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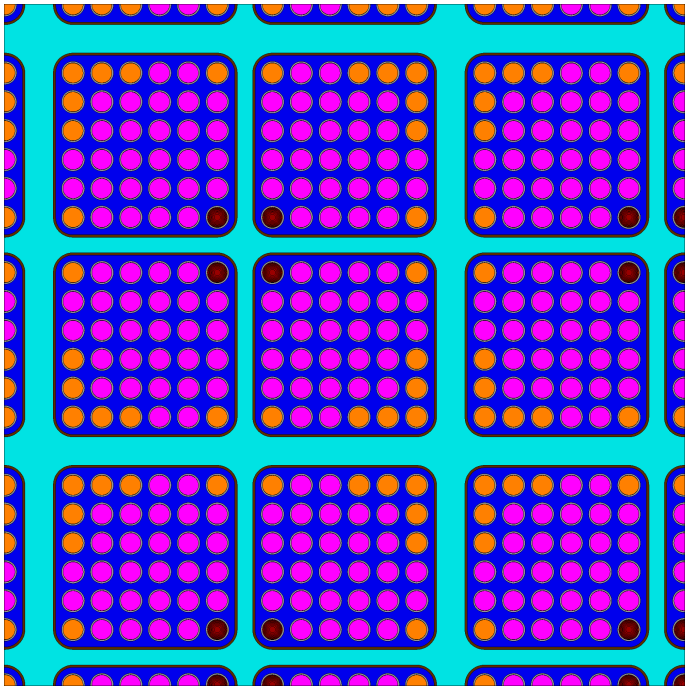

Gundremmingen-A assembly B23 sample I2680 depletion calculation with Serpent 2

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VTT

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Summary

The work described in this report presents simplified and advanced calculations in EU project EURAD work package 8 Spent Fuel Characterization and Evolution Until Disposal (SFC) subtask 2.1. The report presents Serpent 2 depletion calculations of one sample in a 6x6 BWR assembly. The Serpent calculated nuclide concentrations are compared to measured concentrations available in SFCOMPO-2.0. Decay heat of the calculated sample is also examined.

The calculations were performed for a two dimensional and a three dimensional assembly using different nuclear data libraries JEFF-3.2 (and JEFF-3.1.1), ENDF/B-VII.1 and JENDL-4.0. All calculations were repeated three times normalizing the reaction rates to different power densities based on different measurements of sample or assembly burnup. The best correspondence to measurement data was achieved using normalization based on ^{148}Nd NDA measurement of sample burnup. The 3D model agreed with the measurements somewhat better than the 2D model particularly for the calculated plutonium and curium concentrations. One explanation to this is probably more realistic neutron spectrum at sample position resulting from more realistic coolant properties as a function of assembly height. Largest differences of the order of 10 % between calculations with different nuclear data libraries occur for ^{244}Cm. Significant differences between 3-6 % are present also for ^{238}Pu and ^{242}Cm concentrations.

Both 2D and 3D calculations agreed with the measurement data within reported measurement uncertainties for nuclides ^{148}Nd, and ^{238}U and in the 3D calculations also for ^{240}Pu and ^{242}Cm. For the other ten/eight nuclides, differences were larger than measurement uncertainties. However, discrepancies in some of the measurement results indicate that the measurements might not be as accurate as claimed at least for some of the nuclides. Sensitivity and uncertainty analysis of the calculated concentrations are conducted in a second report related to SFC task 2.1.

Confidentiality	Public

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Contents

1. Introduction

The work reported in this report is related to EU project EURAD (European Joint Programme on Radioactive Waste Management) work package number 8 SFC (Spent Fuel Characterization and Evolution Until Disposal). The work package consists of five tasks including i) coordination and training, ii) fuel characterization and related uncertainty analysis, iii) fuel and cladding behaviour and interaction after discharge, iv) accident scenario and consequence analysis and v) civil society interaction. This work is related to task 2 subtask 2.1.

The main objective of task 2 is to produce experimentally verified procedures to determine reliable source terms of spent nuclear fuel (SNF), including realistic uncertainties. The different subtasks include both computational and experimental spent fuel characterization and method development. Nuclide inventory of the fuel cladding is also investigated. Subtask 2.1 consists of benchmark calculations, sensitivity and uncertainty analysis and identification of the significant irradiation history parameters influencing the SNF properties. This report focuses on the benchmark calculation, but some minor sensitivity analysis is also included. The benchmark calculation includes two parts: simplified calculation and advanced calculation. The simplified calculation uses standard procedures often applied at VTT for fuel inventory calculations. The advanced calculation applies a more accurate three dimensional model of the fuel assembly. All calculations have been perfomed with Serpent 2 [1].

The benchmark calculation involves comparison of burnup calculation results to experimental values. Each participant could choose the assembly being calculated from 10 different choices including PWRs and BWRs. The assembly chosen by VTT was a BWR assembly number B23 irradiated in the German Gundremmingen-A BWR reactor between 25.8.1969-5.5.1973. The assembly is described in the SFCOMPO-2.0 database [2]. The database includes nuclide composition data on seven samples cut out of six fuel rods of the assembly B23. At VTT, the focus of the calculations was on one sample cut out of the upper part of a fuel rod (A1) located at the corner of the assembly, sample I2680. The purpose was to concentrate on this one sample and to model it as precisely as possible.

This report is structured as follows. In section 2, the modelled sample and fuel assembly are described based on the data in SFCOMPO-2.0 and in section 3 the Serpent models for the 2D and 3D calculations are presented. Section 4 compares the calculated nuclide concentrations to the measured concentrations and presents calculation data on sample decay heat and section 5 gives a short summary and conclusions.

2. Description of Gundremmingen-A assembly B23

Gundremmingen-A was a German boiling water reactor operated between the years 1967-1977. The reactor thermal and net electric capacity was 801 MWt and 237 MWe [3]. Two assemblies, B23 and C16, irradiated for four and three cycles, respectively, were chosen for post irradiation experiments [4,5]. Seven samples from six fuel rods were cut out of the B23 assembly and five samples out of four fuel rods out of the C16 assembly. This study concentrates

solely on one sample from B23 assembly.

