference case. As shown, the nuclear data affect the CIPN net neutron count rates with a standard deviation of about 1%. The CIPN net neutron count rate is mainly driven by the multiplication of the assembly, which is defined by the geometry and the concentrations of the major actinides and fission products in the fuel. The relatively low impact on net neutron count rate is consistent with the small standard deviations found in the major fissile nuclides (e.g., ^{235}U and ^{239}Pu) and major actinide neutron absorber (e.g., ^{240}Pu), as shown in Figure 5.

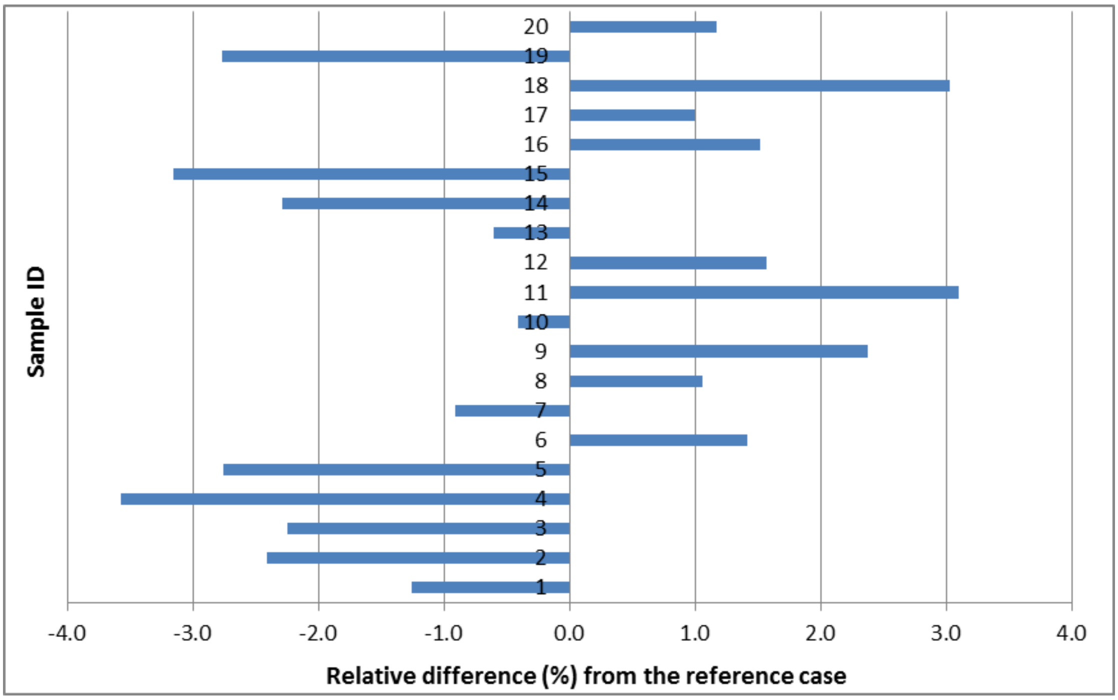

Figure 8. Relative difference of the CIPN passive gamma count rate of the samples from that of the reference case.

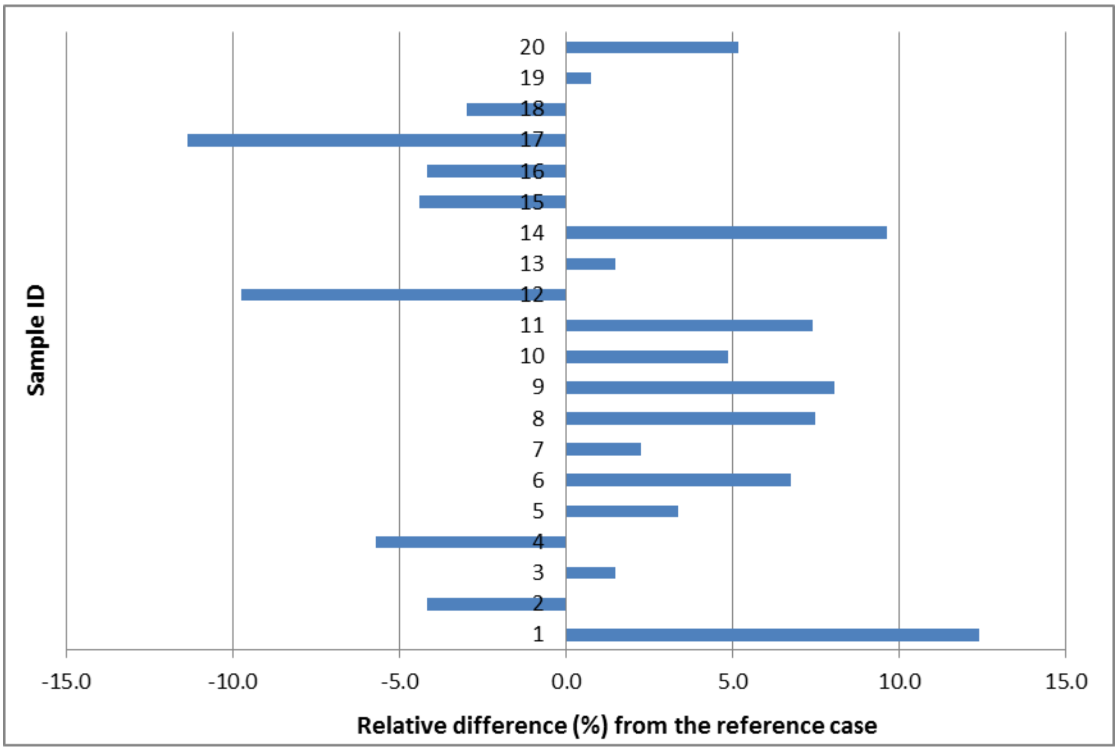

Figure 9. Relative difference of the CIPN passive neutron count rate of the samples from that of the reference case.

