ISOTOPIC PREDICTION SIMULATIONS APPLIED TO HIGH BURNUPS SAMPLES IRRADIATED IN VANDELLOS-II REACTOR CORE

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ABSTRACT

Isotopic content assessment has a paramount importance for safety and storage reasons. During the latest years, a great variety of codes have been developed to perform transport and decay calculations, but only those that couple both in an iterative manner achieve an accurate prediction of the final isotopic content of irradiated fuels. Needless to say, them all are supposed to pass the test of the comparison of their predictions against the corresponding experimental measures.

SCALE6.0 is one of the most well-known and largely used code packages in such calculations through its module TRITON and it has become a reference for new codes designed with the same purpose, what give us confidence in applying it to our case of study: Spanish Vandellos-II pressurized water reactor. For the present paper, a specific fuel rod isotopic content evolution throughout Vandellos-II cycles 7-11 is reproduced for several increasing burnups and compared to experimental measures. First of all, our model considers all the available information concerning the position of the rod within its host fuel assembly as well as these assembly surroundings. These results are then compared to those obtained from a model for which the surroundings have not been taken into account. This way it is possible to elucidate what level of description is needed in order to reproduce isotopic content accurately and shed light on the real importance of the models precision, the involved magnitudes knowledge and, over all, the impact on isotopic calculations of position and surroundings of a specific fuel rod.

An improved version of MONTEBURNS2.0 and LINK, a new code designed to coordinate MONTEBURNS2.0 executions and update materials from one case to another, were developed at the Department of Nuclear Engineering. Both of them are here introduced and used to deal with the same problem in order to validate the options and capabilities implemented in the pursuit of new calculation methodologies.

Key Words: burnup credit, isotopic prediction, SCALE 6.0, MONTEBURNS 2.0, Vandellós-II

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1   INTRODUCTION

An issue of utmost importance in the nuclear power development is the management of the radioactive products and waste due to safety, and security, questions. An accurate control over the spent nuclear fuel content is essential for its safe and optimized transportation, storage and management. The existing transport and storage facilities are designed following a conservative philosophy in order to guarantee the fulfilment of the safety margins, that is, assuming that the fuel they store has not been irradiated, that are fresh. However, taking into account burnup credit, it is possible to design alternative transport and storage arrays more compact, economical and capable of storing a higher number of spent assemblies without renouncing safety. Because of this, it is absolutely important to determine accurately the reactivity of spent fuel and its isotopic content.

Nowadays, to predict isotopic evolution throughout irradiation and decay periods is not a problem thanks to the development of powerful codes and computational methodologies. The most realistic choice to handle this problem is to couple neutron transport and depletion calculations in an iterative manner because, this way, it is possible to reproduce the effect of the isotopic change on the neutronics of the system throughout the entire irradiation history. To perform this kind of coupled calculation it is necessary to write coupling codes to automate the information flow and perform the manipulations that it may require. SCALE 6.0/TRITON [1] module and MONTEBURNS 2.0 [2] are examples of codes that follow a coupling methodology.

To predict the isotopic content measured for samples burnt up to different and increasing burnups in Vandellós-II Spanish pressurized water reactor, both of them are used. The changing position of the sample of interest can be chosen as the main characteristic of the problem, which demands to follow isotopic content trough changing geometries and irradiation histories. An external module, LINK, is designed to automate the procedure and manage all the information related to the isotopic content from one cycle to the following one, allowing a flexible and reliable tracking of the composition of interest. For the present study, MONTEBURNS 2.0 is updated with new capabilities focused on an improved reproduction of the physical conditions under which the sample was irradiated, especially on the power at which the experimental measures suggest it was burnt. We study the effect of our improvements by analyzing the simulation results for several burnups and apply them to a study involving the importance of modelling the surroundings to achieve acceptable predicted values.

2   PROBLEM DESCRIPTION

We focus our study on the Spanish PWR Vandellós-II operation time between June 1994 and September 2000, corresponding to its cycles 7th to 11th. Enriched up to 4.5 wt% ^{235}U, fresh fuel rods were placed at the beginning of the 7th cycle in different locations within the reactor core. The assemblies that hosted them changed within the reactor core from one cycle to the following and, finally, for the 11th cycle rods of interest became part of an assembly placed near the center. After their extraction and cooling, the isotopic content of these test rods was measured by different techniques in two campaigns that took place at the Studsvik laboratories in 2003 and 2006 [3].

