RESEARCH REPORT

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Separate effect of decay and fission yield data uncertainty on spent nuclear fuel source term

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Summary

The effect of decay and fission yield data uncertainties on spent nuclear fuel source term have previously been studied with Serpent in two different works. In the previous studies the effect of the uncertainties of both data types have been studied simultaneously, i.e. investigations on their effects have not been conducted separately. This work aims to fill this gap by performing similar calculations as before, but with one set of calculations only sampling the uncertainties of the decay data and the other set of calculations only sampling the uncertainties of the fission yield data.

The analysis is performed with a 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 are ignored in this study, and only fixed, nominal depletion conditions are considered.

It was found out that the uncertainties caused by the fission yield uncertainties were clearly more notable for the studied components, excluding all spontaneous fission rate results and the nuclide masses at very long decay times.

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Contents

1. Introduction

This work utilizes an 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] introduced in Refs. [2, 3, 4]. This work uses the same studied fuel assembly type, calculation settings and presents the same results as Ref. [3]. However, the difference between these studies is that in the previous study, the uncertainties in both the decay and the fission yield data were studied simultaneously. In this work, two separate calculation sets are made, and the results are compared. In the first calculation set, only the uncertainties of the decay data are sampled. In the latter calculation set, only the uncertainties of the fission yield data are sampled.

The scope of this work comprises of a single VVER-440 fixed fuel assembly type with burnup calculations up to 80 MWd/kgU and decay calculations from multiple burnups up to 10^7 years.

2. Methodology

The nuclear data random sampling methodology utilized in this study is thoroughly described in Ref. [3]. The Serpent calculation settings and geometry model is also the same, and therefore these descriptions are not repeated here. The only difference with the earlier study is that only the decay data (DEC) or the fission yield data (NFY) is random sampled in each calculation.

The studied case is a second-generation TVEL VVER-440 fixed fuel assembly with a 30° symmetry [5], which was also studied in Refs. [2, 3]. The fuel assembly has three different fuel pin types having different U-235 enrichments. One pin type contains also Gd_2O_3. N-14 and Cl-35 are added as 10 ppm impurities in all fuel materials.

The burnup calculations were performed from 0 MWd/kgU to 80 MWd/kgU. Decay calculations were run from 1 d to 10^7 years from burnups 20 MWd/kgU, 40 MWd/kgU, 60 MWd/kgU and 80 MWd/kgU.

3. Results

The selected source term components are studied separately for the burnup and decay calculations. The studied components are decay heat (DH), activity (A), spontaneous fission rate (SF) and photon emission rate (PE). During the decay calculations, masses of C-14, Cl-36, Mo-93, Ag-108m, I-129 and Pu-239 are also examined.

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 for DEC results and with a purple line for NFY results. The blue line represents the mean of the calculations with the nuclear data uncertainty sampling turned on for DEC results. The blue shaded area around the blue line is the total one standard deviation uncertainty of these results. Additionally, the 5th percentile, median and 95th percentile of the nuclear data uncertainty calculation results are plotted in the figures with dashed blue lines. The similar notation for NFY results is presented in yellow. The

Table 1. Legend for the results figures.

red solid line	Mean of 10 repeats, uncertainty sampling off (DEC)
purple solid line	Mean of 10 repeats, uncertainty sampling off (NFY)
blue solid line	Mean of 300 repeats, uncertainty sampling on for decay data (DEC)
yellow solid line	Mean of 300 repeats, uncertainty sampling on for fission yield data (NFY)
blue dashed line	Uncertainty sampling on 5, 50 and 95 percentiles for decay data (DEC)
yellow dashed line	Uncertainty sampling on 5, 50 and 95 percentiles for fission yield data (NFY)
light blue solid line	Relative nuclear data uncertainty for decay data (DEC, right axis)
green solid line	Relative nuclear data uncertainty for fission yield data (NFY, right axis)

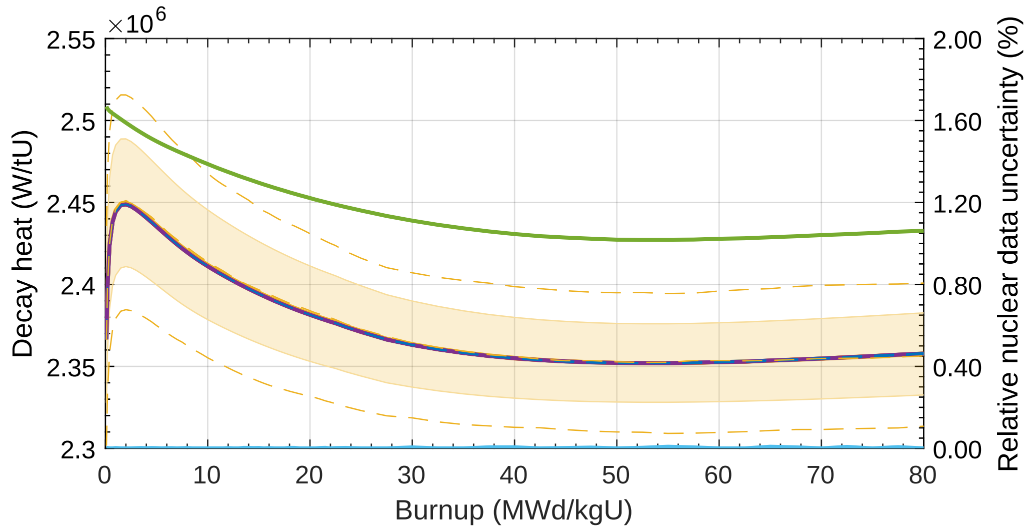


Figure 1. Decay heat during the burnup calculation.

light blue line represents the one standard deviation relative nuclear data uncertainty for DEC results. This uncertainty is defined as the ratio of one standard deviation nuclear data uncertainty and the mean value calculated from the results of the calculations with the nuclear data uncertainty sampling turned on. The similar notation for NFY results is presented with a green line. These two values are presented on the right y axis, whereas all other values are presented on the left y axis. The line styles described here are also shown in Tab. 1.

Due to the employed calculation chain, the results without uncertainty sampling were calculated separately for both calculation sets, even though the calculations were practically the same, excluding the used random number seeds.

3.1 Burnup calculation

The studied source term components are plotted in Figs. 1–4 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 relative differences between the means (Δ_{mean}), the one standard deviation relative nuclear data uncertainties (σ_{ND}) and the base Monte Carlo one standard deviations (σ_{MC}) are presented in Tab. 2 for the studied variables during the burnup calculation.

The σ_{ND} for decay heat, activity and photon emission rate caused by DEC is practically zero during the burnup calculations. For spontaneous fission rate, the DEC component is dominant, however the NFY component is also non-negligible. It is worth noting, that for decay heat,

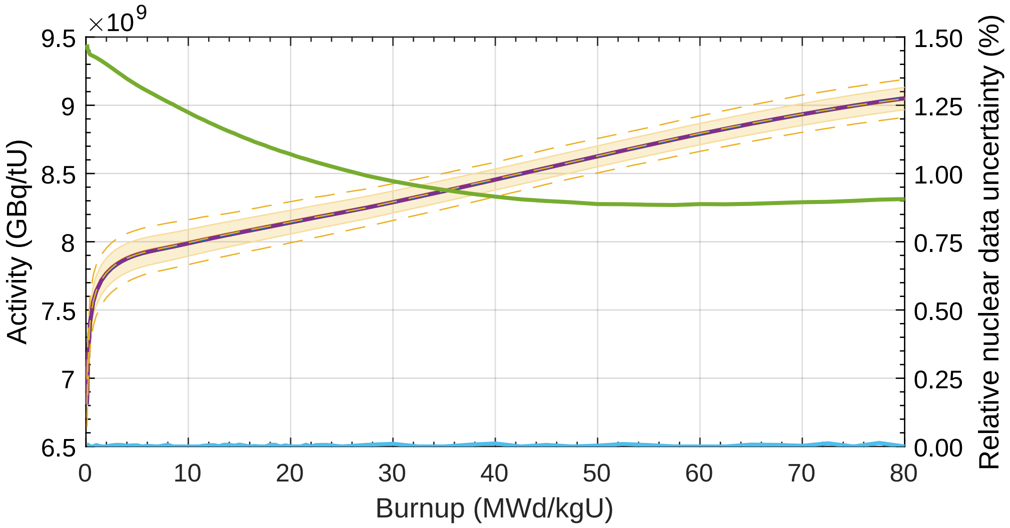


Figure 2. Activity during the burnup calculation.

