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ARTICLE

Modeling of Reprocessed Uranium Enrichment for Nuclear Scenarios Simulations

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Reprocessed uranium recycling in PWR is a key component of the uranium and plutonium multi-recycling strategies in PWR foreseen in the French fleet by the year 2050. Reprocessed uranium (RepU) recovered at UOX spent fuel reprocessing usually contains a significant fraction of ^{235}U. However, RepU enrichment process also results in an increase of the concentration of other Uranium isotopes, such as ^{234}U and ^{236}U. To acquire insights on RepU recycling strategies in PWR and their potential deployment in the French nuclear fleet, nuclear scenario simulations are performed. This paper focuses on the development of a RepU enrichment model for the nuclear scenario simulation code CLASS. This model is based on iterative bisection method calculating the ^{235}U enrichment in enriched reprocessed uranium (ERU) fresh fuel required to reach a discharge target burn-up in PWR depending on the RepU isotopic composition. This algorithm relies on an artificial neural network estimator of the maximum achievable burn-up depending on ERU fresh fuel composition, based on a PWR ERU depletion calculation database. This algorithm is then applied to a sample of RepU isotopic compositions from spent cooled PWR UOX fuels, highlighting the impact of ^{236}U fraction in RepU on the required over-enrichment in ERU fresh fuel.

KEYWORDS: nuclear fuel cycle simulation, fuel loading model, PWR, uranium recycling, enriched reprocessed uranium

I. Introduction

The current French nuclear fleet is based on Pressurized Water Reactors (PWR) mostly loaded with UOX and MOX fuels. UOX spent fuels reprocessing at La Hague plant leads to the extraction of plutonium (used to produce MOX fuel), minor actinides and fission products (vitrified in waste canisters) and reprocessed uranium. Starting in the 1990's, reprocessed uranium (RepU) has been enriched to produce Enriched Reprocessed Uranium (ERU) fuels loaded in several units of PWR 900 MWe. After being stopped for about a decade, the manufacturing and loading of ERU fuel managements in French PWR has resumed in early 2024 and could be enhanced in the next decades. Indeed, recycling RepU in ERU fuel managements is at the heart of the uranium and plutonium multi-recycling strategies in EPR foreseen in the French fleet by the year 2050, as it could allow to stabilize RepU inventory and to reduce natural uranium consumption.[1)]

To evaluate these nuclear material management strategies, nuclear scenarios focusing on the evolution of the French reactor fleet and fuel cycle are performed.[2)] These studies rely on a nuclear scenario simulation tool, such as CLASS (Core Library for Advanced Scenario simulation)[3)] developed at CNRS/IN2P3 since 2011. CLASS models various reactors, fuels, and cycle units, calculating isotopic inventories and material flows over time. It relies on reactor meta-models (fuel loading and irradiation models) developed from a transport depletion calculation databank, performed upstream of the dynamic nuclear fuel cycle simulation.

This paper focuses on the development of a RepU enrichment model for CLASS to perform Uranium recycling strategy simulation. This model aims to calculate the ERU fresh fuel composition, especially ^{235}U enrichment, to achieve a PWR target discharge burn-up. Model building relies on the creation of a PWR ERU calculation database used to train an artificial neural network (ANN) estimator, forming the basis of the enrichment algorithm.

II. PWR ERU Assembly Calculation Databank

1. Modeling Reprocessed Uranium Recycling

RepU recovered at UOX spent fuel reprocessing usually contains an isotopic fraction of ^{235}U (around 1%) higher than in natural Uranium (0.7%).[4)] However, RepU enrichment process will also result in an increase in the concentration of other Uranium isotopes, such as ^{234}U and ^{236}U, produced during UOX fuel irradiation in PWR and cooling.

As ^{234}U and ^{236}U are neutron absorbing isotopes, their presence implies an increase of the ^{235}U enrichment in ERU fuel, compared to equivalent UOX fresh fuel, to reach the same PWR target discharge burn-up.[5)] However, in France, Uranium enrichment is limited to 5% of ^{235}U in the current Orano enrichment plant Georges Besse II.[6)] The RepU enrichment operations will also result in an increase of the concentration of these uranium isotopes in the deriving ERU fresh fuel.[4)]

The available RepU isotopic composition to produce ERU fresh fuel depends on the features of the cooled spent UOX fuel at reprocessing. Thus, as ^{236}U is mainly produced by (n, γ) reaction on ^{235}U, its content in RepU depends on the initial

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^{235}U enrichment in UOX fresh fuel and the PWR discharge burn-up. ^{234}U fraction in UOX fresh fuel derives from the enrichment process of natural uranium and therefore depends on ^{235}U enrichment in UOX fresh fuel. As ^{234}U is also produced by radioactive decay of ^{238}Pu (half-life of 87.7 yr), its content in RepU depends on UOX spent fuel cooling time.

