EXECUTIVE SUMMARY OF THE THESIS

Development of a data-driven surrogate model for helium production in fast reactor Am-bearing fuels

TESI MAGISTRALE IN NUCLEAR ENGINEERING – INGEGNERIA NUCLEARE

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1. Introduction

Recent developments in research regarding Generation IV fast reactors with Americium-bearing mixed oxide (MOX) fuel and their related fuel cycle technologies dictate a need in detailed understanding of the fuel behavior under irradiation and in storage conditions. One important aspect of the behavior of Am-bearing MOX fuel is the helium production in the fuel itself and the eventual helium release into the fuel free volume, causing an increase of the fuel pin internal pressure. The standard tools applied to estimate helium production in the fuel are fuel depletion codes, which enable a reliable estimation of the evolution of the actinides content contained within the fuel. Monte Carlo depletion codes can track the rates of change of the isotope inventory of the nuclear fuel with high accuracy and fidelity by considering the sequential steady state neutronics solution at each burnup step. In this approach, the solution of the neutron transport problem is the main process which requires high computational time.

Fuel performance codes, dedicated to thermo-mechanical analysis of fuel pins, typically employ less computationally expensive approaches to estimate helium production rates. These codes use either dedicated burnup modules, directly solving the Bateman equations with simplified cross sections, or correlations directly calculating the helium production rate from a set of input parameters. Each of these two approaches has its respective advantages and disadvantages: (1) burnup modules have greater accuracy at the expense of an increased computational cost, whereas (2) stand-alone correlations are less accurate and specific to certain fuel/reactor combinations, but fast running and inherently numerically stable. Burnup modules used in fuel performance codes hence represent a midpoint between neutronic depletion codes and correlations, in terms of required computational effort.

In view of their limited computational time and wide range of applicability (given a fuel/reactor combination paired with a verification and validation range) it is possible to apply burnup modules to populate synthetic datasets with the purpose of constructing data-driven surrogate models to then be applied directly in fuel

performance codes. This methodology represents a typical application of surrogate models which is widely used in other sectors and is herein proposed for fuel performance.

In this work the burnup module of the SCIANTIX code [1,2] is extended in tandem with the SERPENT Monte Carlo code [3] to generate macroscopic cross-section lookup tables for two relevant fuel/reactor combinations: a sodium cooled fast reactor (SFR) and a lead bismuth eutectic-cooled fast reactor (LBE-FR). The burnup module of SCIANTIX is extended to a finer grid of burnup and plutonium enrichment steps, and up to a plutonium content up to 51%, which is of interest for several Generation IV reactor core concepts [5]. The extended burnup module is then used to generate synthetic datasets covering a wide range of initial composition of Am-bearing fuel and irradiation conditions, and a helium production rate surrogate model is developed based on non-linear, multivariate regression performed on the datasets.

The proposed model is intended to be used as a stable fast-running tool implemented in fuel performance codes with focus on reduced computational loads with respect to the burnup module itself.

2. Extension of the SCIANTIX burnup module

The methodology used in this work to extend the SCIANTIX burnup module follows a standardized development procedure, regarding a user-specified fuel/reactor combination, and can be summarized as follows:

- A set of SERPENT simulations is performed for an initial fuel composition vector, corresponding to the fuel/reactor combination under evaluation.

- For each initial composition step e_i, and each burnup step bu_i, the respective cross-section values are extracted, evaluated by the reaction rate integrals in the SERPENT simulation.

- An e_i by bu_i matrix is created containing the microscopic cross-section values, with each row representing the initial enrichment steps and each column the burnup steps for each nuclide and each reaction type.

- The matrices are implemented in the SCIANTIX burnup module as lookup tables

corresponding to the relevant fuel/reactor combination.

This procedure occurs once and for all, and the produced lookup tables are incorporated in the SCIANTIX burnup module. The user can then dictate the choice of the respective tables corresponding to the fuel/reactor combination to simulate.

