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Original Article

Simulation of low-enriched uranium burnup in Russian VVER-1000 reactors with the Serpent Monte-Carlo code

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ABSTRACT

This work deals with the assessment of the burnup capabilities of the Serpent Monte Carlo code to predict spent nuclear fuel (SNF) isotopic concentrations for low-enriched uranium (LEU) fuel at different burnup levels up to 47 MWd/kgU. The irradiation of six UO_2 experimental samples in three different VVER-1000 reactor units has been simulated and the predicted concentrations of actinides up to ^{244}Cm have been compared with the corresponding measured values. The results show a global good agreement between calculated and experimental concentrations, in several cases within the margins of the nuclear data uncertainties and in a few cases even within the reported experimental uncertainties. The differences in the performances of the JEFF3.1.1, ENDF/B-VII.1 and ENDF/B-VIII.0 nuclear data libraries (NDLs) have also been assessed and the use of the newly released ENDF/B-VIII.0 library has shown an increased accuracy in the prediction of the C/E's for some of the actinides considered, particularly for the plutonium isotopes. This work represents a step forward towards the validation of advanced simulation tools against post irradiation experimental data and the obtained results provide an evidence of the capabilities of the Serpent Monte-Carlo code with the associated modern NDLs to accurately compute SNF nuclide inventory concentrations for VVER-1000 type reactors.

1. Introduction

The safe, secure and sustainable management of the spent nuclear fuel (SNF) coming from the irradiation in commercial reactors is a topic of paramount importance since it affects the design and optimization of interim and long term fuel storage facilities while reducing the associated radiological risks [1]. Moreover, in nuclear criticality safety studies involving SNF, burn-up credit analyses are being performed and implemented as a means of more accurately and realistically determining the system reactivity by taking into account a decrease in the SNF reactivity during irradiation. Such implementation of burn-up credit has gained a world-wide interest during the last twenty years since many countries are including it as part of their criticality safety licensing strategy. Therefore the capability to accurately calculate nuclide inventories for SNF is fundamental to many areas of nuclear plant operations including storage and transportation, reprocessing, conditioning and disposal [2]. Thanks to the continuous improvement of the computational methodologies for reactor analysis and of the associated nuclear data libraries (NDLs) the prediction capabilities of the neutronics and burnup calculations have improved drastically. Numerous numerical benchmarks have been undertaken that involve comparing different code results for reference calculations as a form of code verification and as a means to evaluate code performance relative to other codes and data used by industry and research [3,4]. However, it is fundamental to validate the performances of the simulation tools through extensive and systematic comparisons between calculated values and experimental data. To address this need, the Nuclear Energy Agency (NEA) in cooperation with Oak Ridge National Laboratory (ORNL) has released the SFCOMPO-2.0 database [5], which contains experimental data for 750 SNF samples from 44 different reactors of 8 different technologies including open source bibliographical references to design specifications and operating information to be used by the scientific community. Within SFCOMPO-2.0, experimentally measured isotopic concentrations of well characterized SNF samples are collected in order to be used to validate the prediction accuracy of the depletion codes for given sets of burnups, initial enrichments, and varying power histories.

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This work in particular deals with the burnup simulations of six fuel samples irradiated in three different VVER-1000 reactor units in Russia during the 1980's. All the necessary data for the modelling have been extracted from SFCOMPO-2.0. In Section 2 the description of the problem is provided together with all the necessary details to build the neutronics model. The modelling approach and assumptions adopted in order to reproduce the actual experimental conditions are described in Section 3 and the detailed results are given and discussed in Section 4.

2. Problem definition

The comparison between calculated and measured SNF isotopic compositions is an important stage of the computer codes validation process. In fact, even such measurements are usually rather difficult to interpret due to the uncertainty of some parameters (i.e. fuel temperature, moderator temperature), to the incomplete information about operation history and to the difficulties to account for of nearby assemblies effect, the information that can be extracted from this comparison provides also an indication of the code accuracy in reference to conditions to real power operation.

As part as the SNF measurements related to VVER-1000 reactors, which are not numerous because of their complexity and high costs, post-irradiation data are available in SFCOMPO-2.0 related to fuel assemblies (FAs) discharged from four different reactors. In particular, in this work the experimental data related to three FAs irradiated in the Russian VVER-1000 reactor units Kalinin-1, Balakovo-2 and Balakovo-3 (one FA for each unit) have been considered. For each of the three FAs, measurement data are provided for two irradiated samples, these belonging to the same fuel rod at different axial locations. For all samples, uranium (^{235}U, ^{236}U, ^{238}U), plutonium (^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu), americium (^{241}Am, ^{243}Am), curium (^{242}Cm, ^{244}Cu) and neodymium (^{142}Nd, ^{143}Nd, ^{144}Nd, ^{145}Nd, ^{146}Nd) concentrations were measured by isotopic dilution techniques in combination with mass-spectrometric analyses. The mass fraction of all these isotopes were measured at the Khlopin Radium Institute several years after discharge and recalculated back at the end of irradiation of each sample and per unit mass of the initial uranium [6]. The experimental data were transmitted to ORNL and reported in Ref. [7].