2.1 Samples Specifications

Rod identified as WZR0058 started its irradiation history with an enrichment of 4.5 wt% ^{235}U. Its position at the outmost part of the host assembly remained unchanged from the 7th to the 10th cycle, while the assembly itself moved from the periphery of the core (7th and 8th cycles) to the center surroundings (9th cycle) and from there to the periphery again, facing this time the water reflector (10th cycle). Finally, the rod of interest was removed from the original assembly and inserted in a new one right in the center of the reactor core (11th cycle). As a result of all these displacements, rod WZR0058 burnup was influenced by different neighboring conditions that, together with the irradiation and decay history and other physical parameters implied, like boron letdown curves, densities and temperatures, determined its final average burnup.

Several samples were extracted from different axial positions of WZR0058. The measures led to the obtaining of four sets of nuclides concentrations, corresponding to the four increasing burnups that the selected axial positions reached according to the axial power profile. Table I summarizes these burnups as estimated by ^{148}Nd and ^{137}Cs measures [4].

Table I. Estimated burnups based on burnup indicators measures

Sample ID	Sample burnup (GWd/MTU)
E58-88	42.489
E58-148	54.820
E58-260	64.624
E58-700	77.013

2.2 Geometrical Levels of Modelization

It is possible to reproduce all these compositions at the end of the irradiation history by modeling the problem with well established simulation codes like those described in the following section. Most of the criticality and depletion codes allow the user to define the problem geometry as detailed as necessary. To assess how the modeling of the rod position impacts on isotopic prediction calculations, the same problem has been approached from two increasing levels of detail. The first one takes into account the position of WZR0058 within the host assembly but it does not include any information about the adjacent assemblies. The second one includes in its definition the presence of the neighbors affecting directly the rod WZR0058. Figure 1 represents the two geometrical levels as well as the position of rod WZR0058.

Since there is no difference between the geometrical definitions at each level of the four samples due to the axial symmetry of the problem, the same geometrical input is used to simulate their burnups. However, needless to say, it has been necessary to vary the temperature, coolant density and power level at which each sample burnt, according to the experimental profiles.
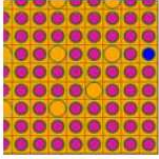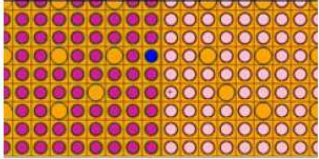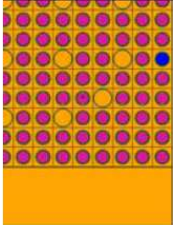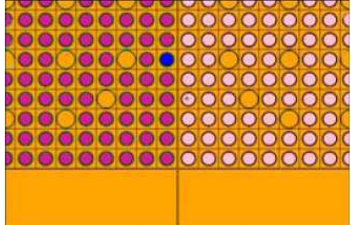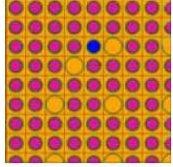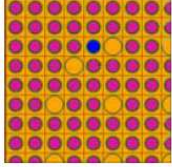
Cycles	Level 1	Level 2
7th to 9th		
10th		
11th		

Figure 1. Modeled levels of geometrical detail

3 USED CODES

SCALE 6.0, developed at the Oak Ridge National Laboratory, has become in recent years a reference code for no few calculations involving nuclear reactor physics, such as criticality and depletion calculations. One of the modules included in the SCALE 6.0 code package is TRITON, which makes easy to study the isotopic evolution of fuel under irradiation through the coupling between 2-D deterministic transport code NEWT and the depletion and decay code ORIGEN-S; the former solves the transport problem and provides the latter with cross-sections and averaged neutron fluxes that it uses in the subsequent depletion calculation, the result of which updates the isotopic content and material composition for the next NEWT calculation. This iterative way to perform the fuel depletion guarantees a realistic monitoring of the isotopic evolution. Another two interesting capabilities of TRITON for the present problem are:

- The power normalization: user can specify the power at which a material burnt and normalize the rest of the materials fluxes according to this requirement. This makes possible to reproduce experimental burnups.

- The isotopic files: once finished the calculation, TRITON prints the isotopic content of a material selected by the user and it is possible to use it as material composition input for another case or geometry. Thanks to this capability, following the isotopic content of the samples throughout the entire irradiation history taking into account their changing surroundings does not represent a problem.

