Evaluating computational methods for modeling off-normal operation of gas centrifuge cascades

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Abstract

This work compares and evaluates different computational approaches for modeling off-normal operation of a gas centrifuge enrichment cascade.

The goal of this work focuses on developing the necessary understanding of potential misuse of enrichment cascades, contributing to more effective international safeguards designs and approaches. While it is straightforward to design a symmetric enrichment cascade under ideal conditions as a function of the theoretical feed, product, and tails assays, it is very difficult to find reliable information about the behavior of a given cascade when the feed assay does not match the design value. Several methods have been developed to assess the behavior of an enrichment cascade in such circumstances. In addition to the $cut(\theta)$ those methods evaluate the feed to product, feed to tails, and the product to tails enrichment ratio, α , β and γ , respectively, as a function of the cascade feed assay. As those four parameters depend on each other, determining two of them fully defines the other. The first approach consists of fixing θ and α recomputing the corresponding assays at each stages of the cascade. The second one maintains the ideal condition of the cascade (α and β fixed across the whole cascade), modifying θ value at each stage accordingly. Both approaches have been implemented into the Cyclus fuel cycle simulator [1, 2]. The third fixes θ and γ , using both α and β at each stage as free parameters. The third method has been investigated in [3].

Following a description of each method and an evaluation of differences between each approach, this work compares the results produced by these methods within scenarios involving misuse of symmetric enrichment cascades simulated using the dynamic nuclear fuel cycle simulator, Cyclus.

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5 1. Motivation

Gas centrifuge cascades are usually designed to operate in an ideal manner, with no losses in separative work. To achieve such ideal configuration, the cascade is designed to be fed with a specific feed assay and produce the target enrichment while rejecting tails at a fixed assay.

With the current international tensions regarding enrichment capabilities, this work aims to measure the effectiveness of a symmetric enrichment cascade when used outside of its designed scope and quantify the attractiveness of such way to build up significant quantity of High Enriched Uranium (HEU).

The present work investigates the performance of an enrichment cycle when chaining gas enrichment cascades tuned for low enrich uranium production from natural uranium to instead produce HEU. As literature on the mater is for obvious reason limited, three behavior models have been implemented and used to evaluate the response of an enrichment cascade when fed with different assays than designed one. This work also takes advantage of the Cyclus[1] fuel cycle capabilities to evaluate the assay values at equilibrium.

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2. Theory

In the following section, equations and algorithms used to model the behavior of a symmetric ideal gas centrifuge cascade from the individual centrifuge property are described. Details will also be provided to model the centrifuge behavior when fed with a different feed enrichment than the design one.

2.1. Centrifuge properties

2.1.1. Separative power

Räetz equation. As described in [4], the separative power of a single centrifuge can be express as an analytical solution [5] of the differential equation for the gas centrifuge:

$$\delta U(L, F, \theta, Z_p) = \frac{1}{2} F \theta (1 - \theta) \left(\frac{\Delta M}{2RT} v_a^2 \right)^2 \left(\frac{r_2}{a} \right)^4 \left[1 - \left(\frac{r_1}{r_2} \right)^2 \right]^2 \qquad (1)$$

$$\left[\left(\frac{1 + L/F}{\theta} \right) (1 - exp[-A_P(L, F, \theta)Z_p]) + \left(\frac{L/F}{1 - \theta} \right) (1 - exp[-A_W(L, F, \theta)(Z - Z_p]) \right]^2,$$
with
$$A_P = \frac{2\pi D\rho}{\ln(r_2/r_1)} \frac{1}{F} \frac{1 - \theta}{(1 + L/F)(1 - \theta + L/F)} \qquad (2)$$

$$A_W = \frac{2\pi D\rho}{\ln(r_2/r_1)} \frac{1}{F} \frac{1 - \theta}{(L/F)(1 - \theta + L/F)} \qquad (3)$$

In this equation, the parameters of average gas temperature, T, peripheral speed, v_a , height, h, diameter, d, pressure ratio, x, feed flow rate, F, countercurrent flow ratio, L/F, are intrinsic to the centrifuge design. To match the cascade design describe in [4] and [3], P1-type centrifuge properties have been chosen (Table 1).

Table 1: Summary of the centrifuge parameters.