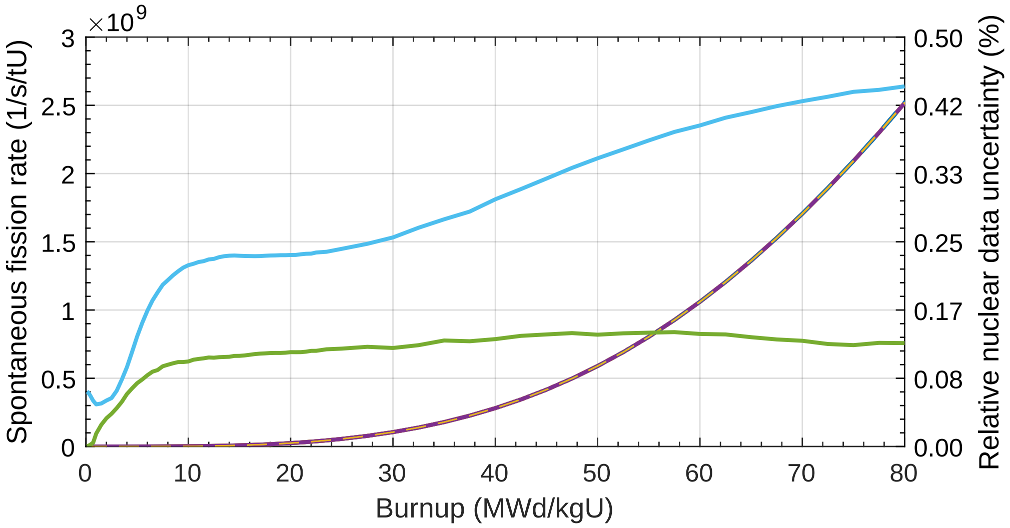


Figure 3. Spontaneous fission rate during the burnup calculation.

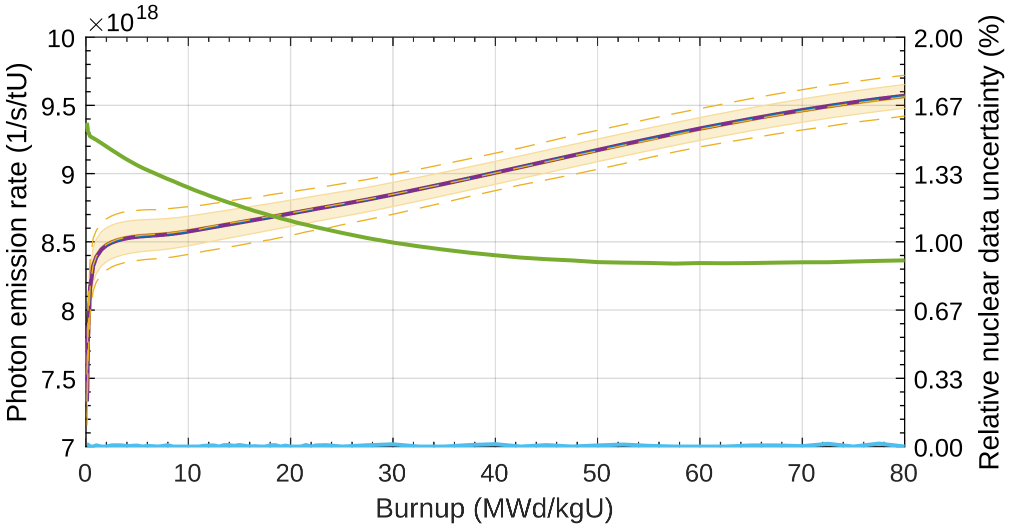


Figure 4. Photon emission rate the burnup calculation.

Table 2. Maximum values during the burnup calculation of the relative difference between the mean values of sampling and base calculations, one standard deviation relative nuclear data uncertainty and base Monte Carlo one standard deviation uncertainty.

	Case	Δ_{mean} (%)	σ_{ND} (%)	σ_{MC} (%)
DH	DEC	0.009	0.01	0.014
	NFY	0.042	1.67	0.016
A	DEC	0.011	0.03	0.024
	NFY	0.064	1.47	0.021
SF	DEC	0.016	0.44	0.064
	NFY	0.021	0.14	0.043
PE	DEC	0.012	0.03	0.026
	NFY	0.058	1.58	0.022

activity and photon emission rate, the maximum σ_{MC} values are in the same order than the maximum value of the DEC σ_{ND} results.

The uncertainties in the fission yield data should not lead directly into increased uncertainties of the spontaneous fission rate, as heavy actinides, and not light fission products tend to have possible spontaneous fission reaction modes. The most likely explanation for these uncertainties is that the NFY sampling calculations have a more uncertain fission product distribution, which leads to uncertainties in the neutron spectrum. The uncertainties in the neutron spectrum lead to uncertainties in the reaction rates which cause the build-up of the heavy actinide nuclides. Therefore the uncertainties of the NFY data can also lead to uncertainties in the spontaneous fission rate.

3.2 Decay calculations

The decay heat, activity, spontaneous fission rate and photon emission rate during the decay calculations are plotted in Figs. 5–8, respectively, for all studied burnups. The maximum relative differences between the means, the one standard deviation relative nuclear data uncertainties and the base Monte Carlo one standard deviations are presented in Tab. 3 for all studied variables during the decay calculations. The decay times of the maximum nuclear data uncertainties are also shown in the table.

For decay heat, activity and photon emission rate the DEC component of σ_{ND} is mostly insignificant. Slight increase of the values are seen at long decay times, where their significance can be higher than that of the NFY component, for example with decay heat. For spontaneous fission rate, the DEC component is dominant. In contrast with the burnup calculation results, the maximum σ_{ND} values for the DEC results are significantly higher than the maximum σ_{MC} values.

The maximum σ_{ND} values of the NFY results of decay heat, activity and photon emission decrease with the increasing burnup. For the DEC results of spontaneous fission rate, the maximum values increase with the increasing burnup. The explanation of the NFY uncertainties in the spontaneous fission rates for the burnup calculations holds also in these decay calculation spontaneous fission rate results.

In general, the photon emission rate has the highest uncertainties, followed by decay heat, activity and spontaneous fission rate.

