Decay Data Uncertainty Propagation in Decay Heat Calculations: A Monte Carlo Approach

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ABSTRACT

A proper characterization of decay heat is essential for nuclear power safety at all stages of the fuel cycle, including reactor operation, spent fuel handling, transportation, reprocessing, and disposal. This work is based on a Monte Carlo approach to quantify the uncertainty propagation of decay data on decay heat calculations using an in-house code named COCODRILO, which is coupled with Serpent2. The calculations are performed for thermal fission pulses of ^{239}Pu and ^{235}U using ENDF/B-VII.1 and ENDF-VIII.0 nuclear decay data libraries. Results are also compared with experimental values at different cooling times. The associated analysis shows the impact of decay data uncertainties on decay heat. Moreover, the study underscores the discrepancies in decay heat uncertainty calculation due to decay data without uncertainty information.

1 INTRODUCTION

Efficient removal of decay heat is critical to preventing core damage and minimizing the risk of radioactive incidents in nuclear reactors and radioactive waste management facilities, ensuring their safe and stable operation. Furthermore, precise decay heat estimation serves as a crucial parameter for safety systems and innovative fuel design of Gen IV reactors. In the context of these advanced reactor concepts, accurate prediction of decay heat is essential for establishing appropriate safety margins. This not only enhances reactor safety but also yields economic benefits. Decay heat can be computed by employing standards such as ANS-5.1[1], ISO [2], and DIN (Deutsch Institute für Normung) [3] or using the summation method [4-5]. Standards offer a parameterized and computationally efficient way to calculate decay heat, particularly made suitable for conventional power reactors in operation. In contrast, the summation method is more extensive in such away that it involves calculating the contribution of radioactive fission products and actinides which are needed to calculate the total decay heat and also the associated uncertainties. Additionally, in the context of decay heat calculations for innovative fuel types, the summation method is appropriate, since there are no established standards for Gen IV reactors. For this work, this second approach is chosen due to its comprehensive and accurate representation of decay heat and associated uncertainties calculations for reactor safety analyses.

The calculation of decay heat using the summation method [Eqn. 1] involves evaluating the decay heat for each isotope $DH_i(t)$ at a given time (t). This calculation requires multiplying the decay rate of an isotope ($N_i\lambda_i$), where N_i is the concentration of isotope i and λ_i is the decay constant of isotope i, by its mean total decay energy ($\overline{E}_i$). The concentration of each isotope i (N_i) can be calculated by solving the Bateman equations, which describe the time evolution of isotopes taking into account the decays and reaction processes using depletion codes such as the Serpent2 Monte Carlo particle depletion codes in this work [6]. Then, the sum of individual isotope decay heat contributions results in the total decay heat, as shown in (Eqn. 1) below.

$$DH(t) = \sum_i N_i \lambda_i(t) \overline{E}_i \qquad \text{(Eqn. 1)}$$

Where,

$\overline{E}_i = \overline{E}_{LPi} + \overline{E}_{EMi} + \overline{E}_{HPi}$

$\overline{E}_{LP,i}$ is the average energy for radiations of light particles for a given nucleus *i*.

$\overline{E}_{EM,i}$ is the average energy for electromagnetic radiation for a given nucleus *i*.

$\overline{E}_{HP,i}$ is the average energy for radiations of heavy charged particles and neutron for a given nucleus *i*.

Decay heat calculations are subject to uncertainties that arise from nuclear data, operational history, and calculation methods [7]. The present work primarily addresses the uncertainties that propagate from nuclear decay data. Investigating these uncertainties is important to understand their sources and minimize their impact. There are two methods for calculating uncertainties associated with decay heat. The first method involves estimating the sensitivity of decay heat to changes in fission yields and decay data [8] as well as the Generalized Perturbation Theory [9]. The second method is a Monte Carlo approach, which is implemented in the present work [10]. This later approach provides distinct advantages, as it takes into account nonlinear effects, and maintains flexibility in handling complex systems but also a relatively simpler alternative to the mathematically intricate GPT approach. The Monte Carlo approach involves generating multiple inputs by sampling from the decay data based on their uncertainties and covariance matrices when they are available, then perform simulations to calculate the mean decay heat value and the corresponding uncertainties. For this, a python-based sampling code named COCODRILO coupled to the Serpent2 code is developed at the SUBATECH Laboratory.

