# DEVELOPMENT OF A 3D DETERMINISTIC FUEL DEPLETION CODE

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## ABSTRACT

Predicting the isotopic evolution and its impact in the core performance becomes crucial for designing and operating nuclear reactors. This work faces the starting point of developing a depletion code using the latest advances in numerical computing. MATLAB will be used to create a depletion program, getting the necessary nuclear data from the SCALE code system. Burn-up chains, managing of the proper nuclear data, creating, and solving ODE systems are analysed in the present work. As a result, a fuel depletion code was created. The code can perform depletion calculations for a single fuel material for a reduced burn-up chain. The relative error between the obtained and the reference nuclides concentrations is considerably low. The general assumptions, requirements and methodology for creating a depletion code have been analysed and verified.

## 1. Introduction

Simulating nuclear reactors involves a whole process of simulating different phenomena at several levels. The study of the whole core, using nodal codes coupled with thermal hydraulics, relies on pre-generated cross section libraries. These libraries are generated by lattice and depletion codes and contain information for any possible scenario under study. The role of depletion codes is to evaluate the isotopic evolution inside reactor cores, specially, in the nuclear fuel.

This work faces the starting point of developing a depletion code. The depletion of a material can be modelled as a first order ordinary differential equation system (ODE system), in which, each equation measures the evolution of a particular nuclide. Starting from an initial concentration of isotopes, the neutron flux, and nuclear data (cross section, yields and decay constants) the ODE is completely defined. In this work, MATLAB [1] will be used to read the nuclear data created by the sequence TRITON/NEWT from the SCALE code system [2], which feeds the depletion module ORIGEN. Then, MATLAB will be used to create and solve the ODE system. ORIGEN's nuclides concentrations will be considered as reference. This work is a first approach to develop a whole depletion code coupled with VALKIN-FVM-Sn [3] for transport calculations and SLEPc, the Scalable Library for Eigenvalue Problem Computations [4], for solving the ODE system.

MATLAB has been chosen to develop a preliminary version of the depletion code. This is because MATLAB provides an easy and comfortable integrated programming environment, making easier the initial development. After the validation of the preliminary version in MATLAB, the code will be translated to FORTRAN, which surpass MATLAB in computation speed and optimization capabilities. The main and final objective of this work is to obtain an in-house lattice-depletion code capable of dealing with arbitrary geometries (not only cartesian or hexagonal) and reading different cross-section data file formats.

## 2. Burn-up

Materials inside a nuclear reactor core are subtle to suffer changes in their isotopy due to two mechanisms mainly: radioactive decay and neutron induced reactions. The element that most experiments this phenomenon is the nuclear fuel; it is naturally radioactive and is the most exposed to the neutron flux, in addition, it is the only element that can experiment fission reactions. Naturally, the isotopic change in the nuclear fuel is the most influential in regards of reactor dynamics. In this work only fuel depletion will be evaluated as it is the most complex and difficult scenario in terms of isotopic evolution.

To study the evolution of an isotope it is possible to stablish a balanced relation between the production and destruction rates of a target nuclide. The evolution of the isotope *i* inside a volume can be described mathematically as follows in Eq. 1, a first order differential equation.

$$\frac{dN_i}{dt} = \sum_{j \neq i} (l_{ij}\lambda_j + \sigma_{j \to i}\phi + \gamma_{j \to i}\sigma_{f,j}\phi)\, N_j(t) - (\lambda_i + \sigma_i\phi)N_i(t)$$

Eq. 1. Depletion equation for nuclide $i$.

where:

- $\frac{dN_i}{dt}$ is the change rate of nuclide $i$ concentration (atoms),
- $N_j(t)$ is the amount of nuclide $j$ at a time $t$ (atoms),
- $\lambda_i$ is the decay constant of nuclide $i$ (1/s),
- $l_{ij}$ is the branching ratio of nuclide $i$ from decay of nuclide $j$,
- $\sigma_{j \to i}$ is the cross section for nuclide $j$ producing nuclide $i$ (barn),
- $\sigma_{f,j}$ is the fission cross section of nuclide $j$ (barn),
- $\sigma_i$ is the removal cross section of nuclide $i$ (barn),
- $\gamma_{j \to i}$ is the independent fractional fission yield of nuclide $i$ from nuclide $j$,
- $\phi$ is the neutron flux (neutrons/cm$^2$-s),

Eq. 1 is a general expression for all kind of isotopes contained in the fuel. However, it is common to differentiate into families of isotopes. The differentiation into families or groups is due to the existence, lack or dominance of some of the terms exposed previously in the target nuclides. The most common and straight forward differentiation is between actinides and fission products.