Modeling RepU enrichment in CLASS therefore requires a model depending on RepU isotopic composition (deriving from UOX fuel properties) and on a target burn-up for PWR 100% ERU fuel management.

^{232}U isotope, although its concentration in RepU is very low (~ppb), is also a subject of interest for the fuel cycle, as it has a high radiological impact on fuel cycle processes. Its content in RepU may lead to the requirement of additional radiological protections for RepU and ERU fresh fuel management.[7] The impact of ^{232}U fraction in RepU is not treated in this work, as it specifically focuses on the impact of RepU isotopic composition on ^{235}U enrichment in ERU fuel. However, it will be the subject of future work.

2. PWR Assembly Calculation

The modeling of PWR loaded with ERU fuel in this paper is based on 300 infinite assembly calculations with reflective boundary conditions.[8] Each depletion calculation is a standard 17x17 PWR assembly loaded with ERU fuel and irradiated until 1500 EFPD (around 55 GWd/t), with a zero-boron concentration. The specific thermal power is set to 36 W/g. These simulations are performed using the SMURE software[9] based on the Monte-Carlo neutron transport code SERPENT 2.1.[10]

Each simulation differs from another by its ERU fresh fuel composition (atomic fractions of ^{234}U, ^{235}U, ^{236}U and ^{238}U in the ERU fuel). In a first approach, a wide ERU fresh fuel isotopic hyperspace, presented in **Table 1**, has been estimated based on guidance from the literature.[4,5,7] 300 ERU fresh fuel compositions have been randomly generated in this hyperspace using the LHS method. ^{238}U is used as a buffer to reach 100% in the uranium vector definition.

Table 1 Enriched reprocessed Uranium composition range (%at) - ^{238}U is used as a buffer to reach 100% in the uranium vector definition

	^{234}U	^{235}U	^{236}U
Min	0 %	3.5 %	0 %
Max	0.5 %	5 %	3.5 %

3. Maximum Burn-up Calculation

For each PWR ERU assembly calculation, the evolution of the infinite multiplication factor as a function of the time is extracted. From the evolution of $k_\infty(t)$, a maximum reachable burn-up (BU_{max}) calculation is performed based on the method from[11]. This approach assumes a linear decrease of the infinite multiplication factor with respect to burnup. The fuel fractionation in the 100% ERU fuel management is set to 3 and a threshold k_∞ of 1.03 has been selected. PWR 100% ERU fuel management modeling will be refined in a future work focusing on calculations at full core scale.

The BU_{max} value is then recorded for each ERU fresh fuel composition. The resulting BU_{max} range for these ERU fresh fuel compositions is [35 – 65 GWd/t]. The distribution of the calculated BU_{max} values as a function of ^{234}U, ^{235}U and ^{236}U fractions in ERU fresh fuel is presented on **Fig. 1**. As expected, the increase of the maximum reachable burnup is directly related to the increase of ^{235}U enrichment in the ERU fresh fuel. The ^{236}U fraction plays also a prominent role on the BU_{max} value. The increase of ^{236}U fraction in ERU fresh fuel limits the BU_{max} at for a given value of ^{235}U enrichment, and leads to an over-enrichment compared to equivalent UOX fuel. However, the impact of the fraction of ^{234}U is not significant, especially since the fraction of this isotope is low (one order of magnitude lower than ^{235}U fraction).