Two fast reactor cases have been simulated, a Sodium cooled Fast Reactor (SFR) and the Liquid Bismuth Eutectic cooled Fast Reactor (LBE-FR). The simulation domain consists of a single fuel pin with cylindrical geometry, composed of the uniform fuel pellet, the fuel-cladding gap, and the cladding, encompassed by the respective coolant material. The depletion calculation was performed in 81 equally distanced burnup steps of 2.5 GWd/t_{HM} up to 200 GWd/t_{HM} and 31 plutonium enrichment (Pu/HM) steps starting from 20 up to 51%.

The SCIANTIX predictive capabilities are verified against the high-fidelity SERPENT results. The verification is performed for each of the 22 nuclides, and for the two fuel/reactor combination cases. A schematic representation of the verification grid used is shown in Fig. 1: such grid has the purpose of testing the performance of SCIANTIX burnup module in correspondence of the burnup/enrichment points used to construct the lookup table and far away from these points as well.

Due to the large amount of data generated in the verification, only the results of one enrichment level (31%) are herein presented. A comparison of the root mean square error, RMSE (averaged in burnup) is depicted in Fig. 2a for the SFR case and in Fig. 2b for the LBE-FR case. The average RMSE for all plutonium enrichment steps is shown in Fig. 3, where it is evident that the extended burnup module has a consistently better predictive performance compared to the previous version, albeit with a small margin for enrichments up to 41% where the margin increases significantly.
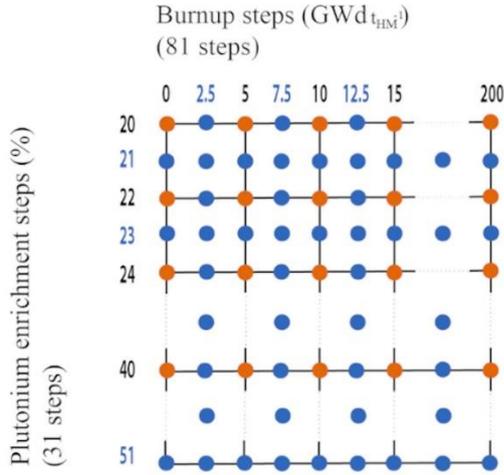
Figure 1: Burnup/enrichment grid used for verification of the SCIANTIX burnup module against SERPENT (blue, this work; orange, Cechet et al. 2021 [2], i.e., previous version of the module itself).
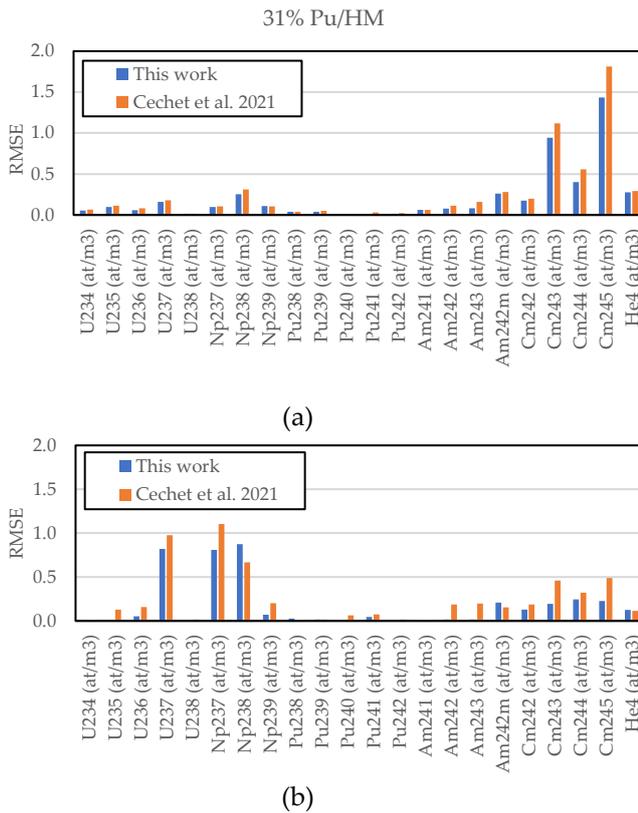