### 2.1 Solving the burn-up equation

The evolution of the isotope inventory in a determined volume can be modelled by a set of coupled depletion equations. All the equations form a first order ODE system. Solving this kind of ODEs is a well-known and straight forward mathematical problem. In the case of the burn-up analysis, the ODE must be solved using numerical methods, due to the large number of isotopes (coupled equations) to consider.

The problem is usually expressed in matrix notation, as in Eq. 2:

$$\frac{dN}{dt} = A\vec{N}(t)$$

Eq. 2. Matrix notation used in depletion ODE systems.

In the matrix form, $A$ is the so called "*transition matrix*" and contains the production and loss ratios for every isotope under study. $\vec{N}(t)$ is a column vector formed by the isotope's concentrations at time $t$.

## 2.2 Burn-up chains and nuclear data

Nuclear fuel under operation contains thousands of nuclides that can experiment dozens of possible neutrons induced reactions and decay transitions. This leads to a huge transition matrix with many relations between nuclides. Some codes, such as SCALE can perform detailed depletion calculation considering most of the nuclides and transitions. However, the dynamics of the reactor are mostly influenced by a reduced number of nuclides and neutron reactions. It is possible to find reduced burn-up chains with a relatively short number of nuclides and neutron reactions. These burn-up chains are suitable to appropriately study the performance of the fuel, this is the case of WIMS-ANL [5], MCDL code [6] and MVP-BURN code system [7]. For this work a burn-up chain model from the MVP-BURN code system, shown in Fig 1. and Fig 2, will be used. The burn-up chain selected is a standard model whose main purpose is to perform burn-up calculations for thermal reactors. It is composed by 27 actinides, 55 fission products and a pseudo fission product.

There are two main ways to consider fission products, explicitly and lumped into a pseudo fission product. The pseudo fission product is created to consider the influence of nuclides that do not have a significant impact on the core individually, but their cumulative effects are not neglectable. The pseudo fission product is created by averaging and weighting the properties of several isotopes into one.

Radiative capture, $(n,\gamma)$, is the major capture reaction. Radiative capture, $(n,2n)$ reaction and fission are the major neutron induced reaction influencing the reactor core. These are the neutron induced reaction to be considered in this work.

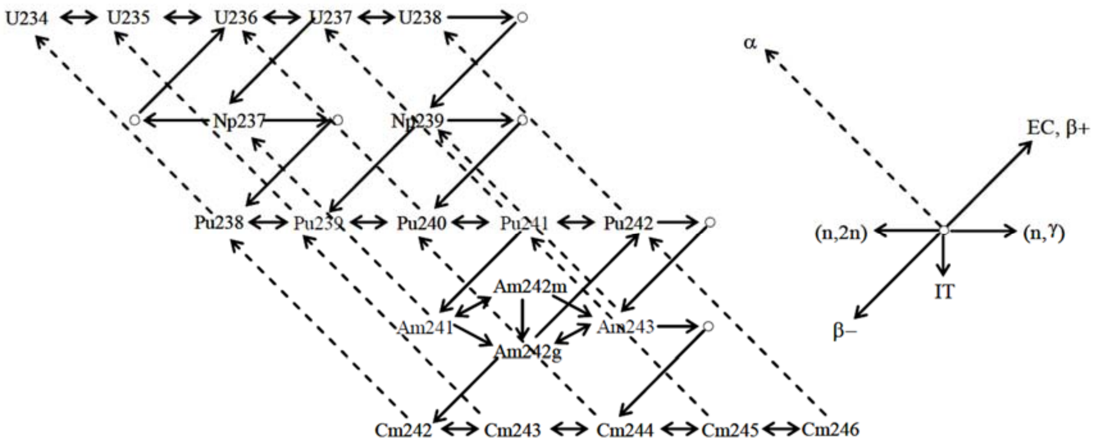

Fig 1. Actinides burn-up chain [7].

## 3. Methodology

ORIGEN performs depletion calculations for 174 actinides, 1149 fission products and 974 activation products. SCALE stores the information required by ORIGEN in libraries and the libraries are stored in binary format files. A file containing the transition matrix (*f33* file) is created for each material to be depleted. In the *f33* file, each row corresponds to a particular transition between nuclides and each column to a depletion step. It contains the cross sections for all neutron reactions, the direct fission yields, the decay constants, and branching ratios for every depletion step. For every depleted material, the neutron flux, the nuclides concentrations, and the time at the end of each depletion step, are stored in a unique binary file (*f71* file).