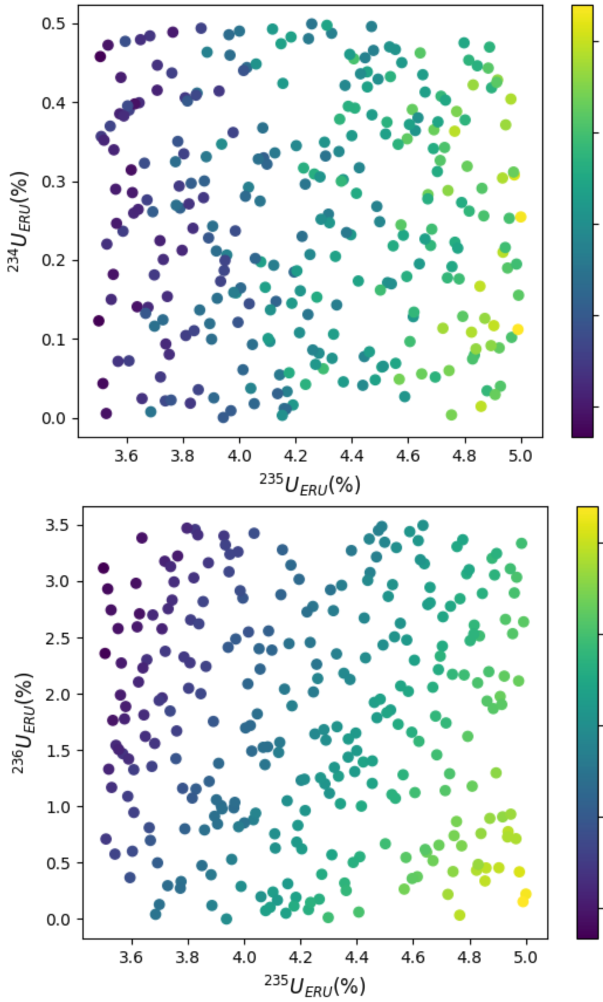

Fig. 1 Distribution of BU_{max} depending on ^{234}U, ^{235}U and ^{236}U contents in sampled ERU fresh fuel isotopic compositions

III. RepU Enrichment Model

1. Artificial Neural Network Building

Calculating the BU_{max} of a fresh fuel composition is at the basis of the fuel loading models developed for CLASS and is usually performed using an Artificial Neural Network.[8]

From the PWR 100% ERU calculations databank, an ANN calculating the BU_{max} from the ERU fresh fuel composition is trained and tested. It is composed of one input layer of 3 neurons containing the ERU fresh fuel composition (%at of

^{234}U, ^{235}U and ^{236}U), one hidden layer of 10 neurons and one output layer of one neuron calculating the BU_{max} value.

The precision of the ANN is determined through a test on an independent sample of ERU fresh fuel compositions, different from the training sample. For each ERU fresh fuel composition in this test sample, an infinite assembly calculation is performed with SMURE/SERPENT as described in section II.2, followed by a maximum burn-up calculation. The ANN is then executed with the ERU fresh fuel composition as an input to calculate the estimated BU_{max}. The distribution of relative differences, between the BU_{max} values calculated by the ANN and by the neutron transport calculations is presented on **Fig. 2**.

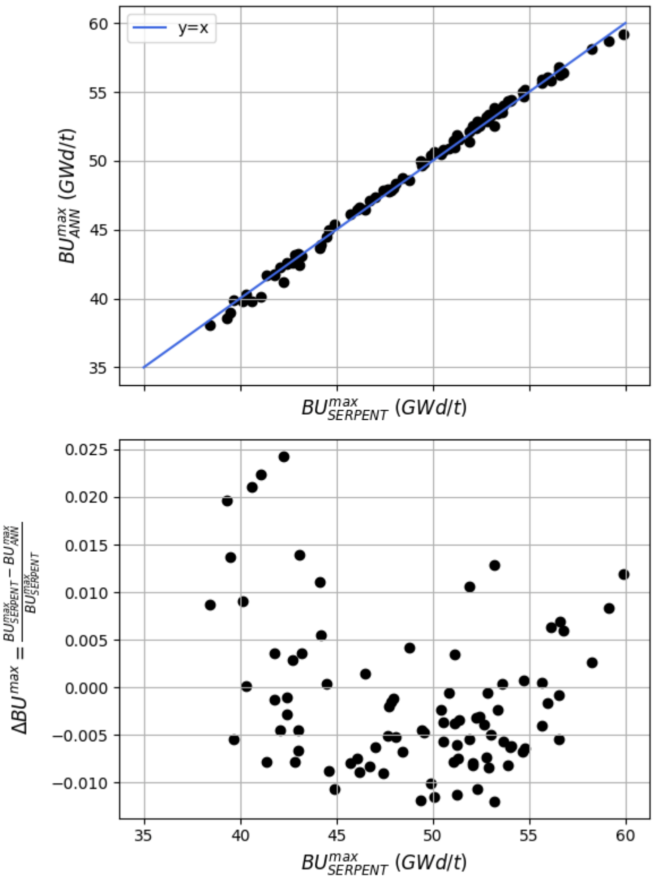

Fig. 2 Distribution of relative errors between the BU_{max} values calculated by the ANN and SERPENT