MONTEBURNS 2.0, developed at Los Alamos National Laboratory, is a coupling code written to automate the process of providing ORIGEN2 with one-group microscopic cross-sections and neutron fluxes from MCNP transport calculations, and providing MCNP with updated material compositions from ORIGEN2 decay calculations. Similarly to TRITON, the calculation strategy followed is iterative throughout the duration of the modeled burnup history. The approximation assumed by MONTEBURNS 2.0 is the middle-of-step constant flux approximation. MONTEBURNS 2.0 broadens its coupling capability by offering the possibility of using CINDER90 as depletion module, and also improves the feed capability by increasing the number of group materials to feed or remove. MCNP geometrical versatility allows us to model the problem introduced above and MONTEBURNS 2.0, to reproduce the physical conditions at which the samples burnt. Nevertheless, when compared to SCALE 6.0, we find two issues to think about:

- MONTEBURNS 2.0 accepts a power value referred to the entire modeled system. It is not possible to burn the sample of interest at a power value without distorting the power of the rest of the assembly.

- Once defined the MCNP geometry, it is maintained till the end of the calculation. This makes mandatory to write several inputs if the simulation of the position and geometrical changes is desired.

These two questions aimed to develop some new tools for MONTEBURNS 2.0 and an external code, LINK, to coordinates the consecutive executions without lack of information.

3.1 Capabilities implemented in MONTEBURNS 2.0

Some calculation procedures and options needed for this problem are not included in the current version of MONTEBURNS 2.0.

3.1.1   Corrections in MONTEBURNS 2.0 feed option and temperature distribution function

Thanks to the MONTEBURNS 2.0 feed option, the varying levels of soluble boron are reproducible by adding or removing fractions of boron in water in a discrete and a continuous way. However, MONTEBURNS 2.0 fails when a continuous removal is selected: no modification of the boron content is carried out. Our version of MONTEBURNS 2.0 fixes this problem and performs executions reproducing correctly the boron letdown curves, designed with both removal options.

In MONTEBURNS 2.0 all the materials evolve at the temperature at which the selected library was generated and it is not possible to draw any temperature distribution, what is another problem that our version fixes. In our case, two continuous-energy ACE format data libraries generated using NJOY-99.259 with 0.01 fractional reconstruction tolerance were used, on the one hand, based on ENDF/B-VII evaluation [6]. The prepared libraries include a total of 432 nuclides at 6 temperatures, but given the temperature conditions of our problem, only isotopes at 600K and 900K, for moderator and fuel respectively, were necessary.

3.1.2 Power normalization methodology

In MONTEBURNS 2.0, for each material, MCNP calculates fluxes normalized to one fission source- neutron; to convert into neutrons per second and cm^2, MONTEBURNS 2.0 multiplies by the constant factor recommended in the MCNP manual [5]

$$C = \frac{\upsilon}{k_{eff}} \frac{P}{Q_{ave}} \frac{10^6 \, W/MW}{1.602 \cdot 10^{-13} \, J/MeV} \tag{1}$$

where P (MW) is the total power of the entire system modelled in MCNP and entered as input in MONTEBURNS, υ is the average number of fission neutrons per fission event, Q_{ave} is the average recoverable energy per fission event (J/fission) and k_{eff}, the eigenvalue of the system. All the materials are, then, equally normalized and MONTEBURNS assigns to each one a power value depending on the volume they occupy, their macroscopic fission cross-section and their neutron flux level. This treatment cannot guarantee the depletion of a pin-cell at the experimental power suggested by burnup indicators when it is modelled as part of a fuel assembly, like in levels 1 and 2 showed in Figure 1. The factor

$$C^* = \frac{P_{norm}}{\sum\limits_{j=n}^{m} \left\{ \Sigma_f^j \varphi_j V_j Q_j \right\}} \frac{10^6 \, W/MW}{1.602 \cdot 10^{-13} \, J/MeV} \tag{2}$$

where

P_{norm}   corresponding total power of all the materials selected as basis of normalization

$j \in [n,m]$ one of the materials, j, part of the basis normalization, made of all the materials from n to m

Σ_f^j   fission macroscopic cross-section for material j

φ_j   unnormalized neutron flux in material j tallied by MCNP

V_j   volume occupied by material j

Q_j   average recoverable energy per fission event in material j

allows the user to select one or more materials as basis of normalization, to introduce in MONTEBURNS the corresponding power and normalize the rest of the materials to the selected fluxes levels. Thus, each material, *i*, is depleted at a power given by

$$P^i = \frac{P_{norm}}{\sum\limits_{j=n}^{m} \left\{ \Sigma_f^j \varphi_j V_j Q_j \right\}} Q^i \Sigma_f^i V^i \varphi^i \tag{3}$$

what makes possible to burn the material of interest at the desired power value.