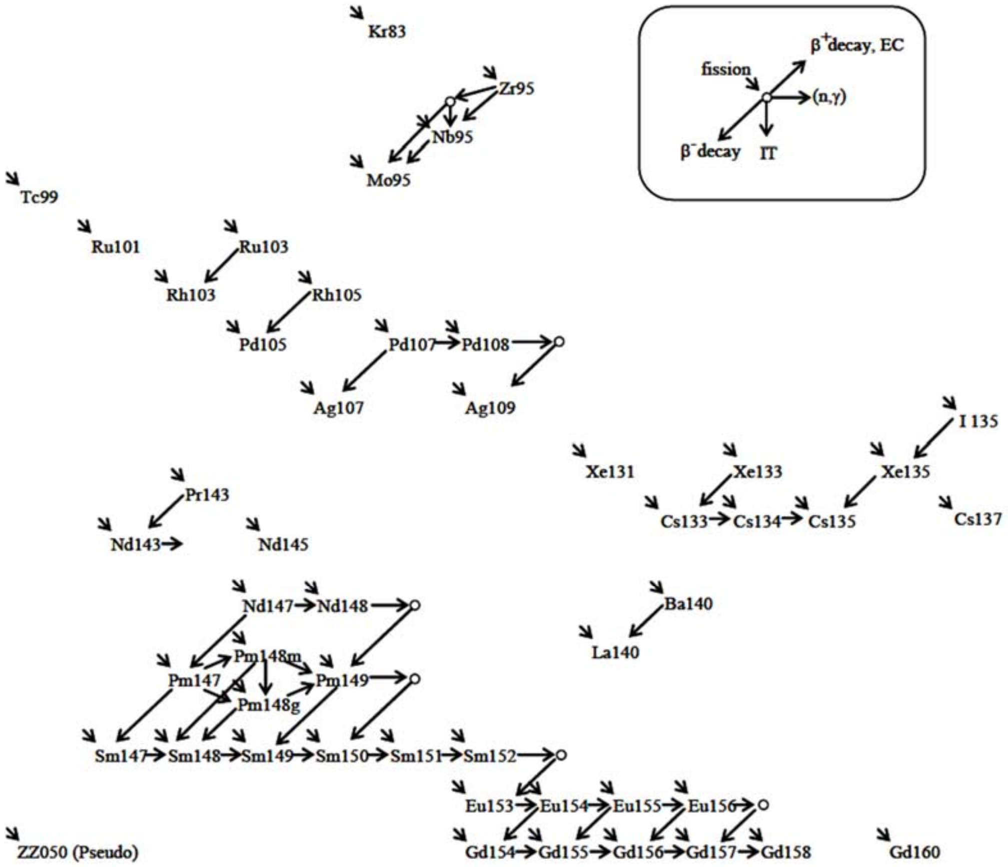

Fig 2. Fission products burn-up chain [7].

## 3.1 SCALE model and data

A fuel rod was modeled and depleted using SCALE. The fuel rod parameters correspond to the TMI-1 PWR unit cell from the UAM benchmark [8]. It was depleted for 365 days with a power density of 0.081 (MW/MTHM), a usual value for LWR fuel rods. All the model information is shown in Tab 1 and Fig 3.

Tab 1. Model data and specifications.

| Parameters | Value |
|---|---|
| Unit cell pitch, [mm] | 14.427 |
| Fuel pellet diameter, [mm] | 9.391 |
| Fuel pellet material | $UO_2$ |
| Fuel density, [g/cm$^3$] | 10.283 |
| Fuel enrichment, w/o | 4.85 |
| Cladding outside diameter, [mm] | 10.928 |
| Cladding thickness, [mm] | 0.673 |
| Cladding material<br>Cladding density, [g/cm$^3$] | Zircaloy – 4<br>6.55 |
| Gap material | He |
| Moderator material | $H_2O$ |
| Fuel temperature, [K] | 900 |
| Cladding temperature, [K] | 600 |
| Moderator (coolant) temperature, [K] | 562 |
| Moderator (coolant) density, [kg/m$^3$] | 748.4 |
| Power density, [MW/MTHM] | 0.081 |

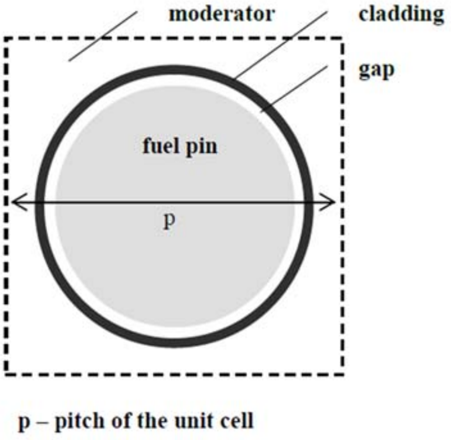

Fig 3. Fuel rod depiction [8].

