Optimization of STREAM Burnup Chain using Depletion Perturbation Theory

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1. Introduction

A method of simplifying depletion chain has been introduced in this research to alleviate the computational burden in nuclear reactor design and analysis. A simplified burnup chain model was created out of the detailed many isotopes matrix using Depletion Perturbation Theory (DPT) [1] approach and redistributing nuclear data (decay fractions, fission yields, etc.) in order to reduce computation time and memory usage for the purpose of calculating effective neutron multiplication factors (k_{eff}) and power distributions. A detailed burnup chain was implemented in the deterministic code STREAM [2] developed at UNIST with a depletion solver based on the Chebyshev Rational Approximation Method (CRAM) [3]. The detailed burnup chain includes 3837 nuclides and 43416 transitions, and is based on ENDF/B-VII.1 nuclear data. However, using the detailed burnup matrix in depletion calculations requires a huge amount of computational time. In order to reduce the computational cost of depletion calculations, a simplified burnup chain model is introduced. The DPT methodology has been applied to generate a 306-nuclide depletion chain optimized for the calculation of the effective neutron multiplication factor (k_{eff}) in light water reactor (LWR) depletion problems.

2. Methods and Results

In this section, a brief description of the proposed method and the depletion chain compression process are given. Depletion calculation using the simplified burn up chain is conducted for a PYREX bearing Fuel Assembly (FA) which is extracted from Virtual Environment for Reactor Application (VERA) benchmarks [4].

2.1 Depletion Perturbation Theory

The burnup equations form a system of first-order linear differential equations that can be written as:

$$\frac{\partial n_j}{\partial t} = \sum_{j \neq i} \lambda_{ij} n_i - \lambda_j n_j, \qquad n_j(0) = n_0, \tag{1}$$

where n_j is the concentration of nuclide j (j=1,2…, N), N is the total number of nuclides, and λ_{ij} are coefficients characterizing the rates of neutron-induced reactions and spontaneous radioactive decay process. Using a matrix notation, Eq. (1) can be expressed as:

$$\frac{\partial \boldsymbol{n}(t)}{\partial t} = \boldsymbol{A}(t)\, \boldsymbol{n}(t), \tag{2}$$

where $\boldsymbol{A} \in \mathbb{R}^{n \times n}$ is the burnup matrix and $\boldsymbol{n} \in \mathbb{R}^{n}$ is the nuclide concentration vector. The diagonal elements $a_{ii} = -\lambda_i$ of the burnup matrix represent the total removal rates and the off-diagonal elements correspond to the production rates. The total number of nuclides commonly depends on the employed nuclear data library. In STREAM using ENDF/B-VII.1 data, the detailed burnup matrix contains all the data regarding transitional paths and branching ratios for 3837 nuclides and 43416 transitions. When constructing the burnup matrix, the nuclides can be indexed in an ascending order with respect to their ZAI index, defined as ZAI = 10000Z + 10A + I, where Z is the atomic number, A is the mass number of the nuclide and I is the isomeric state number. With this order, the non-zero elements are concentrated around the diagonal, except for the fission product production rates located in the upper triangular part of the matrix. The pattern of the burnup matrix for the detailed burnup chain of STREAM is used as data source (branching ratio and chain transmutation information) to generate a simplified depletion chain.

In order to quantify and identify vital nuclides for a simplified burn up chain, the importance of the nuclide number densities (NND) of all nuclides during depletion calculation is evaluated by the adjoint NND (ANND) derived from the depletion perturbation theory (DPT) for burnup-related nuclear properties [5] to identify important fission products. A brief description of the DPT application for defining nuclides of interest for burnup chain simplification are given in the following section.

2.1.1 Derivation of the contribution factors

The depletion calculation in the interval of $[t_i, t_{i+1}]$ is taken into consideration. The purpose of this section is to evaluate the importance of NND of an arbitrary nuclide k at t_i , $n_k(t_i)$, to another nuclide j at t_{i+1} , $n_j(t_{i+1})$. While the traditional approach requires many calculations repeatedly, this process can be simplified by applying DPT as follows.