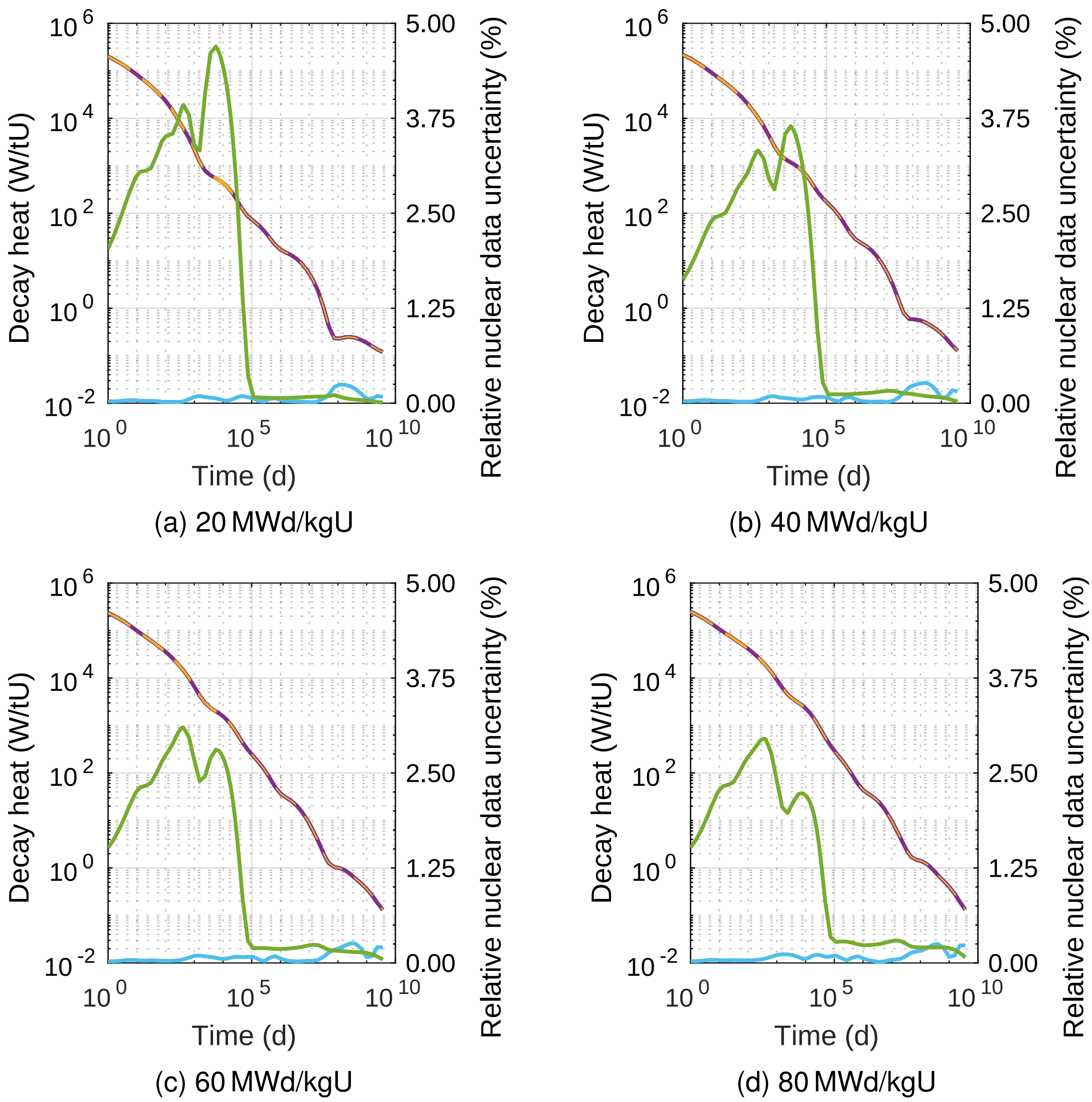
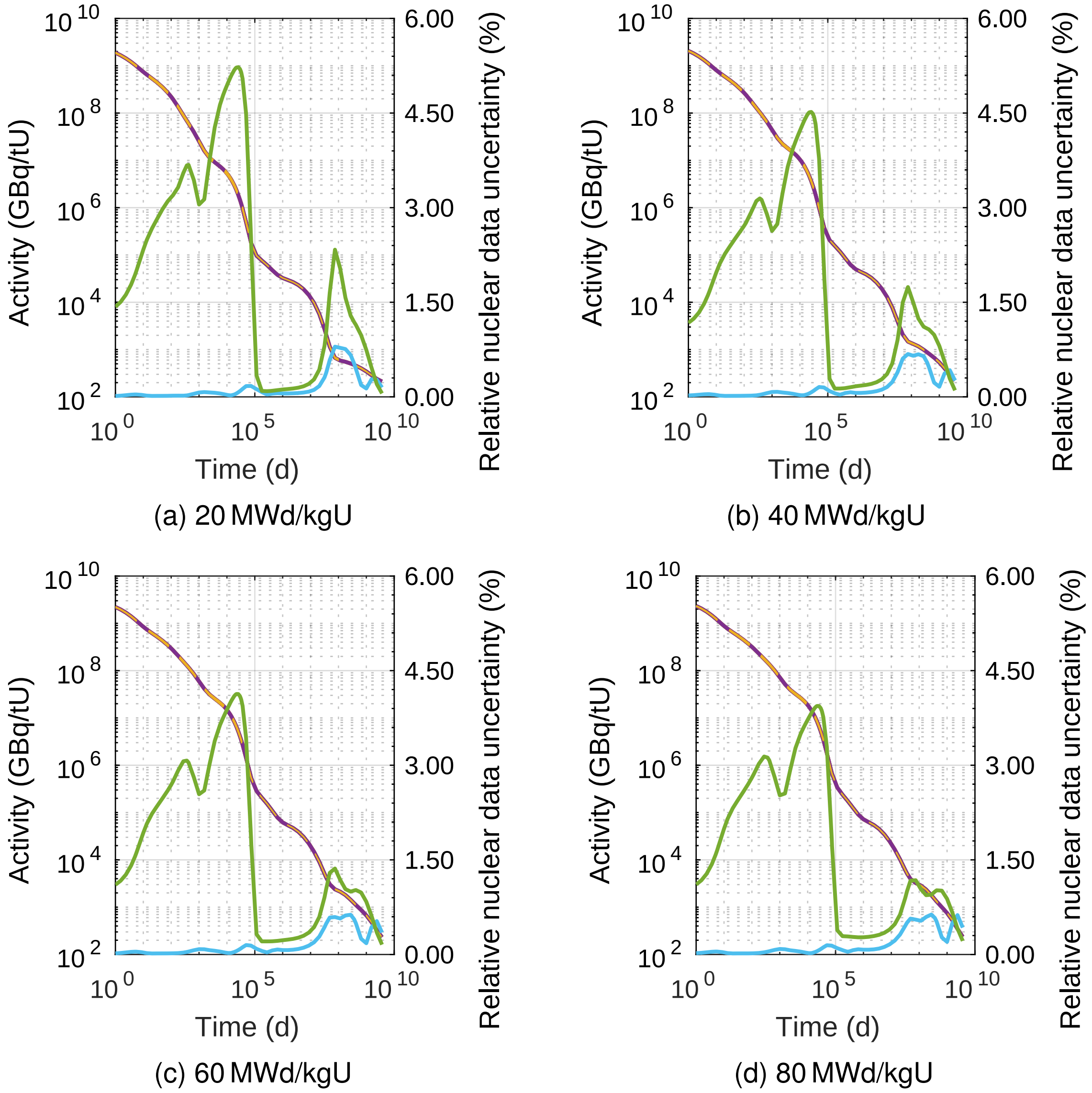


Figure 5. Decay heat during the decay calculation at different burnups.



(a) 20 MWd/kgU (b) 40 MWd/kgU (c) 60 MWd/kgU (d) 80 MWd/kgU

Figure 6. Activity during the decay calculation at different burnups.