T[K]	$v[\mathrm{m/s}]$	h[m]	d[m]	x	F[mg/s]	L/F
320	320	1.8	0.105	10^{3}	13	2

The variable Z_p is the rectifier length, or the location of the feed point, has an optimal axial location as defined by [5]:

$$Z_p = \frac{(1 - \theta)(1 + L/F)}{1 - \theta + L/F} Z \tag{4}$$

This optimizes the recitifer length based on the cut, θ , which is an expression of the fraction of the centrifuge feed that is output as product, and the countercurrent flow, L/F. In practice, this value is a design parameter that is updated in the model during the design of the centrifuge cascade.

The parameters r_1 and r_2 are the separation radii of the enriched material (here ^{235}U vs. ^{238}U), r_1 being the withdrawal radius for the lighter isotope and r_2 for the heavier isotope. Räetz's two-shell model looks for optimal values between these two, as defined from the hydrodynamic equations. The radii ratio is optimized using the following relationship:

$$\max\{[1-(\frac{r_1}{r_2})^2]^2 \times [\ln(\frac{r_2}{r_1})]^{-1}\}$$
 (5)

The ratio can further be constrained by approximating r_2 as being very close to the inner centrifuge wall with radius a:

$$\left(\frac{r_1}{r_2}\right) \approx \left(\frac{r_1}{a}\right) = \sqrt{1 - \frac{2RT}{M}(\ln x)\frac{1}{v_a^2}}$$
 (6)

Here the gas constants are molar mass, M, temperature T, universal gas constant, R, and the pressure ratio, x (typically 1000:1). This relationship is valid when $v_a > 380\frac{\rm m}{\rm s}$. Otherwise, the relationship can be approximated $\{\frac{r_1}{r_2} \approx 0.534 \mid v_a \leq 380\frac{\rm m}{\rm s}\}$. In order to decompose the ratio into each individual radius, knowledge on one of them is required. Glaser [4] states that r_2 typically ranges from 96% to 99% the value of the inner centrifuge wall radius a. The exact behavior of r_2 between these two values in operational conditions is unknown, but a linear approximation can be made. It is assumed that r_2 is always at the midpoint of these two extremes (i.e. $r_2 = 0.975a$). Then, r_1 can be found by simply multiplying this value by the ratio.

With this, all parameters of the separative power equation 1 are defined and a value can be calculated. δU has units similar to the feed flow, that of [mg/s].

First principle. It can be shown ?? that the separative power of a single centrifuge can be derive from the first principle and expressed as a function of the feed to product enrichment factor, α , the cut, θ , and the feed rate, F:

$$\delta U = \frac{F}{2} \frac{\theta}{1 - \theta} (\alpha - 1)^2 \tag{7}$$

2.2. Centrifuges basic properties and definition

The outputs of a centrifuge relative to its input can be described with ratios of the abundance $(R = \frac{N}{1-N})$ where the feed, product, and tail are N, N', N'' respectively. Enrichment factions of α (feed to product), β (feed to tail), and γ (tail to product) can then be defined:

$$\alpha = \frac{1 - N}{N} \frac{N'}{1 - N'} \tag{8a}$$

$$\beta = \frac{1 - N''}{N''} \frac{N}{1 - N} \tag{8b}$$

$$\gamma = \alpha.\beta \tag{8c}$$

2.3. Cascade Design

vo 2.3.1. Symmetric Cascade

A symmetric cascade is a cascade where a stage's feed, F, with a tail, T, of the next stage and product, P, of the previous one. For the cascade feeding stage an external feed, F_{ext} :

$$F_i = T_{i+1} + F_{i-1} \ (+F_{ext}) \tag{9}$$

2.3.2. Symmetric Ideal Cascade

This model constructs cascades as symmetrical and ideal, with no losses in separative work. This means that the tail assay of the next stage (N''_{i+1}) is the product assay of the previous stage (N'_{i-1}) , which can be expressed as:

$$\forall i \ N_i = N'_{i-1} = N''_{i+1} \quad \Leftrightarrow \quad \forall (i,j) \ \alpha_i = \beta_j$$
 (10)

2.4. Building the cascade

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The method used to design a symmetric ideal cascade starts with the feeding stage. As all the enrichment factors are equal across all the cascade, the feeding stage is used to determine all subsequent stages. The feed assay of the feeding stage, N_0 , is fixed by the external feed assay provided as an input.