This distribution of the deviation is characterized by a mean value of 0.1% and a standard deviation of 0.8%, within the range of estimator quality from reactor models previously created for CLASS.[12)]

2. RepU Enrichment Equations

The isotopic composition of fresh ERU fuel (fractions of $^{234}U_{ERU}$, $^{235}U_{ERU}$, $^{236}U_{ERU}$ and $^{238}U_{ERU}$) depends on the isotopic composition of RepU before enrichment (fractions of $^{234}U_{RepU}$, $^{235}U_{RepU}$, $^{236}U_{RepU}$ and $^{238}U_{RepU}$). The equations determining the isotopic composition of fresh ERU fuel as a function of the isotopic composition of RepU also depend on the enrichment process.

In this work, low elementary enrichment processes, such as gaseous diffusion or ultra-centrifugation, are considered. In these cases, the simplified RepU enrichment equations derive from the mass differences between the different Uranium molecules,[4)] resulting in the equation system (1), with M_{23jU} the atomic weight of the Uranium isotope ^{23j}U. The equation system (1) is expressed in terms of atomic fractions. This method allows to deduce the ERU fresh fuel composition obtained through the enrichment process of a RepU composition. Its application enables the efficient retrieval of ERU fresh fuel compositions derived from reference RepU compositions given in.[7)]

$$(1)\quad \begin{cases} \epsilon_i = \dfrac{^{23i}U_{ERU} - ^{23i}U_{RepU}}{^{23i}U_{RepU}(1 - ^{23i}U_{ERU})} & \text{with } i = 4, 5, 6 \\ \epsilon_j = k_j \epsilon_5 & \text{with } j = 4, 6 \\ k_j = \dfrac{M_{238U} - M_{23jU}}{M_{238U} - M_{235U}} & \end{cases}$$

Figure 3 presents the application of the simplified Uranium equation system (1) to calculate fractions of $^{236}U_{ERU}$ with different fractions of $^{235}U_{ERU}$, $^{235}U_{RepU}$ and $^{236}U_{RepU}$. The accumulation of $^{236}U_{ERU}$ during the enrichment process is enhanced by the increase of $^{235}U_{ERU}$, the increase of $^{236}U_{RepU}$ and the decrease of $^{235}U_{RepU}$.

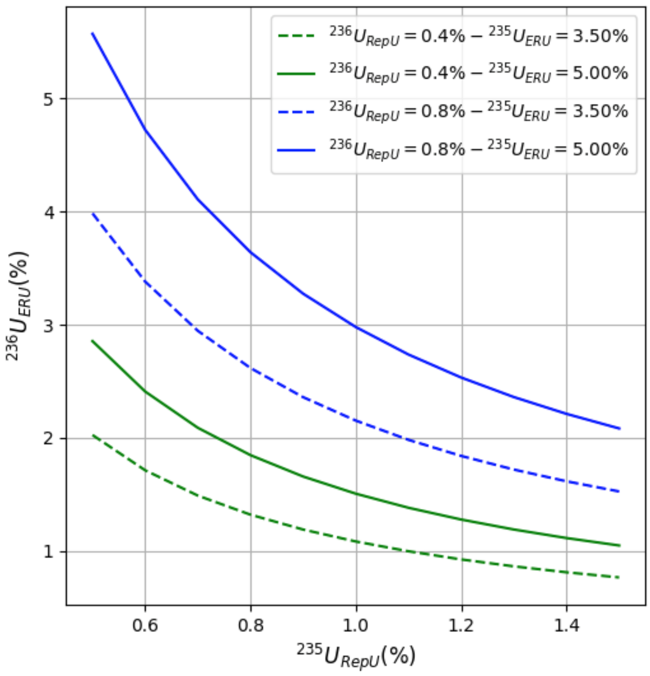

Fig. 3 ^{236}U fraction in ERU fresh fuel as a function of $^{235}U_{RepU}$, for different values of $^{236}U_{RepU}$ and $^{235}U_{ERU}$