2.1. Model description

All the three reactor units have a core consisting of 163 hexagonal fuel assemblies arranged in the hexagonal lattice with a pitch of 23.6 cm. Each assembly contains 312 UO_2 fuel pins, 18 guide thimbles for control rods or burnable poisons and a central guide/instrumentation tube all arranged in a hexagonal lattice with a pitch of 1.275 cm. The fuel rods are made of annular UO_2 pellets with zirconium alloy cladding. Three types of pin cells are located in each VVER: the fuel pin cell, the burnable absorber pin cell, and a central water hole. For the burnable absorber cell, the central cylindrical portion consists of absorber material followed by a gap, cladding, water, and a cylindrical guide tube. The circular part of the central water hole consists of water surrounded by a cylindrical guide tube. The burnable absorber material consists of pellets containing natural boron in a matrix that is primarily composed of aluminium. The complete geometrical data necessary to build the neutronics model are provided in Table 1.

While the Balakovo-3 FA consisted of 312 fuel pins of uniform 4.4-wt% ^{235}U enrichment, in the FAs irradiated in the Kalinin-1 and Balakovo-2 units the fuel enrichment was non-uniform and they contained 246 pins with 4.4 wt% ^{235}U and 66 pins with 3.6 wt% ^{235}U. The Kalinin-1 fuel assembly is designated as ED-0623 and the related experimental samples are referred as 33 and 448. These samples were taken from the fuel rod 312 which was one of the 3.6 wt % ^{235}U rods on the periphery. The Balakovo-2 and Balakovo-3 FAs were designated as ED-1476 and E−1591 respectively and the experimental samples in these assemblies are referred as 6 and 15 for Balakovo-2 and as 912 and 581 for Balakovo-3. Samples 6 and 15 were taken from the 4.4 wt% ^{235}U fuel rod 42 and samples 912 and 581 were taken from the 3.6 wt% ^{235}U fuel rod 23. All specifications for the isotopic concentrations are taken from Ref. [7] and a scheme of the two different FAs with the locations of the experimental rods is given in Fig. 1.

Table 1
Dimensions of the VVER-1000 components.

Fuel assembly	**[cm]**
Distance between FA centers	23.6
Fuel pin lattice pitch	1.275
Inner diameter/thickness of guide thimbles	1.09/0.08732
Central instrumentation tube inner diameter/thickness	0.96/0.0826
Fuel rod	**[cm]**
Inner clad diameter/Clad thickness	0.772/0.07220
Fuel pellet diameter	0.755
Central hole diameter	0.23
Burnable absorber pin	**[cm]**
Absorber diameter	0.758
Clad inner diameter/Clad thickness	0.772/0.069

In the Kalinin-1 case, the sample rod was exposed for one reactor cycle and the sample burnups were estimated in this work to be 13.7 and 15.5 GWd/t. In the case of both Balakovo-2 and Balakovo-3, the samples were irradiated for three reactor cycles and all burnup values were estimated to be in the range of 44.7 and 47.1 GWd/t. The detailed irradiation histories with actual cycle numbers as used by the reactor operators for the six experimental samples are summarized in Table 2.

3. Modeling approach

The large and cheap computing power available nowadays is fostering the increasing use of Monte Carlo methods for reactor core analysis and as a consequence continuous Monte Carlo depletion analysis is also attracting considerable attention for burnup calculations [8,9]. In this study the irradiation of the six experimental samples has been simulated with the Serpent Monte Carlo code [10]. Serpent is a three-dimensional continuous-energy Monte Carlo reactor physics code developed at VTT Technical Research Centre of Finland since 2004. The objective of our study is to assess the prediction capabilities of Serpent to reproduce the measured actinides concentrations at discharge for the six experimental samples. Furthermore, in order to compare the performances of different NDLs, the simulations of the irradiation of each sample have been performed with three different sets of NDLs, namely the JEFF3.1.1 [11], the ENDF/B-VII.1 [12] and the newly released ENDF/B-VIII.0 [13].

Calculations of SNF isotopic compositions are usually carried out for single-fuel assembly configurations with operational parameters averaged over time under the assumption that taking into account for their real time dependence does not practically influence the calculation. As a rule, the influence of nearby assemblies on nuclide compositions is also neglected. In addition, the actual operational data for most of the lattice parameters (such as fuel temperature, local power density, etc.) are generally not available and these should be obtained from other calculations. This situation introduces additional uncertainties but remains the best possible approach to take profit of the measured data. Therefore, our study reflected the same approach used in Ref. [7] where a benchmark specification was formulated within the approach of an isolated

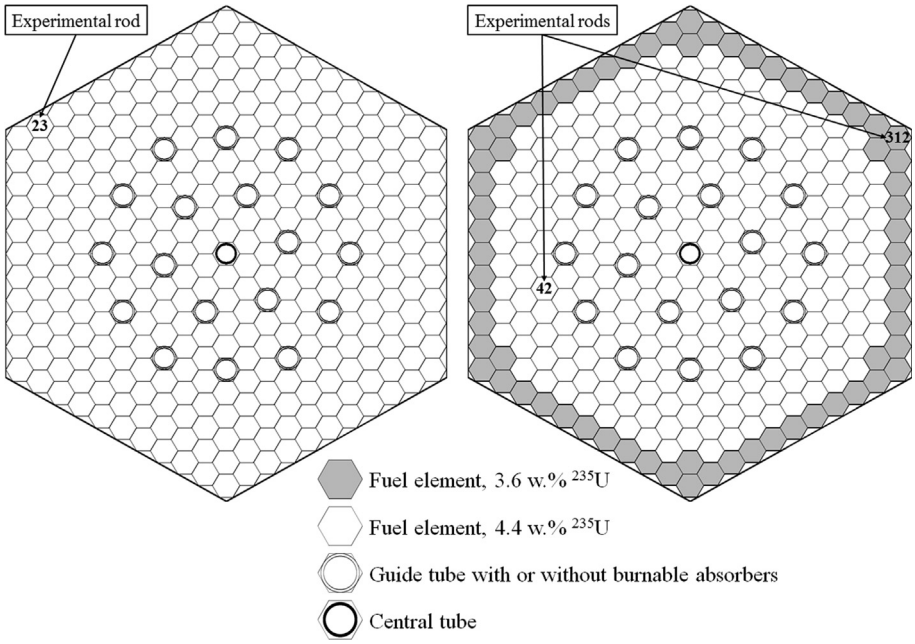

Fig. 1. Fuel assemblies E−1591 (left), E−1476 (right) and E−0623 (right) with locations of the experimental fuel rods.