From equation (1) and (7) it is possible to express α as a function of the feed rate F, the separative performance $\delta U(\theta)$ and the cut θ :

$$\alpha = \sqrt{\frac{2\delta U(\theta)}{F} \frac{1-\theta}{\theta}} + 1 \tag{11}$$

From equations (8), the product assay can be expressed as:

$$N' = \frac{\alpha R}{1 + \alpha R} \tag{12}$$

Then, from mass conservation, $N = \theta N' + (1 - \theta)N''$ and equation (12), it is possible to express β as a function of only the feed abundance, R, the cut θ and α :

$$\beta = \left(1 - \frac{N - \theta N'}{1 - \theta}\right) \left(\frac{R}{\frac{N - \theta N'}{1 - \theta}}\right) \tag{13a}$$

$$\beta = R \left(\frac{1 - \theta}{\frac{R}{R+1} - \theta \frac{\alpha R}{1 + \alpha R}} - 1 \right)$$
 (13b)

Finally from equation (11) and (13b) it is possible to determine the cut, θ required to build an ideal cascade:

$$\theta_{i} = \frac{N_{i} - \frac{1}{1 + \beta/R_{i}}}{\frac{\alpha R_{i}}{1 + \alpha R_{i}} - \frac{1}{1 + \beta/R_{i}}}$$
(14)

To construct an ideal stage, a cut (θ) must first be computed. This is found iteratively, searching for the optimal cut value where $\alpha = \beta$ for a given feed and centrifuge parameters. With a separative power calculated, and α and β value can be determined for an initial cut guess. This model assumes that the ideal cut for a stage should be between 0.1 and 0.9. The two enrichment factors are compared, and the higher or lower cut value is chosen by which pair of factors are closer. A new cut is determined from the chosen factors, a new separative power is computed, and new α and β values are compared. This process continues until, to some precision, the resulting enrichment factors are approximately equal. As illustrated in Fig. 1, for a given input feed assay, the ideal feeding stage typically has a θ value between 0.45 and 0.525 when $\alpha = \beta$.

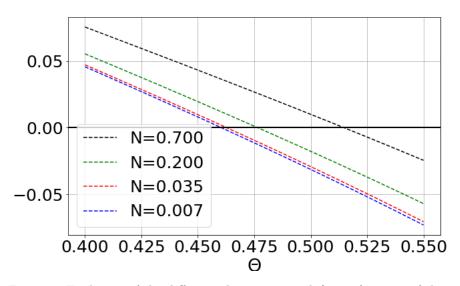


Figure 1: Evolution of the difference between α and β as a function of the cut value for different of feed assays, 0.007 (black), 0.035 (red), 0.2 (green), 0.7 (black).

The number of machines required to construct a stage can then be computed using equation (7) to solve for the centrifuge feed flow:

$$F_c = \frac{2\delta U}{(\alpha - 1)^2} \frac{M}{M_{238}} \frac{1 - \theta}{\theta} \tag{15}$$

Where the molar mass ratio $\frac{M}{M_{238}}$ accounts for the molar mass differences between the feed gas, UF_6 , and the individual uranium isotopes being separated. The stage feed flow can then be divided by the individual centrifuge feed flow, equation (15), to find the exact number of machines needed for the ideal stage. In practice, this number is rounded up to account for fractional machines required.

In a cascade, as α_i and β_i remain constant, only the value of the cut, θ_i , changes across the different stages of a cascade. This algorithm assumes that the corresponding separative power δU (not re-computed) can be achieved with the chosen centrifuge design, tuning other operational parameter such as the rotation speed, the counter-current flow ratio, etc. Once θ_i is determined, it is possible to compute the product and the tail assay.

The design of the ideal symmetric cascade is performed through 2 steps. First, the configuration and number of stages is determined, adding stages until the product assay of the final stage is greater or equal the product targeted assay, and similarly the tails assay is less or equal the tails desired assay. This determines the number of enriching and stripping stages as well as their enrichment properties $(N_i, N'_i, N''_i, \theta_i)$.

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The second step determines the relative flows at each stages, solving the linear flow equation, (16). The cascade can then be populated with actual machines

until the maximum number available of machines is reached.

2.5. Misuse models

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Little information is available about optimising an existing enrichment cascade that is being fed with a feed enrichment that does not match the design enrichment. Here, 3 different methods will be investigated.

The first one assumes that no changes are being made to the cascade, i.e δU , F and θ are fixed across all stages. The second one assumes the cut value at each stage is retuned to maintain the ideal state of the cascade, α and β remain fixed. The last one, described in [3] assumes the tails to product enrichment factor and the cut remain constants (eq. (8c): $\gamma = \alpha \beta$). Models behaviors and assumptions are summarized in Tab. 2.

Table 2: Summary of misuse model properties.