The COCODRILO code and the associated sampling methodology are described in the next section. A brief overview on the available decay data is also included in Section 3. Section 4 presents the decay heat uncertainty results for thermal fission pulses of ^{239}Pu and ^{235}U using ENDF/B-VII.1 and ENDF-VIII.0 nuclear decay data libraries, by evaluating the uncertainty propagation contribution of each component of the decay data.

2 METHODOLOGY

The COCODRILO code is a Monte Carlo code developed to calculate decay heat uncertainties associated with decay data, currently using a Gaussian sampling method with an aim to include other sampling approaches in the future and the use of covariance matrices. The code also offers the flexibility to either sample from the entire array of available nuclides or selectively target a specific list of nuclides. This flexibility provides an opportunity to study the effect of dominant isotopes at a given cooling time on decay heat. Additionally, it is possible to investigate the decay heat uncertainty contribution of each component or the combined effect of the components, because COCODRILO provides the option to sample from different components of decay data (i.e. ELP, EEM, EHP, and Lambda), be it individually, in partial combinations, or as a whole. All the sampled values are independent as we used the ENDF-6 files for nuclear decay data [11]. The algorithm of the code involves generating multiple decay data files, preparing input files, and coupling with the Serpent2 Monte Carlo particle transport code to compute decay heat values with associated uncertainties.

For the work presented here, multiple simulations were performed by selecting different components and combinations from the decay data libraries in order to investigate their impact on the decay heat calculations of fission pulses. Fission pulses were chosen as an initial example due to their suitability for disentangling the effect of the uncertainties of the cross-sections and the decay data. Indeed, the fission event is considered as immediate, and only the decaying processes of the initial fission yields distribution is taken into account. The decay heat calculations were compared with experimental data from Dickens et al. [12-13], and experimental data compiled by Tobias [14]. The fission pulse experiment carried out by Dickens et al. involved irradiating a target sample for a short period with thermal neutrons in a research reactor facility at Oak Ridge National Laboratory. In these experiments, beta and gamma energies were measured separately using spectroscopy with different scintillator types for each particle type. The measured values were then integrated to estimate the total decay heat, with a measurement uncertainty of 4 to 7% for ^{235}U in the cooling time range 4-5 10^3 s, and between 2 and 5% for ^{239}Pu in the cooling time range 2-1.2 10^4 s. These uncertainties are attributed to detector efficiency, calibration, statistics, background subtraction uncertainties, and gaseous fission product losses. Fission products with very short half-lives pose challenges in measurement, contributing to higher uncertainties. On the other hand, for the experimental data compiled by Tobias, the decay heat uncertainty for both fission pulses of ^{235}U and ^{239}Pu ranges from 10% to 5% in the cooling time range 5-10^5 s.

For each computational analysis in this work, 1000 independent samples were taken, and the simulations were performed using thermal neutrons with an energy of 0.025 eV at a rate of 10^{22} particles/sec. Simulations with best estimate values were also performed as a reference for decay heat uncertainty evaluation.

3 NUCLEAR DECAY DATA OVERVIEW

Before proceeding further into the uncertainty propagation, it is essential to thoroughly examine the decay data utilized. ENDF/B-VII.1 [15] and ENDF/B-VIII.0 [16], both of which use ENDF-6 formats, are employed for the calculations. However, it is worth noting that the incompleteness of these decay data and the associated uncertainty information, results in underestimation of the decay heat uncertainty. A total of 3821 isotopes with different levels of information comprised in both data libraries. Of these isotopes, 80 % have ELP and EEM decay data, 36% have EHP decay data, and 93% have their decay constants measured in both ENDF/B-VII.1 and ENDF/B-VIII.0 libraries. However, as shown on Fig.1 in terms of uncertainty values, almost 20% of the ELP and EEM data, 63% of the EHP data, and 5% of the decay constants do not have evaluated uncertainties in both ENDF/B decay libraries. It is also worth mentioning the updates included in ENDF/B-VIII.0 decay data library. Uncertainty values for ELP and EEM components are included for ^{82}Ge, ^{83}Ge, and ^{91}Br. Moreover, updates in ENDF/B-VIII.0 include ELP data with uncertainties for 31 isotopes, EEM data with uncertainties for 63 isotopes, and two updates for EHP (^{83}Ge and ^{91}Br) and decay constant in one isotope (^{82}Ge).