Figure 2: Comparison of the RMSE (averaged in burnup) between SCIANTIX burnup module and SERPENT reference results for the SFR case (a) and for the LBE-FR case (b), at 31% initial plutonium enrichment.
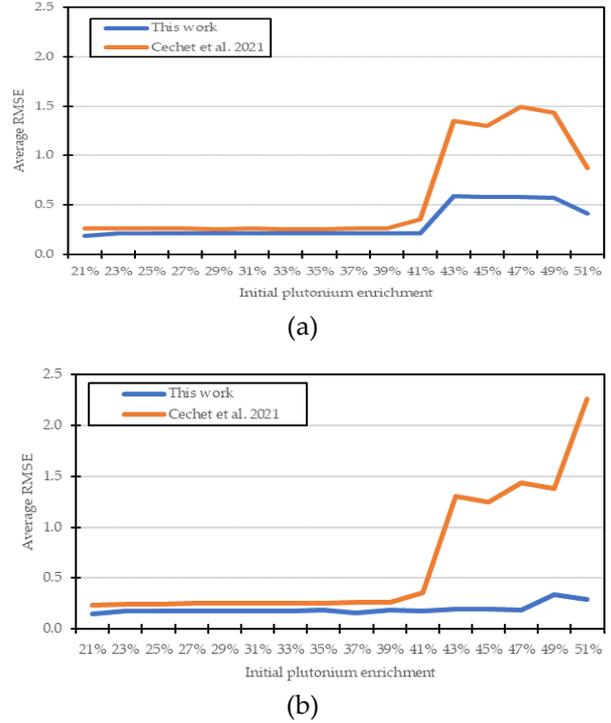

Figure 3: Comparison of the RMSE (averaged in burnup and on all nuclides) between SCIANTIX burnup module and SERPENT reference results for the SFR case (a) and for the LBE-FR case (b), as function of initial plutonium enrichment.

3. Development of the surrogate model for the helium production rate

The extended SCIANTIX burnup module is used to populate several synthetic datasets with calculated helium concentration values as a function of the input variable matrix. The output vector $Y_{N,n}$ (i.e., the helium concentration) is calculated by the code as a function of a selection of n values of N input variables constituting the input matrix $I_{N,n}$, sampled from predefined ranges. By examining the probability distribution of the output $Y_{N,n}(I_{N,n})$ the desired information of the dependence of helium concentration on each input variable can be obtained. The number of input variables N and number of values sampled n dictate the computational time for the development of the synthetic datasets (e.g., for the generation of a 10'000 values dataset, the computational time required is around three hours). To ensure that each of the input variables $I_{i,n}$ has all portions of its distribution represented by input values, the Latin hypercube sampling (LHS)

technique is adopted: the range of each $I_{i,n}$ is divided into M strata of equal marginal probability $1/M$ and sampled only once from each stratum [4]. Using this technique, the advantage of limiting the amount of input values per each input variable and represent them in a fully stratified manner is evident, without a biased projection of the values that end up being important in the final statistical model. For example, Fig. 4 reports the Pu/HM input variable distribution for 881 data points, highlighting the dense coverage of values over the sampling space.
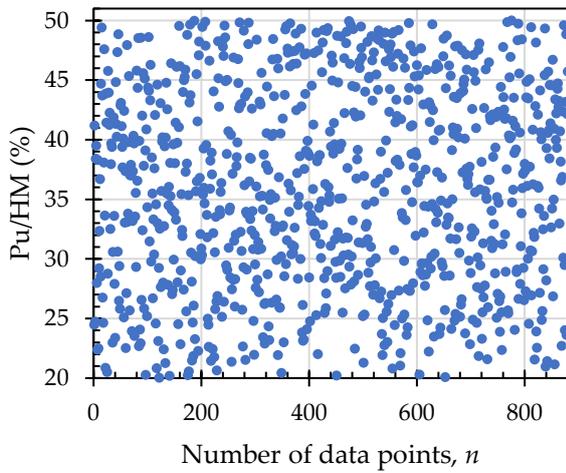

Figure 4: Scatterplot produced using the Latin hypercube sampling technique for 881 data points of Pu/HM values as an example of an input variable coverage density.