Assembly B23 is depicted in Figure 1 [2]. It is a 6x6 assembly with axially uniform fuel. The measured sample I2680 was cut out of fuel rod A1 at the height of 268 cm. Rod A1 is located in a corner position and presented in green colour in the figure. During the first three cycles, the assembly was located at the same location in the core. For the fourth cycle, the assembly was shuffled into another location. Fuel rod A1 was always located in a corner with narrow water gap on both sides. The position of the cruciform control rod is indicated by the black lines in Figure 1.

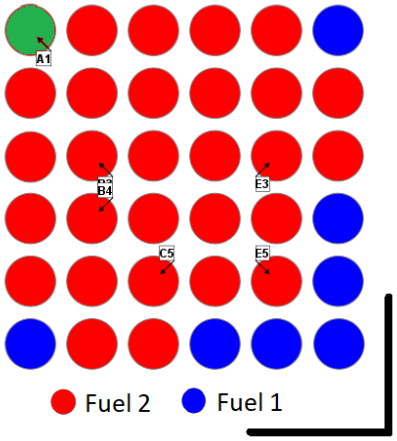

Figure 1. Modelled assembly B23 [2]. The calculated sample is presented in green colour.

The assembly and fuel rod dimensions are given in Table 1 [2, 4, 5]. The fuel density is 10.5 g/cm^3 [4]. The assembly comprises two different fuel types with U-235 enrichments of 1.87 % (fuel 1) and 2.53 % (fuel 2). Rod A1 is of fuel type 2. Fuel cladding is composed of Zircaloy-2 [4, 5] and the channel wall of Zircaloy-4 [2, 4, 5]. The absorber material of the cruciform control rods is B_4C powder and the cladding material SS 304 [2, 4, 5].

Table 1. Assembly and fuel rod dimensions for the Gundremmingen-A assembly B23 [2, 4, 5].

Assembly		Fuel rod	
Parameter	Value [cm]	Parameter	Value [cm]
Channel outer diam	11.352	Pin pitch	1.78
Channel inner diam	11.052	Fuel diam	1.224
Assembly pitch wide	13.098	Gas gap	0.01375
Assembly pitch narrow	12.303	Clad thickness	0.0889
Active length	330.2	Pin diam	1.428

The irradiation history of the assembly B23 is presented in Table 2 and some operating history parameters are given in Table 3 [2, 4, 5]. There is no information on the control rod operating history.

The measurement data given in SFCOMPO-2.0 [2] is taken from references [4, 5] that describe the post-irradiation analysis of the Gundremmingen-A assemblies B23 and C16 performed in two JRC laboratories Ispra and Karlsruhe. The fuel rods were first subjected to non-destructive analysis (NDA) with gamma scanning. For destructive analysis three 1 cm thick samples were cut out of each position. One sample was sent to Karlsruhe, one to Ispra and one was kept for reserve. Mass and alpha spectrometry were used to determine the concentrations and isotopic

Table 2. Irradiation history of the Gundremmingen-A assembly B23 [2, 4, 5].

Cycle of operation	Time period	Duration [EFPD]	Burnup increment [MWd/kgU]
Second	25.8.69-30.5.70	279	5.839
Shut down	31.5.70-24.7.70	56	
Third	25.7.70-12.6.71	323	6.131
Shut down	13.6.71-15.7.71	33	
Fourth	16.7.71-30.4.72	290	5.483
Shut down	1.5.72-30.6.72	61	
Fifth	1.7.72-5.5.73	309	5.174

Table 3. Operating history parameters of the Gundremmingen-A assembly B23 [2, 4, 5].

Parameter	Value	Unit
Void at 268 cm	50	%
Void at 44 cm	0	%
Coolant inlet temp	539	K
Coolant outlet temp	559	K
Fuel temp	923	K
Coolant pressure	69	bar

compositions of uranium and some heavier elements and Nd-148 concentration. Gamma spectrometry was used for the measurement of radioactive fission products. Sample burnup was determined with three methods ^{148}Nd, ^{137}Cs (NDA) and destructive (DA) ^{137}Cs measurements. The burnups based on the measurements of sample I2680 are listed in Table 4.

Table 4. Sample I2680 burnups based on measurements with different methods [4, 5].

Method:	^{148}Nd	^{137}Cs (NDA)	^{137}Cs (DA)
Burnup [MWd/kgU] :	27.40	27.75	23.83

The measurement data in SFCOMPO-2.0 contains also uncertainty estimates. However, many of the differences between the two laboratories for many of the measured components in some of the studied samples [4] are considerably larger than the reported uncertainty values [2]. Most of the measured quantities, including nuclide concentrations, have been normalized to reactor shut down date [2, 4–6].

3. Serpent 2 model

All calculations were performed using Serpent 2, the continuous-energy Monte Carlo particle transport code, developed at VTT [1]. This chapter describes the Serpent models used in the calculations of the Gundremmingen-A B23 assembly. Calculations were first conducted in two dimensions (simplified calculation) and then a three dimensional model of the fuel assembly was constructed for advanced calculations. The fuel and assembly geometry and materials in both models are based on the data given in chapter 2. In the xy-plane at the axial position of the measured sample I2680 the dimensions of both models were identical.

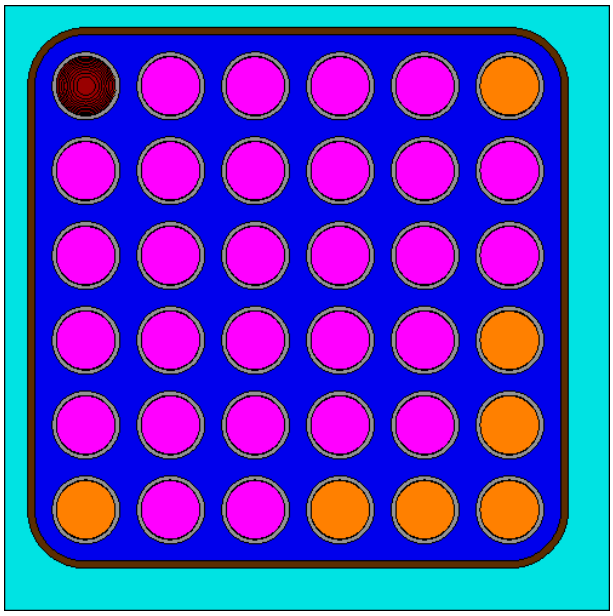

Figure 2. Two dimensional Serpent model of the Gundremmingen-A assembly B23. The dark red fuel pin is sample I2680.

