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Evaluating the Effect of Decay and Fission Yield Data Uncertainty on Spent Nuclear Fuel Source Term Using Serpent 2

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ABSTRACT

Knowledge of spent nuclear fuel (SNF) source term (decay heat, reactivity, nuclide inventory and other relevant properties of SNF) is essential in the safe handling and final disposal of SNF. For example, decay heat power determines how densely the fuel canisters can be packed in the final disposal tunnels. Computational characterization of SNF involves numerous uncertainty sources, one of which is the uncertainty in nuclear data. Different nuclear data libraries are known to yield significant differences in nuclide inventory calculations. How these uncertainties affect each component of the source term is not generally known. This work aims to identify the major uncertainty components in decay and fission yield data in relation to the SNF source term.

The uncertainty propagation from decay and fission yield data uncertainties into the uncertainties of the source term was conducted using a sampling based technique and an extended version of the Monte Carlo particle transport code Serpent 2. Normally, Serpent 2 only uses the tabulated values for the fission yield and radioactive decay data from ENDF-6 format data files. In the extended version, the nominal values are perturbed based on random sampling using the uncertainty data present in the ENDF-6 format files. Producing the SNF source term several times using different randomly perturbed nuclear data allows the propagation of the input uncertainties to all output quantities included in the SNF source term.

The analysis investigated the decay and fission yield data related uncertainties in the SNF source term of a TVEL second-generation type VVER-440 fixed assembly with an average enrichment of 4.37 % U-235 and six gadolinium-doped fuel rods with 3.35 % Gd_2O_3. Other nuclear data related uncertainties were ignored in this study, and only fixed, nominal depletion conditions were considered.

1 INTRODUCTION

This work introduces ENDF-6 file format uncertainty random sampling technique for fission product yield and radioactive decay data in the Serpent Monte Carlo particle transport code [1]. Uncertainties of some spent nuclear fuel source term components are studied. The scope of this work is limited only to a single fuel assembly type with burnup up to 20 MWd/kgU and a decay calculation up to 10^7 years. Studies with higher burnups will be conducted in subsequent work.

2 METHODOLOGY

The fission product yield data and radioactive decay data of ENDF-6 files contain both best estimate values and one standard deviation uncertainty estimates. Usually, only the best estimate values are read and used in Serpent burnup and decay calculations. In a special version of Serpent 2.1.31 used in this work, an input option to use values sampled from normal distribution based on the uncertainty value for each nuclide was added. The option can be enabled separately for the fission product yields and the radioactive decay data. Currently the sampling applies for all nuclides, i.e. separate nuclide lists can not be given.

The values are sampled during the reading of the ENDF-6 files, and therefore the sampled values stay constant during a single Serpent calculation. All sampled values are independent, as the ENDF-6 file format does not contain covariance data for these values. This described functionality will be included in the next Serpent 2 update. During this work, negative sampled values were replaced by the nominal value. This procedure causes the sampled mean and variance to be different than the values given in the ENDF-6 files. The effect of this decision instead of for example sampling from a truncated normal distribution or log-normal distribution was not studied during the work. In the future, the distribution will be studied further.

To begin assessing the uncertainties in the spent nuclear fuel source term, the methodology is demonstrated using a single example case, where both the fission product yield data and radioactive decay data is random sampled for all nuclides. First, the calculation is divided into subsequent burnup and decay calculations. The effect of the nuclear data uncertainty and the uncertainty arising from the use of the Monte Carlo method is separated by performing calculations both without and with the nuclear data uncertainty sampling. The variance caused by the Monte Carlo method and the nuclear data uncertainty are assumed to be independent so that the total variance in the latter calculation can be calculated as

$$\sigma_{\mathrm{tot}}^2 = \sigma_{\mathrm{MC}}^2 + \sigma_{\mathrm{ND}}^2 \tag{1}$$

where σ_{MC}^2 is obtained from the former calculations. The calculations without the uncertainty random sampling are performed 10 times, and the calculations with the sampling are repeated 100 times. All cases are run with different random number generator seed values. The subsequent burnup and decay calculations are run with the same seeds.