For instance, a RepU with a $^{236}U_{RepU}$ of 0.8% should contain at least 1.2% of ^{235}U, to limit the fraction of $^{236}U_{ERU}$ at 2.5% in ERU fresh fuel with a $^{235}U_{ERU}$ enrichment of 5%. With a RepU containing 0.4% of $^{236}U_{RepU}$, the content of

$^{235}U_{RepU}$ could decrease to 0.6% and still guarantee a maximum fraction of $^{236}U_{ERU}$ of 2.5%. Similar trends are observed for $^{234}U_{ERU}$ evolution with RepU composition and $^{235}U_{ERU}$ enrichment.

3. Enrichment Algorithm

The ANN model calculating BU_{max} for ERU fresh fuels has been included in a RepU enrichment algorithm for the nuclear scenario code CLASS, presented on **Fig. 4**.

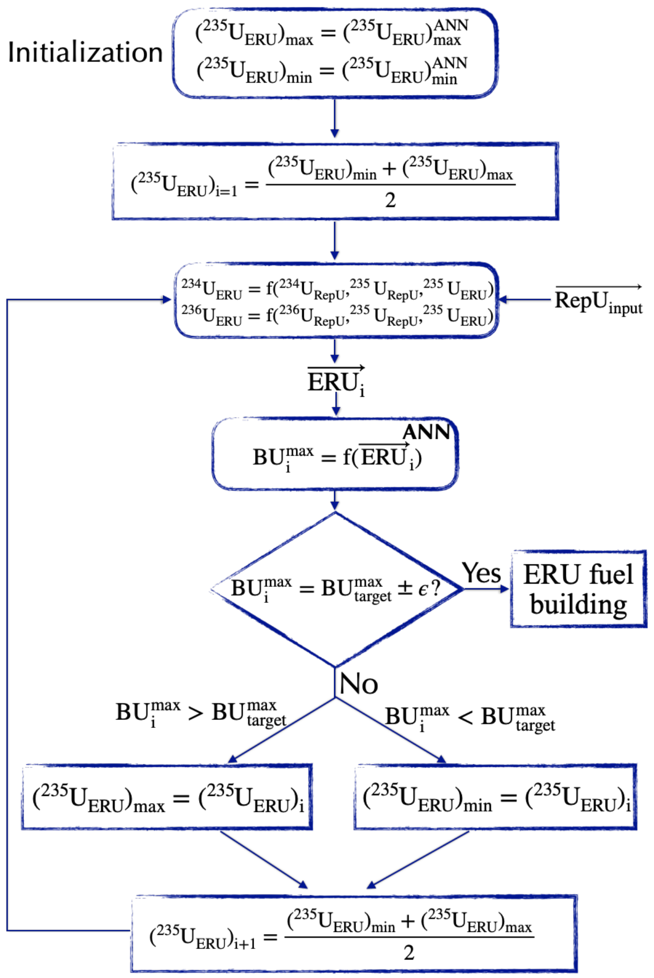

Fig. 4 Reprocessed uranium enrichment algorithm

In uranium mono-recycling scenarios in PWR 100% ERU, the input isotopic vector for Uranium recycling in PWR 100% ERU fuel managements is the RepU recovered at UOX cooled spent fuel reprocessing. This available RepU is then enriched in an enrichment plant to reach a target BU_{max} set by the user. Considering a target BU_{max} and a RepU composition, the RepU enrichment algorithm has therefore to calculate the required ^{235}U enrichment from which the ERU fresh fuel composition derives.

This algorithm is based on an iterative bisection method. $(235UERU)_{min}$ and $(235UERU)_{max}$ are initialized at the beginning of the algorithm within the range of 235U of the ANN. At the first step, 235U enrichment in ERU fresh fuel is set at the midpoint of the range. An ERU fresh fuel composition is calculated from the simplified system of enrichment equations (1) presented in the previous section. Depending on the resulting BU_{max} calculated by the ANN and its comparison with the target BU_{max}, $(^{235}U_{ERU})_{min}$ or $(^{235}U_{ERU})_{max}$ is updated. The ^{235}U enrichment is then adjusted for the next step, until the target BU_{max} is reached, within a tolerance ε set by the user (usually coherent with the ANN precision).