The burnup Eq. (2) for $[t_i, t_{i+1}]$ time interval can be written as:

$$\frac{\partial \boldsymbol{n}(t)}{\partial t} = \boldsymbol{A_i}(t)\, \boldsymbol{n}(t), \qquad t_i \leq t \leq t_{i+1}, \tag{3}$$

where $\boldsymbol{A_i}$ is the corresponding burnup matrix at t_i, and $\boldsymbol{n}(t_i)$ is given in advance. Let us assume a perturbation $\delta n(t_i)$ is added into the system. The perturbed burnup equation is given as:

$$\frac{\partial \boldsymbol{n}'(t)}{\partial t} = \boldsymbol{A}_i'(t)\boldsymbol{n}'(t), \quad t_i \le t \le t_{i+1}, \tag{4}$$

with $\boldsymbol{n}'(t) = \boldsymbol{n}(t) + \delta\boldsymbol{n}(t)$ and $\boldsymbol{A}_i'(t) = \boldsymbol{A}_i(t) + \delta\boldsymbol{A}_i(t)$. The term $\delta\boldsymbol{A}_i(t)$ describes the corresponding perturbation of the burn up matrix due to the change in NND $\delta\boldsymbol{n}(t)$ at t_i. If the higher order is neglected, Eq. (4) can be expressed as:

$$\frac{\partial \delta\boldsymbol{n}(t)}{\partial t} = \boldsymbol{A_i}(t)\delta\boldsymbol{n}(t) + \delta\boldsymbol{A_i}(t)\boldsymbol{n}(t). \tag{5}$$

Multiplying a vector $\boldsymbol{w^T}$ (superscript "T" represents for the transpose operator) to Eq. (5) then integrating over $[t_i, t_{i+1}]$ yields:

$$\begin{aligned}\boldsymbol{w}^T(t)\delta\boldsymbol{n}(t)|_{t_i}^{t_{i+1}} &= \int_{t_i}^{t_{i+1}} \delta\boldsymbol{n}^T(t)\left(\boldsymbol{A_i^T}(t)\boldsymbol{w}(t) + \frac{\partial \boldsymbol{w}(t)}{\partial t}\right)dt \\ &+ \int_{t_i}^{t_{i+1}} \boldsymbol{w}^T(t)\delta\boldsymbol{A_i}(t)\boldsymbol{n}(t)dt\,. \end{aligned} \tag{6}$$

If one defines the vector $\boldsymbol{w}$ as the solution of Eq. (7) with the condition $\boldsymbol{w}(t_{i+1}) = \boldsymbol{e}_j$:

$$\frac{\partial \boldsymbol{w}(t)}{\partial t} = -\boldsymbol{A_i^T}(t)\boldsymbol{w}(t), \qquad t_i \le t \le t_{i+1}, \tag{7}$$

where $\boldsymbol{e}_j$ is a unit column-vector with all zero elements except for the element at the index *j* is equal to one. The matrix $\boldsymbol{A}_i^T$ is the adjoint matrix of the burnup matrix $\boldsymbol{A_i}$ thus vector $\boldsymbol{w}(t)$ can be considered as a "adjoint nuclide number density" vector. Using this definition, Eq. (6) can be rewritten as in Eq. (8):

$$\delta n_j(t_{i+1}) = \boldsymbol{w}^T(t_i)\delta\boldsymbol{n}(t_i) + \textstyle\int_{t_i}^{t_{i+1}} \boldsymbol{w}^T(t)\delta\boldsymbol{A_i}(t)\boldsymbol{n}(t)dt, \tag{8}$$

where

- $\boldsymbol{w}^T(t_i)\,\delta\boldsymbol{n}(t_i)$ is related to the direct effect of the change in number density,
- $\int \boldsymbol{w}^T\delta\boldsymbol{A}^i\boldsymbol{n}\,dt$ is related to indirect effects including neutron flux-spatial distribution effect and reactor power normalization effect, whose detailed derivation can be found in [1] and is given as:

$$\begin{aligned}\int_{t_i}^{t_{i+1}} \boldsymbol{w}^T(t)\delta\boldsymbol{A_i}(t)\boldsymbol{n}(t)dt &= -\overbrace{\langle \delta B^i \Gamma^{i^*}\phi^i\rangle}^{\text{Neutron flux-spatial distribution effect}} \\ &\quad -\underbrace{\frac{P^{i^*}}{V_f}\sum_j \kappa_j \delta n_j(t_i)\langle\sigma_{f,j}^i\phi^i\rangle}_{\text{Reactor power normalization effect}}, \end{aligned} \tag{9}$$

where

- δB^i is the perturbated term for the operator $\boldsymbol{B^i} = \boldsymbol{M^i} - \frac{1}{k_{eff}^i}\boldsymbol{F}^i$ with $\boldsymbol{M^i}$ and $\boldsymbol{F}^i$ denote neutron destruction and generation from fission operators in $[t_i, t_{i+1}]$,
- Γ^{i^*} is the generalized adjoint neutron flux,
- $\boldsymbol{\phi}^i$ is the neutron flux,
- P^{i^*} is so-called the adjoint operator to the reactor power,
- V_f is the fuel region volume,
- κ_j and $\sigma_{f,j}^i$ are the emitted energy by one fission reaction and a microscopic fission cross section of nuclides j respectively,
- and brackets denote the integration over all the energy groups and whole volume of the system.