Table 2
Irradiation histories for the VVER samples.

Reactor-Unit	Assembly Number of pins	Rod	Sample	Cycle	Cycle length/downtime [days]	Sample burnup [MWd/kgU]
Kalinin-1	ED-0623	312	33	5	250/NA	13.7
	246 pins (4.4%^{235}U)		48	5	250/NA	15.5
	66 pins (4.4 wt%^{235}U)					
Balakovo-2	ED-1476	42	6	3	283/189	44.7
	246 pins (4.4%^{235}U)			4	322/76	
	66 pins (4.4%^{235}U)			5	359/NA	
			15	3	283/189	46.5
				4	322/76	
				5	359/NA	
Balakovo-3	E−1591	23	581	2	297/94	47.1
	312 pins (4.4t%^{235}U)			3	350/78	
				4	413/NA	
			912	2	297/94	45.4
				3	350/78	
				4	413/NA	

fuel assembly with a zero-current boundary condition and averaged operational characteristics. These characteristics (such as moderator density and temperature, fuel temperature and boron acid concentration) were obtained from NPP operational data and from calculations simulating the whole core operation performed with the BIPR-7 and the TOPRA-S codes [14]. Since the detailed operational data corresponding to the irradiation of the six samples are not available, in our work we considered the same values for the fuel temperature, moderator density and acid boron content as calculated in Ref. [14] and reported in Ref. [7]. Among the six irradiated samples, the burnup conditions varied slightly with differences in fuel composition, location in the VVER assembly, and burnup amount. Our analysis has been performed by developing individual Serpent models for each irradiated sample.

The Kalinin-1 samples were irradiated for one reactor cycle of 250 days duration. No doubt, the reactor power level varied during this cycle. However, due to the absence of any specific data on the actual power variations and assuming these variations to be not extreme, in our simulations a constant power over the entire cycle has been assumed. The amount of burnup experienced by the samples is of primary importance in the prediction of final nuclide concentrations. Determining each sample's burnup can be difficult, and uncertainties in burnup are a serious hindrance when assessing the validity of a particular simulation approach because the process is reasonably sensitive to variations in burnup [15]. During the measurements, fuel burnups were determined through the concentrations of the neodymium isotopes due to their well know behavior as burnup indicators [6]. In particular, the ^{145}Nd + ^{146}Nd build up method was used. This method is based on the fact that this sum is invariant to the neutron flux (it does not require correction of the mass balance for the (n,γ) reaction) [16]. In our simulations, in order to properly reproduce the actual experimental burnup experienced by each sample, the power density level was also tuned on the experimental ^{145}Nd + ^{146}Nd concentrations. In practice, for each sample it was used a burnup value that produced a calculated-to-measured value that was on average unity for the sum ^{145}Nd + ^{146}Nd.

For the Balakovo-2 and Balakovo-3 experimental samples the irradiation was over three reactor cycles and an average power density was reported from the reactor operators for each sample

location and for each cycle. The length of each cycle was also reported. Thus, the irradiation histories for the Balakovo-2 and -3 samples were constructed by requiring that (1) the $^{145}Nd + {}^{146}Nd$ concentrations to be used as indicators of total burnup as in the case of Kalinin-1, (2) each cycle be of the appropriate duration (and with the reported downtime between cycles), and (3) the power levels for the three cycles for any one sample be in the same ratios as the power densities reported. The values for the power densities used in our simulations resulting from the criteria above together with the values for moderator density, fuel temperature and boron acid content are summarized in Table 3. All these parameters have been taken from Ref. [7] and consistently a moderator temperature of 575 K was assumed for all the simulations.

The version 2.1.30 of the Serpent code has been used and each transport calculation in each burnup step has been performed by running $2 \cdot 10^7$ neutron histories distributed over 200 cycles (10^5 neutron/cycle). In order to achieve a proper convergence of the fission source 20 inactive cycles have also been considered. The Serpent code uses built-in calculation routines for the burn-up calculation which is usually divided into two steps [17]. The first step is the transport cycle in which the rates of all neutron-induced transmutation reactions are calculated. These data are then combined with radioactive decay constants and fission product yields read from NDLs. In our study for the depletion calculations with each NDLs, the corresponding versions of radioactive decay and neutron fission yield data have been used. In the second stage the Bateman equations describing the isotopic changes in the irradiated materials are solved by means of the CRAM method and after that the process is repeated using the updated material compositions. Furthermore, in our Serpent simulations in order into account for the spatial and mutual self-shielding effect, the experimental fuel pins have been divided into ten annular subregions of equal area. As result of an optimization study aiming to assess the impact of the length of the depletion steps on the final calculated values for the isotopic concentrations, a number of steps ranging from 20 to 28 was used for each irradiation cycle, depending on its specific length.

