31st International Conference Nuclear Energy for New Europe
NENE 2022 September 12-15 PORTOROŽ SLOVENIA

A Monte Carlo Fuel Assembly Model Validation Adopting Post Irradiation Experiment Dataset

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ABSTRACT

The 3D Monte Carlo code Serpent is currently being validated for Light Water Reactor's (LWR) fuel cycle simulations. This work chose the Takahama-3 Post Irradiation Experiment (PIE) dataset as a test case. Having key information related to the history of the plant, it was possible to compare the Serpent's results against more than 35 isotopic species' concentrations, measured following a destructive analysis of two fuel rods (SF95, SF97) at the end of their irradiation cycle. Nevertheless the presence of systematic sources of uncertainties related to the geometry, the results show a good agreement with the experimental data. Also, it is shown how the prediction capability may be increased up to +8% adopting a realistic temperature mesh for the fuel and the coolant.

1 INTRODUCTION

The ability to precise reproduce the dynamic evolution of the fuel is a fundamental task in the design and control of nuclear plants. In this framework, Monte Carlo (MC) routines represent an attractive method for facing this kind of operations due to the explicit geometry treating, the usage of continuous energy cross sections and the physical statements simplification given by the random neutron sampling.
The depletion capability of a neutronic code may take advantage from Spent Nuclear Fuel (SNF) data[1]; in the past 50 years, experimental campaigns have been put in place in order to acquire data relatively to isotopic compositions of irradiated fuels through the adoption of destructive radiochemical techniques. Up to now, they are considered as internationally established benchmarks.

This paper aims to present the validation of a fuel assembly model relying on the available Post Irradiation Experiment (PIE) dataset from the Takahama-3 facility[2]. In this work, up to

~ 230 concentrations, measured after different depletion periods, will be compared against the predictions from the Monte Carlo code Serpent[3]. The analysis will be conducted considering two figures of merit: first, the reliability of the single isotopic prediction will be quantified by the calculated-to-experimental (C/E) ratio. Also, each C/E evaluation will be accompanied by its uncertainty due to the Monte Carlo bias and by the experimental measurement technique. Then, the whole simulation will be characterized by the Success Rate (SR): considering the presence of systematic uncertainties, an acceptance threshold for the results is set. By so doing, the SR is defined as the ratio between the number of C/E points inside the acceptance band and the total number of comparisons.
The analysis will also test different features, seeking which is able to increase the SR and, therefore, the goodness of the prediction. In particular, it will be considered: two neutronic libraries (ENDF/VII.0 and JEFF-3.1.1), a different temporal mesh for the neutronic solver ($\Delta t_1 = 25$ [d], $\Delta t_2 = 50$ [d] and $\Delta t_3 = 100$ [d]) and two fuel temperature approaches ($T_{fuel} =$ const and $T_{fuel} = f(z,t)$).
This paper is organized as follows: Section [2] describes the Takahama-3 experiment, focusing on the irradiation history and the assembly's geometry; Section [3] will focus on the comparison analysis, listing both the uncertainty sources as well as the sensitivity cases; in Section [4] the results of this work will be compared against the literature references and Section [5] sums up the principal results and future improvements.

2 MODEL DESCRIPTION

The Takahama-3 facility is a Pressurized Water Reactor (PWR) operated by Kansai Electric Power Company in Japan since 1984[4]. Its nominal thermal power is 2652 [MW] and operates with a 17x17 fuel assembly design. Three fuel pins (SF95, SF96 and SF97) were investigated belonging to two different assemblies (NT3G23 and NT3G24). In particular, each pin was destroyed at the end of its operational cycle and five to six samples were drilled from each one; totally, 16 samples were collected from the three rods. From each sample, both actinides as well as fission products were measured through destructive analysis: slices of 0.5 [mm] were sectioned and dissolved from the depleted pins. Then, the concentrations were measured through different radiochemical techniques.

2.1 Irradiation History

This work focuses on the simulation of rods SF95 and SF97 due to the absence of burnable gadolinium in the fuel. Future validations will take into account also the SF96 rod.
The irradiation history of the assemblies follow operation cycles based over on/off periods in which the fuel is shuffled in the core disposition after a cooling period. In particular, the assembly NT3G23 (and so, fuel rods SF95 and SF96) was irradiated for two cycles; assembly NT3G24 (i.e., rod SF97) was irradiated for three cycles reaching an higher burnup level with respect the previous two.