3.1 Two dimensional model

The Serpent model in the two dimensional calculations is presented in Figure 2. Sample I2680 is indicated with dark red colour. Pink colour is used for the other fuel pins of the same type with 2.53 % ^{235}U enrichment. The orange pins are type 1 with ^{235}U enrichment 1.87 %. Dark blue indicates coolant and light blue moderator. The used references did not include information on the rounded corners of the channel walls although e.g. figure 1.33 in reference [5] clearly indicates that the corners are rounded. The radius of the rounded corners was calculated so that the distance from the flow channel outer wall to the fuel pin surface remained constant. Thus, the inner and outer radius of the rounded corners in the model were 1.076 cm and 1.226 cm, respectively.

No information was given on the ^{234}U content in the fuel, so it was omitted and the fuel in the model contained only ^{235}U, ^{238}U and ^{16}O. Oxygen making 11.85 wt-% of the fuel. Zircaloy-2 and Zircaloy-4 compositions were taken from reference [7]. The elemental compositions given in the reference were decomposed into isotopic compositions using Serpent's "-elem" command option. The elemental compositions from [7] are presented in table 5.

Table 5. Elemental compositions of Zry-2 and Zry-4 in the Serpent calculations.

Element	Zircaloy-2 [%]	Zircaloy-4 [%]
Sn	1.5	1.5
Fe	0.15	0.2
Cr	0.1	0.1
Ni	0.05	-
Zr	98.2	98.2

The fuel was divided into pin-wise depletion zones using Serpent's automated depletion zone division. Additionally, sample I2680 was divided in ten equal size radial depletion zones.

In order to make a Monte Carlo simulation meaningful, the integral reaction rate estimates

calculated by the code must be normalized by a given constant. In Serpent, this can be done by a few different options described in Serpent Wiki [8]. None of the possible parameters for normalization were directly available in the SFCOMPO-2.0 database. However, burnup and effective power days (EFPD) of reactor operation were given and these can be used for calculation of power density. Therefore, power density was used for normalization. Burnup however, is not unambiguously defined by the SFCOMPO-2.0 data. Three different burnups were given for the sample depending on the measurement technique (see Table 4). Additionally burnup for the whole assembly was given for each cycle (Table 2). Since normalization can have a significant effect in the calculations, two of the three measured sample burnups and the assembly burnup were used to determine power densities and applied in the calculations. Sample burnup 27.75 MWd/kgU measured with ^{137}Cs (NDA) was ignored since it was nearly the same as the burnup from ^{148}Nd measurement. Power densities calculated from the sample burnup from ^{148}Nd and ^{137}Cs destructive measurements are presented in Table 6 together with average power densities for the entire assembly during each cycle based on the reported assembly burnup (Table 2).

Table 6. Average power densities for the whole assembly at each cycle and for sample I2680 over all cycles.

	Assembly, cycle of operation				I2680, all cycles	
	Cycle 2	Cycle 3	Cycle 4	Cycle 5	^{148}Nd	^{137}Cs (DA)
Power density [kW/gU x 10^{-3}]	20.928	18.981	18.907	16.744	22.814	19.842

As for other operating history data, fuel temperature was assumed constant at the value 923 K throughout the calculation. Coolant and moderator temperatures were estimated as the average of inlet and outlet temperatures yielding 549 K. Additionally Serpent needs information on the water densities. These were obtained by creating a CASMO-4E model [9] of the assembly and making a calculation in 69 bar pressure and 50 % void. The coolant and moderator densities could then be obtained from the CASMO output file. The CASMO calculation was repeated with the different power densities used, but the power densities yielded no visible differences in the water densities. Thus, the densities for coolant and moderator were 0.397 g/cm^3 and 0.759 g/cm^3, respectively. Information on control rod movements was not available so they were assumed to be fully withdrawn during the whole operation and were omitted from the model.

Altogether 69 burnup steps plus three decay steps between cycles were applied to irradiate the fuel for 1201 effective power days plus 150 days of shutdown between the cycles. Each cycle was started with a depletion step of 3 days which corresponds to less than 0.1 MWd/kgU. The step length was gradually incresed to 20 days. The length of the step was never more than double the previous step length. In two of the irradiation cycles, the last step was larger, 26 or 30 days.

All calculations were first run with JEFF based nuclear data. Cross sections based on JEFF-3.2 and fission yield and radioactive decay data based on JEFF-3.1.1 were used. Thermal scattering data for light water was based on JEFF-3.1. In order to make the results more comparable to the results of other participants who might calculate the same sample, a commonly agreed library ENDF/B-VII.1 was used in another set of calculations. For the sake of more complete investigation on the effect of nuclear data on the results also JENDL-4.0 libraries were applied. In the case of JENDL-4.0 libraries, JEFF-3.1 based thermal scattering data for light water was applied because of the the lack of readily available thermal scattering data based on JENDL-4.0.

The assembly was modelled in an infinite lattice using reflective boundary conditions. The substep method developed for Serpent [10] was used in the burnup calculation applying linear extrapolation in predictor and linear interpolation in corrector with 10 substeps in both predictor and corrector. Doppler-broadening rejection correction was used for some uranium and plutonium nuclides. All calculations with the JEFF-based nuclear data libraries were repeated six times with different random number generator seed values to get an idea of the variance caused by the Monte Carlo method on the results. The calculations with the other libraries were also repeated six times when using power density calculated from the burnup based on Nd-148 measurement.

In every 500 active cycles and 50 inactive cycles, 50 000 neutron histories were modelled yielding altogether 25 000 000 neutron histories. The statistical uncertainty in k_{inf} with this population was approximately 10 pcm over the whole irradiation period. The calculations were performed on two different Linux clusters with CentOS Linux version 7 using Intel Xeon 2.2 GHz (cluster 1) and 3.2 GHz (cluster 2) nodes and openMP parallelization. In both clusters, calculations with 20 CPUs took $\sim$ 62 h.