It is also important to note that the depletion solver in the Serpent code is a deterministic module and therefore there is no statistical error associated to the Serpent inventory calculations. Statistical errors in Monte Carlo solutions are introduced into nuclide number densities through various types of reaction rates and the nuclide number densities errors are then propagated though the burnup. However, there have been several studies in the frame of statistical error propagation in Monte Carlo depletion calculations and the major conclusions were that the propagated errors are negligible with respect to the nuclear data uncertainties [8,18]. In addition, the burnup capabilities of the Serpent code were also successfully verified within the framework of a numerical benchmark exercise in Refs. [3,4] through an extensive comparison with deterministic simulations performed with the SCALE code. The results of this study showed a remarkable agreement between Monte Carlo and deterministic results on the final nuclide concentrations for VVER-1000 FAs loaded with LEU fuel and irradiated up to ~40 MWd/kgHM. Therefore, in this study the statistical error associated to the final nuclide concentrations computed with Serpent is considered to affect the results by a negligible amount. The Serpent models of the VVER-1000 FAs types are shown in Fig. 2.

4. Discussion of results

Calculated-to-experimental ratios (C/E's) of nuclide concentrations evaluated with the JEFF3.1.1, ENDF/B-VII.1 and ENDF/B-VIII.0 NDLs for the six irradiated samples are listed in Table 3. The experimental errors associated to each measurement as reported in Ref. [7] are given in Table 4. The agreement between computed and measured concentrations can be considered as satisfactory for the majority of the isotopes since it falls mostly within the range of the nuclear data uncertainties and in a few cases even within the experimental uncertainties (see Table 5). Moreover, the general tendency of the calculations to under-predict the experimental data can be observed. However, for a few cases one can also observe opposite trends (under-/over-prediction) in the C/E's for two samples belonging to the same fuel rod. This is for example the case of the ^{235}U prediction in samples 581 and 912. Exactly the same trend was found also in Ref. [7] which seem to indicate either a problem on the measurements or that the adopted modeling assumptions (i.e. the values for the state variables reported in Ref. [7]) for samples 581 and 912 do not correctly represent the actual physics of the experiment. The qualitative trends for the C/E's of this study are consistent with the ones reported in Ref. [7]. However, for most of the isotopes one can also observe an increased accuracy of the simulations performed with the Serpent code and modern NDLs with respect to the ones that used the HELIOS code with ENDF/B-VI data in Ref. [7] (see Fig. 3 through 8). In particular it is worth to highlight the strong increase in the accuracy of the ^{244}Cm predictions in our study. The predictions for fuel rod 42 (Balakovo-2) are of higher accuracy with respect to the ones for fuel rods 312 (Balakovo-3) and 23 (Kalinin-1). This is probably due to the fact that this rod is an internal one, suggesting that it might be possible to obtain a better accuracy when modeling rods 312 and 23 by including adjacent FAs in the Serpent model in order to take into

Table 3
Relevant parameters for the six VVER-1000 samples as used in the Serpent simulations.

Reactor-Unit	Assembly/Rod	Sample	Cycle	Power density [W/g]	Moderator Density[a] [g/cc]	Fuel Temperature[a] [K]	Boric acid content[a] [g/kg]
Kalinin-1	E−0623/312	33	5	50.90	0.68	963	4.01
		448	5	57.53	0.74	988	4.01
Balakovo-2	E−1476/42	6	3	58.96	0.72	1002	2.52
			4	50.53	0.72	877	2.45
			5	38.28	0.73	809	2.72
		15	3	55.53	0.69	966	2.52
			4	49.50	0.69	874	2.45
			5	37.14	0.71	811	2.72
Balakovo-3	E−1591/23	581	2	41.27	0.74	891	2.10
			3	42.77	0.74	892	2.35
			4	37.89	0.74	822	2.62
		912	2	34.72	0.70	853	2.10
			3	44.46	0.69	881	2.35
			4	37.44	0.70	824	2.62

[a] Values extracted from [7].

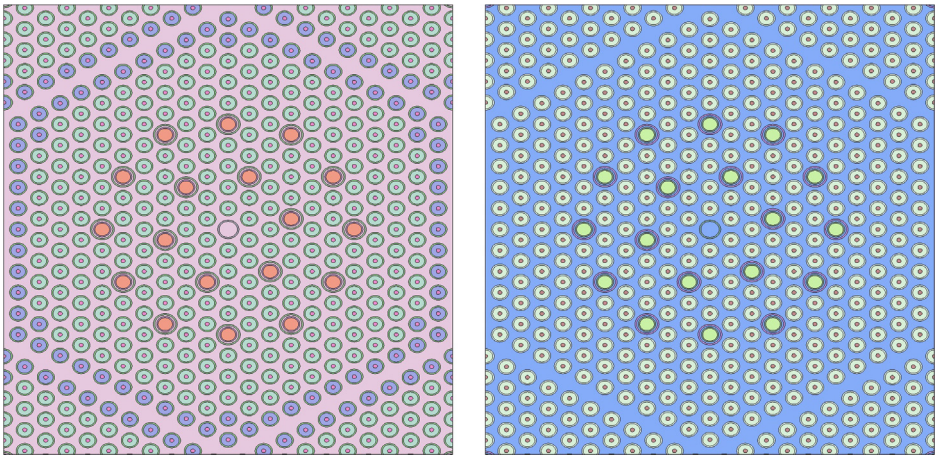

Fig. 2. Serpent models of the E−0623 (left), E−1476 (left) and ED-1591 (right) fuel assemblies.

Table 4
Calculated/Experimental (C/E) ratios of isotopic concentrations computed with different NDLs.