Tab. [1] resumes the reactor time operations and Tab. [2] sums up the average specific powers for the rod in the cycles.

2.2 Geometry

The assemblies share the same geometric disposition: a 17x17 rod bundle with 250 standard pins, 14 pins with burnable absorbers and 25 guide tubes. Fig. [1] shows the relative position of the drilled rods inside the relative assembly.

Start	Stop	Cycle	State	Time interval [d]	Rods
26/01/1990	15/02/1991	5	On	385	SF95, SF96, SF97
15/02/1991	14/05/1991		Cool	88	SF95, SF96, SF97
14/05/1991	19/06/1992	6	On	402	SF95, SF96, SF97
19/06/1992	20/08/1992		Cool	62	SF97
20/08/1992	30/09/1993	7	On	406	SF97

Table 1: Irradiation history of Takahama-3 experiment. Data from [2].

	SF95	SF97
Cycle 5	0.04576	0.037147
Cycle 6	0.04013	0.039128
Cycle 7	-	0.034402

Table 2: Average Specific Power for Takahama-3 experiment. Entries are in $\left[\frac{kW}{g}\right]$.

Referring to Fig. [2], the axial division of the fuel rods has been modelled as follows:

- Nozzle: 12 [cm] of Stainless Steel. It has been considered an apparent density of $\rho_{nzl} = 3.416\left[\frac{g}{cm^3}\right]$[5];
- Cladding cap: 2 [cm] of Zircalloy-4;
- Upper plenum: 13.9 [cm] oh Helium;
- Active zone: 364 [cm] of fuel. In the simulation, has been chosen an axial division of 20 slices. The top and bottom's four with a length of 8.1 [cm] each and a central zone of twelve slices of 25 [cm] each;
- Lower plenum: 3.6 [cm] of Helium;

A reflector of 20 [cm] of light water has been consider in the axial direction. Then, black boundary conditions applies.
For what concern the radial boundary conditions, an infinite lattice has been considered. A more detailed discussion about the implications of che boundary conditions' choice will be faced in Sec. [3].

3 ANALYSIS

The result analysis is based on the evaluation of the *calculated-to-experimental* (C/E) ratio: considering the $i-th$ isotope in the $j-th$ axial position, then $C_{i,j} = \mathcal{N}_{ij}^{C}$ is the Serpent's predicted concentration and $E_{i,j} = \mathcal{N}_{ij}^{E}$ represents the experimentally measured value. The

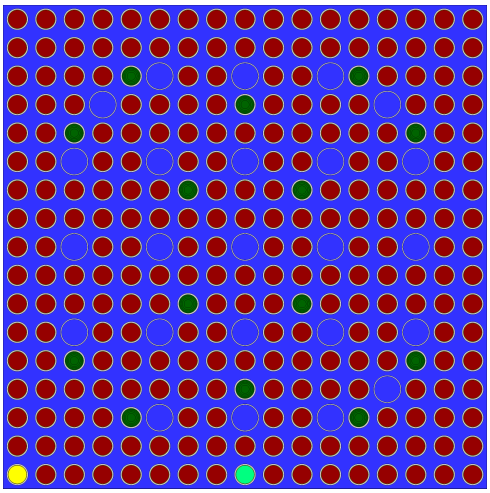

Figure 1: Top view of Takahama assembly design. Light green rod indicates the position of SF97 while the yellow rod indicates the SF95 position.

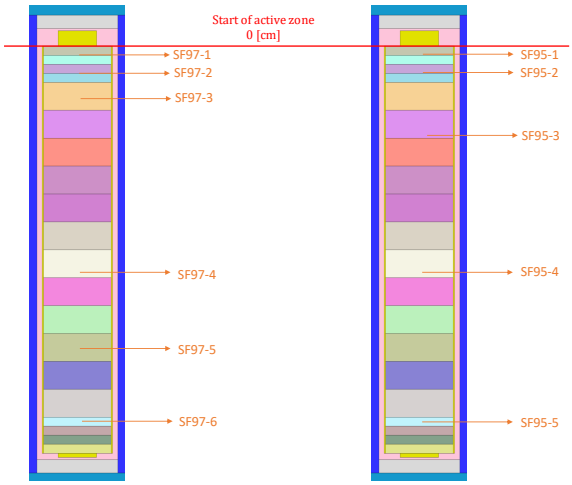

Figure 2: Lateral view of SF95 and SF97 models. Both the rods have been divided in 20 axial zones: 8 (4+4) more refined at the ends and 12 larger in the center.