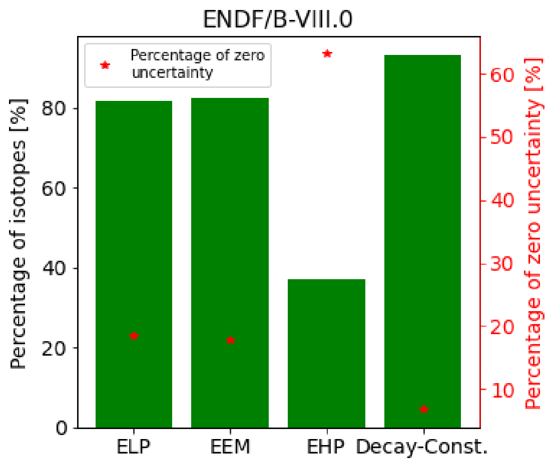


Figure 1: Percentage of Isotopes with decay data and the corresponding zero uncertainty percentage for ENDF/B-VIII.0 decay data library.

Figure 2, shows the distribution of relative uncertainties in percentage for cases where data is available for the ENDF/B-VIII.0 decay data library. The relative uncertainty is below or equal to 20% for 97% of the ELP data, 98% of the EEM data, 98% of the EHP data, and 91% of the decay constants. Moreover, the relative uncertainty is 100% for ^{74}Ga for the ELP data.

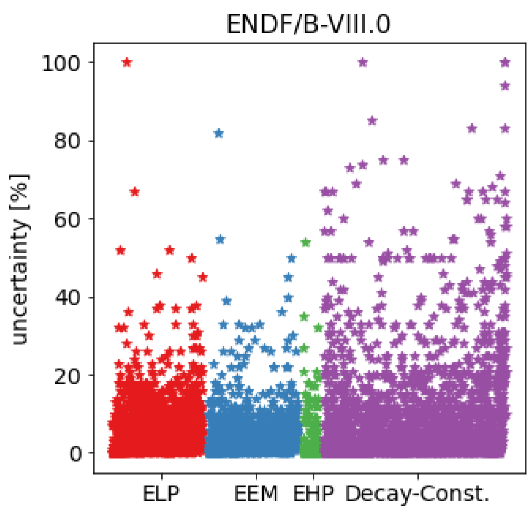


Figure 2: Uncertainty distribution for decay energies and decay constants for ENDF/B-VIII.0 decay data library.

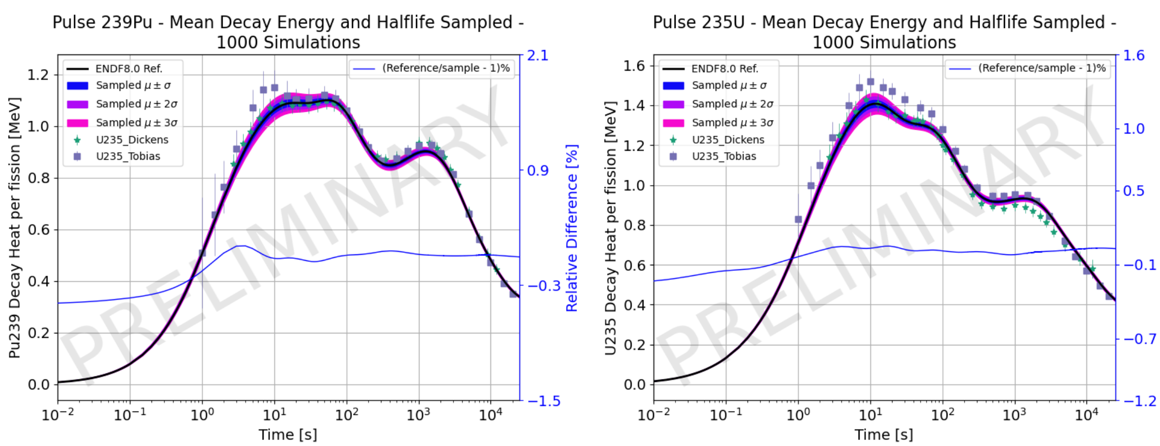
The impact of missing uncertainty data is investigated in this work by conducting a comparative analysis of uncertainty propagations within decay data libraries. Two approaches were followed for the comparison: one that does not account for zero uncertainty values and another that treats zero uncertainties with 100% uncertainty, as recommended by [8] (see Table 1). The number of isotopes requiring this treatment is 2232 for ENDF/B-VII.1 without considering the EHP data which are not used here; with EHP, it amounts to 3644. In the case of ENDF/B-VIII.0, this number is 2229 isotopes without EHP, and the same as ENDF/B-VII.1 with EHP.