3.1.1 Premininary sensitivity analysis

Because of some approximations and assumptions needed in the model, some preliminary sensitivity calculations were made to estimate the effect and necessity of these assumptions. As expected, power density had a significant effect of several percents on the calculated nuclide concentrations and this was the reason why the actual calculations were repeated with several power densities. Other investigated parameters were the water temperature, sample depletion zone division, flow channel corners and inclusion of impurities in the fuel. In the following, effect on Pu-236 concentrations is ignored since it is rather difficult to calculate and its concentrations in the measurements were at the limit of detection.

The water temperature was approximated by making a linear fit as a function of axial distance based on the inlet and outlet temperatures yielding the temperature of 555 K instead of 549 K with simple averaging. The effect of this change was negligible and it was decided to simply use the average of inlet and outlet temperatures.

In the tests the sample was divided in two depletion areas, a 0.3 mm thick outer rim and the rest of the fuel. In one test the division was ignored and the sample was depleted as one single material. This had a small effect of the order of 1-2 % to the concentrations of Pu-242 and Cm-244. Therefore finer depletion zone division was decided for the calculations.

Since the radius of the rounded corners of the flow channel was not given in the SFCOMPO-2.0 definitions, the effect of having rounded versus sharp corners was investigated. This generally had an effect of 1-2 % on many of the measured and calculated isotopic concentrations. The fact that the calculated sample is located at the corner of the assembly probably plays a role on the effect of the corners. If the sample had been in the centre of the assembly, the effect would probably have been clearly less.

There was no information on impurities in the fuel in the SFCOMPO-2.0 or the other references. Therefore, impurities were not included in the calculations at this point. In order to see if this decision has any effect on the quantities of the measured nuclides 10 ppm of N-14 and 10 ppm of Cl-35 were added in the fuel. The addition of these impurities had no effect on the concentrations of the measured nuclides.

3.2 Three dimensional model

The three dimensional model was similar to the 2D-model in the xy-plane along the active fuel length of the assembly. The 3D-model in the xz-plane is presented in Figure 3. The pink pillars in the figure represent the fuel pins and the thin red horizontal slab between the pink pillars in the upper part of the assembly represents the axial level of the measured sample I2680. The fuel was divided into burnup zones radially pin by pin and axially according to the lines seen in Figure 3 along the axial length of the fuel pins. Additionally, sample I2680 was divided in ten equal area rings like in the 2D calculation. The greyish and violet slabs above and below the fuel pins represent the plenum area (greyish area right above the fuel rods) and bottom and top tie plates and end plugs.

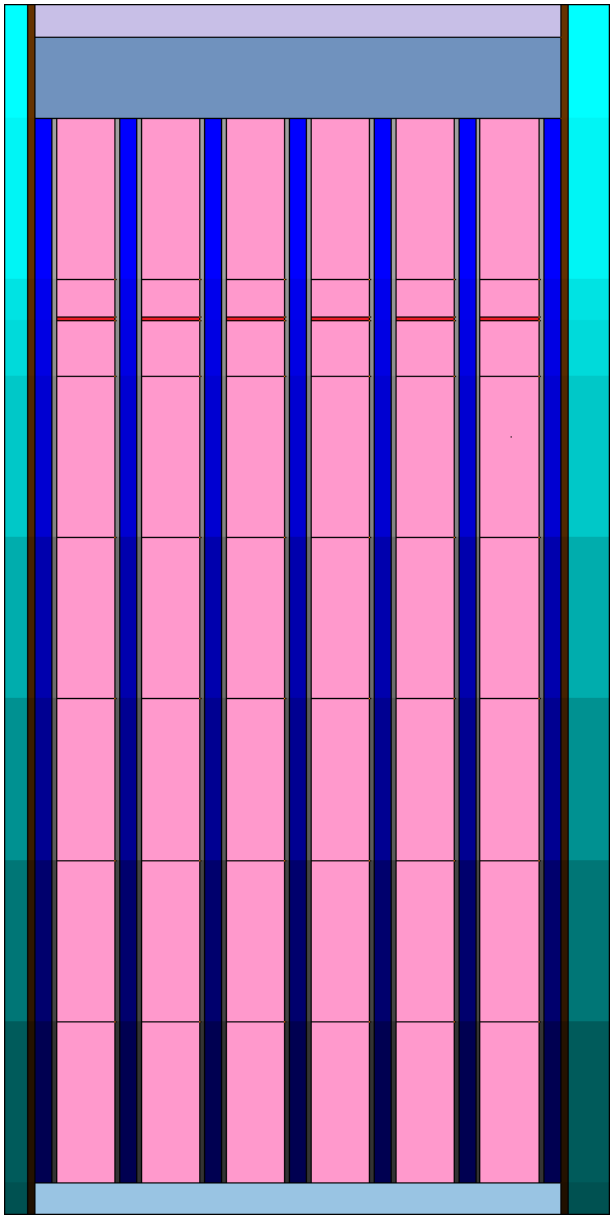

Figure 3. Three dimensional Serpent model of the Gundremmingen-A assembly B23 in xz-plane. The pink pillars represent the fuel and the red thin discs between the pink pilars picture the axial level of sample I2680.

No information was available on the Gundremmingen-A assembly top and bottom structures

and fuel plenum area. Therefore, these structures were approximated based on information in reference [11]. The reference describes general structures and material compositions for BWR and PWR assemblies for ORIGEN model creation. In the Serpent model, the area below the fuel is a homogenized mixture of the materials in the lower tie plate and end plug. The area above the fuel is a homognized mixture of the gas plenum and spring and the area above the plenum is a homogenized mixture of the materials in the upper end plug and tie plate. The length of these areas is scaled down from those given in reference [11] based on the active fuel height of the Gundremmingen-A fuel and the fuel depicted for a BWR in reference [11]. Then the height of the material areas are further rounded to a nicer number, since the scaled number is in any case only a rough estimate. The purpose of modelling the end structures, especially the top of the assembly, is to have an estimate of neutron reflection from the end structures back to the fuel instead of simply applying black boundary conditions above and below the active fuel. The used material compositions and heights are presented in table 7

Table 7. Material compositions of the bottom and top structures of the B23 assembly used in the Serpent model.