Isotope	Kalinin-1/FA E−0623					
	C/E's - Sample 33			C/E's - Sample 448		
	JEFF3.1.1	ENDF/B-VII.1	ENDF/B-VIII.0	JEFF3.1.1	ENDF/B-VII.1	ENDF/B-VIII.0
^{235}U	0.97	0.97	0.97	1.01	1.01	1.00
^{236}U	0.82	0.82	0.84	0.82	0.82	0.84
^{238}U	0.99	0.99	0.99	1.00	1.00	1.00
^{238}Pu	0.77	0.75	0.79	0.69	0.71	0.72
^{239}Pu	0.93	0.93	0.93	0.92	0.92	0.92
^{240}Pu	0.94	0.93	0.93	0.91	0.91	0.91
^{241}Pu	0.96	0.96	0.97	0.92	0.91	0.92
^{242}Pu	1.00	0.98	0.98	0.90	0.88	0.89
^{243}Am	1.03	1.13	1.05	0.86	0.92	0.84
^{244}Cm	0.64	0.72	0.73	0.96	1.04	1.07
Isotope	**Balakovo-2/FA E-1476**					
	C/E's - Sample 6			C/E's - Sample 15		
	JEFF3.1.1	ENDF/B-VII.1	ENDF/B-VIII.0	JEFF3.1.1	ENDF/B-VII.1	ENDF/B-VIII.0
^{235}U	0.95	0.94	0.92	0.99	0.99	0.97
^{236}U	0.93	0.94	0.95	0.91	0.92	0.93
^{238}U	0.99	0.99	0.99	0.99	0.99	0.99
^{238}Pu	0.83	0.80	0.89	0.82	0.78	0.87
^{239}Pu	0.96	0.97	0.98	0.99	1.00	1.01
^{240}Pu	1.00	0.99	1.01	1.02	1.02	1.02
^{241}Pu	0.94	0.95	0.96	0.97	0.98	0.99
^{242}Pu	1.04	1.03	1.05	1.02	0.99	1.01
^{241}Am	0.78	0.88	0.80	1.10	1.23	1.13
^{243}Am	1.01	1.11	1.01	1.03	1.10	1.00
^{242}Cm	1.01	0.97	1.02	1.11	1.04	1.11
^{244}Cm	0.98	1.08	1.12	0.94	1.01	1.04
Isotope	**Balakovo-3/FA E-1591**					
	C/E's - Sample 581			C/E's - Sample 912		
	JEFF3.1.1	ENDF/B-VII.1	ENDF/B-VIII.0	JEFF3.1.1	ENDF/B-VII.1	ENDF/B-VIII.0
^{235}U	1.08	1.09	1.06	0.93	0.93	0.91
^{236}U	0.89	0.90	0.91	0.91	0.91	0.93
^{238}U	0.99	0.99	0.99	0.99	0.99	0.99
^{238}Pu	0.78	0.73	0.82	0.73	0.70	0.78
^{239}Pu	0.95	0.96	0.95	0.92	0.93	0.93
^{240}Pu	0.96	0.96	0.96	0.95	0.95	0.95
^{241}Pu	0.94	0.94	0.94	0.92	0.92	0.92
^{242}Pu	0.88	0.86	0.88	0.96	0.94	0.95
^{241}Am	0.66	0.74	0.67	0.85	0.94	0.85
^{243}Am	0.88	0.94	0.83	1.00	1.07	0.96
^{242}Cm	1.04	0.98	1.03	1.05	0.99	1.04
^{244}Cm	0.78	0.84	0.84	0.89	0.96	0.97

Table 5
Experimental error in the measurements of the actinides final concentrations [6,7].

Isotope	Kalinin-1/FA E–0623		Balakovo-2/FA E–1476		Balakovo-3/FA E–1591	
	Sample 33	Sample 448	Sample 6	Sample 15	Sample 581	Sample 912
	Experimental error [%]		Experimental error [%]		Experimental error [%]	
^{235}U	0.4	0.4	0.7	0.7	0.7	0.6
^{236}U	0.7	0.6	0.8	0.6	0.6	0.8
^{238}U	0.1	0.1	0.09	0.1	0.1	0.1
^{238}Pu	6.2	4.5	6.4	3.3	3.1	6.0
^{239}Pu	6.3	8.2	8.2	6.6	8.0	8.2
^{240}Pu	1.1	1.9	1.1	0.6	1.1	0.6
^{241}Pu	2.4	1.9	1.1	0.6	1.1	0.6
^{242}Pu	2.2	1.5	2.8	1.4	1.3	1.1
^{241}Am	–	–	3.0	4.2	3.7	2.8
^{243}Am	3.7	2.0	2.5	2.8	2.6	2.8
^{242}Cm	–	–	8.3	9.5	9.5	8.7
^{244}Cm	10.2	9.0	8.5	13.9	8.5	8.9
^{145}Nd	1.0	0.8	0.9	0.7	0.8	0.7
^{146}Nd	1.1	1.0	0.8	0.6	0.8	0.8

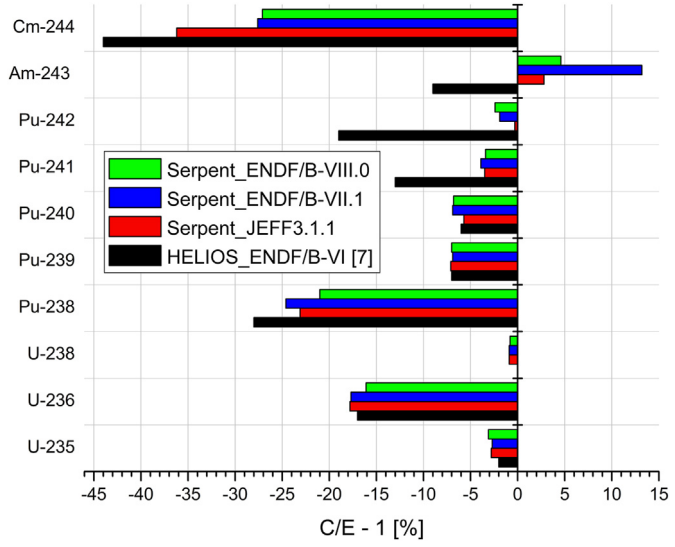

Fig. 3. C/Es for the actinides concentrations computed with Serpent with different NDLs and compared with HELIOS results [7] for the Kalinin-1/Sample-33.