## 3.2 Program development

It was stated in Eq. 1 all the data needed to evaluate the nuclide evolution in a volume. All this data is available in both, *f33* and *f71* SCALE files. These files are contained in the temporary folder, which must be stored. Once the files are stored, the "*obiwan"* tool provided by SCALE must be used to translate the information from binary into ascii format. With all this data and a burn-up chain it is possible to create a MATLAB program to perform depletion calculations. The program will read and process the data proceeding from SCALE, then, create and solve the ODE system for every depletion step. There are available several first order ODE system solvers in MATLAB, in this work **ode45** is used.

Depletion calculations are usually performed for long time periods, several months or years. It is usual to deplete the materials in intermediate steps inside the period of interest. During the steps, the transition matrix is considered to be constant. This is the case of SCALE and the program under development. As in SCALE, the transition matrix was divided in two. One containing the neutron induced reactions, that needs to be multiplied by flux and changes with time, and one containing the decay transitions, Eq. 3.

$$\frac{dN}{dt} = (A_{\sigma,n}(t)\phi_n(t) + A_\lambda)\vec{N}(t)$$

Eq. 3. Split matrix notation.

An advantage of this approach is that the decay matrix must be created only once. On the other side, the neutron induced matrix and flux must be updated at each depletion step, as the concentration vector. The concentration vector from SCALE is only used at time zero, then it is used the new concentration obtained by MATLAB at the end of each depletion step. The program workflow is shown in Fig 4.

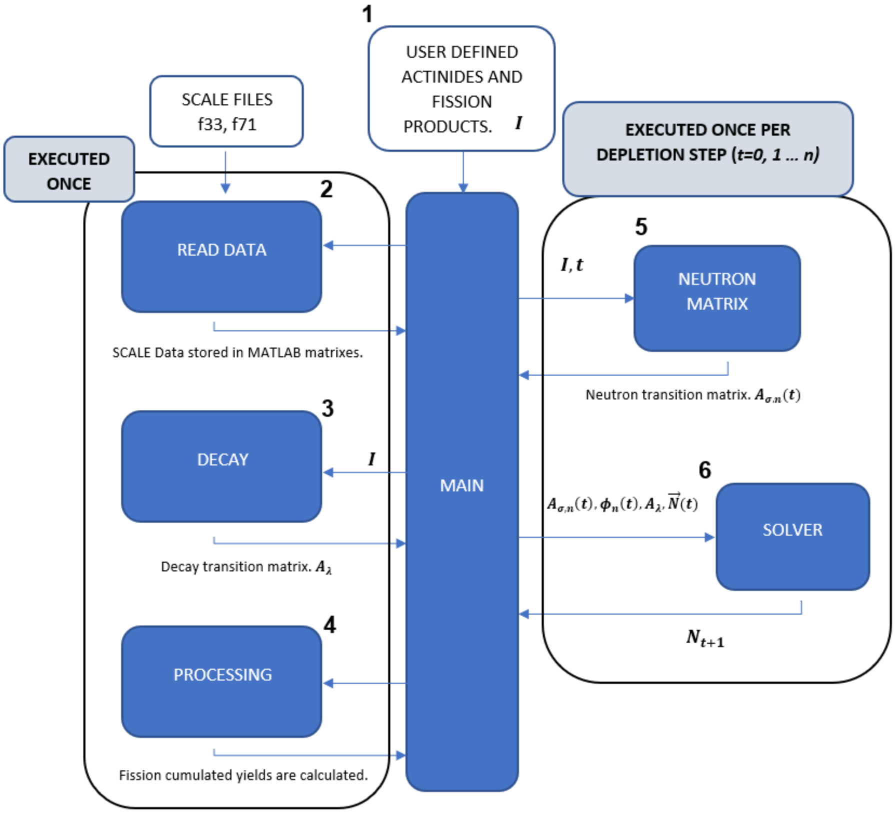
1
USER DEFINED ACTINIDES AND FISSION PRODUCTS. $I$
SCALE FILES f33, f71
EXECUTED ONCE
EXECUTED ONCE PER DEPLETION STEP (*t=0, 1 … n)*
2
READ DATA
SCALE Data stored in MATLAB matrixes.
3
DECAY
$I$
Decay transition matrix. $A_\lambda$
4
PROCESSING
Fission cumulated yields are calculated.
MAIN
5
$I, t$
NEUTRON MATRIX
Neutron transition matrix. $A_{\sigma,n}(t)$
6
$A_{\sigma,n}(t), \phi_n(t), A_\lambda, \vec{N}(t)$
SOLVER
$N_{t+1}$

Fig 4. Program workflow.