3.1.3 LINK: isotopic management for transport and decay calculations

As explained before, reproducing the geometries of each cycle obliges us to model them separately and, then, to execute them with MONTEBURNS 2.0 one by one. That means to take the resulting compositions at the end of each cycle and to update the initial compositions of the following one by hand. This process implies a lot of time and the possibility of making mistakes. Our external code, LINK:

- automates a serial of executions
- carries out automatically the material managing between the inputs to execute, that is, takes final compositions and writes them as part of the initial composition of the desired geometrical models.
- manages the entire inventory: the isotopes considered for the MCNP transport calculations and the isotopes decayed by ORIGEN. LINK does not allow any lack of information in the updating process.
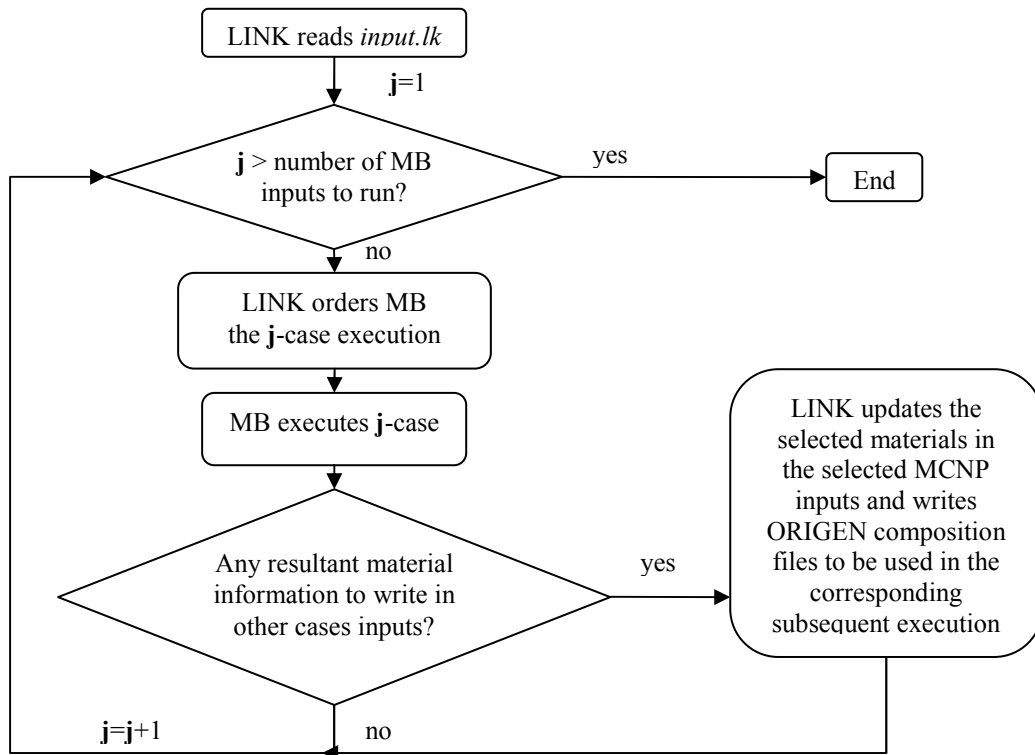
Figure 2 summarizes the process.

Figure 2. LINK conceptual flow chart

4 RESULTS

To validate our methodologies, cycles 7-11 modelization series were executed with TRITON, our uptdated version of MONTEBURNS 2.0. TRITON calculations were performed with the SCALE 44-group cross-section library based on ENDF/B-V data and following the two-dimensional depletion sequence, which calls NEWT as transport code, ORIGEN-S as depletion code and NITAWL as cross-section processor. MONTEBURNS 2.0 included the library-at-temperature selection option and was executed by LINK, emulating, then, TRITON execution flow. For MONTEBURNS 2.0 executions, processed libraries at 600 K and 900K based on ENDF/BVII were chosen. PWRU ORIGEN library was used.

The Vandellós-II problem served us to probe, firstly, the impact of each of the capabilities presented above on the isotopic content calculation; secondly, how much the prediction is affected by them with burnup; and, finally, the importance of the surroundings on this isotopic prediction calculation. We devote the next three sections to describe the executions that were performed, the results we obtained and the conclusions they suggest.