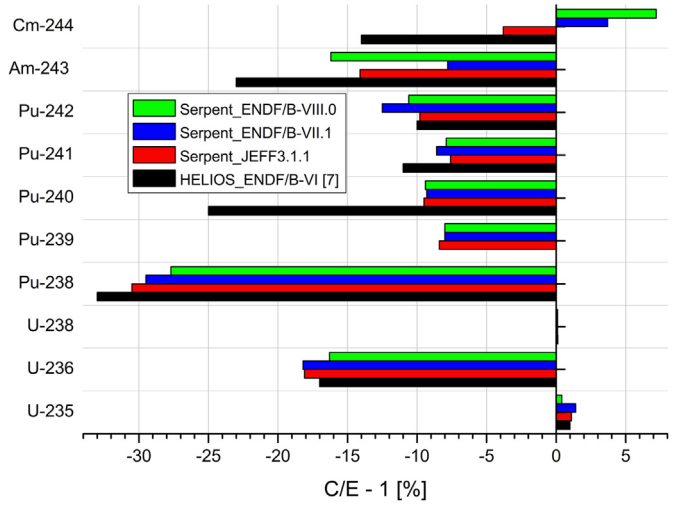

Fig. 4. C/Es for the actinides concentrations computed with Serpent with different NDLs and compared with HELIOS results [7] for the Kalinin-1/Sample-448.

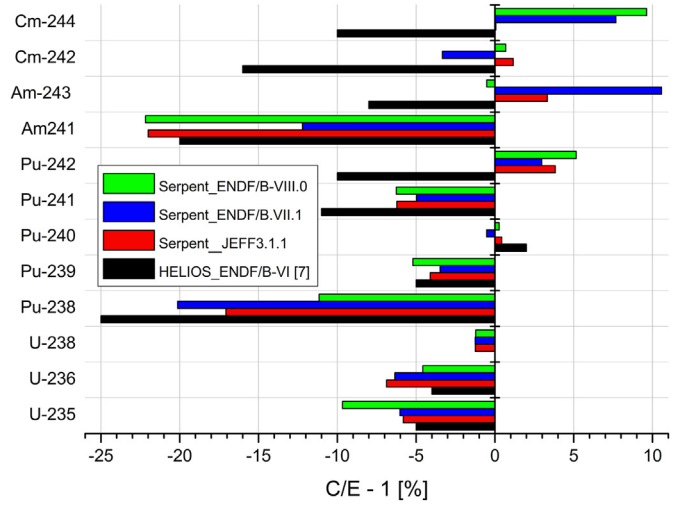

Fig. 5. C/Es for the actinides concentrations computed with Serpent with different NDLs and compared with HELIOS results [7] for the Balakovo-2/Sample-6.

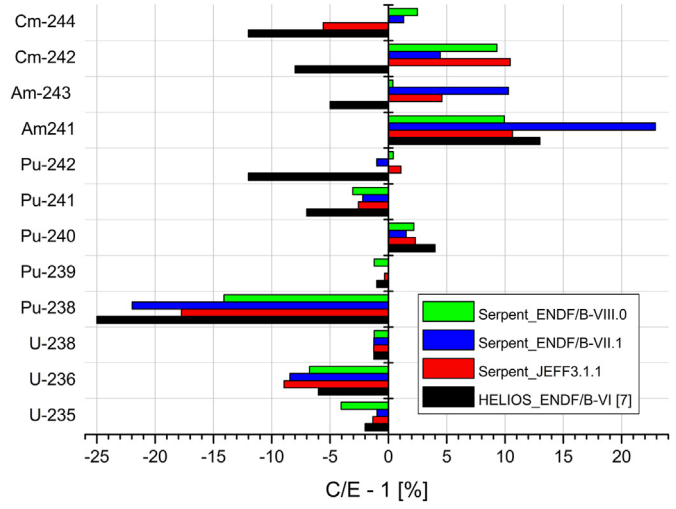

Fig. 6. C/Es for the actinides concentrations computed with Serpent with different NDLs and compared with HELIOS results [7] for the Balakovo-2/Sample-15.

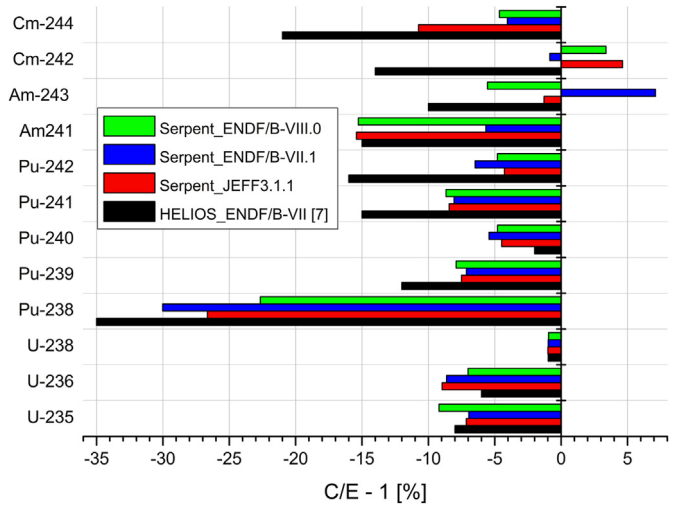

Fig. 7. C/Es for the actinides concentrations computed with Serpent with different NDLs and compared with HELIOS results [7] for the Balakovo-3/Sample-912.

account for the so called "neighboring effect". However, this option was out of the scope of this study.