latter may be taken from literature references[1] in the unit of $\left[\frac{g}{ton\,U}\right]$.
Then, for all (i, j) it is possible to evaluate the punctual figure of merit:

$$R_{ij} = \frac{C_{i,j}}{E_{i,j}} \tag{1}$$

As R_{ij} is close to one, as the isotopic prediction is accurate. However, this evaluation suffers from uncertainty over both C_{ij} and E_{ij}. The next section will treat the different uncertainty sources that affects this model.

3.1 Uncertainty propagation

This work assumes that the variability of the result is dependent by three main contributors and it can be expressed as follows:

$$\sigma^2_{tot} = f(\sigma^2_{MC}, \sigma^2_{exp}, \sigma^2_{sys}) \tag{2}$$

Where σ^2_{tot} is the total variance, σ^2_{MC} takes into account the stochastic effect due to the transport procedure, σ^2_{exp} is the experimental (known) variance and σ^2_{sys} represents the model's systematic

errors. Another important source of uncertainty comes from the nuclear libraries. However, the present work does not propagate its contribution. Future improvements will take into account also this factor.

Monte Carlo

The Serpent code is not able to furnish the standard deviation for the calculated concentrations. This is due to the fact that the Bateman Equations are solved through the exact direct CRAM method[6].
For this reason, in order to quantify σ^2_{MC}, ten different simulations have been launched with the same input files, resulting in ten outputs $\mathcal{N}_l$. Then, for each nuclide, the average composition in each drilled position has been evaluated $\langle\mathcal{N}\rangle$. For each run ($l = 1 : 10$) it was possible to evaluate the standard deviation of the l-th result with respect to the average $\sigma^2_l = (\mathcal{N}_l - \langle\mathcal{N}\rangle)^2$. Considering all the different runs: $\sigma^2_{MC} = \frac{1}{10}\sum_l \sigma^2_j$.
It turns out that with the adopted statistic of $n = 400000$ neutron per cycle, $c_a = 100$ active cycles and $c_i = 100$ inactive cycles, the relative standard deviation remains at sub-percent level for almost all the considered isotopes.

Experimental

The experimental uncertainty for each nuclide is related to the measurement procedure. Different radiochemical techniques have been used to characterized the samples (e.g., Isotopic Dilution Mass Spectrometry, α and γ spectrometry). Literature references report[2] the relative uncertainty for each isotope, which ranges from 0.1% to 10%.

Systematic

From a theoretical level, a 3D model may seem the best way to reproduce the fuel's depletion in a rod bundle. In fact, this approach is able to track the spacial evolution of the neutron flux from a fresh fuel cosine shaped to a depleted fuel flat profile. However, the lack of some geometrical information may lead to an inaccurate knowledge of the neutron flux in the axial's end.
In particular, the Takahama-3 experiment suffers from the presence of a systematic error given by three main aspects:

- **Z boundary**: the axial end behaviour depends mostly by the backscattering capability of the reflector. For the Takahama-3 reactor, no data were present for either the reflector's length and its composition. In the present work, it has been hypothesized light water having width of 30 [cm].

- **X-Y boundary**: between one operation cycle and another, the assemblies are *shuffled* in the reactor's lattice in order to allows an uniform irradiation. Both the the initial assembly position as well as the shuffled disposition were unknown for the presented case. For this reason, this work adopts a reflective boundary condition in the radial direction, introducing an overestimation/underestimation of the neutron flux.

- **Sample location**: as reported in [7], the estimated error in the sampling position is < 2 [cm]. In the central zone this is a negligible uncertainty, while in the ends, even this short distance may change the neutron flux significantly.