Once the *f33* and *f71* files are translated to ascii, by means of the *"obiwan"* tool, all the information needed for every depletion step is available and the code can be executed. In the first place, the user must define the actinides and fission products to be used. Secondly, the code will automatically read the files and store the information for the whole depletion. The information is stored into tables and vectors, where each column corresponds to a depletion step, including the initial point. Thirdly, the decay transition matrix is created for the specified nuclides. Then, the fission cumulated yields are computed. The next two steps are repeated once per depletion step. The following step is to create the neutron transition matrix for the specified nuclides at the current depletion step. With all the previous information, the flux and concentrations at the moment (information read from the *f71* file), the ODE can be solved and the concentrations at the end of de depletion step (beginning of the following) is obtained.

## 4. Results

The program was developed and verified in two phases, the first one considering only actinides, and the second one with fission products. The MVP-BURN burn-up chain is chosen to evaluate the program, nevertheless, the program is prepared to consider any actinide or fission products combination defined by the user. In this work, the pseudo fission product is not considered.

To evaluate the results, the relative error between the concentrations from SCALE and the MATLAB program was calculated at the end of every depletion step, Eq. 4. Positive error means a greater SCALE concentration than MATLAB. It is also shown the evolution of relevant nuclides during the depletion.

$$Rel. error\ \% = \frac{N_{i,SCALE} - N_{i,MATLAB}}{N_{i,SCALE}} * 100$$

Eq. 4. Rel. Error equation.

### 4.1 Actinides

There are 27 actinides in the burnup chain. They are numbered as stated in Tab 2, the results will be shared using that notation. Four depletion calculations were performed for 365 days with 4, 16, 32 and 64 uniformly distributed depletion steps.

Tab 2. Actinides identifications.

| Id | Nuclide | Id | Nuclide | Id | Nuclide | Id | Nuclide |
|---|---|---|---|---|---|---|---|
| 1 | U-234 | 8 | Np-237 | 15 | Pu-241 | 22 | Am-244 |
| 2 | U-235 | 9 | Np-238 | 16 | Pu-242 | 23 | Cm-242 |
| 3 | U-236 | 10 | Np-239 | 17 | Pu-233 | 24 | Cm-243 |
| 4 | U-237 | 11 | Np-240 | 18 | Am-241 | 25 | Cm-244 |
| 5 | U-238 | 12 | Pu-238 | 19 | Am-242m | 26 | Cm-245 |
| 6 | U-239 | 13 | Pu-239 | 20 | Am-242 | 27 | Cm-246 |
| 7 | Np-236 | 14 | Pu-240 | 21 | Am-243 | | |

The results obtained with the MATLAB program match very well with SCALE as seen in Fig 5. The relative error for the 27 actinides is considerably low.

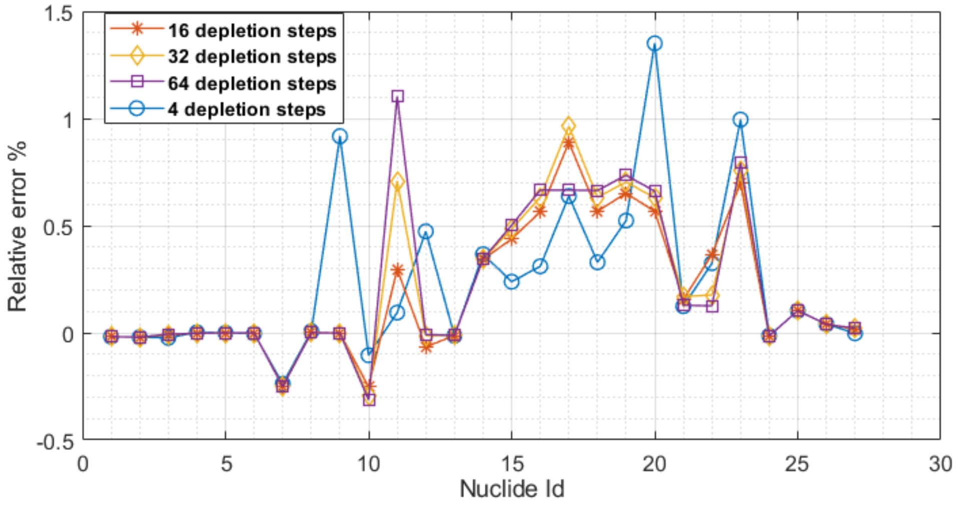

Fig 5. Results for actinides at the end of the depletion, 365 days.

## 4.2 Fission products

In the burn-up chain, there were considered 55 fission products. They have been numbered as in Tab 3.