Zone	Length	Material volume fractions [%]					
	[cm]	Water	SS 304	Zry-2	SS 302	Inconel X-750	He
Bottom	10	75.82	24.18				
Plenum	25	59.7		7.8	2.0		30.5
Top	10	75.18	15.24	6.94		2.64	

The coolant and moderator temperatures and densities and cladding and channel wall temperatures were defined for different elevations using Serpent's multiphysics interface. The temperatures and densities were defined separately for the same axial zones as the depletion zone division indicated with thin lines in the fuel and colour differences in the materials in Figure 3. The temperature changes were assumed linear assuming inlet and outlet temperatures at the bottom and top of the model. the void fraction was also assumed to behave linearly using void 0 % at the height of 44 cm and below and void 50 % at the height of sample I2680 (268 cm) as specified in SFCOMPO. This means that the water temperature and therefore density at the sample position were somewhat different from the 2D model, namely 555 K and 0.391 g/cm^3 (coolant), 0.747 g/cm^3 (moderator).

The assembly was modelled in an infinite lattice in the radial direction (reflective boundary conditions) just like in the 2D case. In the axial direction black boundary conditions were applied. The total number of neutron histories run was 40 000 000 using 200 active cycles and 200 000 neutrons in each cycle. Number of inactive cycles had to be increased from 50 used in the 2D calculation to 180 in order to reach convergence in the fission source. The statistical uncertainty in k_{inf} was approximately 20 pcm during the whole irradiation period. Calculations in cluster 1 with 40 CPUs took $\sim$ 70 h and in cluster 2 with 20 CPUs $\sim$ 8 d.

4. Results

In the following inspection of results, results are given for all three cases where the calculation was normalized to power density determined from effective power days and the three different burnups where sample burnup was based on ^{148}Nd (^{148}Nd) or ^{137}Cs (DA) (^{137}Cs) measurements or the assembly averaged burnup (Assembly). in all three cases results are presented

using the three different nuclear data libraries JEFF-3.20 (JEFF), ENDF/B-VII.1 (ENDF/B) and JENDL-4.0 (JENDL).

4.1 Results of the simplified calculations

Table 8 presents the calculated sample and assembly burnups. The sample burnups based on measurements of ^{148}Nd and ^{137}Cs (DA) are 27.40 and 23.83 MWd/kgU and assembly averaged burnup based on measurements is 22.627 MWd/kgU. The correspondence between measured and calculated burnups is excellent when comparing measured burnup with the calculation where power density was derived from the corresponding sample or assembly burnup. Differences between libraries are mostly negligible. When normalizing to assembly burnup, there is a small 0.1 % difference in the sample burnups between calculations with the different libraries. When normalizing to sample burnup, the calculated assembly burnup with the ENDF/B-VII.1 libraries differs approximately 0.1-0.2 % compared to the other libraries.

Table 8. Sample and assembly burnups calculated with different nuclear data libraries and normalizations to power density calculated from different measurements of sample and assembly burnup.

	^{148}Nd			^{137}Cs (DA)			Assembly		
	JEFF	ENDF/B	JENDL	JEFF	ENDF/B	JENDL	JEFF	ENDF/B	JENDL
sample	27.41	27.41	27.41	23.84	23.84	23.84	25.87	25.83	25.85
assembly	24.022	24.054	24.032	20.814	20.849	20.827	22.627	22.627	22.627

Table 9 presents the calculated / experimental ratios of the nuclide concentrations. The experimental values are those measured at Ispra laboratory. Not all samples in SFCOMPO, including I2680, included measurement data from both laboratories (Ispra and Karlsruhe). Percentual differences between calculated and measured values (calculated/measured-1) are presented in Figures 4-6. The y-axis limits in all three figures has been set to [-40 – 10 %] for easier comparison. For ^{244}Cm the differences between calculations normalized to sample burnup from ^{137}Cs and measurement are 55, 50 and 51 % for the libraries JEFF-3.2, ENDF/B-VII.1 and JENDL-4.0, respectively, and are off the scale in Figure 5. In addition to the nuclides presented in Table 9 and Figures 4-6, also ^{236}Pu and ^{241}Am nuclide concentrations were measured, but are not included in this study. ^{241}Am is left out because of large uncertainties related to the measurements. In some cases for the Gundremmingen-A samples, two different laboratories got significantly different measurement results (over 80 %) [4]. ^{236}Pu was left out because the measured concentrations were at the lower limits of detection [4]. Similar approach has been taken also e.g. in previous SCALE calculations of the Gundremmingen-A samples [6].

Overall, best correspondence with the measurements is clearly achieved when ^{148}Nd measured burnup is used. The only exceptions are ^{137}Cs that has the best agreement with measurements with ^{137}Cs measured burnup and ^{235}U that has the best agreement when assembly burnup was used in the calculations. The calculated results mostly tend to underestimate the measured nuclide concentrations. Clear exceptions to this is ^{137}Cs when ^{148}Nd burnup was used in the calculations and ^{235}U when ^{137}Cs measured burnup was used.

The largest differences between nuclear data libraries of the order of 10 % occur for ^{244}Cm. Differences between 3-6 % between libraries occur also for ^{238}Pu and ^{242}Cm. The differences are mostly between JEFF-3.2 and the other two libraries. Differences between ENDF/B-VII.1 and JENDL-4.0 are generally much smaller except in the case of ^{244}Cm when the differences between these libraries is 44 %. Differences of the order of 1-1.5 % occur for ^{148}Nd and ^{240}Pu between JENDL-4.0 and JEFF-3.2 and for ^{241}Pu between ENDF/B-VII.1 and JEFF-3.2. All

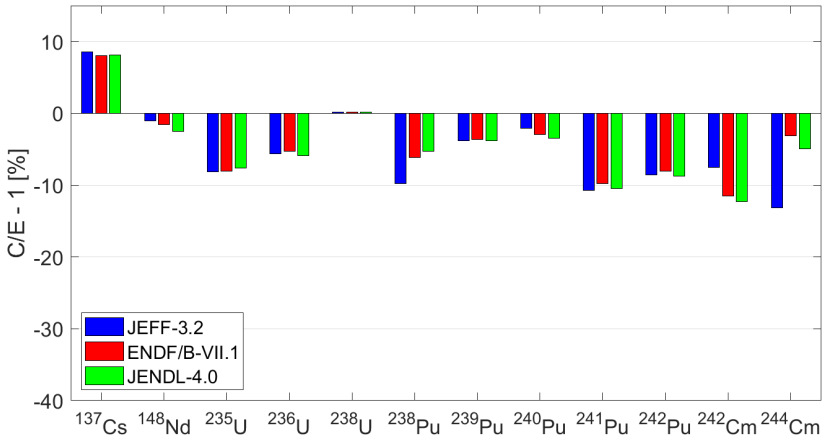

Figure 4. Calculated/Measured - 1 isotopic concentrations in % when normalizing to sample burnup from ^{148}Nd measurement.