Due to the previously listed uncertainties, this work bases its validation only on those samples which are sufficiently distant from the rod's end. For this reason, it has been excluded the SF95-1 and SF97-1 data-set (Fig. [2]).
Only the experimental standard deviation will be considered, since the Monte Carlo contribution is negligible with the applied statistics:

$$\sigma_{tot}^2 = f(\sigma_{exp}^2) \tag{3}$$

The systematic error is not directly quantifiable; according to literature [8], an uncertainty related to the neutron flux introduces a variability from 10% to 20%. This work places an *acceptance threshold* of $\pm 15\%$ from the unity in the C/E analysis. The ratio between the C/E points inside the threshold and the total samples defines the *success rate*, able to characterize the global goodness of a simulation. Fig. [3] shows the C/E distribution for the SF95 data with the acceptance threshold.

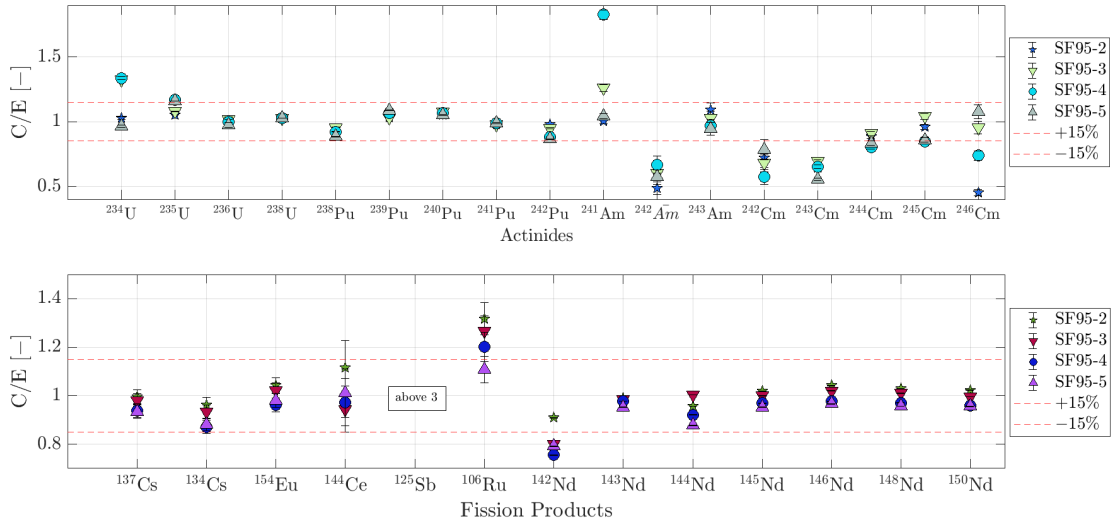

Figure 3: Calculated-to-experimental ratio for the SF95 data-set. The top row shows the prediction for the actinides, while the bottom row contains the FPs.

3.2 Sensitivity

In order to seek which model design option best increases the success rate, a sensitivity analysis has been performed. This work tests three input features: the choice of the nuclear library, the time span for the neutronic solver and the temperature treatment for the fuel. The following sections briefly explain those features while the results are summed up in Sec. [4].

Nuclear library

This work compares two nuclear databases: the ENDF/B-VII and the JEFF-3.1.1. Both the cases exploit a constant fuel temperature of $T = 900\,[K]$ and an axial division of each pin in 40 zones.

Temporal mesh

A neutronic simulation is carried out coupling of two equations sets': depending on the burnup scheme adopted, the neutron flux is kept constant at a certain calculated value and the

Bateman equations are solved. The choice of how frequently the constant neutron flux value has to be updated is a non-trivial task.
The first 48 [h] of depletion after a startup are treated separately due to the poisons' presence. Then, three regular time step's length has been considered: $\Delta t_1 = 25$ [d], $\Delta t_2 = 50$ [d] and $\Delta t_3 = 100$ [d].
Comparing the success rates obtained from these cases, the optimal Δt may be chosen, ensuring the higher accuracy whith the lowest computational time.

Temperature treatment

This work considers two approaches for the fuel and coolant temperature evaluation. The former keeps constant values of $T_f = 900$ [K] and $T_c = 560$ [K] for the whole burnup period. The latter, imposes a temperature $T_f(t, z)$ and $T_c(t, z)$, variable both in the z-dimension as well as in time. The rods have been divided in five non-regular zones where it has been applied a fuel temperature proportional to the thermal neutron flux. The coolant temperature has been thereby modified adopting an effective heat transfer coefficient. Also, the temperatures have been updated in each zone every ~ 200 [d], to keep track of the change in the fuel depletion.