The studied case is a second-generation TVEL VVER-440 fixed fuel assembly with a 30° symmetry [2]. The Serpent geometry plot of the case is presented in Fig. 1. The problem has three different fuel pin types with different U-235 enrichments. One pin type also contains Gd_2O_3. The fuel pins containing Gd_2O_3 are divided into 10 equal volume burnup zones. N-14 and Cl-35 are added as 10 ppm impurities in all fuel materials to estimate the production of C-14 and Cl-36 due to the activation of these impurities. This estimation is based on Ref. [3].

The burnup calculations were performed with rather small burnup steps, having a total of 43 steps when reaching 20 MWd/kgU. The linear extrapolation with quadratic interpolation using 10 substeps in both the predictor and corrector was used as a depletion algorithm. A population with 100000 neutrons per generation with 100 active and 25 inactive generations was used.

The unresolved resonance probability table sampling was used, energy grid thinning was set to 5×10^{-6} tolerance, and Doppler-broadening rejection correction (DBRC) was used for three uranium isotopes and two plutonium isotopes. Upcoming Serpent 2 JEFF-3.2 cross section data files were used with Serpent 1 distributed JEFF-3.1.1 fission product yield and radioactive decay data files. The power density was set to 37.9 W/gU.

The burnup calculations were run up to 20 MWd/kgU, and decay calculations were run from 1 d to 10^7 years from this maximum burnup.

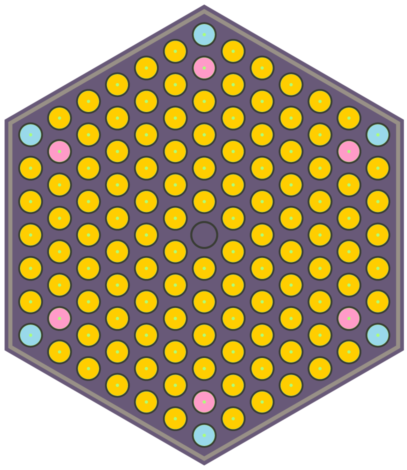

Figure 1: Serpent geometry plot of the studied assembly. The yellow pins have 4.4 % U-235 enrichment, the light blue pins have 4.2 % U-235 enrichment, and the pink pins have 4.0 % U-235 enrichment and 3.35 % Gd_2O_3 content. The center pin is an instrumentation tube.

3 RESULTS

The source term components were studied separately for the burnup and decay calculations. The studied components are total decay heat, activity, spontaneous fission rate and photon emission rate. Additionally, the masses of some interesting nuclides were studied separately during the decay calculation. The nuclide Pu-239 was chosen due to proliferation standpoint. The other studied nuclides are C-14, Cl-36, Mo-93, Ag-108m and I-129 as they are the nuclides propagating to biosphere in the TURVA-2012 safety case of Posiva during a 10000 year study time of spent nuclear fuel final disposal in Finland [4].

In all presented figures, the coloring and notation is as follows. The mean of the base calculations without uncertainty sampling is plotted with a red line. The red shaded area around the red line is the one standard deviation of the base calculation results. This value is so small in the presented results that it is practically indistinguishable in the figures. The blue line represents the mean of the calculations with the nuclear data uncertainty sampling turned on. The blue shaded area around the blue line is the total one standard deviation uncertainty of these results. The green line represents the one standard deviation relative nuclear data uncertainty. This uncertainty is defined as the ratio of σ_{ND} solved from Eq. (1) and the mean value calculated from the results of the calculations with the nuclear data uncertainty sampling turned on, or the value represented with the blue line.

The studied source term components are plotted in Figs. 2–5 for the burnup calculation. For all components, the mean values calculated from the sampling calculations were different than the mean values calculated from the base calculations. The maximum difference between the means ranged from 0.04 % of the spontaneous fission rate to 0.32 % of the total decay heat. The one standard deviation relative nuclear data uncertainties were within 2.0 % for all values. The base Monte Carlo one standard deviation was under 0.04 % of the respective means for all variables.