$\gamma_i = \alpha_i \beta_i, \theta_i$ α_i, β_i
blended unchanged
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2.5.1. Model A

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The tuning method A does not re-optimize θ_i keeping the same flow as the ideal configuration. From equation (11), maintaining δU and F while θ is unchanged implies α remains unchanged as well. According to equation (13b), when α and θ are fixed, if the feed assay (N) changes, β will change accordingly. This breaks the ideal status of the cascade, i.e. $N_i \neq N'_{i-1} \neq N''_{i+1}$.

In order to compute the proper product and tails assay at each stage, the tails and the product from the next and the previous stage respectively must be blended in order to determine the correct stage feed assay. All feed assays are iteratively updated, blending the proper product and tails, then using the updated feed assay, the new product and tails assays are recomputed. This process is repeated until the sum of the square difference in assays is smaller than 10^{-8} . As the cut remains fixed at each stage, the flows do not need to be recomputed.

This model assumes that it is possible to maintain the separative power of a centrifuges, δU , for any feed assays N while maintaining its cut θ and feed flow F.

2.5.2. Model B

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Using the second method, the cut value at each stage, θ_i , is retuned in order to maintain the α_i and β_i at their original values (equation (14)). Since the cascade remains ideal, the product and tails assay at each stage can easily be determined using equations (8).

As the cut values change, the relative flow rates between the different stages are recomputed using equation (16). Under this model, the flow at each stage of the original ideal cascade is assumed to be the maximum flow allowed at that stage. Therefore, all of the recomputed flow rates are scaled together to ensure that no stage experiences a flow rate larger than that of the original cascade. Some stages may now experience flow rates much lower than the original cascade.

This model assumes that it is possible to tune a centrifuge separative power δU , for any feed assay N, cut θ and feed flow F, in order to maintain its constant feed to product enrichment factor α .

65 2.5.3. Model C

The last model assumes that the tails to product enrichment factor remains constant regardless to the feed assays. To compute the response of the cascade

one need to determine α and β such that their product and θ remain fixed. From equations (8) and the assay conservation equation $N = \theta N' + (1 - \theta)N''$ it is possible to express the product, N', dependent on the feed assay N, γ , and the cut, θ , as one solution of the second order equation (17):

$$\theta(\gamma - 1)N'^{2} + ((N + \theta)(\gamma - 1) + 1)N' - N\gamma = 0 \tag{17}$$

The only solution allowing product assay to range between 0 and 1 is the following :

$$N' = \frac{N+\theta}{2\theta} + \frac{1-\sqrt{\gamma^2(N-\theta)^2 + 2\gamma(N^2 + N - \theta^2 + \theta) + (N+\theta+1)^2}}{2\theta(\gamma-1)}$$
(18)

Once the product assay is known, one can trivially determine the tails assay, α and β , using equations (8) and mass conservation.

Similar to model A, because the cut values remain constant, the flows do not need to be recomputed and the correct assays, α and β , are determined through iterative blending of the product assays of the previous stage and the tails assay of the next stage using equation (18).

This model assumes that it is possible to tune the centrifuge separative power δU in order to maintain for any feed assay N and its tails to product enrichment factor γ while maintaining its cut θ and feed flow F.

3. The experiment

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This work focuses on comparing the different misuse models to a reference calculation in which a single large cascade is build and designed to directly produce HEU from natural uranium. This works uses the Cyclus fuel cycle simulator to allow material exchange between facilities. The enrichment cascade algorithm have been implemented in the *mbmore* package [2]. In each cases, 5060 centrifuges have been used and spread across up to 30 different gas centrifuge enrichment cascades.

3.1. The cascade configuration

3.1.1. Reference

As mentioned, all the further calculations will be compared to the most favorable configuration to produce HEU, where all the available centrifuges are used in a single large ideal symmetric cascade designed to directly produce HEU from natural uranium, with a tails assay close to 0.3w%. The design characteristic of the reference cascade are summarized in Table 3.

3.1.2. Default cascade

The default cascade is the ideal symmetric cascade designed for normal civilian enrichment operation, enriching natural uranium to about 3.5w%, with a tails assay close to 0.3w%. This cascade will be layered and fed with uranium at higher enrichment to evaluate the possibility to use them, with little or no tuning, to produce HEU. The characteristics of the default cascade are summarized in Table 3.