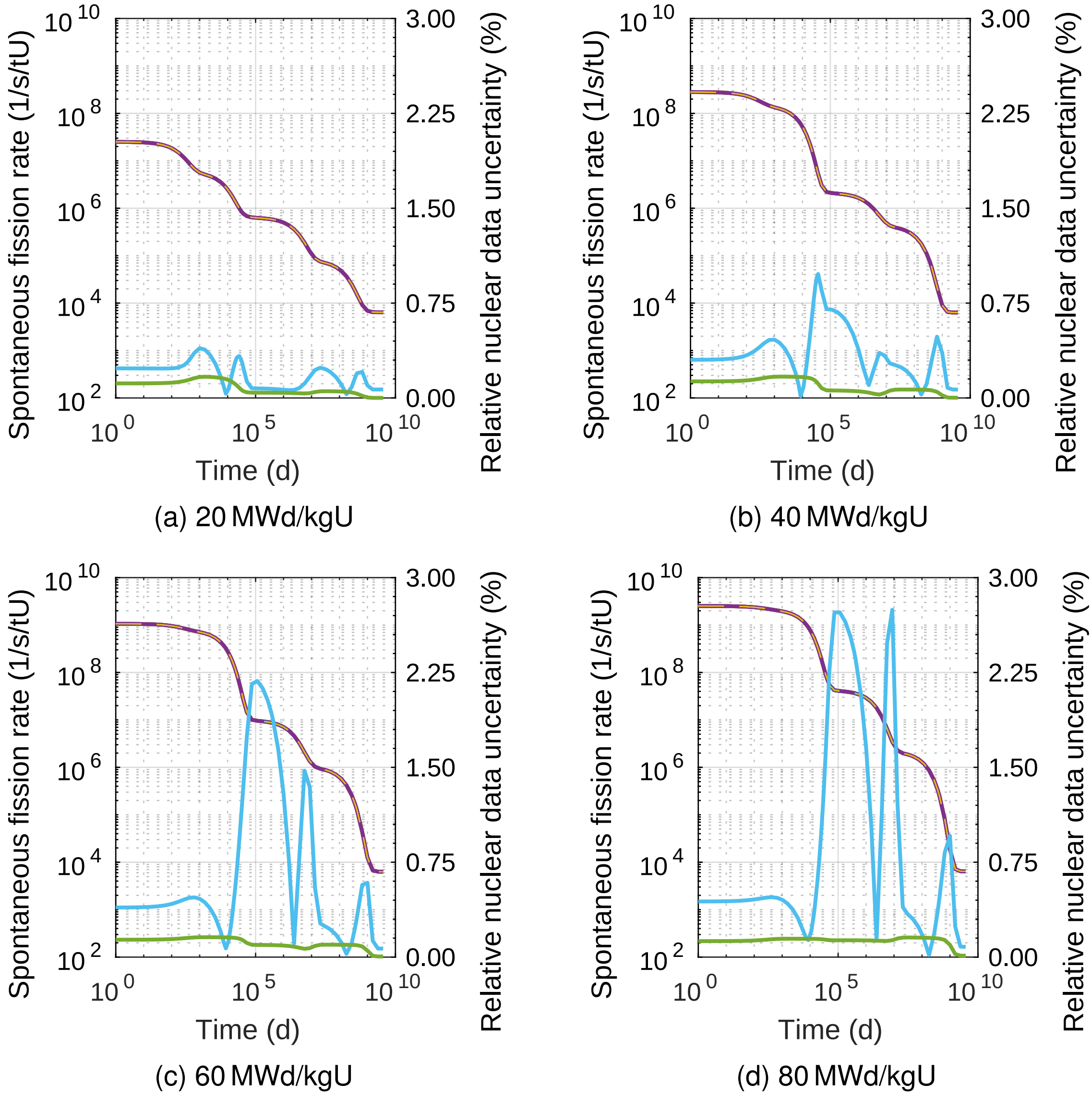
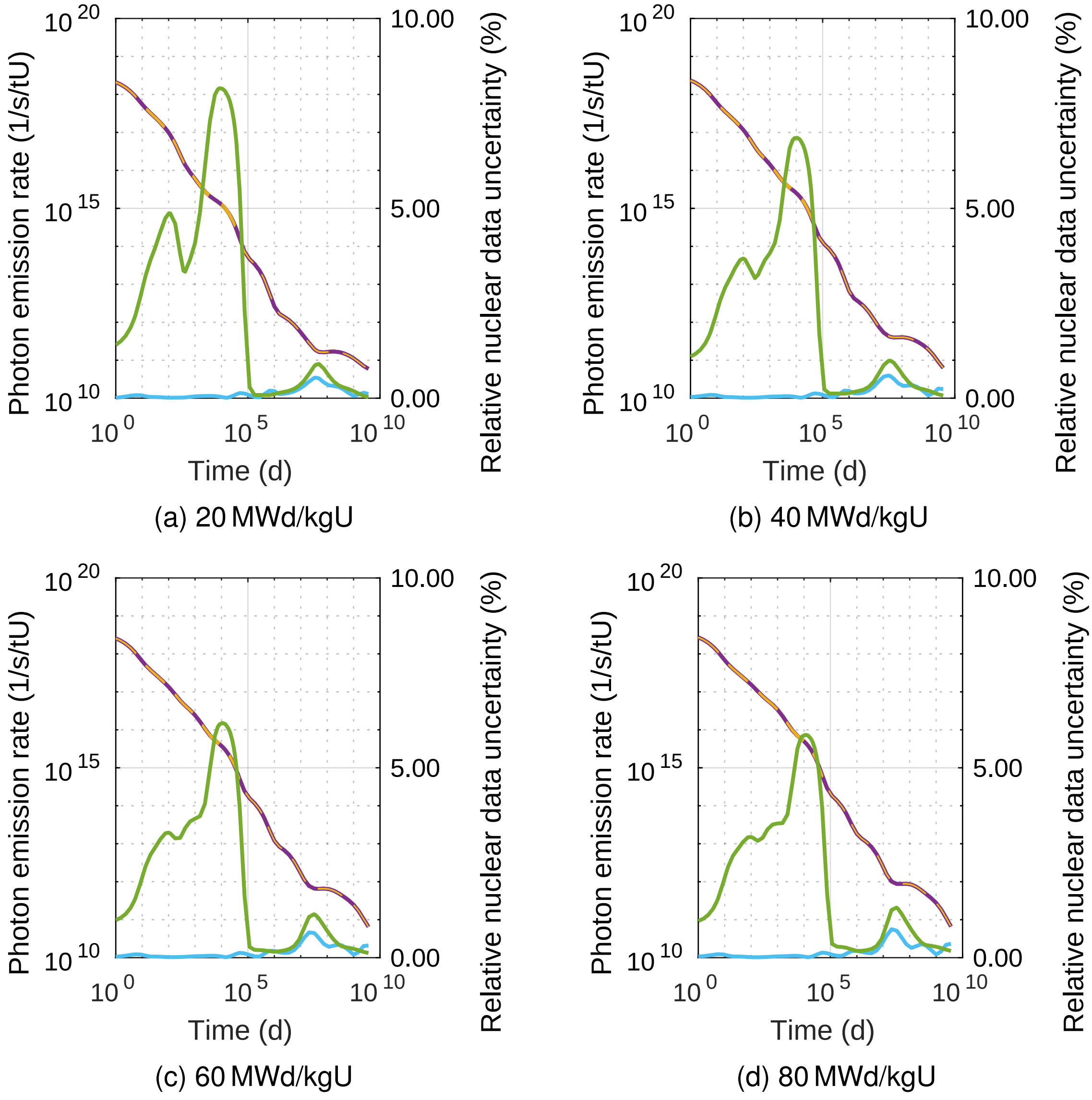


Figure 7. Spontaneous fission rate during the decay calculation at different burnups.



(a) 20 MWd/kgU

(b) 40 MWd/kgU

(c) 60 MWd/kgU

(d) 80 MWd/kgU

Figure 8. Photon emission rate during the decay calculation at different burnups.

Table 3. Maximum values during the decay calculation for the relative difference between the mean values of sampling and base calculations, one standard deviation relative nuclear data uncertainty and base Monte Carlo one standard deviation uncertainty.