The studied source term components excluding the selected nuclide masses are plotted in Figs. 6–9 for the decay calculation. The relative nuclear data uncertainties grow greater in the decay calculations than in the burnup calculations. The uncertainties peak between 10^3 d and 10^5 d of decay time. The uncertainty of decay heat stays under 5.0 % for this study. It falls rapidly until reaching a relatively low, almost constant, value after some 10^5 d. In contrast, the

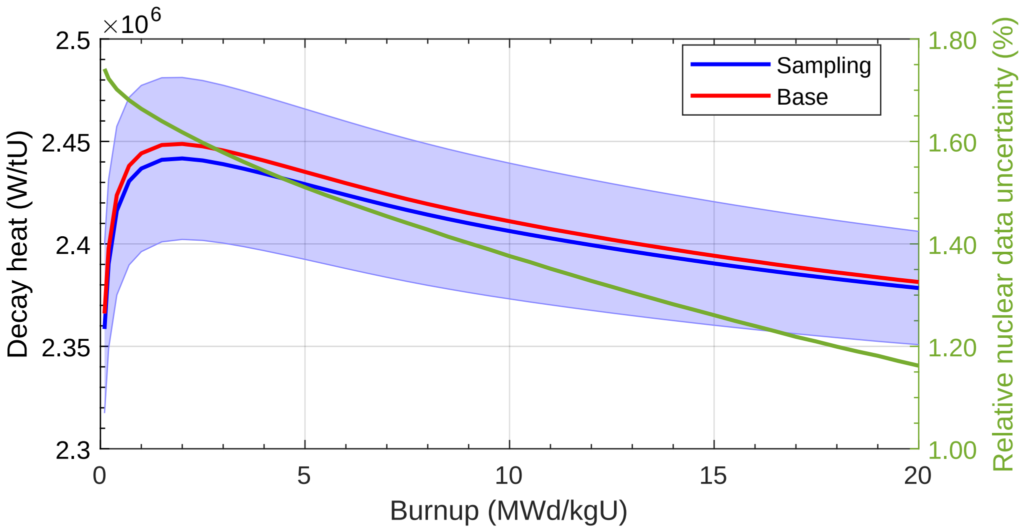

Figure 2: Decay heat during the burnup calculation.

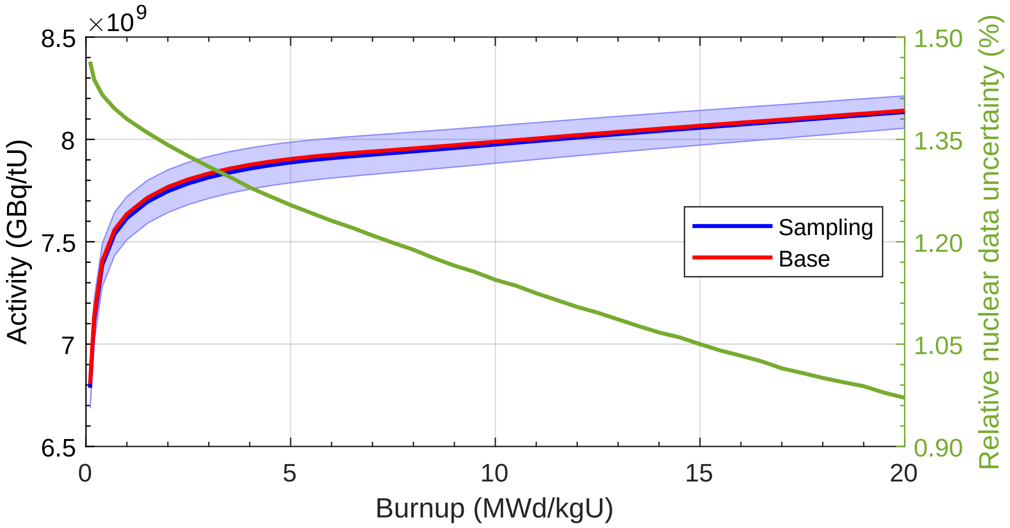

Figure 3: Activity during the burnup calculation.

uncertainty of activity has an another lower peak around 10^8 d. The base Monte Carlo one standard deviation uncertainties were under 0.05 % of the means for all studied components. The maximum difference between the means of the base and the uncertainty sampling calculations ranged from 0.09 % of the spontaneous fission rate to 0.43 % of the decay heat.