Tab 3. Fission products identifications.

| Id | Nuclide | Id | Nuclide | Id | Nuclide | Id | Nuclide |
|---|---|---|---|---|---|---|---|
| 28 | Kr-83 | 42 | Ag-107 | 56 | Nd-145 | 70 | Sm-152 |
| 29 | Zr-95 | 43 | Ag-109 | 57 | Nd-147 | 71 | Sm-153 |
| 30 | Nb-95m | 44 | I-135 | 58 | Nd-148 | 72 | Eu-153 |
| 31 | Nb-95 | 45 | Xe-131 | 59 | Nd-149 | 73 | Eu-154 |
| 32 | Mo-95 | 46 | Xe-133 | 60 | Pm-147 | 74 | Eu-155 |
| 33 | Tc-99 | 47 | Xe-135 | 61 | Pm-148m | 75 | Eu-156 |
| 34 | Ru-101 | 48 | Cs-133 | 62 | Pm-148 | 76 | Eu-157 |
| 35 | Ru-103 | 49 | Cs-134 | 63 | Pm-149 | 77 | Gd-154 |
| 36 | Rh-103 | 50 | Cs-135 | 64 | Pm-150 | 78 | Gd-155 |
| 37 | Rh-105 | 51 | Cs-137 | 65 | Sm-147 | 79 | Gd-156 |
| 38 | Pd-105 | 52 | Ba-140 | 66 | Sm-148 | 80 | Gd-157 |
| 39 | Pd-107 | 53 | La-140 | 67 | Sm-149 | 81 | Gd-158 |
| 40 | Pd-108 | 54 | Pr-143 | 68 | Sm-150 | 82 | Gd-160 |
| 41 | Pd-109 | 55 | Nd-143 | 69 | Sm-151 | | |

First, the direct fission yield was used. In contrast to the actinides, considering only the 55 fission products is not enough to get a good match between the SCALE and MATLAB results, Fig 6. This is because there are many contributions neglected to the considered nuclides. Theses nuclides are produced by two main sources, directly by fission, and by decay from nuclides also produced by fission. Whereas the nuclides considered explicitly are stable or have a relatively large half-life, most of their parents have short half-lives. The decay contributions to most of the fission products of the burn-up chain are considerably larger than the direct fission contributions. A common approach to consider the decay contributions to the explicit fission products is to obtain the cumulative fission yield, adding the parent's yield to the considered fission products yield. The parent's yield must be multiplied by the branching ratio of the decay transition before been added to the daughter's yield.

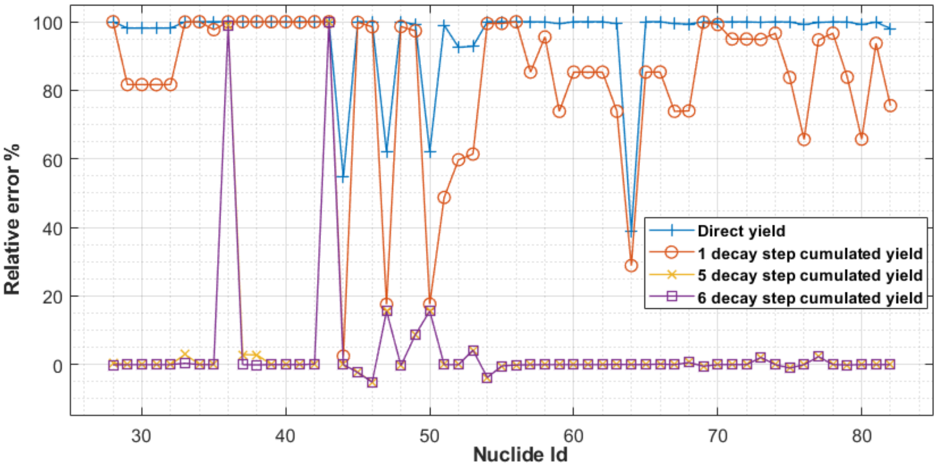

Fig 6. Results at the end of the depletion (365 days) for fission products and several cumulative fission yields.

It was determined to cumulate the yields for transitions with a decay constant greater than 1e-7 ($s^{-1}$). A nuclide may be produced by several parents from different decay mechanisms. Considering only the immediate parents of the fission products to generate the cumulated yields may not be enough. It was studied the response of cumulating the yield, not only from the explicit fission products parent nuclides, but from the parents' ancestors as well. The cumulative yield was obtained for several decay steps backwards. The results are shown in Fig 6. It is evident that, the most decay ancestors are considered to compute the cumulative yield, the better results are obtained. The difference between considering 5 or 6 decay steps backwards is very low, 6 steps backwards is used for now on. By considering 6 decay steps backwards, 55 nuclides are considered explicitly and 552 implicitly into the cumulative yields.

### 4.3 Final results

In Fig 6, most errors (for 6 decay steps backwards cumulated yield) are between -6 % and 5% for all fission products, except for the Rh-103 and Ag-109, which have a relative error near 100%, and Xe-135 and Cs-135, with a relative error over 15%. These four nuclides were found to have isomeric states with great neutron induced contributions. As seen in Fig 7, adding Rh-103m, Ag-109m, Xe-135m and Cs-135m to the burn-up chain considerably improves the results. All relative errors are now between +4% and -6% for 4 depletion steps, and between +2% and -2.5% for 64 depletion steps. Tab 4 shows the new nuclide identifications for the fission products.