Table 1: Zero uncertainty treatment

Reference calculation	Uncertainty treatment
E + ΔE: where, ΔE = 0 λ + Δλ: where, Δλ = 0	E + ΔE: where, E = ΔE λ + Δλ: where, λ = Δλ

4 RESULTS

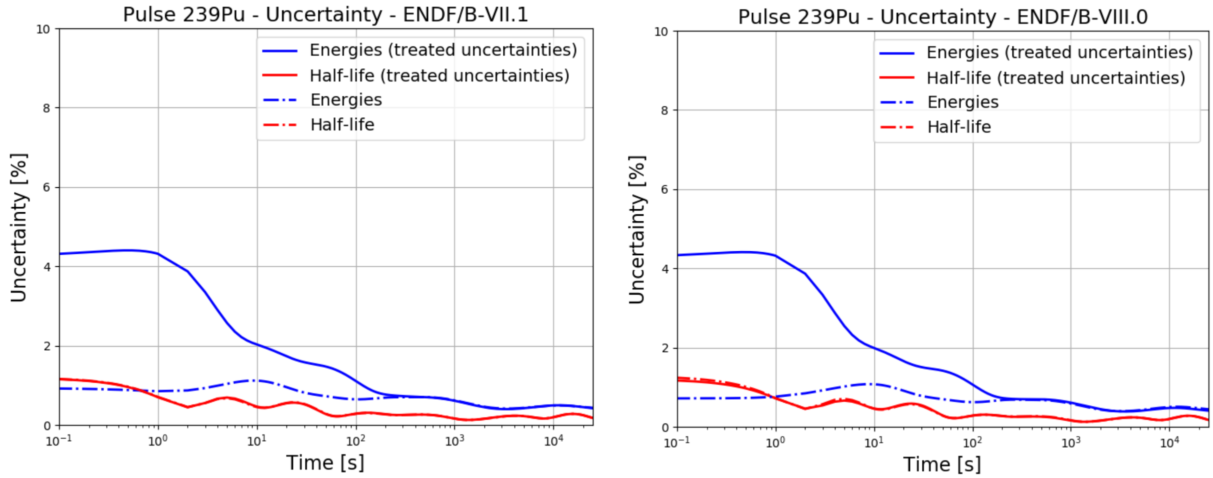
In this section, the result of decay heat per fission over cooling time, taking into account associated uncertainties from decay data, are illustrated. The uncertainty propagation to decay heat for both ^{239}Pu and ^{235}U fission pulses are calculated by the COCODRILO code coupled to Serpent2. Additionally, the effects of the absence of decay data uncertainty information on the overall decay heat uncertainty calculations are investigated.



(a) (b)