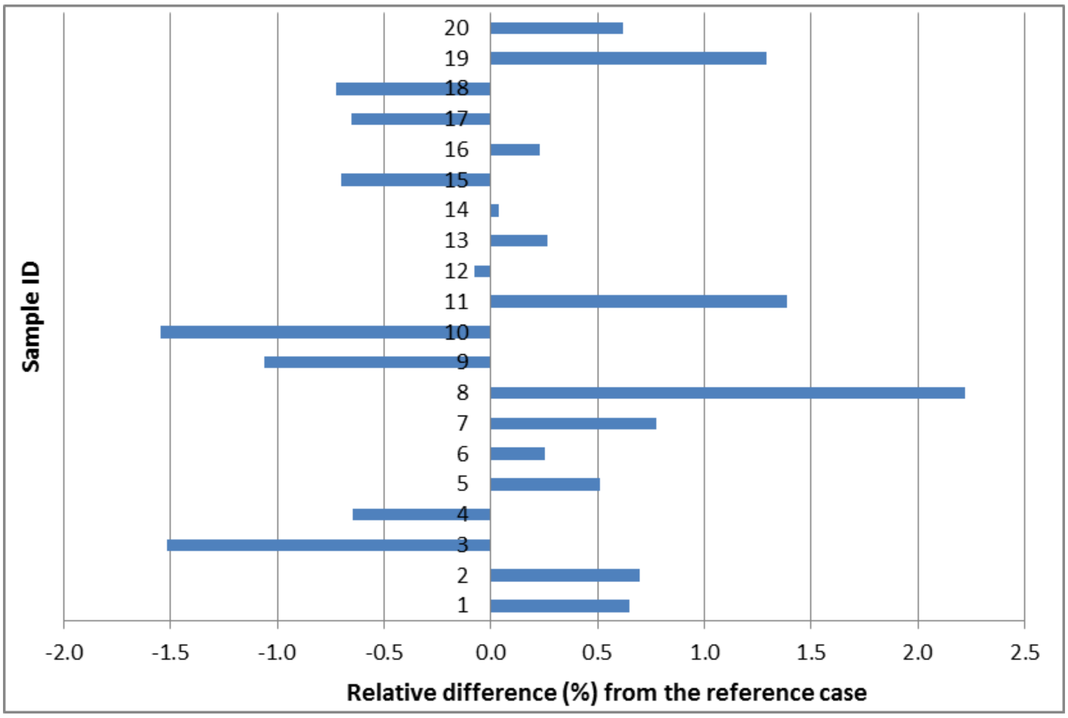

Figure 10. Relative difference (%) of the CIPN net neutron count rate of the samples from that of the reference case.

6. Summary and conclusions

This work has examined the impact of nuclear data uncertainties on nuclide concentrations in spent fuel and the resulting NDA response of the CIPN instrument. Uncertainties in the nuclide concentrations were estimated based on burnup calculations using 120 sets of perturbed nuclear data libraries generated using stochastic sampling of the uncertainty and covariance data. The resulting nuclide concentrations in each case were compared to that of the reference case, in which the nuclear data were not perturbed. To study the impact on the CIPN instrument response, a subset of 20 perturbed sets of assembly nuclide concentrations was imported into the MCNPX model to simulate the uncertainties in the CIPN count rates.

Analysis of the uncertainties is important to the NGSI project because modelling and simulation of the spent fuel assembly concentrations have been extensively used to predict instrument performance, and spent fuel calculations will be required for instrument calibration. The uncertainties in the nuclear data used by the codes represent estimates of the minimum uncertainties that can be realistically expected due to limitations in the accuracy of the basic nuclear data used in the simulations. An alternate and more direct approach to the determination of bias and uncertainties associated with the modelling and simulation would be by experimental benchmarking. However, in the case of the new advanced NGSI instruments, there is a lack of destructive analysis measurements of the spent fuel assembly compositions, and thus no such benchmarks exist. The quantification of uncertainties associated with the nuclear data used by the codes represents one of several options for uncertainty analysis.

The impact of nuclear data uncertainties on the concentrations of major plutonium isotopes in spent fuel is estimated to be approximately 1%, and the impact on most other actinides is less than 4%. For ^{244}Cm, the most important source of passive neutrons in spent fuel, the uncertainties are greater (~7%). Uncertainties in calculated fission product concentrations are greater than those for actinides due to larger uncertainties in fission yield data. As noted previously, the fission product uncertainties are overestimated based on a preliminary version of the fission yield covariance file applied in this study. The impact on the CIPN passive neutron count rates were the largest (~6%), followed by passive gamma (~2%), and net neutron (~1%). The sensitivity of other NDA instruments to nuclear data will vary due to the different responses of the instruments. The assembly nuclide concentrations generated based on the perturbed nuclear data can be used to study the sensitivity of other NDA instruments. This work provides quantitative assessments of the nuclear data uncertainties on nuclide concentrations in spent fuel and also on NDA instrument responses. These values provide a realistic assessment of the impact of nuclear data uncertainties on instrument performance, and represent the expected minimum level of uncertainty in many cases since these uncertainties exclude other sources of uncertainty associated with the NDA measurements.

Finally, in addition to the assessment of total uncertainties in the modelling and simulation due to nuclear data, the methods described in this work may also be applied to evaluate the impact of different types of nuclear data and specific nuclides on the application. Such an approach may be useful to identify specific areas where improved nuclear data would result in lower uncertainties in the advanced NDA instrument performance.

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