In this study, no coupling effect was assumed to exist between the neutron field and the nuclide field. In other words, the second term in the right-hand side (RHS) of Eq. (8) can be neglected:

$$\delta n_j(t_{i+1}) = \boldsymbol{w}^T(t_i)\delta\boldsymbol{n}(t_i) = \sum_k^N w_k^T(t_i)\,\delta n_k(t_i)\,. \tag{10}$$

By dividing both sides of Eq. (10), the relative changes in nuclide j can be observed:

$$\frac{\delta n_j(t_{i+1})}{n_j(t_{i+1})} = \frac{\sum_k^N w_k^T(t_i)\,\delta n_k(t_i)}{n_j(t_{i+1})}. \tag{11}$$

In order to evaluate the importance of a nuclide k to other nuclide j, let us quantify the effect of the removal of nuclide k at t_i to nuclide j at t_{i+1}, *i.e.*, the term $\delta n_k(t_i)$ in Eq. (11) is replaced with $n_k(t_i)$. Thus, the contribution factor (CF_k) can be introduced as the importance of nuclide k at $t = t_i$ to a particular depletion nuclear property, which is the number density of nuclide j at the end of depletion calculation ($t = t_{EOC}$) in this study, and is defined as follows:

$$CF_k(t_i) = \frac{w_k^T(t_i)\,n_k(t_i)}{n_j(t_{EOC})} \quad , \quad w_k^T(t_{EOC}) = e_j^T\,. \tag{12}$$

In this study, nuclides j are nuclides whose number density should be reproduced using optimized chain and determined beforehand based on their importance in a real core analysis and reactivity calculation scenario. The adjoint nuclide number densities and depletion dependence of contribution factors of ^{155}Eu in a UO_2 fuel pin cell at 60 MWd/kg burnup is illustrated in Figs. 1 and 2. This nuclide is the precursor of ^{155}Gd, a crucial nuclide in reactor depletion calculations because of its large capture cross-sections. In order to achieve the ANND as the solution for Eq. (7), backward calculation is conducted with the final condition of $\boldsymbol{w}(t_{EOC}) = \boldsymbol{e}_j$, where *j* is ^{155}Eu. This means the ANND of ^{155}Eu is equal to one at the end of the depletion process.

As discussed above, CF can be interpreted as the importance of one nuclide NND on a burnup related characteristic, thus it is unitless. In Fig.1, one can understand that the contribution of nuclide ^{153}Gd to the production of nuclide ^{155}Eu is relatively low compared to ^{152}Eu and ^{153}Eu. Therefore, it can result in removal of ^{153}Gd in the depletion chain. By observing in Fig. 2, although the ANND of ^{152}Sm cannot be evaluated because of small quantity, its CF is not omittable, which means it is necessary to include ^{152}Sm in the generation chain of ^{155}Eu.

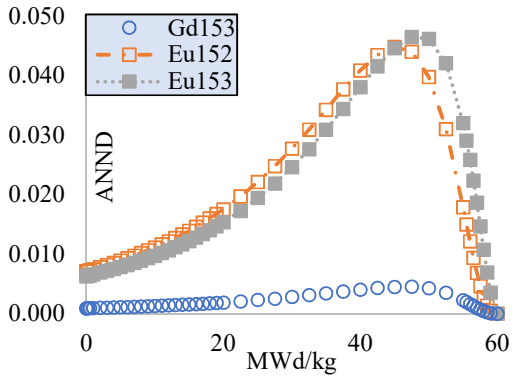

Figure 1. ANNDs of three isotopes to ^{155}Eu after depletion.

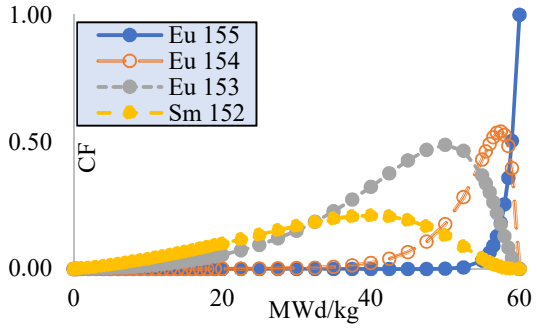

Figure 2. CFs of four isotopes to ^{155}Eu after depletion.