4.1 Effect of the implemented capabilities on isotopic prediction

The most important features of our system LINK / [MONTEBURNS 2.0]* are the selection of mixtures as basis of power normalization and the automatic management of isotopic contents for MCNP inputs and ORIGEN 2.1 inputs. To analyze the impact of these two improvements, several executions have been carried out for the sample that reached the lowest burnup, that is, E58-88, burnt up to 42.5 GWd/MTU. Specifically, the results are referred to:

- an execution with the normalization methodology and ORIGEN 2.1 isotopic management deactivated: $_a[42.5]$

- an execution with ONLY the isotopic management activated: $_a[42.5]^i$

- an execution with ONLY the normalization capability, with the sample of interest selected as basis of normalization, activated: $_a[42.5]^n$

- an execution with BOTH capabilities activated: $_a[42.5]^{n,i}$

An execution of the problem with SCALE 6.0, as explained before, was performed in order to have an external reference for our results. The sub index "a" stands for "adjacent" and it means the geometries considered correspond to detail level 2 in Figure 1.

From an inspection of Table II, that presents the deviation between the calculated and experimental [Grams of isotope]/[Grams of U-238] % values, it is possible to define three categories of isotopes. The first one would be the family the prediction of those is improved by the activation of the normalization methodology. Examples of this family are U-234, U-235, Pu-239, Pu-242, Am-241, neodymium, (but Nd-142), Cs-133, Ce-140, Ce-142 and Sm-151. Since the sample burns under a more realistic flux level, it is reasonable the improvement in U-235 and Pu-239 prediction, and the corresponding effect on some fission products. The second family would be those isotopes for which the prediction improves because of the activation of the ORIGEN 2.1 isotopic management from one cycle to the following; few of them have been found: Pu-239, Am-243, Cm-244, Nd-142, Nd-143, Sm-150, Sm-151, Eu-154, Eu-155, Gd-156 and La-139. Finally, there are isotopes for which the prediction improves when both capabilities are activated: U-235, Pu-238, Pu-241, Pu-242, neodymium, cesium, cerium (but Ce-144) and europium (but Eu-153). Rare are the isotopes for which none of the capabilities improve the prediction separately but they do when are applied at the same time: Pu-238, Pu-241. However, it can be seen that most of them improve mainly because of the application of the normalization method, so we conclude that this capability is the main responsible of the obtained accuracy.

Table II. Comparison (C/E-1)*100% for sample E58-88 at 1101 days from discharge with different capabilities activated

Isos.id	$_a[42.5]^{n,i}$	$_a[42,5]^i$	$_a[42.5]^n$	$_a[42.5]$	Scale6.0
U-234	8.04	8.04	5.16	7.32	4.09
U-235	0.58	3.64	0.85	1.64	-4.03
U-236	7.39	6.46	7.39	6.77	8.94
Pu-238	-8.10	-11.85	-9.97	-10.60	0.60
Pu-239	-3.50	-2.13	-3.50	-5.13	1.39
Pu-240	2.72	4.85	5.55	0.59	8.56
Pu-241	-1.92	-6.50	-4.97	-3.57	-0.45
Pu-242	0.64	-3.34	-0.89	-3.03	9.09
Np-237	-7.37	-11.87	-7.90	-6.58	-1.54
Am-241	21.50	20.14	18.78	21.50	24.29
Am-243	36.24	22.09	36.61	30.47	51.41
Cm-244	76.86	62.05	87.05	76.86	107.97
Cm-246	46.36	30.09	22.44	11.92	-9.65
Nd-142	28.49	22.37	-66.50	-67.19	25.64
Nd-143	-3.10	-3.87	-3.87	-4.64	-0.67
Nd-145	-2.81	-3.57	-3.00	-3.38	1.42
Nd-146	-0.12	-2.52	-0.49	-1.97	2.61
Nd-148	-1.30	-3.05	-1.30	-2.35	0.60
Nd-150	-3.38	-5.56	-3.38	-4.83	-0.10
Cs-133	-1.47	-2.51	-1.36	-2.51	3.33
Cs-134	-20.98	-26.48	-23.01	-23.88	-72.72
Cs-135	-14.31	-15.90	-14.71	-16.49	-10.62
Cs-137	-9.72	-11.38	-9.72	-10.94	-11.39
Ce-140	-2.71	-4.27	-3.34	-4.38	2.09
Ce-142	-2.44	-4.02	-2.44	-3.46	0.19
Ce-144	14.38	7.30	14.38	7.68	-91.81
Sm-147	1.55	2.17	0.92	-0.32	-1.87
Sm-148	-1.94	-5.44	-1.94	-1.94	4.05
Sm-149	-5.71	-6.21	-6.70	-5.22	0.36
Sm-150	6.01	3.48	6.01	4.49	5.57
Sm-151	-1.55	-0.63	-1.90	-4.32	24.47
Sm-152	5.38	3.08	4.17	1.33	26.91
Sm-154	7.84	5.41	7.84	5.81	6.68
Eu-153	-6.89	-8.04	-7.20	-6.79	-6.86
Eu-154	5.42	3.18	6.17	6.17	6.03
Eu-155	7.90	6.75	9.04	9.33	-24.57
Gd-154	42.67	42.27	44.06	44.46	41.84
Gd-155	21.05	19.87	22.23	22.82	-8.99
Gd-156	6.60	1.43	6.78	3.09	9.34
Gd-158	33.19	29.02	21.74	19.66	27.76
Gd-160	11.25	8.20	11.38	8.71	-6.23
Ru-106	-22.92	-27.83	-23.20	-28.11	-89.37
La-139	2.81	1.16	2.81	1.71	5.88
Tc-99	2.35	0.81	2.01	0.64	7.09