The studied nuclide masses during the decay calculation are presented in Fig. 10. The figures are cropped so that a maximum of six orders of magnitude are shown. The relative uncertainties are plotted for the whole interval where the nuclide masses were above zero. The Monte Carlo base calculation relative standard deviation was under 0.1 % of the mean values for other nuclides than Mo-93, for which the uncertainty was as high as 3.5 % during the whole decay calculation. The actual magnitudes of the C-14 and Cl-36 masses depend on the original mass of the parent nuclides N-14 and Cl-35 assumed to be in the fuel as impurities.

The relative one standard deviation nuclear data uncertainties largely vary from nuclide to nuclide. As the nuclide masses start decreasing, the relative uncertainties increase. The uncertainties of Mo-93 and Ag-108m are rather high even when their masses are relatively constant. When interpreting the uncertainties, it has to be remembered that all nuclear data was random sampled, and not just that of the specific nuclides.

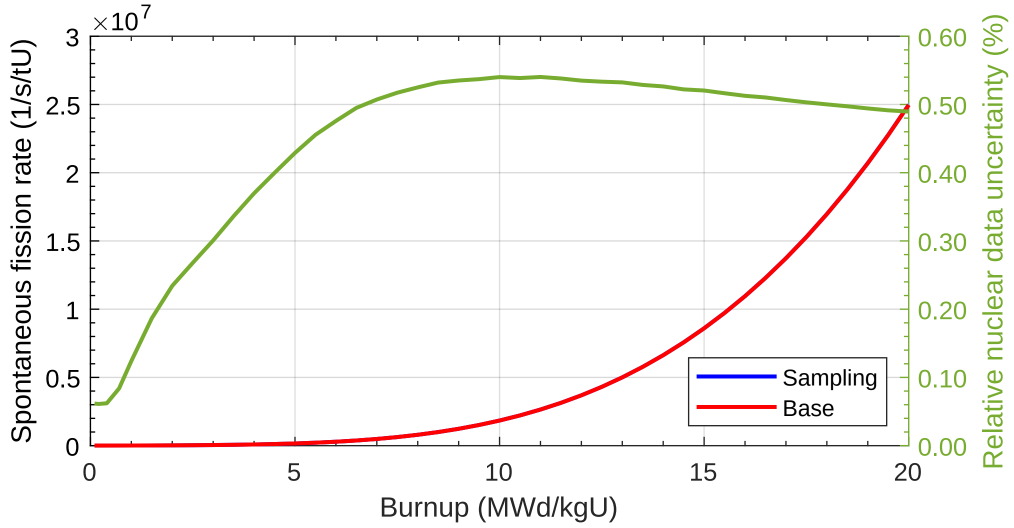

Figure 4: Spontaneous fission rate during the burnup calculation.

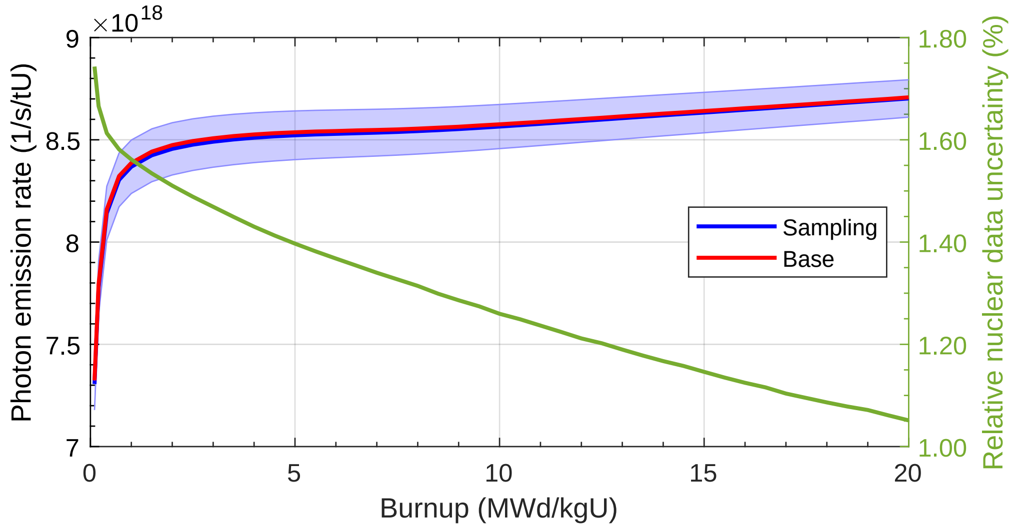

Figure 5: Photon emission rate during the burnup calculation.