2.2 Identification of Important Nuclides for $\boldsymbol{k_{eff}}$ *and Reactor Power.*

The method to identify the nuclides of interest relies on the calculation of the contribution factors as presented in the previous section 2.1.1. The simplification procedure can be expressed as follows. The candidate nuclides for a simplified depletion chain are determined by conducting a detailed burnup chain calculation for Fuel Assemblies (FAs) that encompass common fuel types and burnable poisons (BPs) and marking the nuclides whose number densities are above a given threshold, *e.g.*, 0.01% of the total number density including all nuclides. In the first step, out of the 3837 nuclides present in the detailed depletion chain, only those nuclides with cross-sections available in the ENDF/B-VII.1 library are retained due to the fact that the current used cross section library is ENDF/B-VII.1. This is considered as a sub-step for the main simplification process. The corresponding number of selected nuclides amounts to 426. In the next step, the number densities of the 426 nuclides are evaluated with the detailed burnup chain at 80 MWd/kg. A number density threshold (10^{-10} % of total number nuclide densities) is then applied for accumulated ND of all nuclides at the end of depletion process and only 281 nuclides are selected out of the 426 nuclides. This sub-step is to identify nuclides whose ND need to be reproduced. Thirdly, the contribution factors of all nuclides to a specific number of target nuclides, which are selected based on their cruciality on reactor core calculation, are calculated and a set of fission products satisfying the initial limit is determined. These target nuclides are organized as follows: ^{83}Kr , $^{103; 105}$Rh, ^{109}Ag, ^{135}I, $^{131; 135}$Xe, $^{133; 134; 135; 137}$Cs, ^{140}Ba, ^{140}La, $^{143; 145}$Nd, $^{147; 148; 148m; 149}$Pm, $^{147; 149; 150; 151; 152}$Sm, $^{153; 154; 155; 156}$Eu, $^{152; 154; 155; 156; 157; 158; 160}$Gd, $^{234; 235; 236; 237; 238; 239}$U, $^{236; 237; 238; 239}$Np, $^{236; 238; 239; 240; 241; 242; 243}$Pu, $^{241; 242; 242m; 243; 244}$Am, $^{242; 243; 244; 245; 246; 247; 248; 249}$Cm, $^{249; 250}$Bk, $^{249; 250; 251; 252}$Cf. By defining a cut-off value (10^{-5}) for the obtained CF_k for all nuclides, a set of 102 important fission products is selected which must be included in the final simplified burn up chain to reproduce the number densities of the target nuclides. The final simplified burnup chain of 306 nuclides is constructed based on the combination of setting a threshold for NND at the end of depletion process and the CF of all nuclides for defined target nuclides. This extraction process is described in Fig. 3.

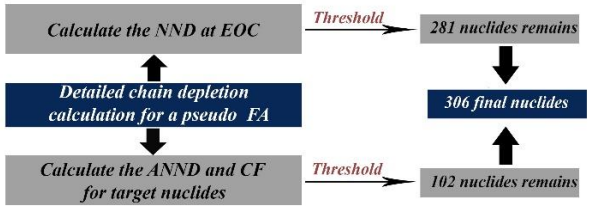

Figure 3. Simplified depletion chain extraction process

A simplified burnup chain consisting of the identified nuclides is then constructed. For the fission yield and the transitional fraction (TF), those values that reflect the probability that one given nuclide transmutes into another nuclide, are redistributed. A general principle for redistributing the fission yield is to add the independent fission yields of neglected FP precursors nuclides to the independent fission yields of the daughter nuclides which are candidate nuclides for the simplified chain. If a daughter nuclide B produced by a transmutation of an identified nuclide A is neglected in the simplified burnup chain and the daughter nuclide B is the precursor of a nuclide C with a specific transitional path, the nuclide C is then treated as a daughter nuclide of the nuclide A directly, yielded by the multiplication of transmutation

value of the nuclide A to nuclide B and nuclide B to C. Eventually, the number of transitions in the simplified burnup chain is condensed from 43416 to 7075.

2.3 Numerical Results

Numerical results are presented for the 2D depletion of a PYREX bearing FA whose geometry and fuel composition can be found in [4].