Cascade Design Reference Default 0.71w%0.71w%Targeted Feed Product 90w%3.5w%Assays Tails 0.3w%0.3w%Product 90.35w%4.13w%Effective Assays Tails 0.29w%0.29w%4 4 Stages Stripping Number Enriching 39 10

Table 3: Summary of cascade design.

3.2. Scenarios

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In the following, cascades can be connected in tandem, where each set of cascade in parallel is called a "level", as illustrated in Figure 2. The reseults from seven different simulations have been compared, to evaluate the effectiveness of an enrichment cascade when used outside of its designed scope:

• one as the reference calculation, with a single cascade designed to directly produce HEU from natural uranium,

- three calculations (one per misuse model) where default cascade are chained to produce HEU, without recycling the tails of each cascade sending their tails to the waste,
- three calculations (one per misuse model) where default cascade are chained to produce HEU, and the tails of each cascade are recycled, blending the tails of one level in the feed of the previous level of cascades (see Figure 2).

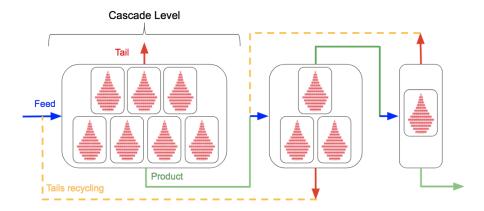


Figure 2: Schematic representation of the chained cascades with three levels, with the feed, product and the tails flows, in blue, green and red, respectively. The dashed orange line represent the alternative tails flow when tails recycling is considered.

3.3. Level population

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In order to assign the optimum number of cascades to each level, a "level cut" as been computed as:

$$\Omega_j = \frac{N_j - N_j''}{N_j' - N_j''},\tag{19}$$

where, j represents a level of cascade and N_j , N'_j and N''_j the feed, product and tails assay, respectively, of the cascades at this level.

A flow equation similar to (??) is then solved to obtain the optimum number of cascade per level. When the tails are not recycled, the $(1-\theta)$ terms are removed from the flow equation. The results of the level population are summarized in Table 4.

As it is not possible to assign a fraction of an enrichment cascade, cascade per level are rounded up for each level but the first one. The remaining available cascades are attributed to the first level.

4. Results

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4.1. Miss-use modeling

Table 4: Summary of cascades level population.

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Model		A/NR	A/R	$\mathrm{B/NR}$	$\mathrm{B/R}$	C/NR	C/R
	Feed	0.71w%	1.3w%	0.71w%	0.94w%	0.71w%	1.33w%
Level 0 Assay	Product	4.13w%	7.7w%	4.13w%	5.43w%	4.13w%	4.82w%
	Tails	0.29w%	0.5w%	0.29w%	0.39w%	0.29w%	0.55w%
Cascades	Impl. (Ideal)	25(26.7)	25(26.4)	25(26.6)	24(25.8)	25(26.7)	25(26.4)
	Feed	4.13w%	11.9w%	4.13w%	6.84w%	4.13w%	12.2w%
Level 1 Assay	Product	22.8w%	55.7w%	20.6w%	30.7w%	22.9w%	58.5w%
	Tails	1.8w%	6.6w%	1.72w%	2.91w%	1.81w%	6.52w%
Cascades	Impl. (Ideal)	3(2.9)	4(3.2)	3(2.9)	4(3.4)	3(2.9)	4(3.2)
	Feed	22.8w%	55.7w%	20.6w%	34.3w%	22.9w%	58.5w%
Level 2 Assay	Product	78.5w%	95.0w%	61.0w%	75.8w%	82.0w%	97.0w%
	Tails	4.13w%	50.9w%	9.56w%	17.5w%	15.7w%	53.8w%
Cascades	Impl. (Ideal)	1(0.3)	1(0.3)	1(0.4)	1(0.6)	1(0.3)	1(0.35)
	Feed	78.5w%	N.A.	61.0w%	75.8w%	82.3w%	N.A.
Level 3 Assay	Product	98.2w%	N.A.	90.4w%	95.0w%	99.1w%	N.A.
	Tails	76.1w%	N.A.	79.3w%	56.1w%	80.3w%	N.A.
Cascades	Impl. (Ideal)	1(0.03)	N.A.	1(0.08)	1(0.2)	1(0.03)	N.A.

As illustrated in Figures 3 and summarized on Table 4, the different models don't have the same effect on the cascade behavior. While models A and C, achieve a quick enrichment gain with the cascades chaining, 4/23/78/98 and 4/23/82/99, respectively, model B, only achieves an enrichment gain of 4/21/61/90. This table also shows the integer number of cascades implemented (Impl.) at each level in the simulation, as well as the non-integer number of cascades that would achieve an ideal configuration (Ideal).