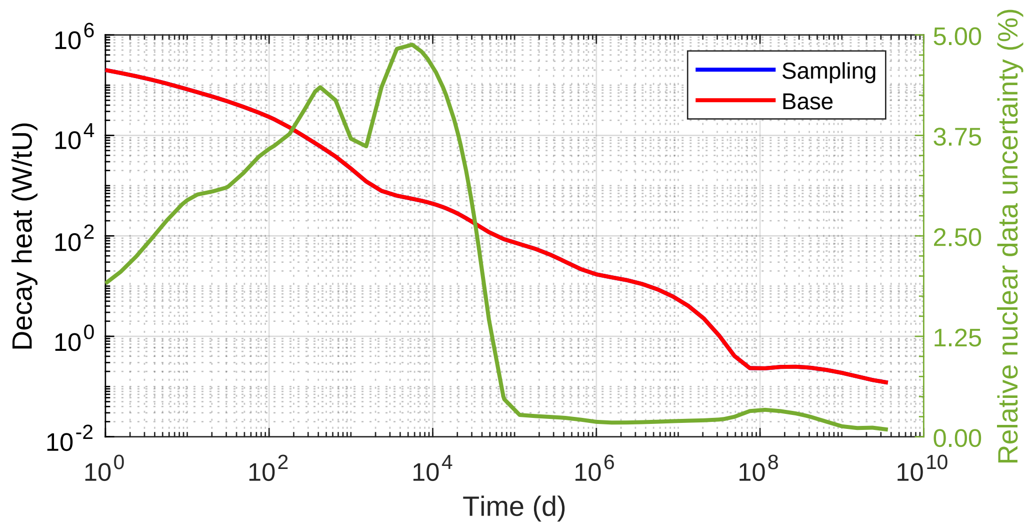

Figure 6: Decay heat during the decay calculation.

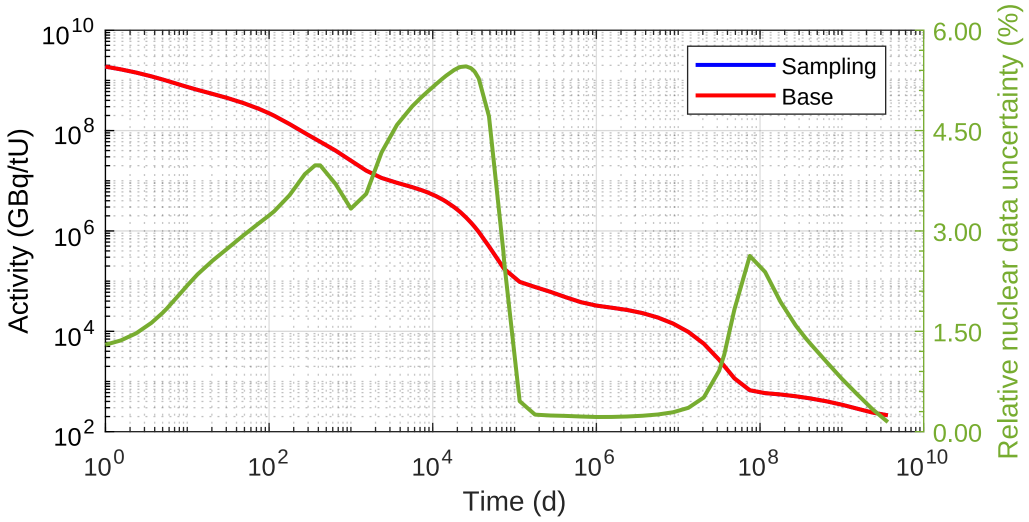

Figure 7: Activity during the decay calculation.