The results for ^{235}U are under-predicted on average within 3% and 8% for the Kalinin-1 and Balakovo-2 cases respectively while for the Balakovo-3 samples an opposite behavior of the C/E's has been found as already mentioned above. Apart from the contribution of the nuclear data uncertainties, the reasons for the discrepancies on the ^{235}U concentrations are mostly attributed to the uncertainties on fuel enrichment (typically in the order of 0.05 wt% [1]) and on the burnup. In fact, the ^{235}U concentration is very sensitive to the burnup value, especially for medium/high burnup. In particular, for our study it is worth to mention that the experimental uncertainty associated to the ^{145}Nd and ^{146}Nd measurements which were used to tune our simulations on the actual burnup is in the order of 0.6–1% depending on the samples [6]. In Ref. [1] it has been shown that for a burnup of 48 MWd/kgHM, an uncertainty of 0.05 wt% on fuel enrichment and 1% in burnup can originate deviations of ~5% in the ^{235}U concentrations. Also the ^{236}U concentrations, which are mostly originated from the ^{235}U (n,γ), are systematically under-predicted particularly at low burnup. A generally good prediction of the concentrations for the plutonium isotopes can be observed, except for the case of ^{238}Pu. This isotope is consistently and quite strongly underestimated for all samples, particularly in the case of Kalinin-1 at low burnup. The strong underestimation of ^{238}Pu has already been reported in several studies as for example in Ref. [19]. Especially at low burnup, 25% of the ^{238}Pu formation is originated from the ^{238}U (n,2n) therefore one of the possible origin for the low ^{238}Pu C/E's could be the underestimation in the NDLs of this cross-section. A lot of cross-sections are involved in the ^{238}Pu production in UOx fuels and in order to identify the main channel contributing to this underestimation a proper sensitivity analysis should be performed. However this goes outside the scope of this work. The ^{239}Pu buildup, very important for SNF criticality safety analysis is well predicted with all the C/E's within the experimental uncertainty. The ^{240}Pu concentrations are also well reproduced, particularly for the Balakovo-2 samples, confirming the correct modeling of the negative reactivity worth of this poisoning isotope. As far as the americium isotopes, one can systematically observe that the C/E's are more accurate for ^{243}Am than for ^{241}Am. The ^{242}Cm concentrations are remarkably well predicted in all cases within the experimental uncertainty. Also the ^{244}Cm concentrations can be considered well predicted due the high experimental error associated to their measurements.

When comparing the performances of the different NDLs one can observe a general very good agreement. In particular, the results obtained with JEFF3.1.1 and ENDF/B-VII.1 are very close to each other for most of the isotopes with the exceptions of ^{243}Am and ^{244}Cm for which the ENDF/B-VII.1 systematically predicts higher values by ~7–9% and ~8–11% respectively and of ^{242}Cm for which the ENDF/B-VII.1 values are lower by ~4–7%. When comparing the two ENDF/B libraries a slightly increased accuracy in the C/E's for some of the isotopes computed with ENDF/B-VIII.0 can be noticed. This release contains reaction libraries for 557 isotopes and thermal scattering libraries for 34 materials and represents a significant improvement with respect to its predecessor ENDF/B-VII.1 [13]. The slightly increased accuracy observed in our study with the ENDF/B-VIII.0 data can be systematically noticed for all plutonium isotopes. In particular, the C/E's on the ^{238}Pu concentrations increase by ~5–12% depending on the samples. As far as the reactivity values, one can clearly observe a systematic lower prediction when using ENDF/B-VIII.0 data compared to ENDF/B-VII.1 data. These discrepancies tend to increase during the irradiation time, as highlighted in

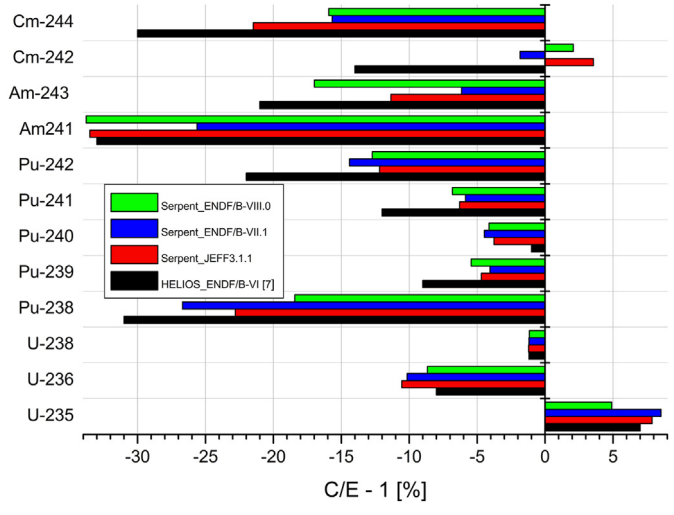

Fig. 8. C/Es for the actinides concentrations computed with Serpent with different NDLs and compared with HELIOS results [7] for the Balakovo-3/Sample-581.