	Burnup (MWd/kgU)	Case	Δ_{mean} (%)	σ_{ND} (%)	σ_{MC} (%)	Time of max σ_{ND} (a)
DH	20	DEC	0.013	0.25	0.013	3.2×10^5
	20	NFY	0.212	4.69	0.011	15
	40	DEC	0.013	0.27	0.020	7.5×10^5
	40	NFY	0.095	3.65	0.019	15
	60	DEC	0.010	0.26	0.025	1.0×10^6
	60	NFY	0.112	3.10	0.022	1.2
	80	DEC	0.012	0.25	0.021	1.0×10^6
	80	NFY	0.139	2.95	0.026	1.0
A	20	DEC	0.043	0.80	0.017	2.1×10^5
	20	NFY	0.186	5.23	0.012	70
	40	DEC	0.041	0.68	0.020	2.1×10^5
	40	NFY	0.062	4.52	0.018	70
	60	DEC	0.042	0.63	0.023	7.5×10^5
	60	NFY	0.070	4.13	0.021	60
	80	DEC	0.040	0.63	0.019	7.5×10^5
	80	NFY	0.107	3.94	0.024	60
SF	20	DEC	0.031	0.39	0.050	2.7
	20	NFY	0.036	0.17	0.047	4.2
	40	DEC	0.036	0.98	0.074	1.0×10^2
	40	NFY	0.031	0.17	0.068	4.2
	60	DEC	0.088	2.18	0.054	3.2×10^2
	60	NFY	0.012	0.16	0.041	4.2
	80	DEC	0.104	2.75	0.048	2.4×10^4
	80	NFY	0.023	0.16	0.043	8.7×10^4
PE	20	DEC	0.032	0.54	0.028	1.0×10^5
	20	NFY	0.347	8.16	0.018	24
	40	DEC	0.028	0.60	0.025	8.7×10^4
	40	NFY	0.223	6.86	0.024	30
	60	DEC	0.037	0.66	0.035	5.6×10^4
	60	NFY	0.174	6.18	0.039	30
	80	DEC	0.036	0.75	0.044	5.6×10^4
	80	NFY	0.236	5.87	0.034	30

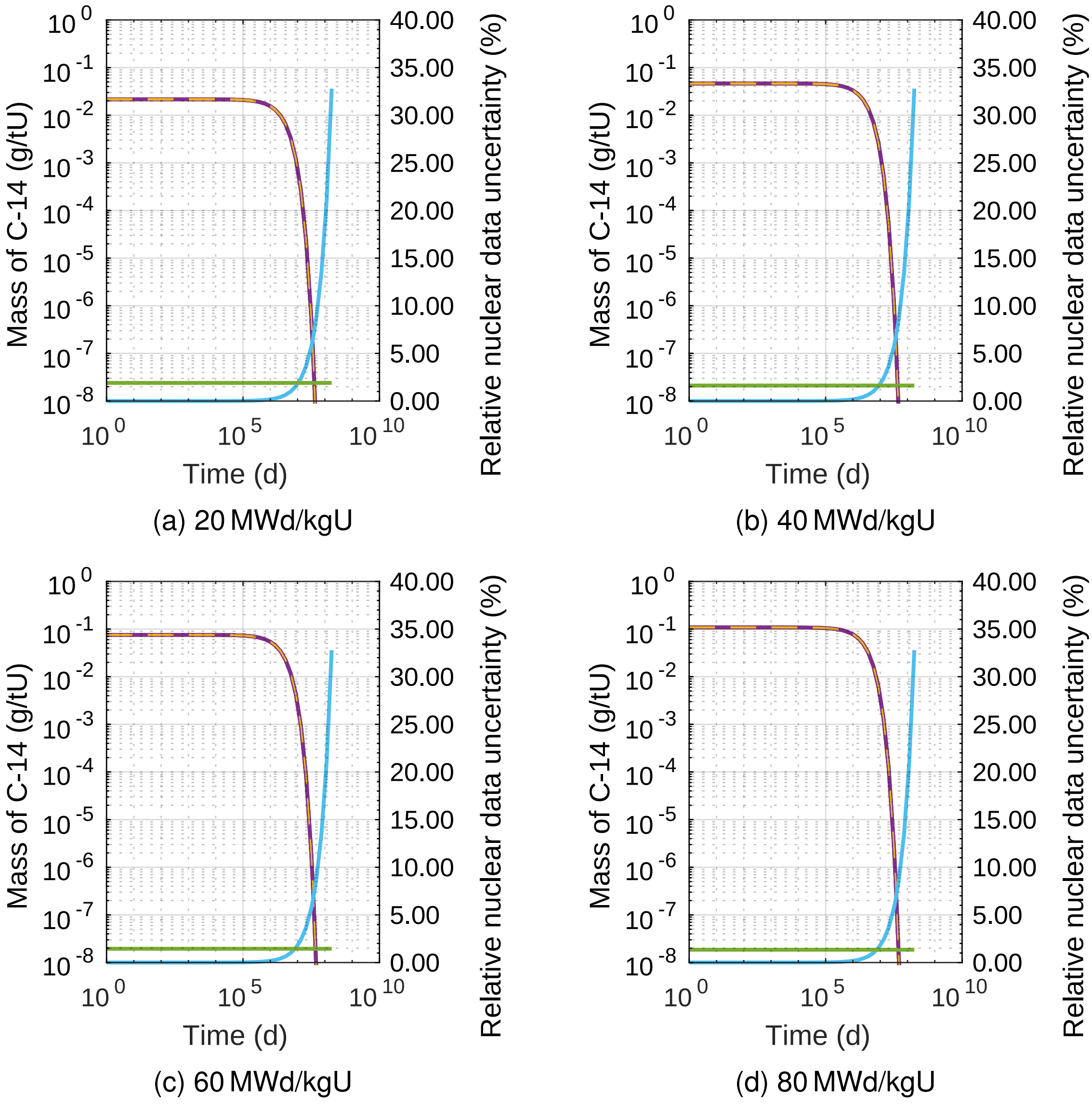


Figure 9. Mass of C-14 during the decay calculation at different burnups.

3.2.1 Nuclide masses

The masses of nuclides C-14, Cl-36, Mo-93, Ag-108m, I-129 and Pu-239 during the decay calculations are shown in Figs. 9–14, respectively. The figures are cropped so that a maximum of eight orders of magnitude are shown. The relative differences between the means, the one standard deviation relative nuclear data uncertainties and the base Monte Carlo one standard deviations are presented in Tab. 4 for all studied nuclides at 1 d decay time.

The behavior of the nuclide masses and their uncertainties are rather similar for the different nuclides and burnups. The σ_{ND} of NFY results is practically constant at all decay times. The σ_{ND} of DEC results is practically zero until an exponential increase begins somewhat before the beginning of the notable nuclide mass decrement. The masses of all nuclides increase with the increasing burnup.

The magnitude of σ_{ND} of the NFY results decrease for C-14 and Ag-108m with the increasing burnup. Cl-36 has a slight increment of the NFY σ_{ND} magnitudes with the increasing burnup.