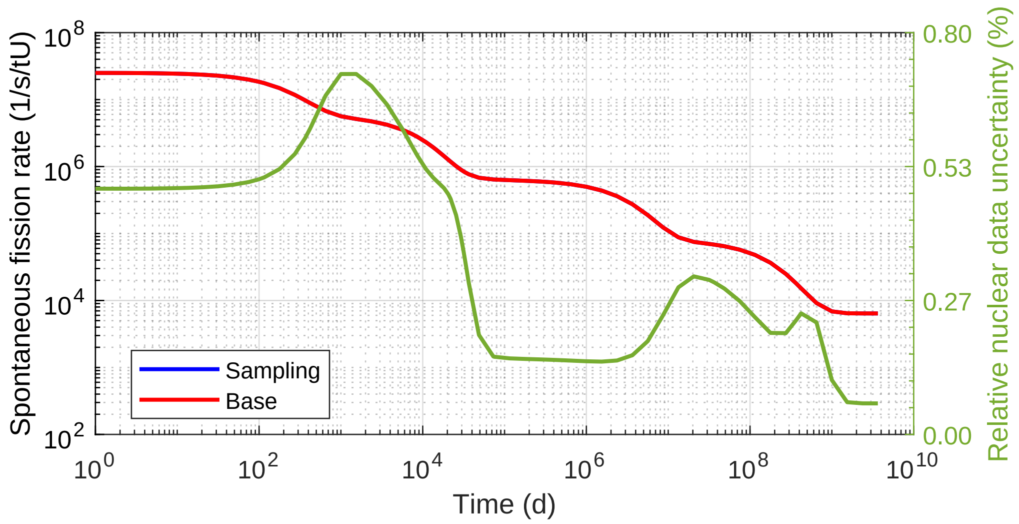

Figure 8: Spontaneous fission rate during the decay calculation.

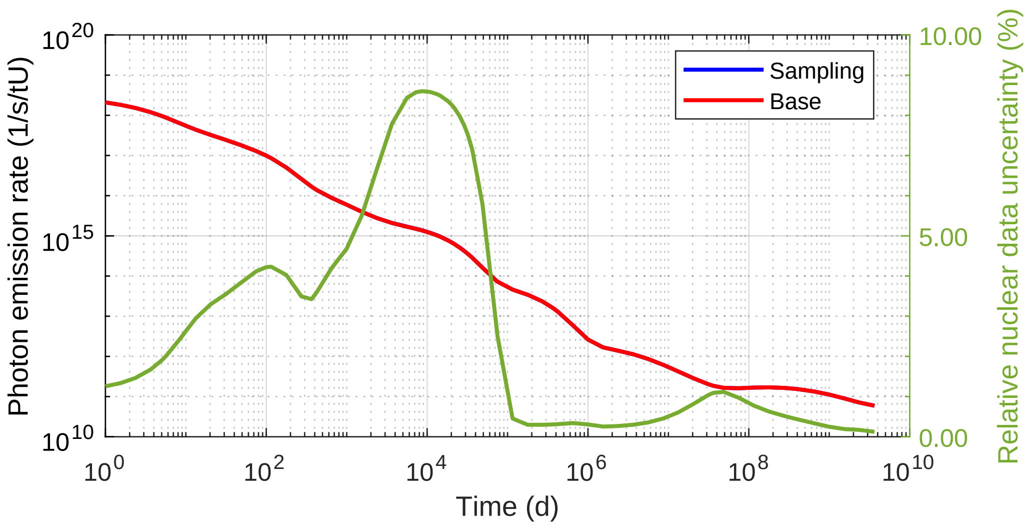

Figure 9: Photon emission rate during the decay calculation.