4.2. Tails recycling

As shown in Figures 3, recycling the tails increases the overall product assay at all the different levels. As the tails assay of a level n + 1 is always higher than the product assay of the level n - 1, recycling the tails of level n + 1 will

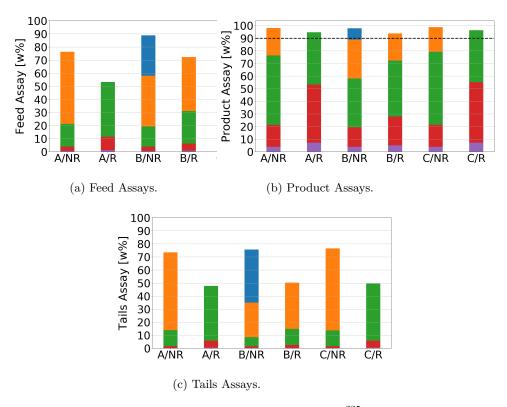


Figure 3: Feed (a), product (b) and tails assays (c) in w% of 235 U, per cascade level from 0 to 3 (red, green, orange, blue), per model (A/B/C) and without/with tails recycling (NR/R). The black dashed line represents the 90w% enrichment threshold.

consequently increase the feed assay of level n (see Table 4). Moreover, with an increased feed assay, tails and product assays increase as well, increasing de facto the feed assays of respectively cascade levels n-1 and n+1, etc. This effect reduces the number of cascade levels required to reach HEU in case A and C.

4.3. HEU Production Rate

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As shown in Figure 4, recycling increases the final HEU production rate, from 2 to almost 20 kg/y when using models A and C, and from 17 to 38 kg/y with the model B. For the reference calculation where all the available cascades

are used within a single large cascade design for direct HEU production, the HEU production rate is slightly over 50 kg/y.

As models A and C, rely on maintaining the cut values at each stages of the cascade and share the same number of levels, have the exact same cascade repartition across the different levels and the same HEU production rate.

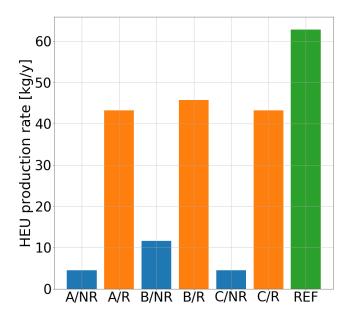


Figure 4: Production rate at equilibrium for the different model configurations, the case without tails recycling (blue), with tails recycling (orange), and the reference one (green). A-B-C represent the model used, and NR-R the case without tails recycling and the case with tails recycling, respectively.

5. Discussion

We can observe that when the cascade is left completely untouched (Model A) or when it is slightly retuned to maintain the tails to product enrichment factor as well as the cut of each centrifuges (Model C), chaining the cascade can achieve large increase of the enrichment at each level. On the contrary, when

retuning the cut of each centrifuges to maintain the ideal state of the cascades (Model B) while chaining them, the HEU production rate is favored over the enrichment gain.

The tails recycling allows each model to achieve a large gain in productivity, even for then model B in which the number of levels required to reach 90w% of ^{235}U in the uranium does not change. Even if no cascade chaining options achieves the same production rate as direct enrichment, the model B with tail recycling reached about 80% of the optimum production rate. Such production rate would allow the accumulation of a Significant Quantity of HEU in less than 8 months...

6. Conclusion and future work

This work has investigated the possibility to chain centrifuge enrichment cascades that are designed to enrich uranium for commercial reactors in order to produce HEU, instead. Three methods have been implemented to model symmetric enrichment cascade behavior when fed with different uranium enrichment than the designed enrichment.

One of these method achieves up to 80% of the production rate of a single large enrichment cascade designed specifically for HEU production using the same number of centrifuges.

This work will be extended to the near future with additional misuse methods, allowing for example, the reconfiguration of the centrifuges in the cascades.

For this study, the use of the Cyclus fuel cycle simulator was not required; it only allows a quick determination of the blending equilibrium. Future studies will make use of the full capability of Cyclus Dynamic Resource Exchange in order to automatically assign the different cascades to the different level as function of the resources availability, optimising the productions rates in each cases.

While mathematically correct, the authors do not guaranty the feasibility of the different misuse tuning methods implemented and are welcoming any insight on the matter.

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