IV. RepU Enrichment Model Application

1. RepU Isotopic Composition Sampling

To test the RepU enrichment model built in this work, an available RepU composition range, as encountered in a uranium mono-recycling in PWR strategy, is determined. Such a RepU composition range has been defined from spent cooled PWR UOX fuel compositions calculated with CLASS and is presented in **Table 2**. UOX fresh fuel in PWR is irradiated until a burnup within the range [30 – 60 GWd/t] and cooled between 0 and 10 years before reprocessing. UOX fresh fuel ^{235}U enrichment is determined by CLASS using an ANN based fuel loading model depending on the sampled burnup value. 1000 RepU compositions have been sampled in the range of Table 2.

Table 2 Reprocessed Uranium composition range (%at) - ^{238}U is used as a buffer to reach 100% in the uranium vector definition

	^{234}U	^{235}U	^{236}U
Min	0.02 %	0.9 %	0.4 %
Max	0.04 %	1.2 %	0.8 %

2. Enrichment Algorithm Application

The RepU enrichment algorithm previously defined is applied for each RepU composition. A target BU_{max} value is randomly sampled within the output range of the ANN. If the ^{235}U enrichment required to reach this BU_{max} value leads to an ERU fresh fuels composition outside the ANN input range (Table 1), the associated sample is excluded. For instance, target BU_{max} values close to the ANN upper limit already leads to ^{235}U enrichments in UOX fresh fuels close to the upper limit of 5%. Considering the presence of ^{236}U in the RepU composition, ^{235}U enrichments higher than 5% would be required to reach high values of burnup. Around 30% of the samples are excluded in our case.

3. Discussion of the Results

For the sampled isotopic compositions, the ^{234}U and ^{236}U fractions in ERU fresh resulting from the application of the RepU enrichment algorithm for a sampled BU_{max} value are presented on **Fig. 5** and **Fig. 6**. ^{234}U and ^{236}U fractions in ERU fresh fuel increase with the ratio of these fractions in RepU with regards to ^{235}U. Higher ratio of ^{234}U and ^{236}U in RepU lead to higher ^{234}U and ^{236}U fractions in ERU fuel, as they are enhanced by the enrichment process.

A higher target BU_{max} value also leads to higher ^{234}U and ^{236}U fractions in ERU fresh due to the increase of the required ^{235}U enrichment. Considering the isotopic range of ERU fresh fuel set for the ANN construction (Table 1), the lower and upper limits of the ^{234}U and ^{236}U fractions are never reached. Thus, the limitation on the isotopic composition of the ERU

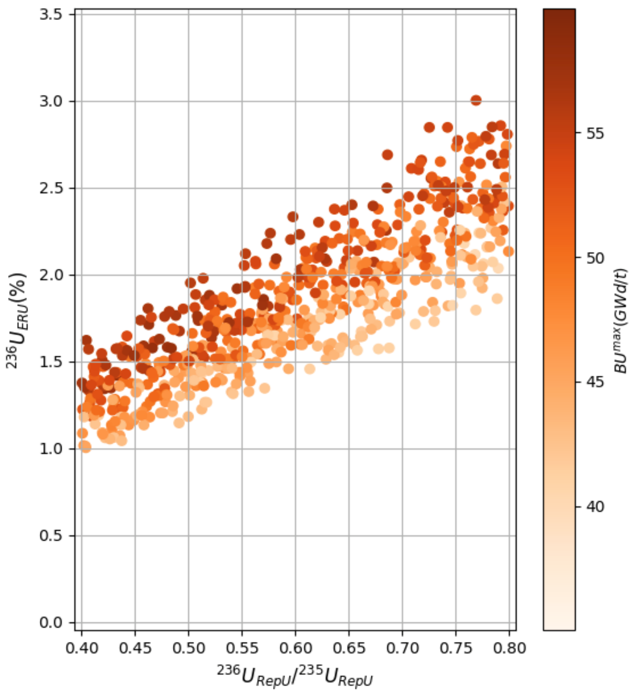

Fig. 5 ^{236}U fraction in ERU fresh fuel as a function of the ratio $^{236}U_{RepU}$ / $^{235}U_{RepU}$. The sampled BU_{max} value is highlighted for each sample as an orange color map.