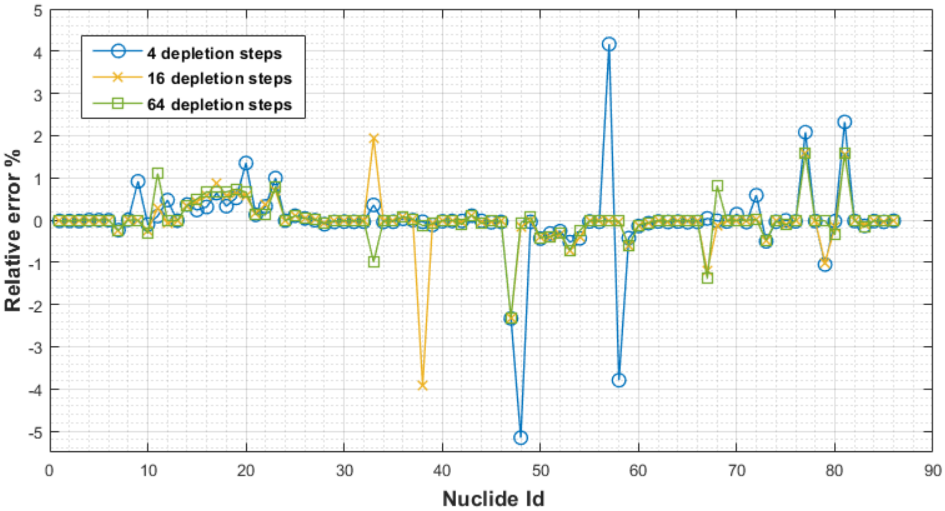

Fig 7. Final results for all nuclides at the end of the depletion (365 days).

Tab 4. Second fission products identifications.

| Id | Nuclide | Id | Nuclide | Id | Nuclide | Id | Nuclide |
|---|---|---|---|---|---|---|---|
| 28 | Kr-83 | 43 | Ag-107 | 58 | Pr-143 | 73 | Sm-151 |
| 29 | Zr-95 | 44 | Ag-109m | 59 | Nd-143 | 74 | Sm-152 |
| 30 | Nb-95 | 45 | Ag-109 | 60 | Nd-145 | 75 | Sm-153 |
| 31 | Nb-95m | 46 | I-135 | 61 | Nd-147 | 76 | Eu-153 |
| 32 | Mo-95 | 47 | Xe-131 | 62 | Nd-148 | 77 | Eu-154 |
| 33 | Tc-99 | 48 | Xe-133 | 63 | Nd-149 | 78 | Eu-155 |
| 34 | Ru-101 | 49 | Xe-135m | 64 | Pm-147 | 79 | Eu-156 |
| 35 | Ru-103 | 50 | Xe-135 | 65 | Pm-148m | 80 | Eu-157 |
| 36 | Rh-103m | 51 | Cs-133 | 66 | Pm-148 | 81 | Gd-154 |
| 37 | Rh-103 | 52 | Cs-134 | 67 | Pm-149 | 82 | Gd-155 |
| 38 | Rh-105 | 53 | Cs-135m | 68 | Pm-150 | 83 | Gd-156 |
| 39 | Pd-105 | 54 | Cs-135 | 69 | Sm-147 | 84 | Gd-157 |
| 40 | Pd-107 | 55 | Cs-137 | 70 | Sm-148 | 85 | Gd-158 |
| 41 | Pd-108 | 56 | Ba-140 | 71 | Sm-149 | 86 | Gd-160 |
| 42 | Pd-109 | 57 | La-140 | 72 | Sm-150 | | |

In Fig 7 is shown that the final results match very well between MATLAB and SCALE. For now, only the results at the end of the depletion have been shown. It is also interesting to study the nuclide evolution during the burn-up, Fig 8.