4 RESULTS

Tab. [3] sums up the success rates (SR) for the presented sensitivity cases. The SR is defined as the ratio between the sum of the C_i/E_i points falling in the defined threshold and the total samples.
The same methodology has been applied to results from other 2D neutronic codes present in literature. In particular, the SWAT code proposes a sensitivity on the fuel temperature while ORIGEN2.1 analyse different libraries [1].

Code	Sensitivity	SF95	SF97
This work	ENDF/B-VII.0	0.758	0.753
	JEFF-3.1.1	0.708	0.747
	$\Delta t_1 = 25$ [d]	0.70	0.78
	$\Delta t_2 = 50$ [d]	0.70	0.78
	$\Delta t_3 = 100$ [d]	0.68	0.75
	T_f = const	0.68	0.75
	$T_f = f(z)$	0.76	0.726
SWAT	T = const	0.63	0.689
	$T = f(t, z)$	0.675	0.726
ORIGEN 2.1	PWR-US	0.616	0.526
	PWR-UE	0.675	0.636
SCALE 5.1	-	0.76	0.78

Table 3: Comparison between the results from this work and the literature references for the Takahama-3 benchmark in terms of success rate.

5 CONCLUSIONS

This work focused on the validation of a 3D fuel assembly model exploiting the Monte Carlo code Serpent. In particular, the features are able to increase the prediction accuracy of the isotopic concentration were investigated. The Takahama-3 PIE data-set was used as a test for different sensitivity cases.
The nuclear libraries are continuously updated: the comparison between ENDF/B-VII.0 and JEFF-3.1.1 has shown how the former better reproduces by $+5\%$ the SF95's samples and by $\sim 0.6\%$ the SF97 ones. From what concern the temporal mesh's choice, has been shown that the success rate for $\Delta t = 100$ [d] is smaller compared with the other two rods. Also, the simulations with $\Delta t = 25$ [d] and $\Delta t = 50$ [d] are characterized by the same success rate. This allowed to increase the time step maintaining the same accuracy and therefore reducing the computational efforts. Finally, the adoption of a temperature field were able to increase the accuracy of the SF95 benchmark in the given acceptance band of +8%. However, the SF97 rod shows a reverse trend, making the constant temperature results more accurate of 3%. This could be explained by the fact that the latter rod is more subjected to the systematic error propagation.
Future works will focus on the inclusion of neutron library-related uncertainty and to the validation of the SF96 bar with burnable absorber. In addition, temperature has been shown to be a feature that can macroscopically increase the accuracy of the analysis. For this reason, the mesh's detail level will be further investigated through external thermal-hydraulic codes.

REFERENCES

[1] I C Gauld. Technical development on burn-up credit for spent lwr fuel. 12 2001.

[2] Charlotta Sanders and Ian Gauld. Isotopic analysis of high-burnup pwr spent fuel samples from the takahama-3 reactor. 03 2022.

[3] Jaakko Leppänen, Maria Pusa, Tuomas Viitanen, Ville Valtavirta, and Toni Kaltiaisenaho. The serpent monte carlo code: Status, development and applications in 2013. *Annals of Nuclear Energy*, 82:142–150, 2015.

[4] *Operating Experience with Nuclear Power Stations in Member States*. Operating Experience with Nuclear Power Stations in Member States. IAEA, Vienna, 2021.

[5] Tatiana Kaznacheevskaia. Validation of the serpent monte carlo burnup code for the takahama-3 benchmark experiment. 2020.

[6] Maria Pusa and Jaakko Leppänen. Solving linear systems with sparse gaussian elimination in the chebyshev rational approximation method. *Nuclear Science and Engineering*, 175(3):250–258, 2013.

[7] Mark Dehart, Ian Gauld, and Kenya Suyama. Three-dimensional depletion analysis of the axial end of a takahama fuel rod. 01 2008.

[8] Y Oyama, C Konno, and Y Ikeda. Measurement techniques for fusion blanket neutronics experiments. 12 1994.