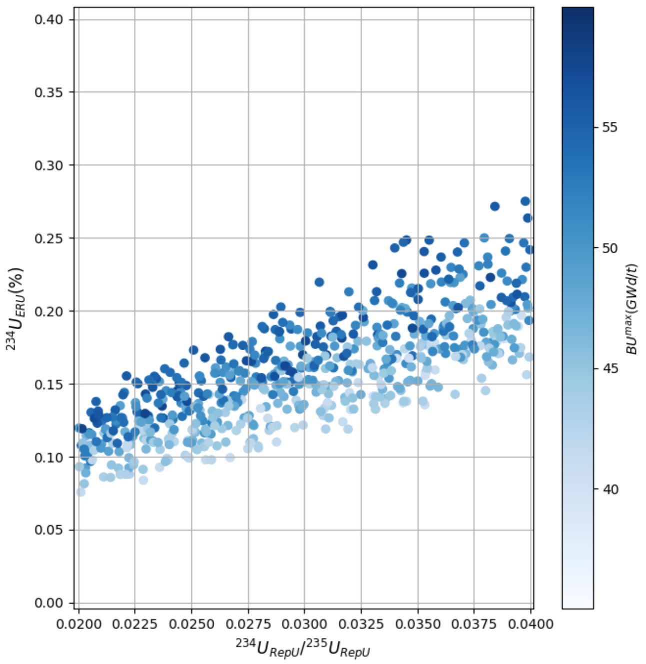

Fig. 6 ^{234}U fraction in ERU fresh fuel as a function of the ratio $^{234}U_{RepU}$ / $^{235}U_{RepU}$. The sampled BU_{max} value is highlighted for each sample as a blue color map.

fresh fuel acceptable for the ANN model is carried by the upper and lower fraction of ^{235}U fractions depending on the sampled BU_{max} value.

Figures 7 and **8** present the ^{235}U enrichment in ERU fresh fuel depending on the target BU_{max}. The ratio of ^{234}U and ^{236}U in RepU with regards to ^{235}U are also highlighted.

As expected, the required ^{235}U enrichment in ERU fresh fuel increases with the target BU_{max}. The required ^{235}U

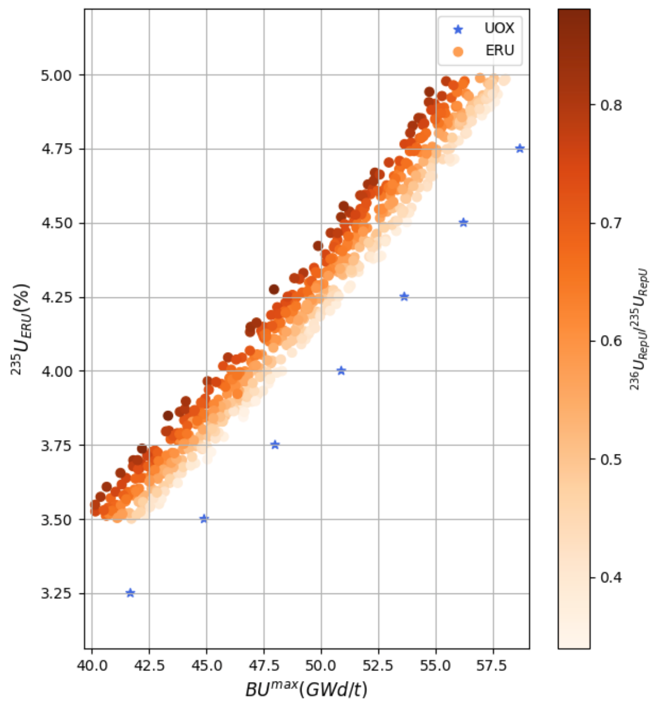

Fig. 7 ^{235}U enrichment in ERU fresh fuel as a function of sampled BU_{max}. The ratio $^{236}U_{RepU}$ / $^{235}U_{RepU}$ is highlighted for each sample as an orange color map.