4.2 Effect of the implemented capabilities on isotopic prediction at increasing burnups

Since Vandellós-II program is focused on high burnup spent fuel, we apply our methodology to the isotopic prediction calculation in samples of higher burnup, those listed in Table I. The samples, extracted from different axial positions, were modeled and executed following the same philosophy than in the previous section: for each one, an execution with a complete selection of our capabilities and another one with none of them selected were performed. A comparison between the results of these calculations is shown in Table III.

As expected, an important improvement in the prediction of U-235 has been achieved. The reduction in the deviation from measured values is clear in the lowest burnup case but it is especially meaningful for samples E58-260 and E58-700, burnt up to 64.7 GWd/MTU and 77 GWd/MTU respectively. However, U-235 content in E58-148 is worse predicted when activating the introduced capabilities. Pu-239, though, is better calculated in the whole range of burnups, even for E58-148. In fact, it is noticeable at a first sight that nuclear fuel and major actinides are the most affected by the application of our methodology, leading in general to better results, but in sample E58-148, the number of actinides improved is lower and there is an inverse estimation of the uranium and some plutonium isotopes in comparison with the rest of the calculations, that is: uranium is overestimated in all cases and underestimated in E58-148.

Regarding to fission products, neodymium, cesium and cerium are generally more accurately estimated, as well as other isotopes at different burnups from which it is not possible to infer some kind of rule of thumb.

It is important to note that our capabilities affect negatively to the prediction of americium and curium. Nevertheless, the results are poor without them, what suggests a wrong treatment of the reactions implied in their formation and decay by the original code or a poor quality in the nuclear data library used for them.

Table III. Comparison (C/E-1)*100% for WZR0058 samples at 1101 days from discharge with and without all the capabilities