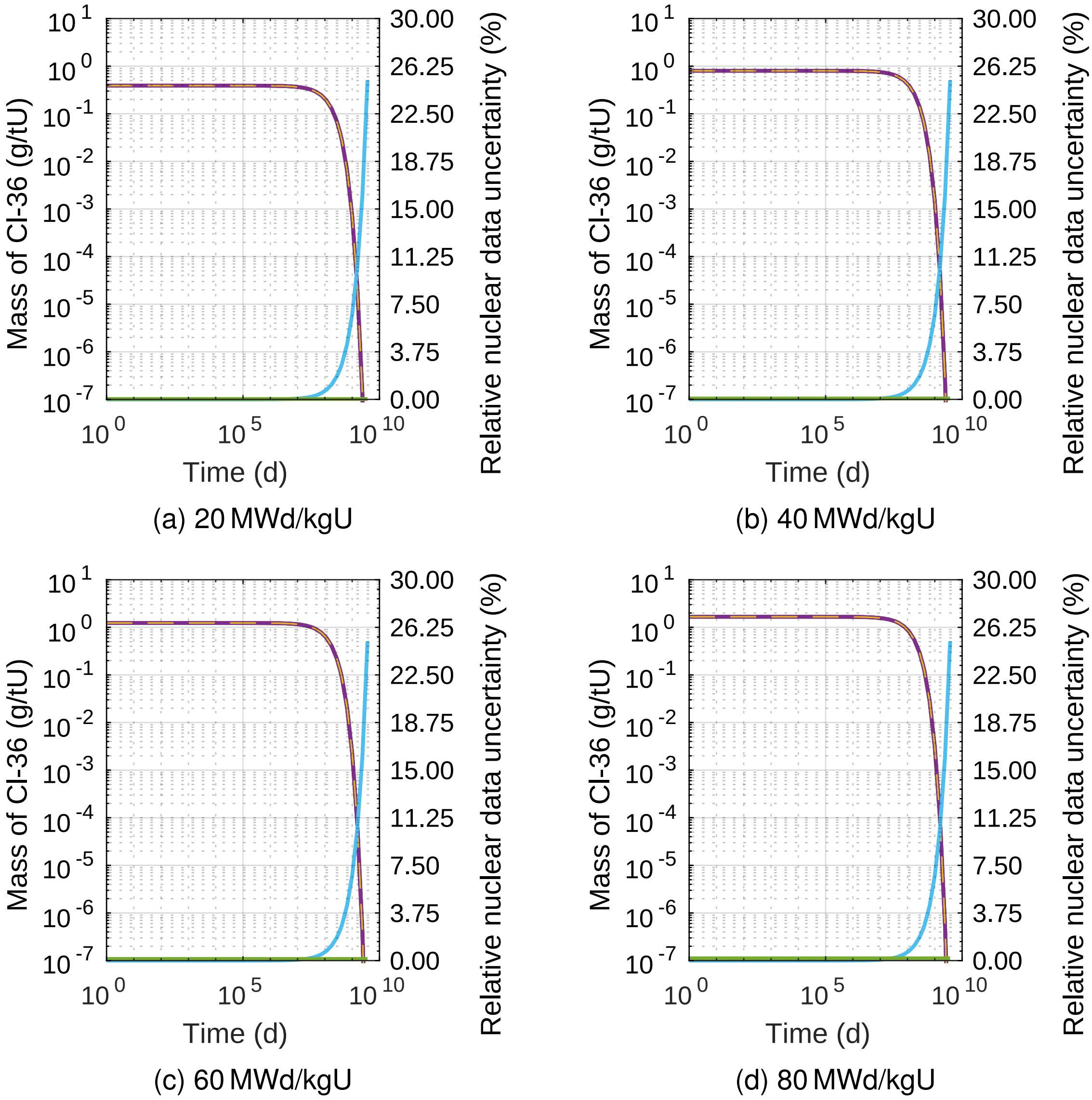
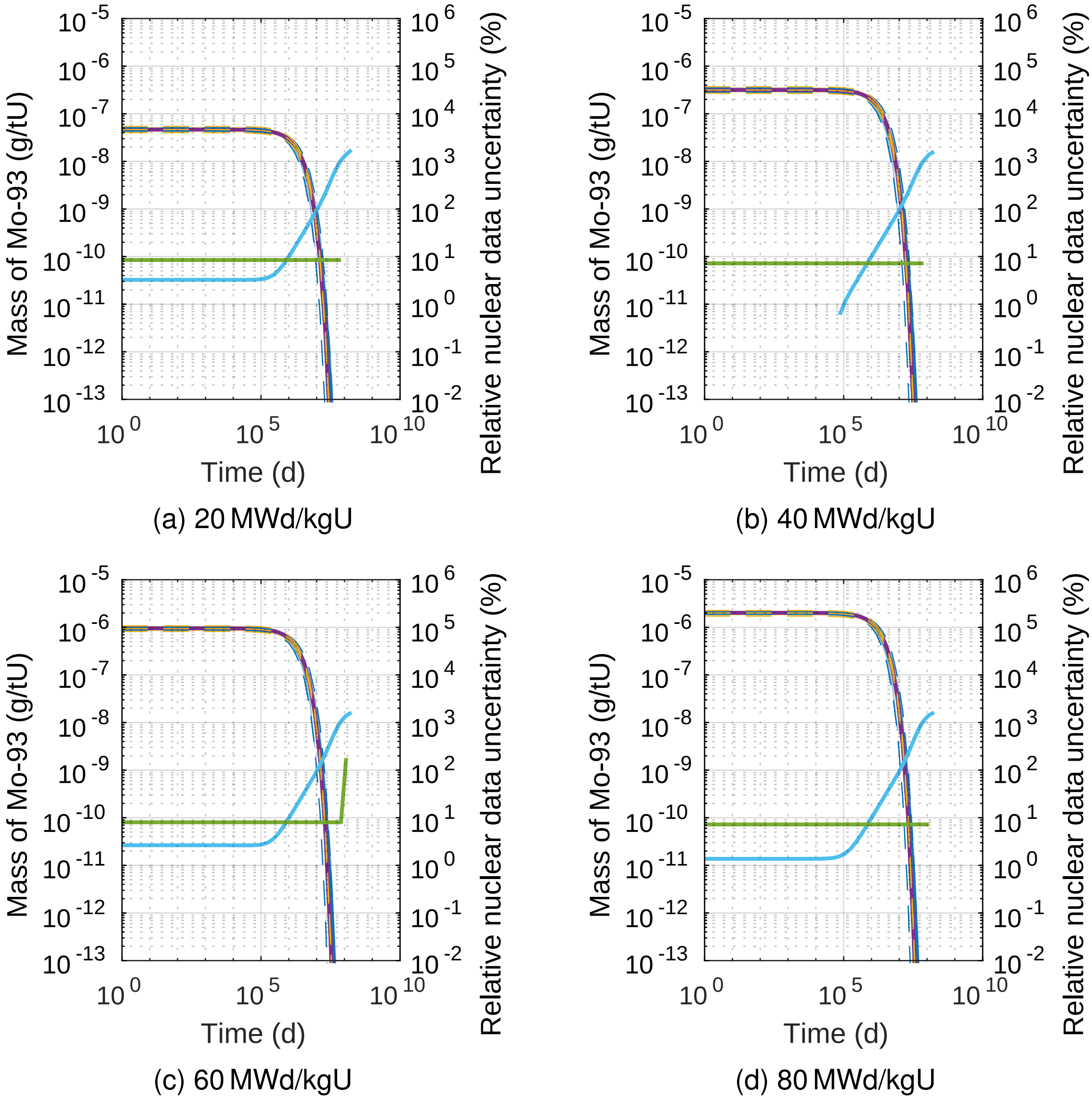


Figure 10. Mass of Cl-36 during the decay calculation at different burnups.



(a) 20 MWd/kgU

(b) 40 MWd/kgU

(c) 60 MWd/kgU

(d) 80 MWd/kgU

Figure 11. Mass of Mo-93 during the decay calculation at different burnups.