The input variables considered in this analysis are the fuel initial fuel composition in terms of all constituent nuclides expressed as the ratio of each nuclide per heavy metal ($^iX/HM$), the initial oxygen to metal ratio of the fuel O/M, the initial fuel density ρ_{fuel}, the fuel temperature T_{fuel}, the irradiation time t_{irr} and the fission rate density $\dot{F}$. The limits are chosen in-line with the predictive capabilities of the extended SCIANTIX burnup module as well as for the fuel specifications of a selection of Generation IV reactors core concepts [5]. The implementation of the LHS technique is performed in MathWorks MATLAB coupled with the SCIANTIX code for the helium concentration calculation.

For the development of the two surrogate models, two training datasets were generated using the LHS method on the extended SCIANTIX burnup module, one for the SFR case of n = 8′538 data

points, and one for the LBE-FR case of n = 8′579 data points.

Apart from the two training datasets, five validation datasets were generated for the SFR case, and four validation datasets were generated for the LBE-FR case, with different sizes and variable limits for the purpose of assessing the predictive capability of the proposed models.

The training dataset is composed of the multidimensional input matrix $I_{i,n}$ representing the contribution of each input variable, and an output vector $Y_{N,n}$ (helium concentration). Subsequently, multivariate non-linear regression and statistical analysis was performed with the JMP statistical package to formulate surrogate models for the two reactor cases of interest, SFR and LBE-FR. The method of stepwise regression employed is that of forward regression. It is performed by considering the p-values of each regressor with respect to the output values in an iterative process, in which each regressor is added to the formulation considering its respective p-value being below the 0.05 threshold. Within this approach, collinearity between input variables is avoided by including only one of the inter-correlated variables in the final model (e.g., irradiation time and fission rate density are heavily correlated with burnup thus the latter is excluded from the final model, given also that the former two variables can be used as input in the SCIANTIX burnup module). At each iteration step, after a variable has been added to the model, the p-values changes accordingly for each variable. Two surrogate models were selected out of the candidate models examined ($> 10^7$), one for the SFR and the LBE-FR case, which are in the form:

$$
\begin{aligned}
\log[^4He] = {}& A \cdot \log[t_{irr}] \\
& + B \cdot \dot{F} \\
& + (C \cdot [Pu] + D \cdot [Pu]^2 + \cdot [Pu]^3) \\
& + F \cdot \rho_{fuel} \\
& + G \cdot [^{241}Am] + H \cdot [^{242}Am] \\
& + I \cdot [^{242}Cm] \\
& + S
\end{aligned}
\tag{1}
$$

In addition to the expression depicted in Eq. 1, an ordinary differential equation for the calculation of the helium production rate $d[^4He]/dt$ is proposed:

$$\frac{d[^4\text{He}]}{dt_{\text{irr}}} = A \cdot t_{\text{irr}}^{A-1} 10^P,$$

$$
\begin{aligned}
P = [&B \cdot \dot{F} \\
&+ (C \cdot [\text{Pu}] + D \cdot [\text{Pu}]^2 + E \cdot [\text{Pu}]^3) \\
&+ F \cdot \rho_{\text{fuel}} \\
&+ G \cdot [^{241}\text{Am}] + H \cdot [^{242}\text{Am}] \\
&+ I \cdot [^{242}\text{Cm}] \\
&+ S]
\end{aligned}
\qquad (2)
$$

where A, B, C, D, E, F, G, H, I and S are coefficients, shown in Table 1 for the two fuel/reactor combination cases along with the respective standard error.

Table 1: Values of the coefficients in the helium production surrogate model for the two fuel/reactor combinations, with the corresponding standard error.