Isos.id	E58-88		E58-148		E58-260		E58-700	
	$_a[42.5]^{n,i}$	$_a[42.5]$	$_a[54.8]^{n,i}$	$_a[54.8]$	$_a[64.7]^{n,i}$	$_a[64.7]$	$_a[77]^{n,i}$	$_a[77]$
U-234	8.04	7.32	-5.85	-3.70	25.41	30.54	19.22	25.09
U-235	0.58	1.64	-10.08	-3.65	5.88	24.33	0.80	13.56
U-236	7.39	6.77	-1.02	-2.50	3.40	1.71	9.49	6.34
Pu-238	-8.10	-10.60	9.13	2.27	-10.47	-19.29	-11.95	-15.74
Pu-239	-3.50	-5.13	0.05	0.71	-0.71	-1.06	-3.08	-3.70
Pu-240	2.72	0.59	3.29	4.32	2.40	-4.84	1.75	3.82
Pu-241	-1.92	-3.57	7.58	2.38	-0.52	-3.77	0.30	-6.43
Pu-242	0.64	-3.03	12.18	3.25	-0.05	-8.78	-3.56	-6.22
Np-237	-7.37	-6.58	-18.86	-18.82	-9.26	-13.02	-8.29	-3.43
Am-241	21.50	21.50	29.82	30.21	18.26	21.10	21.47	18.26
Am-243	36.24	30.47	13.77	4.10	17.50	-1.71	27.11	4.07
Cm-244	76.86	76.86	72.56	65.66	59.63	26.16	86.79	46.51
Cm-246	46.36	11.92	141.64	71.80	68.10	-15.88	85.04	27.35
Nd-142	28.49	-67.19	32.51	-66.70	20.45	-71.87	16.49	-27.41
Nd-143	-3.10	-4.64	-0.25	-2.13	-0.81	-3.44	-0.93	-3.64
Nd-145	-2.81	-3.38	-0.95	-3.37	-3.16	-8.12	-4.36	-9.05
Nd-146	-0.12	-1.97	1.98	-2.30	0.45	-7.15	-0.25	-5.26
Nd-148	-1.30	-2.35	7.09	3.95	0.73	-5.50	-2.83	-7.56
Nd-150	-3.38	-4.83	5.05	1.29	-2.70	-9.45	4.15	-1.07
Cs-133	-1.47	-2.51	1.89	-1.23	-7.25	-12.32	4.63	1.01
Cs-134	-20.98	-23.88	-9.33	-17.55	-9.97	-24.22	-2.60	-15.25
Cs-135	-14.31	-16.49	-18.76	-21.50	-24.22	-28.30	-6.85	-14.65
Cs-137	-9.72	-10.94	-4.19	-7.09	-5.44	-11.86	-2.98	-8.56
Ce-140	-2.71	-4.38	1.21	-2.34	-4.28	-10.71	-2.05	-7.08
Ce-142	-2.44	-3.46	-2.20	-5.03	-3.22	-8.80	-3.21	-7.39
Ce-144	14.38	7.68	20.65	6.58	18.23	0.46	21.94	5.18
Sm-147	1.55	-0.32	5.27	4.48	12.78	13.01	10.64	8.34
Sm-148	-1.94	-1.94	2.10	-5.83	5.99	-5.19	6.75	0.59
Sm-149	-5.71	-5.22	-3.46	-12.23	-1.15	-4.31	9.48	-3.61
Sm-150	6.01	4.49	3.91	1.84	5.93	-0.33	3.45	-1.02
Sm-151	-1.55	-4.32	4.09	5.18	3.82	3.01	-2.40	-9.02
Sm-152	5.38	1.33	6.18	-1.87	0.05	2.96	0.72	-4.38
Sm-154	7.84	5.81	19.71	14.74	25.37	13.92	15.59	7.42
Eu-153	-6.89	-6.79	-6.56	-8.32	-8.62	-15.55	-7.47	-8.98
Eu-154	5.42	6.17	7.97	7.78	15.31	3.14	33.53	28.62
Eu-155	7.90	9.33	-0.43	-1.52	10.22	-4.15	17.85	12.81
Gd-154	42.67	44.46	21.20	25.64	49.48	39.91	30.64	28.52
Gd-155	21.05	22.82	8.11	7.18	27.78	11.90	14.82	10.32
Gd-156	6.60	3.09	2.60	-3.17	4.04	-12.89	-1.20	-11.56
Gd-158	33.19	19.66	34.82	11.46	44.52	3.93	46.66	2.47
Gd-160	11.25	8.71	29.45	23.39	31.56	18.36	31.35	21.32
Ru-106	-22.92	-28.11	-12.20	-22.17	-11.51	-26.97	-5.42	-19.48
La-139	2.81	1.71	-	-	9.33	2.66	4.23	-0.61
Tc-99	2.35	0.64	-	-	-	-	-	-

No experimental data available for those isotopes marked with (-)

4.3 Effect of the sample surroundings on isotopic prediction at increasing burnups

Tested the validity of our normalization method and isotopic management, LINK / [MONTEBURNS 2.0] system is used to assess the importance of the adjacent assemblies in this isotopic calculation exercise. Due to a lack of information related to the specific irradiation history of the neighboring assemblies, their burning cycles description is based on hypotheses that may make them general and inaccurate. It is, then, interesting to know what isotopes and how much are meaningfully influenced by the inclusion of the adjacent assemblies in the model. In order to identify them, we compare the results (g/cc) obtained from the models that include the neighbor assemblies against the results obtained from the models that only represent the host assembly. Table IV shows, for each burnup, the deviation of isotopic content calculated without adjacent assemblies from that calculated taking them into account. The quantities compared are the ones printed by LINK/ [MONTEBURNS 2.0] at the end of the last calculation step, that is, at 1101 days from discharge.

Generally, the agreement between both levels is good but for the 54.8 GWd/MTU case, what points out that something is mistaken in the model used: the model itself or the provided input data. The 42.5 GWd/MTU case provides differences mainly below 1% of deviation. Note that deviations increase slightly with burnup, so a larger number of isotopes is above this percentage value. Nevertheless, deviations do not show a clear trend as burnup increases isotope by isotope. We mean, U-235 deviation increases with burnup, and Pu-239 deviation too, as it is foreseeable, but for other isotopes it is not possible to define clearly a general trend.