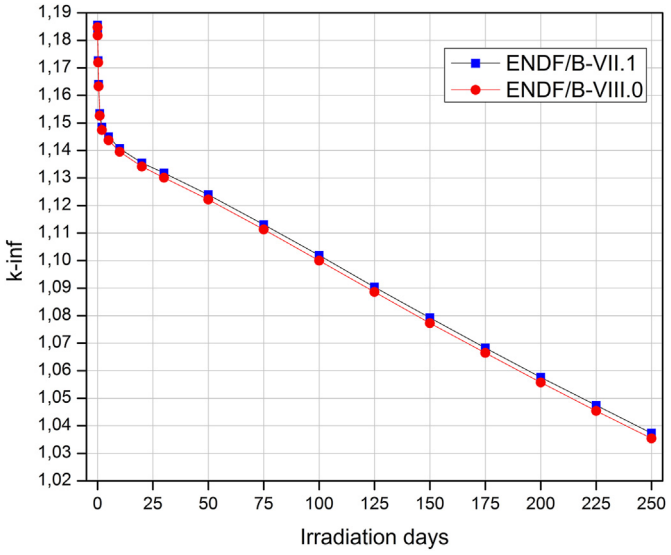

Fig. 9. k-inf vs. burnup for the Kalinin-1/Sample-33.

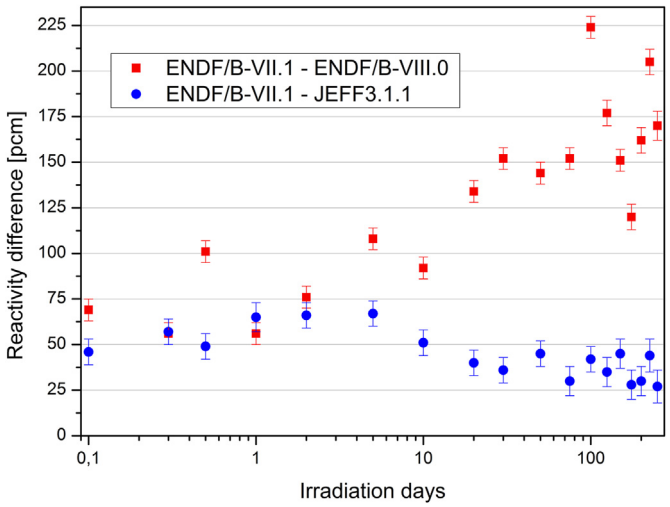

Fig. 10. Differences in reactivity calculated by different NDLs for the Kalinin-1/Sample 33.

Fig. 9 which shows the k-infinity variation vs. burnup for the Kalinin-1/Sample 33. In this case the reactivity differences at BOC and EOC are 56 ± 5 pcm and 179 ± 8 pcm respectively. The RMS difference over burnup was found to be 127 pcm (see Fig. 10). The same behavior in terms of reactivity differences when comparing the two ENDF/B libraries was found for all the experimental samples. These results are consistent with recent data reported in Refs. [20,21]. One of the main reason for such reactivity difference is attributed to the increased ^{16}O (n,α) reaction evaluation in ENDF/B-VIII.0 and to its contribution in absorbing neutrons and decrease the reactivity [22]. Reactivity values predicted with ENDF/B-VII.1 data are also slightly higher than the corresponding ones predicted with JEFF3.1.1. In this case the differences are nearly constant during the burnup steps. The RMS reactivity difference over burnup was found to be 47 pcm (see Fig. 10).

Despite the additional uncertainties due to modeling parameters, most of the C/E's obtained in this study are within the margins of the uncertainties due to the nuclear data as reported in literature for similar cases of burnup values, fuel enrichment and neutron spectra [15,23–26]. Moreover, for a few isotopes the discrepancies between the computed and the measured concentrations are within the corresponding experimental errors as for example in the case of the ^{242}Pu, ^{242}Cm, ^{244}Cm for Balakovo-2 and ^{242}Cm for Balakovo-3. Apart from the possible compensation effects that can certainly play a role for such remarkable agreements, this can also be considered as a positive indication on the global good quality of the simulations performed and of the adopted modeling assumptions.

5. Summary and conclusions

The SFCOMPO-2.0 database is of high interest for depletion calculation validation and benchmarking and in this work some of the few available post irradiation data related to VVER-1000 reactors have been investigated. The goal of the study was to investigate the capability of the Serpent Monte Carlo code to predict LEU SNF isotopic compositions at different burnup levels covering the range from 13 to 47 MWd/kgU. The irradiation of six UO_2 experimental samples with enrichment varying from 3.6 wt% to 4.4 wt% in three different VVER-1000 reactor units has been simulated and the predicted concentrations of actinides up to ^{244}Cm have been compared with the corresponding measured values at discharge. For most of the actinides the results showed a relatively good agreement between calculated and experimental concentrations, often within the range of the nuclear data uncertainties and in a few cases even within the reported experimental uncertainties. The performances of different NDLs have also been compared and the newly released ENDF/B-VIII.0 has shown an increased accuracy in the prediction of the C/E's for some of the actinides, particularly for the plutonium isotopes. Future studies will deal with detailed sensitivity analyses to evaluate the possible correlations existing between the computed C/E's and also to quantify the impact of the modelling assumptions on the output parameters to better understand the modelling complexity required to achieve a desired level of accuracy. In any case, this work represents a step forward towards the validation of advanced simulation tools against experimental data. It provides an evidence of the capabilities of the Serpent Monte-Carlo code with the associated modern NDLs to compute SNF nuclide inventory concentrations for VVER-1000 reactors with accurate precision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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