Coefficient	Variable	SFR	Standard error	LBE-FR	Standard error
A	$\log[t_{\text{irr}}]$	0.66984	0.00383517	0.6771483301	0.00375305
B	$\dot{F}$	2.58123×10^{-21}	6.0452×10^{-23}	2.6631×10^{-21}	5.8679×10^{-23}
C	$[\text{Pu}]$	-0.90146	0.01363592	-0.92536684	0.01313788
D	$[\text{Pu}]^2$	0.0237788448	0.00039144	0.0243855707	0.0003769
E	$[\text{Pu}]^3$	-0.000204616	3.64727×10^{-6}	-0.000209562	3.50935×10^{-6}
F	ρ_{fuel}	0.0000311663	3.48953×10^{-6}	0.000037483	3.38246×10^{-6}
G	$[^{241}\text{Am}]$	0.0680062668	0.00398778	0.0736685629	0.00378016
H	$[^{242}\text{Am}]$	0.1686687076	0.00397259	0.1611164143	0.00386775
I	$[^{242}\text{Cm}]$	0.1849589901	0.00305256	0.1793083124	0.00298431
S	-	34.06157919	0.15778665	34.280047124	0.15130481

Each surrogate model comprises seven dependencies: (1) the irradiation time t_{irr} (h), (2) the fission rate density $\dot{F}$ (fiss m^{-3} s^{-1}), (3) the total plutonium content Pu/HM (at%), (4) the fuel density ρ_{fuel} (kg m^{-3}), (5) the initial americium-241 concentration ^{241}Am/HM (at%), (6) the initial americium-242 concentration ^{242}Am/HM (at%), and (7) the initial curium-242 concentration ^{242}Cm/HM (at%).

The quality of the final fit and the accuracy of the models were quantified in terms of the RMSE, the coefficient of determination R^2 and the adjusted coefficient of determination R^2_{adj}. The metrics for each dataset are shown in Table 2.

The helium production surrogate model performance for the SFR case is shown in Fig. 5 for the training and one validation dataset (b). The model exhibits the best performance for the training dataset (a) as is to be expected. For the dataset (b) to (e) the correlation exhibits relatively good predictive capability, particularly considering the wide applicability range in terms of irradiation conditions, with most points being concentrated around the 45° diagonal, and with RMSE ranging from around 0.20 to 0.28. The R^2 and

R^2_{adj} statistic metrics range from around 0.60 to 0.71, indicating a good fit of the correlation to the values produced from the extended SCIANTIX burnup module. Figure 6 reports the surrogate model performance for the LBE-FR case, where the proposed model exhibits a better fit with the generated datasets compared to the SFR case.