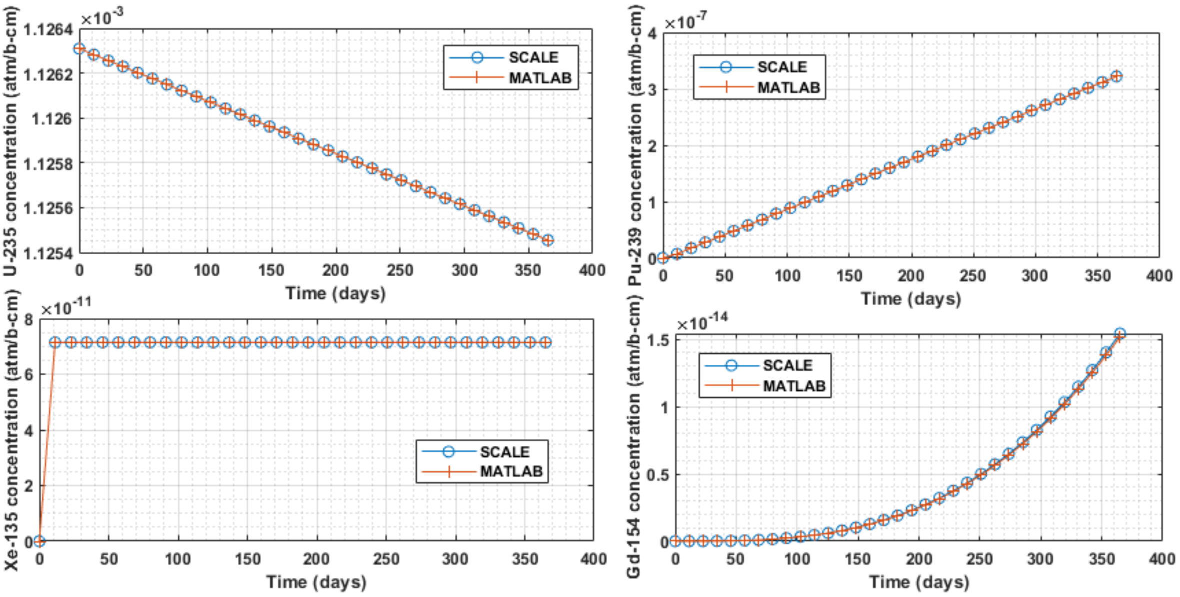

Fig 8. Nuclide's concentration evolution for 64 depletion steps.

## 5. Conclusions and future works

Depletion data from SCALE, which perform burn-up calculations using a large number of nuclides and reactions, has been used to feed a MATLAB depletion program. The program is able to perform depletion calculations for a set of actinides and fission products determined by the user, it is automatized for working with any number of depletion steps. The final burn-up chain contained 27 actinides and 59 fission products compared to the 174 actinides and 1149 fission products used in SCALE. There were considered only three possible neutron induced transitions, fission, $(n, 2n)$ and $(n, \gamma)$. The cumulated yield was calculated for all the fission products.

It has been proven that, once the proper cumulative yield is obtained for the fission products, the most important nuclides in reactor dynamics can be properly calculated for a reduced burnup chain. The MATLAB solver provides good results, with relative errors below 5% in absolute value for 4 depletion steps, and below 2.5% for 64 depletions steps. The methodology followed to read the data, create, and solve depletion ODE systems is validated.

After the developing of this work, it is now possible to translate the validated methodology to FORTRAN. The SLEPc ODE solver could be used, this will allow to consider more nuclides explicitly. The program will be automatized to deplete several materials and it will be coupled with VALKIN-FVM. In this way it will be possible to perform depletion calculations for complex geometries in 2-D or even 3-D.

## 6. Acknowledgments

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## 7. References

1. MATLAB. (2010). *Version 9.9.0.1592791 (R2020b) Update 5*. Natick, Massachusetts: The MathWorks Inc.
2. T. Rearden and M.A. Jessee, Eds., *SCALE Code System, ORNL/TM-2005/39, Version 6.2.3*, Oak Ridge National Laboratory, Oak Ridge, Tennessee (2018).
3. Bernal, et al., (2018), VALKIN-FVM: A modal finite volume method for solving the transient neutron diffusion equation on unstructured meshes, PHYSOR 2018: reactor physics paving the way towards more efficient systems.
4. V. Hernandez, J. E. Roman, and V. Vidal. *SLEPc: A scalable and flexible toolkit for the solution of eigenvalue problems.* ACM Trans. Math. Software, 31(3):351-362, 2005.
5. WIMS Library Update Project WLUP, http://www.nds.iaea.org/wimsd/, 2001.
6. Cuong Kien NGUYEN, et al. *The development of depletion program coupled with monte carlo computercode*, https://inis.iaea.org/collection/NCLCollectionStore/_Public/47/065/47065286.pdf?r=1&r=1, 2021.
7. Keisuke OKUMURA, Yasunobu NAGAYA and Takamasa MORI, *MVP-BURN: Burn-up Calculation Code using a Continuous-energy Monte Carlo Code MVP*, JAEA, Japan, Jan. 2005MVPBURN.
8. Kostadin Ivanov, et al., (2013). Benchmark for uncertainty analysis in modeling (UAM) for design, operation and safety analysis of LWRs (Vol. 1). OECD Nuclear Energy Agency.