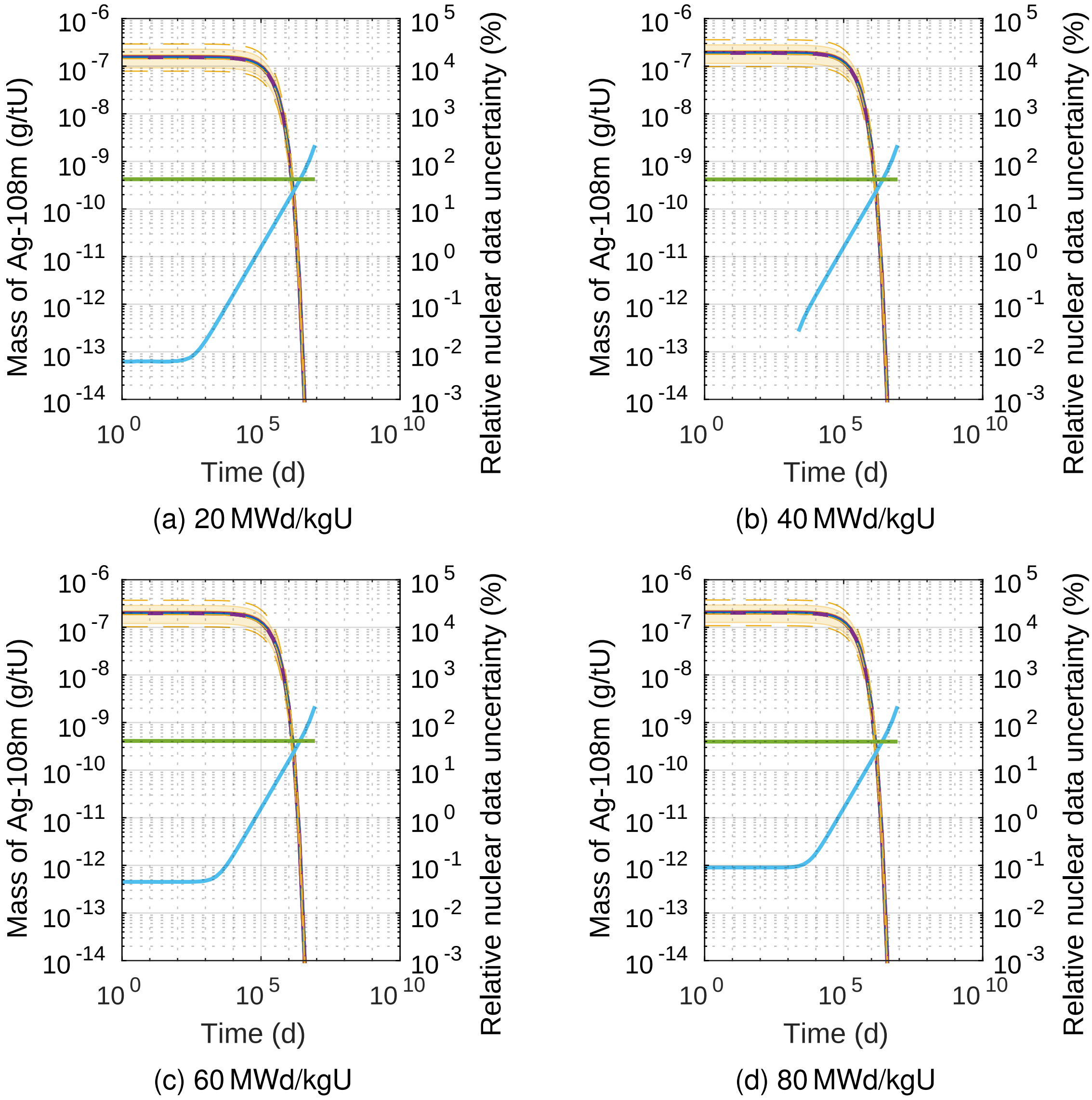


Figure 12. Mass of Ag-108m during the decay calculation at different burnups.

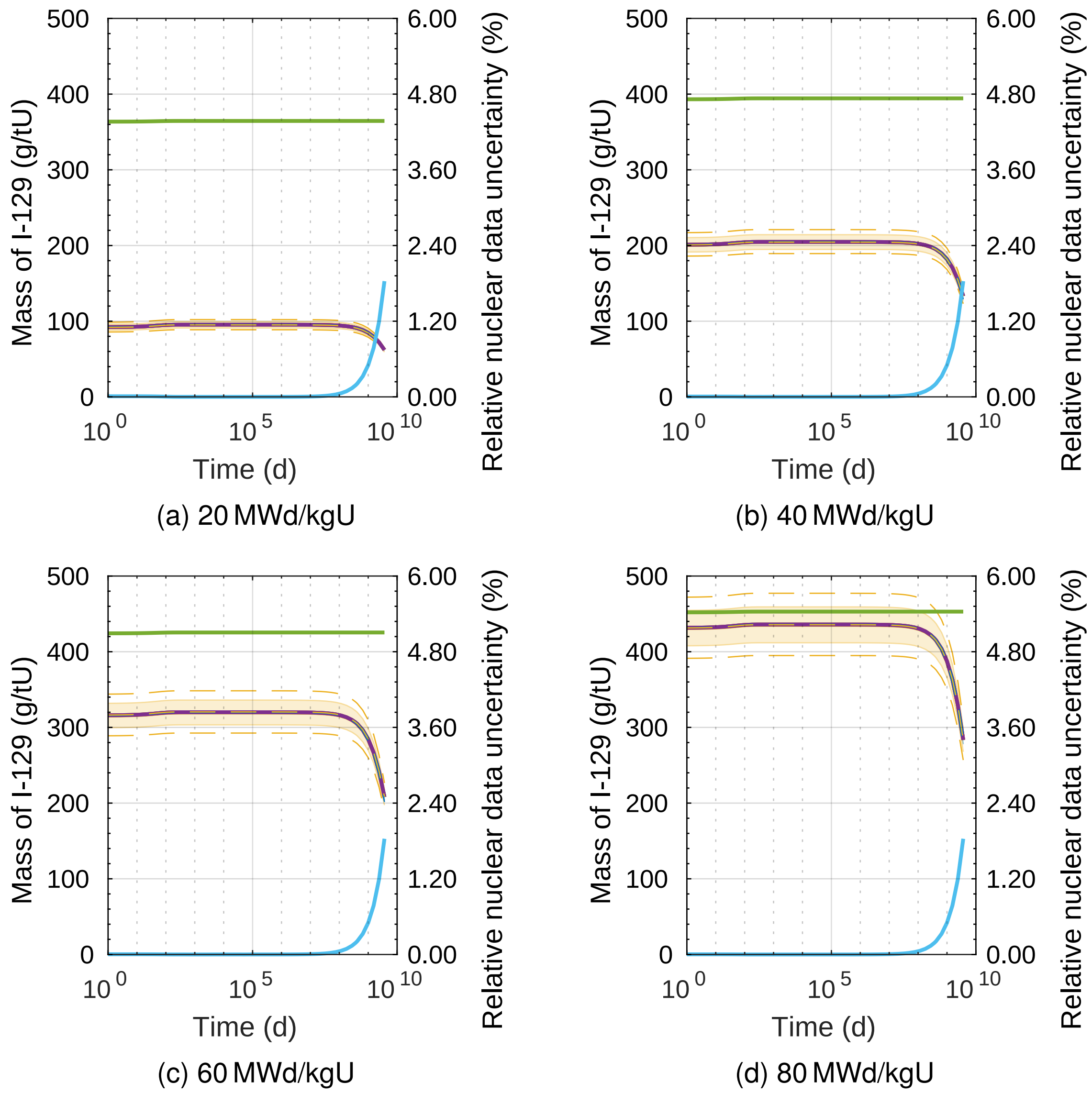


Figure 13. Mass of I-129 during the decay calculation at different burnups.