The variance associated with the predicted values occurs due to the high number of input variable combinations which lead to different burnup values for each initial fuel composition. For higher number of data points in each dataset, better agreement is observed as a denser matrix of input values is generated.
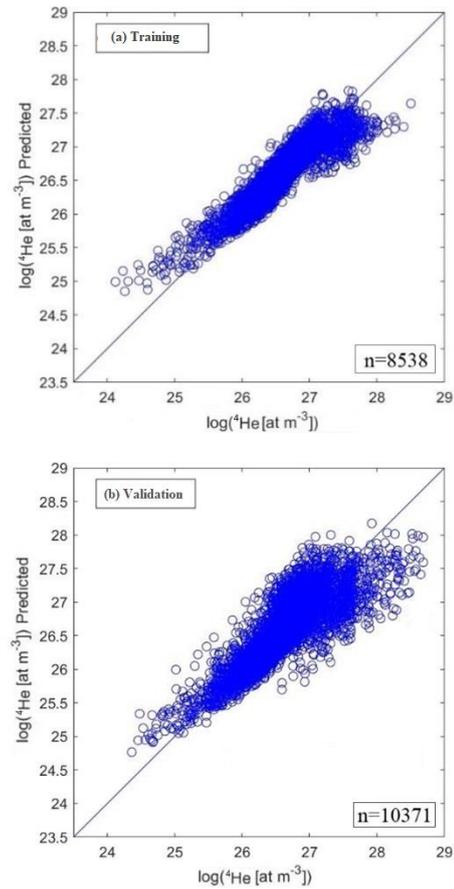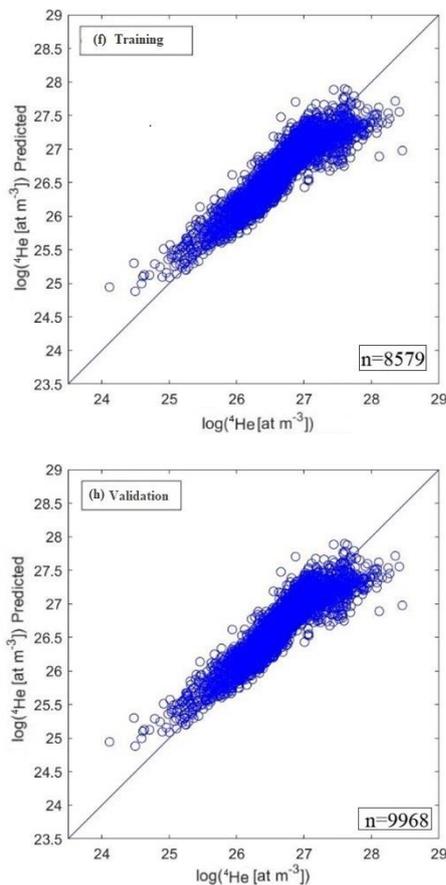

Figure 5: Evaluation of the proposed helium production surrogate model against the values calculated by SCIANTIX for two datasets in the SFR case, where (a) is the training dataset and (b) is one of the validation datasets.

Table 2: Statistic validation metrics of the proposed helium production surrogate model each dataset on the SFR and LBE-FR cases.

SFR case

Dataset	(a) Training	(b)	(c)	(d)	(e)
n	8'538	10'371	99'445	86'332	9'97
RMSE	0.1650	0.2759	0.1986	0.2059	0.19
R^2	0.8123	0.6019	0.7106	0.6889	0.70
R^2_{adj}	0.8122	0.6017	0.7104	0.6889	0.70

LBE-FR case

Dataset	(f) Training	(g)	(h)	(i)
n	8'579	84'648	9'968	313
RMSE	0.1486	0.1504	0.1818	0.3094
R^2	0.8441	0.8455	0.7542	0.5136
R^2_{adj}	0.8440	0.8455	0.7540	0.5094

Figure 6: Evaluation of the proposed helium production surrogate model against the values calculated by SCIANTIX for two datasets in the LBE-FR case, where (f) is the training dataset and (h) is one of the validation datasets.

4.   Conclusions

In this work the SCIANTIX burnup module has been extended and verified for two fuel/reactor combinations, sodium-cooled fast reactor and lead-bismuth eutectic cooled fast reactor. The depletion calculations made by the burnup module has been compared to the high-fidelity results from the SERPENT depletion code, in terms of the root-mean square error metric. The extended burnup module is found to perform well in its whole definition range, i.e., burnup from 0 to 200 GWd/t_{HM} and Pu/HM from 20 to 51%.

Using the extended burnup module as a surrogate model, datasets were generated using the Latin hypercube sampling technique with several input vectors covering a wide span of initial fuel compositions and properties correlated to the production of helium in the fuel. Non-linear multivariate regression was performed to obtain two helium surrogate models corresponding to the sodium and lead-bismuth eutectic fast reactor cases, respectively. The direct use of these models allows to bypass entirely the need of a burnup module for the assessment of the helium production rate in the fuel, thus allowing for a very fast computational time, which is of potential interest for fuel performance codes targeting efficient thermo-mechanic calculations.

The proposed surrogate models for helium production have been verified against secondary generated datasets and found to be reliable in the considered ranges of input variables.

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5.   Acknowledgements