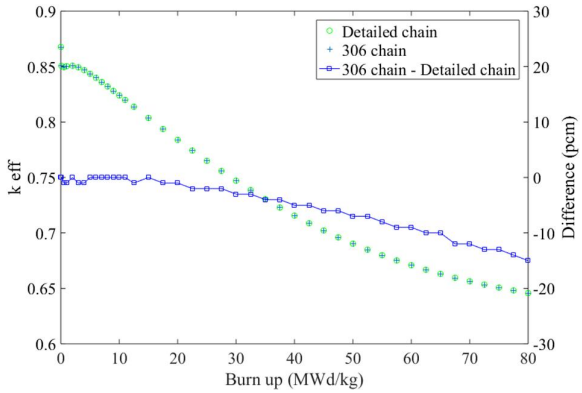

Figure 4. Neutron multiplication factor generated by STREAM with different depletion chains

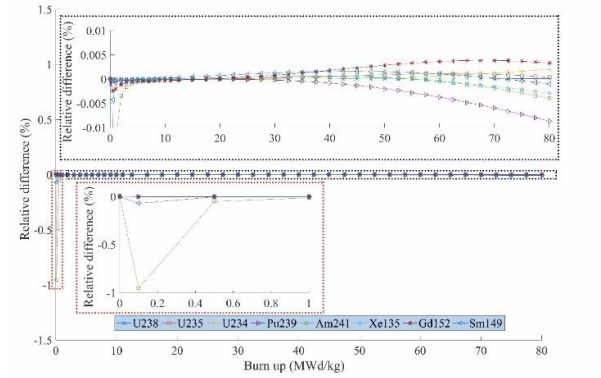

Figure 5. Number density comparison between 306 chains and detailed burnup chain for PYREX bearing FA case

Table I. Computational time for different burnup chains

	Detailed chain	306-nuclide chain
Total simulation time (s)	1831	562
Depletion time (s)	1432	32

As shown in Fig. 4, the maximum value of k_{eff} difference reaches 14 pcm at the end of cycle. Furthermore, it can be seen in Fig. 5 that the number density discrepancies for the nuclides are all smaller than 0.01% except for ^{149}Sm and ^{241}Am. The maximum disparity for these two nuclides is at the second burn up step amounts to 0.11% and 0.97%, respectively. The result in the computational time reduction is given in Table I.

3. Conclusions

A method of simplifying depletion chain was introduced in this research based on the DPT to determine important nuclides in reproduction of nuclide number densities. The simplified chain contains 306 nuclides with the adjusted fission yield and transitional fraction out of more than 3000 nuclides in ENDF/B-VII.1 decay library. By using this burnup chain model, the k_{eff} variation of a PYREX bearing FA depletion calculation agrees well with that of the detailed chain, with speed-up factors of 40 and 3 in depletion calculation and total simulation, respectively. The simplified burnup chain shows satisfactory performance in reproducing number densities throughout depletion, the maximum disparity of 0.97% for ^{241}Am. Due to the overestimation of strong neutron absorbing fission products (^{152}Gd, ^{149}Sm), the k_{eff} from the simplified chain is slightly lower by ~20 pcm than the one from the detailed chain at 80 MWd/kg. With its high order of fidelity, this method can be used to generate reduced-order depletion chains for further updating of STREAM data with a high degree of fidelity. Furthermore, it can be modified to suit desired performances such as source term calculation, decay heat and criticality.

Acknowledgement

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REFERENCES

[1] G. Chiba, M. Tsuji, T. Narabayashi, Y. Ohoka, and T. Ushio, "Important fission product nuclides identification method for simplified burnup chain construction," *Journal of Nuclear Science and Technology,* vol. 52, no. 7-8, pp. 953-960, 2015.

[2] S. Choi, C. Lee, and D. Lee, "Resonance treatment using pin-based pointwise energy slowing-down method," *Journal of Computational Physics,* vol. 330, pp. 134-155, 2017.

[3] M. Pusa, "Rational Approximations to the Matrix Exponential in Burnup Calculations," *Nuclear Science and Engineering,* vol. 169, no. 2, pp. 155-167, 2011.

[4] J. Park, A. Khassenov, W. Kim, S. Choi, J. Yu, D. Lee*, "Comparative Analysis of VERA Depletion Benchmark through Consistent Code-to-Code Comparison," *Annals of Nuclear Energy*, Under review (2018)

[5] M. L. Williams, "Development of Depletion Perturbation Theory for Coupled Neutron/Nuclide Fields," *Nuclear Science and Engineering,* vol. 70, no. 1, pp. 20-36, 1979.