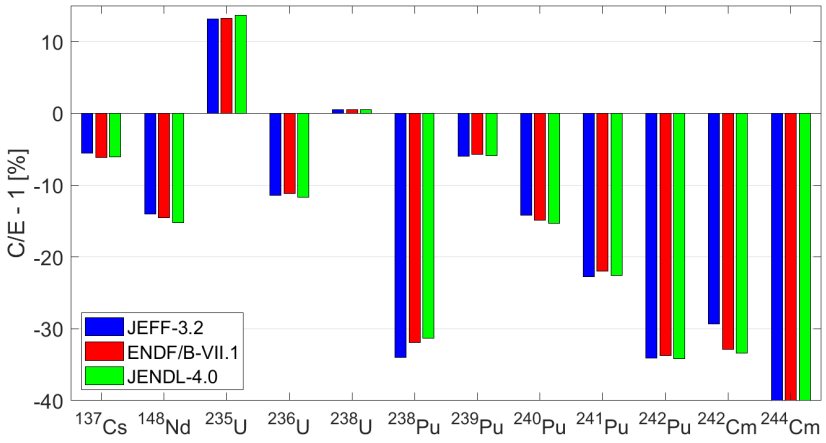

Figure 5. Calculated/Measured - 1 isotopic concentrations in % when normalizing to sample burnup from ^{137}Cs (DA) measurement.

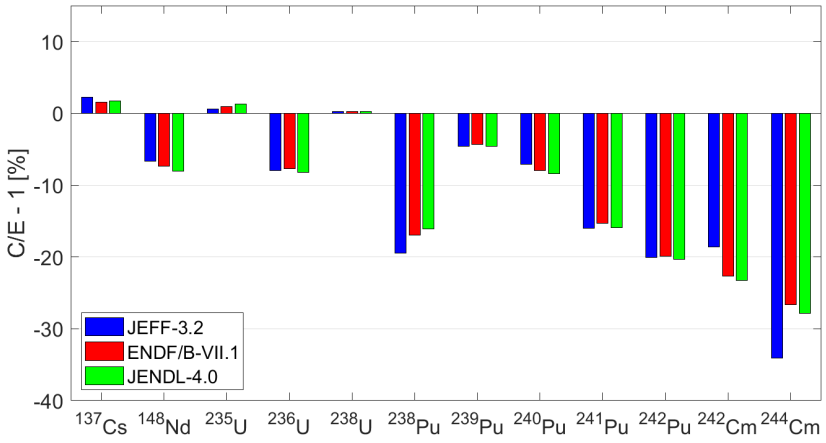

Figure 6. Calculated/Measured - 1 isotopic concentrations in % when normalizing to measured assembly burnup.

Table 9. Ratios C/E of the isotopic concentrations with all libraries and power densities in the 2D calculations. The E values have been measured at Ispra laboratory.

	^{148}Nd			^{137}Cs (DA)			Assembly		
Nuclide	JEFF	ENDF/B	JENDL	JEFF	ENDF/B	JENDL	JEFF	ENDF/B	JENDL
^{137}Cs	1.09	1.08	1.08	0.94	0.94	0.94	1.02	1.02	1.02
^{148}Nd	0.99	0.98	0.98	0.86	0.85	0.85	0.93	0.93	0.92
^{235}U	0.92	0.92	0.92	1.13	1.13	1.14	1.01	1.01	1.01
^{236}U	0.94	0.95	0.94	0.89	0.89	0.88	0.92	0.92	0.92
^{238}U	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
^{238}Pu	0.90	0.94	0.95	0.66	0.68	0.69	0.81	0.83	0.84
^{239}Pu	0.96	0.96	0.96	0.94	0.94	0.94	0.95	0.96	0.95
^{240}Pu	0.98	0.97	0.97	0.86	0.85	0.85	0.93	0.92	0.92
^{241}Pu	0.89	0.90	0.90	0.77	0.78	0.77	0.84	0.85	0.84
^{242}Pu	0.91	0.92	0.91	0.66	0.66	0.66	0.80	0.80	0.80
^{242}Cm	0.92	0.89	0.88	0.71	0.67	0.67	0.81	0.77	0.77
^{244}Cm	0.87	0.97	0.95	0.45	0.50	0.49	0.66	0.73	0.72

other differences between libraries are less than 1 %. Differences between libraries in ^{238}U concentration are negligible ($<$ 0.006 %).

Very similar differences were achieved between SCALE calculations and measurements [6] for nuclides ^{148}Nd, ^{235}U, ^{238}U, ^{239}Pu and ^{240}Pu when comparing to Serpent calculations with ^{148}Nd measured burnup. The power density in the Serpent calculations was the same as the average power density over all cycles used in the SCALE calculations. Serpent agreement with measurement was slightly better for ^{241}Pu and ^{242}Pu and SCALE results were slightly better for ^{236}U. Serpent calculated clearly better results for ^{242}Cm and SCALE succeeded clearly better for ^{244}Cm. The ^{137}Cs results are not comparable since the measured concentration reported in the SCALE calculations is different than that reported in SFCOMPO-2.0.