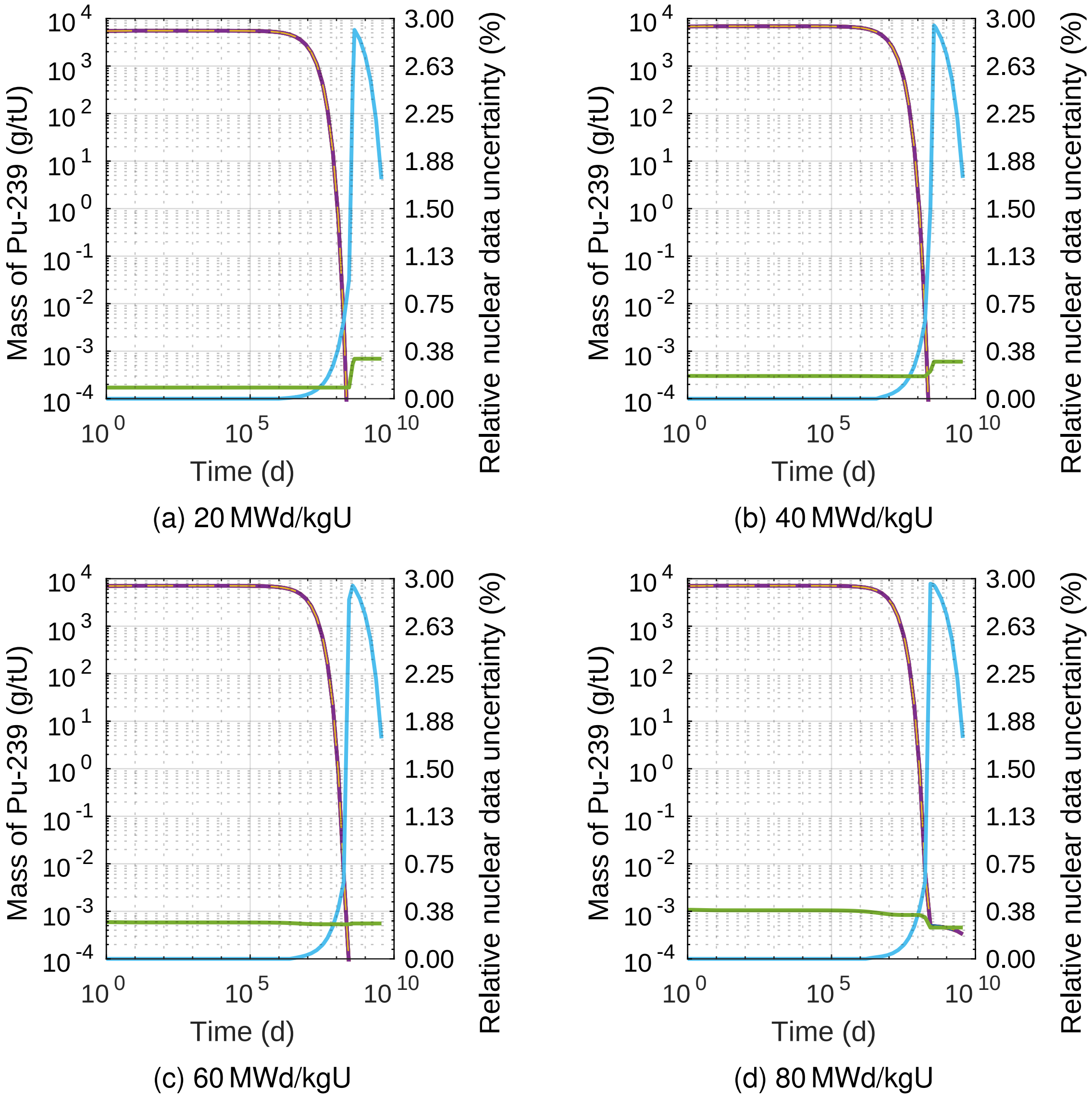


Figure 14. Mass of Pu-239 during the decay calculation at different burnups.

Table 4. Values of the relative difference between the mean values of sampling and base calculations, one standard deviation relative nuclear data uncertainty and base Monte Carlo one standard deviation uncertainty for the studied nuclide masses at 1 d decay time.

	Burnup (MWd/kgU)	Case	Δ_{mean} (%)	σ_{ND} (%)	σ_{MC} (%)
C-14	20	DEC	0.001	0.00	0.002
	20	NFY	0.163	1.90	0.001
	40	DEC	0.001	0.00	0.002
	40	NFY	0.178	1.63	0.003
	60	DEC	0.001	0.00	0.005
	60	NFY	0.181	1.45	0.006
	80	DEC	0.000	0.00	0.006
	80	NFY	0.178	1.34	0.009
Cl-36	20	DEC	0.000	0.00	0.002
	20	NFY	0.002	0.03	0.001
	40	DEC	0.002	0.00	0.003
	40	NFY	0.002	0.07	0.004
	60	DEC	0.001	0.00	0.006
	60	NFY	0.004	0.13	0.007
	80	DEC	0.001	0.00	0.007
	80	NFY	0.002	0.18	0.009
Mo-93	20	DEC	0.577	3.25	3.821
	20	NFY	0.555	8.47	3.985
	40	DEC	0.925	0.00	6.360
	40	NFY	0.509	7.21	6.921
	60	DEC	0.145	2.65	3.916
	60	NFY	1.497	8.03	2.660
	80	DEC	1.351	1.37	3.291
	80	NFY	1.169	7.23	3.144
Ag-108m	20	DEC	0.003	0.01	0.014
	20	NFY	3.649	42.18	0.011
	40	DEC	0.005	0.00	0.042
	40	NFY	3.620	41.82	0.027
	60	DEC	0.000	0.05	0.029
	60	NFY	3.548	41.06	0.019
	80	DEC	0.007	0.09	0.034
	80	NFY	3.385	39.79	0.024
I-129	20	DEC	0.001	0.01	0.001
	20	NFY	0.081	4.36	0.001
	40	DEC	0.000	0.01	0.001
	40	NFY	0.086	4.72	0.001
	60	DEC	0.000	0.00	0.001
	60	NFY	0.091	5.09	0.001
	80	DEC	0.000	0.00	0.001
	80	NFY	0.095	5.42	0.001
Pu-239	20	DEC	0.004	0.00	0.010
	20	NFY	0.008	0.09	0.004
	40	DEC	0.009	0.00	0.023
	40	NFY	0.014	0.18	0.021
	60	DEC	0.006	0.00	0.023
	60	NFY	0.002	0.29	0.024
	80	DEC	0.005	0.00	0.027
	80	NFY	0.000	0.39	0.030

The σ_{ND} of the NFY results increase for I-129 with the increasing burnup. Pu-239 has a slight increment of both σ_{ND} results with the increasing burnup.

The nuclide C-14 is barely born in the fission reactions, and Cl-36 is not a fission yield nuclide. Similarly, Pu-239 is not born in fission reactions. The most likely explanation of the NFY data uncertainties in the masses of these nuclides, especially for Pu-239, is due to the uncertainties in the neutron spectrum caused by the uncertainties of the fission yield distributions.

4. Summary

A previously implemented radioactive decay data and fission yield uncertainty sampling method was utilized to estimate the uncertainties in spent nuclear fuel source term components. This study was performed to estimate the effect of these uncertainties separately. It was found out that the uncertainties caused by the fission yield uncertainties were clearly more notable for the studied components, excluding all spontaneous fission rate results and the nuclide masses at very long decay times.

References

[1] Leppänen, J., Pusa, M., Viitanen, T., Valtavirta, V., and Kaltiaisenaho, T. "*The Serpent Monte Carlo code: Status, development and applications in 2013.*" Annals of Nuclear Energy 82 (2015), 142–150.

[2] Rintala, A. "*Evaluating the Effect of Decay and Fission Yield Data Uncertainty on Spent Nuclear Fuel Source Term Using Serpent 2.*" In proc. 28th International Conference Nuclear Energy for New Europe. (2019).

[3] Rintala, A. *Evaluating the effect of decay and fission yield data uncertainty on spent nuclear fuel source term using Serpent 2 — continued study*. Research report VTT-R-00209-20. VTT Technical Research Centre of Finland Ltd, 2020.

[4] Rintala, A. "*Evaluating the Effect of Decay and Fission Yield Data Uncertainty on BWR Spent Nuclear Fuel Source Term.*" In proc. NENE 2020 Conference Proceedings. (2020).

[5] Lahtinen, T., Antila, M., and Saarinen, S. "*Review of core design and operating experience in Loviisa.*" In proc. 20th Symposium of AER on VVER Reactor Physics and Reactor Safety. (2010).