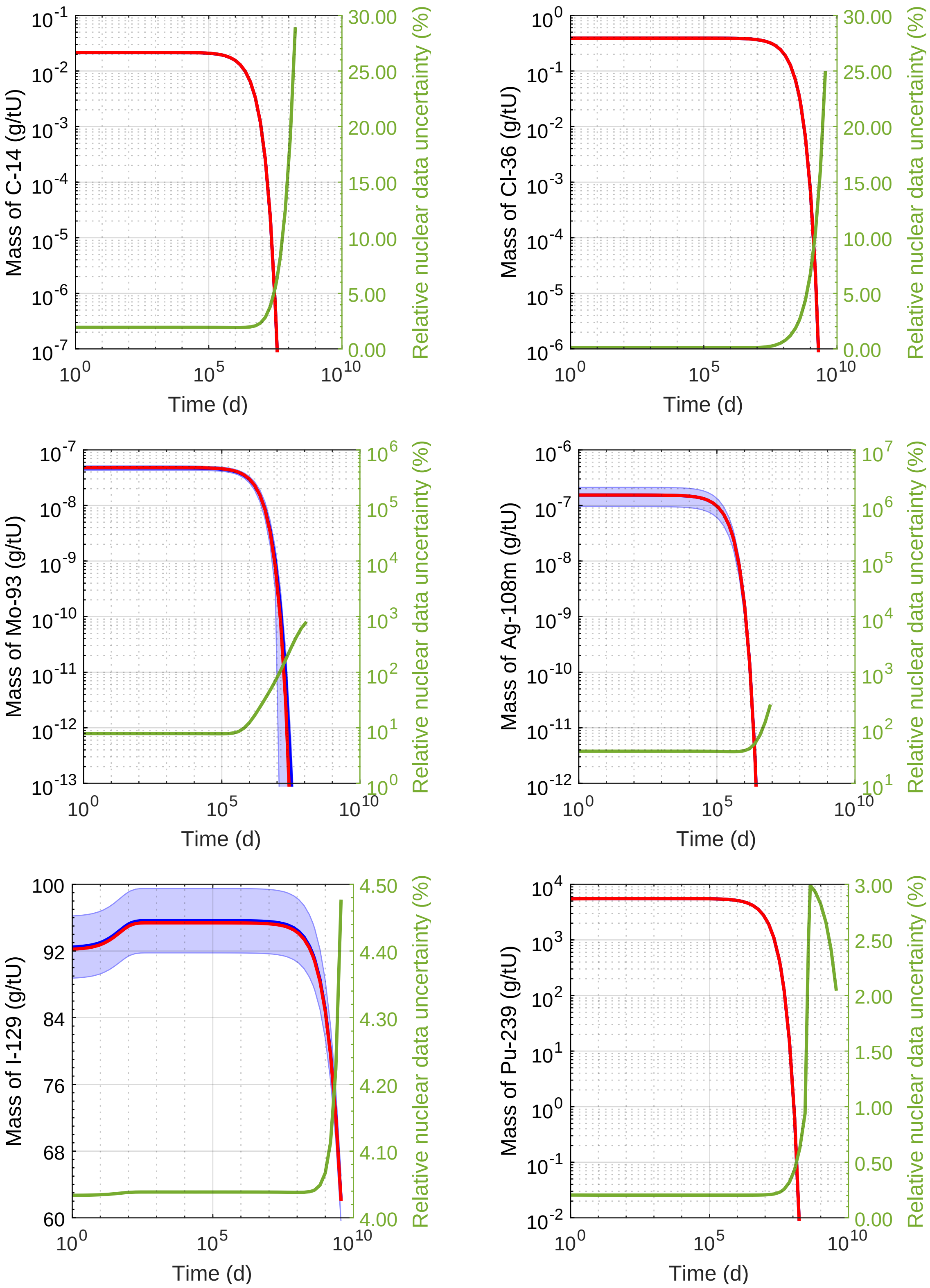

Figure 10: Masses of selected nuclides during the decay calculation.

4 SUMMARY AND FUTURE WORK

A fission yield and radioactive decay data uncertainty random sampling option was added to the Monte Carlo particle transport code Serpent. The methodology was demonstrated with a single assembly type by performing a burnup calculation and a subsequent decay calculation after the considered maximum burnup.

The relative nuclear data uncertainties of studied spent nuclear fuel source term components, excluding the masses of the selected nuclides, were under 2 % during the burnup calculation and under 9 % during the decay calculation. The relative uncertainties of the studied nuclide masses during the decay calculation were higher, especially after the masses began rapidly decrease.

This work will be expanded for other assembly types, longer burnup times and decay calculations from different discharge burnups. Additionally, the most important nuclides regarding the uncertainties of major spent nuclear fuel source term components should be identified. One obvious expansion of this work would be to study the uncertainties of the nuclear data of these nuclides separately, i.e. not randomizing all the nuclear data, but only the data of these nuclides one at a time.

The methodology should be improved to better take into account the possibility of negative values produced by random sampling. Additionally, the convergence of the obtained uncertainty values should be confirmed, for example by calculating ten times more calculations to see if the obtained uncertainties increase or decrease.

ACKNOWLEDGMENTS

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