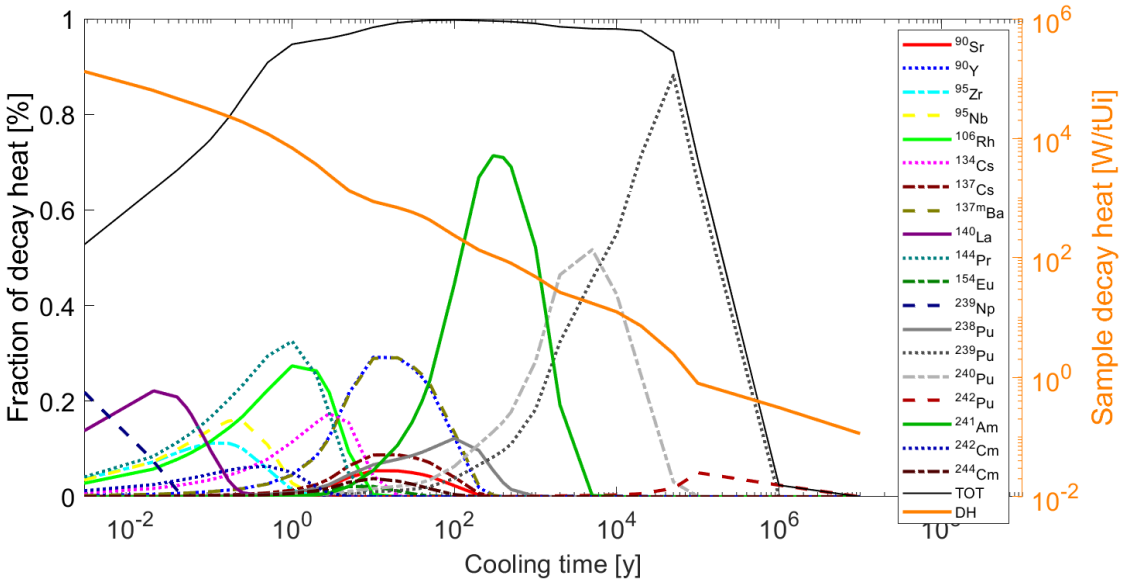

Figure 7. Contribution to sample I2680 decay heat by the most significant decay heat producing nudlices. TOT stands for the total fraction of decay heat from all the nuclides presented in the figure.

Figure 7 presents sample I2680 decay heat as a funtion of decay time and the percentual contribution of the most important decay heat contributors from the calculations based on burnup from ^{148}Nd measurement and JEFF based nuclear data. The nuclides presented in the figure are those responsible for more than 99 % of the total decay heat between 30 – 500 years after irradiation and some shorter lived nuclides whose contribution is significant at shorter time periods after irradiation. Between 30 – 100 y after irradiation, the four most important decay heat producers are ^{137m}Ba, ^{90}Y, ^{241}Am and ^{238}Pu who contribute around 80 % of the total decay heat between this period. After 100 years the importance of ^{137m}Ba and ^{90}Y begins to diminish as their precursors ^{137}Cs ($T_{1/2}$=30.1 y) and ^{90}Sr ($T_{1/2}$=28.8 y) decay. After this period ^{239}Pu and ^{240}Pu become more important.

4.2 Results of the advanced calculations

The 3D calculations were done for the three different power density normalizations like the 2D calculations. Different libraries were not repeated except for the calculations based on ^{148}Nd measured burnup. In this case calculations were repeated using the ENDF/B-VII.1 libraries. Table 10 presents the ratios of calculated isotopic concentrations (3D) divided by the measured concentrations. Ratios from 2D calculations based on ^{148}Nd measured burnup with JEFF libraries are also presented for comparison. Figure 8 presents the differences to measurements in percentage in all 3D calculated cases.

Table 10. Ratios C/E of the isotopic concentrations in all cases in the 3D calculations and in the 2D calculation when normalizing to ^{148}Nd measured burnup and applying JEFF-3.2 cross sections.

	^{148}Nd 2D	^{148}Nd		^{137}Cs (DA)	Assembly
Nuclide	JEFF	JEFF	ENDF/B	JEFF	JEFF
Cs-137	1.09	1.10	1.09	0.96	0.88
Nd-148	0.99	1.00	1.00	0.87	0.80
U-235	0.92	0.91	0.91	1.13	1.25
U-236	0.94	0.95	0.95	0.89	0.85
U-238	1.00	1.00	1.00	1.00	1.01
Pu-238	0.90	0.94	0.98	0.69	0.58
Pu-239	0.96	0.98	0.98	0.96	0.94
Pu-240	0.98	1.00	0.99	0.87	0.80
Pu-241	0.89	0.91	0.93	0.80	0.73
Pu-242	0.91	0.95	0.94	0.69	0.57
Cm-242	0.92	0.96	0.92	0.74	0.62
Cm-244	0.87	0.96	1.10	0.48	0.34

Similarly to 2D calculations, best agreement with measurement results is achieved using the burnup based on ^{148}Nd measurement. The only exception being ^{137}Cs concentration that has the best agreement with measurements when applying ^{137}Cs (DA) measured burnup. Again, calculated concentrations mostly underestimate the measured values. Exceptions to this are ^{137}Cs concentration (both libraries) and ^{244}Cm (ENDF/B-VII.1) based on burnup from ^{148}Nd measurement and ^{235}U concentration in calculations based on ^{137}Cs measured burnup or assembly burnup.

The 2D and 3D calculation differences to measurements together with measurement uncertainties reported in SFCOMPO-2.0 [2] are presented in Figure 9. Clearly increased accuracy in the 3D calculations compared to 2D is observed for the heavier nuclides starting from ^{238}Pu. For the lighter nuclides, the differences to measured values are rather similar. One explanation