Reader can identify, though, some isotopes for which the neighbor presence seems to determine considerably the result of the prediction calculation, especially for the highest burnups. See, for example, U-234, Np-237, Am-243, Cm.-246, Eu-155 and Gd-155.

It is important to realize that for isotopes that are important for the criticality of the spent fuel, the exclusion of the neighbor assemblies leads to an overestimation of the predicted values, as higher as burnt the sample is. However, we emphasize the good agreement between both models.

Table IV. Comparison $([Burnup]^{n,i}/_a[Burnup]^{n,i} -1)*100\%$ for WZR0058 samples at 1101 days from discharge. Libraries based on ENDF/B-VII

Isotope	[42.5]	[54.8]	[64.65]	[77]
U-234	-2.67	6.56	-5.36	-3.92
U-235	0.26	15.12	0.65	1.14
U-236	-0.58	-2.11	0.77	-2.52
U-238	0.00	0.36	0.00	0.00
Pu-238	0.00	-10.20	-0.89	2.79
Pu-239	-0.85	-0.84	1.10	1.69
Pu-240	2.07	3.55	2.07	-1.92
Pu-241	0.52	-5.81	0.97	3.64
Pu-242	-0.61	-14.90	1.81	1.41
Np-237	0.57	-5.08	-3.44	6.18
Am-241	0.00	0.96	0.93	3.65
Am-243	-4.51	-14.10	-7.41	-2.33
Cm-244	-1.05	-18.26	-1.71	-0.85
Cm-246	1.31	-23.78	-12.44	-8.25
Nd-142	-1.19	-13.38	-0.24	-2.64
Nd-143	0.00	-1.57	0.41	0.53
Nd-145	0.39	-3.88	0.58	-0.26
Nd-146	-0.56	-6.67	-0.69	0.00
Nd-148	0.00	-5.51	0.00	0.00
Nd-150	0.00	-6.25	-0.47	0.39

Cs-133	0.12	-5.00	-0.85	0.00
Cs-134	-1.10	-13.59	0.65	-1.50
Cs-135	0.46	-1.06	0.81	-0.19
Cs-137	0.00	-5.81	0.00	0.00
Ce-140	0.00	-5.83	0.00	0.00
Ce-142	0.00	-4.55	0.00	0.00
Ce-144	0.00	-18.41	0.00	0.00
Sm-147	-1.23	0.00	-1.14	-2.30
Sm-148	1.79	-8.77	1.33	2.37
Sm-149	2.09	-11.27	-0.47	-0.43
Sm-150	0.48	-5.47	-0.62	0.27
Sm-151	-0.59	-0.89	4.64	1.92
Sm-152	1.16	-9.16	3.39	1.89
Sm-154	0.00	-7.69	-0.21	-0.16
Eu-151	0.00	0.90	4.72	2.39
Eu-153	-2.03	-1.71	0.00	0.64
Eu-154	1.42	-4.90	-1.57	-3.65
Eu-155	3.72	-6.79	-10.79	-1.55
Gd-154	0.84	-0.96	-1.52	-1.90
Gd-155	3.41	-6.12	-10.57	-1.34
Gd-156	-0.17	-12.73	-0.59	-0.39
Gd-158	0.00	-14.54	-1.45	-2.33
Gd-160	-0.11	-9.02	0.00	0.00
Ru-106	0.00	-19.20	0.00	0.47
La-139	0.00	-5.04	0.00	0.00
Tc-99	-0.33	-4.86	-0.12	-0.31

(Table IV continuation)

CONCLUSIONS AND FUTURE WORK

In order to reproduce the measured isotopic content of samples irradiated in a changing position within the reactor core up to increasing values of burnup, several capabilities have been developed. Firstly, and external module, LINK, to perform the executions and manage the complete information related to the isotopic content of the materials of interest. Without it, it is not possible to reproduce the changing surrounding geometry throughout the irradiation history. Secondly, a new version of MONTEBURNS 2.0 has been developed. Its main feature is the possibility of selecting a specific material as basis of normalization and, this way, reproducing the experimental burnup values more accurately. In this work, it has been proved the importance of them all and discovered the huge impact of the normalization method on the prediction of major actinides and more specifically of U-235 at high burnups. The relative importance of the adjacent assemblies in the model has been assessed. The results suggest the low importance of their presence except for certain isotopes, for which the trend should be considered if a right criticality calculation is desired. The 54.8 GWd/MTU case needs to be studied again and the provided irradiation data reviewed. Other geometrical configurations and burnups will be studied to continue with the characterization of the implemented capabilities.

AKNOWLEDGMENTS

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