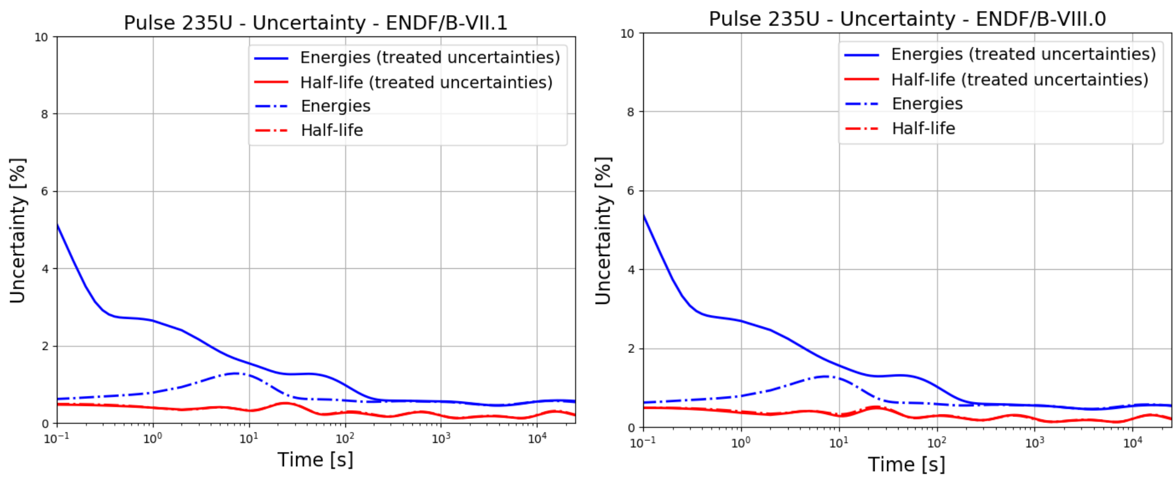
Figure 3: Total decay heat per fission as a function of cooling time. (a) ^{239}Pu fission pulses with ENDF/B-VIII.0 decay data library and (b) ^{235}U fission pulses with ENDF/B-VIII.0 decay data library.

Figure 3(a) & (b) above, show the total decay heat per fission trends over cooling times for ^{239}Pu and ^{235}U thermal fission pulses using ENDF/B-VIII.0 decay data library in comparison with experimental pulse decay heat data. The reference plots in both cases are outputs of simulations using the nominal decay data values. The different shades of colors indicate the uncertainty ranges at 1σ , 2σ, and 3σ respectively. The right y-axis shows the relative difference in percentage between the reference and the mean of the sampled decay heat values. The comparison with experimental data for ^{239}Pu [14-15] reveals an underestimation of calculated values between cooling times of 1-10 s and between 3.5 10^2-5 10^3 s. The ^{235}U fission pulse decay heat calculation depicts underestimation for cooling time below 10^2 s compared to experimental data [12-14].



(a) (b)

Figure 4: (a) Decay heat per fission as a function of cooling time for ^{239}Pu with treated uncertainty. (b) Comparison between reference and treated uncertainty



(a) (b)

Figure 5: (a) Decay heat per fission as a function of cooling time for ^{235}U with treated uncertainty. (b) Comparison between reference and treated uncertainty

Figure 4 and Figure 5 present the effect of the uncertainty treatment applied as described in Section 3. Figures 4(a) and 5(a) show the uncertainties in decay heat calculations as a function of cooling time using the ENDF/B-VII.1 decay data library, while figures 4(b) and 5(b) depict the uncertainties from the ENDF/B-VIII.0 decay data library for ^{235}U and ^{239}Pu fission pulses. In both cases, the uncertainties in decay heat calculations with treated decay data are significantly high, especially in the 10^{-1}- 10^{1} s cooling time range. This is due to the absence of uncertainty data for the decay energies of most isotopes with short half-lives, leading to their substitution with 100% uncertainty. From these analyses, it can be deduced that the calculation of decay heat uncertainty can be improved by new measurements of the decay energy data and associated uncertainties.

5 CONCLUSIONS and OUTLOOKS

The impact of decay data uncertainties on decay heat uncertainty was first performed using the Gaussian sampling method by coupling COCODRILO to the Monte Carlo particle transport code Serpent2. The methodology was demonstrated by performing decay heat calculations for ^{239}Pu and ^{235}U thermal fission pluses. It was also shown that ~20% of the ELP and EEM data are without uncertainty values. The impact of the zero uncertainty values was also presented by comparing the uncertainty propagation of decay data components with treated zero uncertainty and taking the uncertainties as tabulated. The uncertainty from the decay energies reached to ~6% at the beginning of the cooling time when calculated with treated uncertainties.

The COCODRILO will continue by including sampling of fission yield data using covariance matrices, and perform parametric studies by employing other decay data libraries, such as JEFF3.3 [17] and JENDL-5 [18]. In addition, the methodology will be applied to calculate decay heat uncertainty on a PWR assembly and one of the GEN IV reactor concepts known as Molten Salt Fast Reactor (MSFR) [19].

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