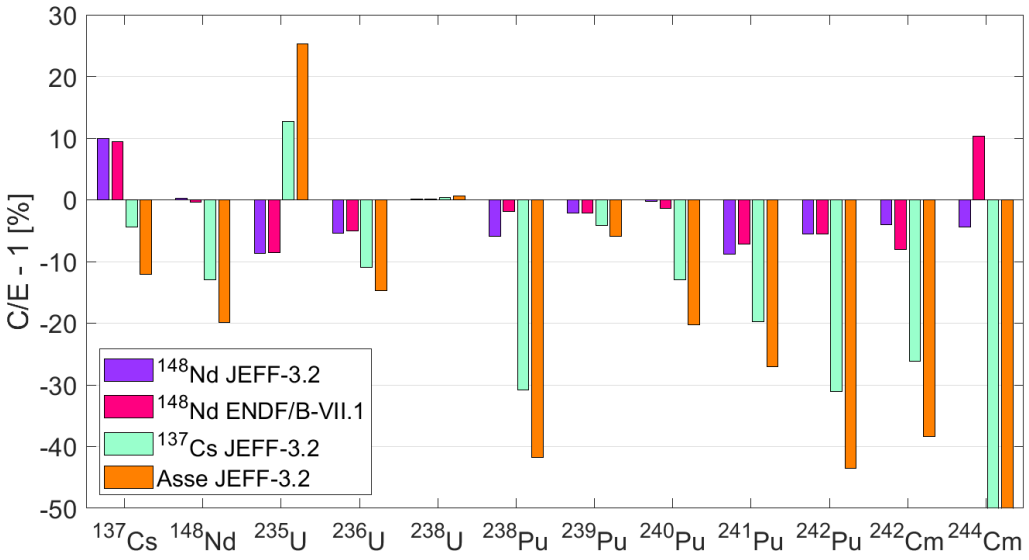

Figure 8. Calculated/Measured - 1 isotopic concentrations in % in the 3D calculations.

to the increased accuracy of the transuranium nuclides is probably related to the slightly harder neutron spectrum in the 3D calculations. Coolant and moderator densities are slightly smaller at sample position (0.391 vs. 0.397 g/cm^3 for coolant and 0.747 vs. 0.759 for moderator) in the 3D calculation. Harder spectrum generally causes more transuranium built-up and in this case reduces the difference between measured and calculated results. The water densities and neutron spectrum in the 3D calculation are likely more realistic than in the 2D calculation since they take into account the gradual change in the coolant properties as a function of assembly height. The differences to measurement results are within reported measurement uncertainties for ^{148}Nd and ^{238}U and in the 3D case also for ^{240}Pu and ^{242}Cm. It is worth to note that in several cases the values measured for the Gundremmingen-A samples at Ispra and Karlsruhe differ considerably more between the two institutions than the reported uncertainty values.

Difference between sample decay heat with the 2D and 3D model is mostly around 2 %. The 2D calculation underestimates the 3D calculation at all times after irradiation. Differences in decay heat from ^{90}Sr and ^{90}Y between 2D and 3D calculations are less than 1 %. Differences for the other nuclides presented in Figure 7 are between 1 – 5 % except for a couple of nuclides when the nuclide concentration rapidly starts to decline.

5. Summary and conclusions

Isotopic concentrations have been calculated with Serpent 2 for one sample I2680 from assembly B23 irradiated in the BWR reactor Gundremmingen-A and compared to measurement results reported in SFCOMPO-2.0 [2]. Calculations were done using a two dimensional assembly model and a three dimensional model. In both cases the calculations were normalized in three ways using power density calculated from effective power days and three different measured burnup values. The burnups were sample burnups based on ^{148}Nd NDA measurement and on ^{137}Cs DA measurement and assembly burnup. Best agreement with measurements was achieved using sample burnup based on ^{148}Nd measurement.

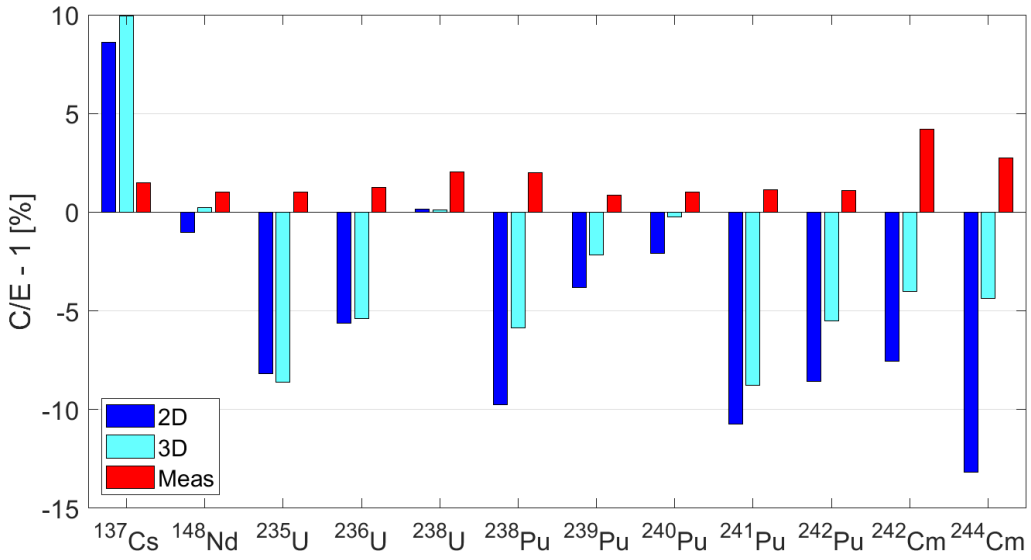

Figure 9. Calculated/Measured - 1 isotopic concentrations in % in the 2D and 3D calculations using ^{148}Nd measured burnup and JEFF-3.2 cross sections. Measurement uncertainties from SFCOMPO-2.0 are presented in red bars.

In the 2D case, three different nuclear data libraries were used. The reference case applied cross sections based on JEFF-3.2 and decay and fission yield libraries based on JEFF-3.1.1. The other cases applied cross section, decay and fission yield libraries based on ENDF/B-VII.1 and JENDL-4.0 evaluated nuclear data. Largest differences between libraries were obtained for ^{244}Cm. Clear differences were also observed for ^{238}Pu and ^{242}Cm concentrations.

Results calculated with the 3D model agreed better with measurements than the 2D results for all studied plutonium and curium isotopes. One explanation to this is likely the more realistic coolant properties as a function of assembly height and hence more realistic neutron spectrum at sample position. However, due to the lack of accurate data in axial direction both for the assembly geometry and operating history, it is hard to say for sure whether this improvement is due to the 3D model or just a happy accident. For example, accurate information on void fraction and water temperature was not available for different elevations and these had to be approximated from inlet and outlet temperatures and void fraction at two elevations. Also, no data on the assembly end structures was available.

The 3D calculations were within reported measurement uncertainties for ^{148}Nd, ^{238}U, ^{240}Pu and ^{242}Cm. An uncertainty estimation for the calculations is conducted in a different report [12].

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