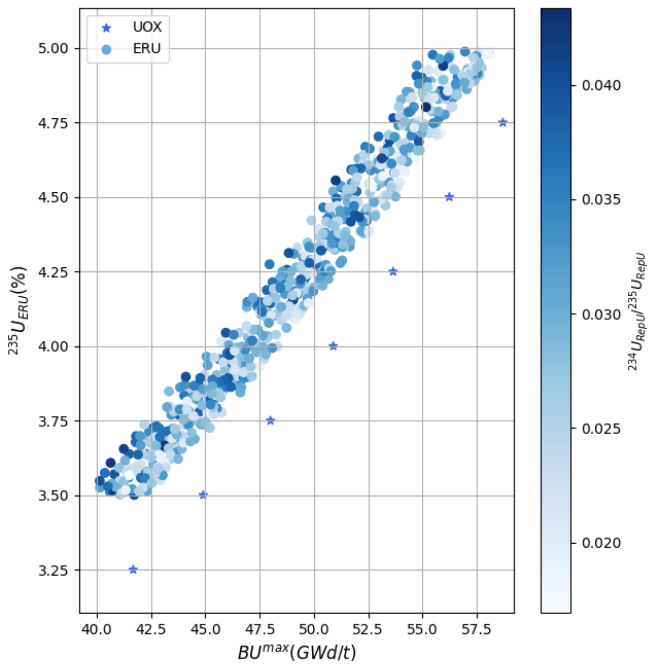

Fig. 8 ^{235}U enrichment in ERU fresh fuel as a function of sampled BU_{max}. The ratio $^{234}U_{RepU}$ / $^{235}U_{RepU}$ is highlighted for each sample as a blue color map.

enrichment in ERU fresh fuel is higher compared to equivalent UOX fresh fuel (up to +0.5% in absolute for a given BU_{max} value).

Figue 7 highlights the impact of ^{236}U fraction in RepU composition on the required ^{235}U enrichment. Indeed, for a given BU_{max}, the increase of ^{236}U in RepU composition implies an increase of ^{235}U enrichment in ERU fresh fuel to compensate neutron absorbing behavior of ^{236}U. A fraction of 0.4% to 0.8% of ^{236}U in RepU leads to a dispersion on ^{235}U enrichment values of +0.25% in absolute for a given BU_{max}, value. This issue should be aggravated through RepU multi-recycling as ^{236}U fraction in RepU would increase at each recycling.

Figure 8 confirms the lesser impact of ^{234}U fraction in RepU on ^{235}U enrichment in ERU fresh fuel.

V. Conclusion

RepU recycling in PWR is a key component of the uranium and plutonium multi-recycling strategies in PWR foreseen in the French fleet by the year 2050. To acquire insights on these nuclear material management strategies and their potential deployment in the French nuclear fleet, nuclear scenario simulations are performed.

These studies rely on a nuclear scenario simulation tool, such as CLASS developed at CNRS/IN2P3. CLASS models various reactors, fuels, and cycle units, calculating isotopic inventories and material flows over time. It relies on reactor models developed using neutron transport codes, upstream of the dynamic cycle simulation.

This paper presents the conception of a fuel loading model for CLASS, dedicated to the enrichment of RepU to produce ERU fresh fuel for PWR.

This model builds a ERU fresh fuel achieving a target discharge burn-up, from a RepU composition. The relation between ERU fresh fuel and RepU compositions is obtained by applying Uranium enrichment equations depending on the enrichment process.

A PWR ERU transport depletion calculation database has been created and used to train an ANN estimator of the maximum reachable burnup depending on ERU fresh fuel composition. This ANN model has been included in a RepU enrichment algorithm, deducing the ERU fresh fuel composition depending on the RepU isotopic composition and the target BU_{max}.

To test the RepU enrichment model, an available RepU composition range has been defined from spent cooled PWR UOX fuel compositions calculated with CLASS. RepU compositions and BU_{max} values have been sampled. The RepU enrichment algorithm is applied for each RepU composition and target BU_{max} value to obtain the associated ERU fresh fuel compositions. The analysis of these ERU fuel compositions highlight the impact of ^{236}U fraction on the required ^{235}U over-enrichment in ERU fuel to reach the same BU_{max} as in an equivalent UOX fuel.

This work will be continued by the construction for CLASS of an irradiation model for ERU fuel in PWR. An analysis of ERU spent fuel isotopic composition will also be conducted, especially concerning the plutonium isotopic composition. Then, the RepU enrichment model will be applied to multi-recycling of plutonium and uranium in PWR strategies modelled with CLASS.

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