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Applying enrichment capacities for multiple recycling of LWR uranium

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Abstract. The use of fissionable materials accumulated during the open fuel cycle faces both challenges and opportunities for modern nuclear power engineering. A growing disproportion between the production and consumption of natural uranium draws attention to the problem of used nuclear fuel. The paper examines the current situation in the field of reuse (single or multiple) of reprocessed uranium (recovered from spent nuclear fuel), which is used for the production of low-enriched fuel (LEU) to fuel the fleet of light water reactors (LWR). The world experience in handling this material has been analysed. This study also gives an overview of the currently proposed methods for enriching the ²³⁵U content in reprocessed uranium to the required level by means of gas centrifugation process, while simultaneously meeting the limitations on the presence of ^{232,234,236}U in commercial LEU. Savings of natural uranium was estimated for repeated recycling of VVER spent fuel. It was supposed that re-enrichment of reprocessed fuel would be done by arranging the most promising for such purposes cascade schemes. The obtained results can be used as a basis for further scientific, technical, and feasibility studies on the large-scale utilization of reusable materials in the different fuel cycles of LWR.

1. Introduction

1.1 Preamble

It is common knowledge that most of light-water-reactors (LWRs) use low enriched uranium fuel with enrichment of U up to 5%. And as nuclear industry poised to grow, we are waiting for a widening gap between uranium mining and its consumption [1]. Therefore, the problems with supply of this resource will appear and it will be difficult to provide enough fuel for LWRs. One of the possible solutions to this is using spent nuclear fuel (SNF) for producing fresh fuel assemblies.

Here we will focus on uranium recovered separately from used fuel. Worth noting, that vast bulk of SNF is represented by uranium (~66% of the total mass and ~95% of the residual fuel), which is effectively extracted in generally used conventional PUREX process [2]. The reuse of such reprocessed uranium (RepU) has a number of potential advantages: (1) natural uranium (NatU) saving; (2) avoidance of disposal costs for a vast volume of SNF that is reprocessed; (3) possible separative work reduction (depends on the ²³⁵U assay of the RepU) [2].



1.2 Study objective

This paper provides an analysis of the factors involved in the multiple reuse of spent nuclear fuel. In our case, uranium is recovering from commercial light-water-reactor (LWR) – Russian VVER-type. It is assumed, that chemical reprocessing allows getting pure uranium from spent nuclear fuel. Such regenerated uranium undergoes re-enrichment and then returns to nuclear fuel cycle. Recycling of uranium in this way could be efficiently done several times [3].

In such a case, disadvantages of uranium recycling are connected mostly with presence of ^{232}U and ^{236}U minor isotopes [3-4] concentrations of which are strictly limited by the ASTM specifications for enrichment feed and product [5]. Complying with these standards, designed for uranium hexafluoride (a compound used in the uranium enrichment process [6]), ensures the fulfillment of the requirements of radiation safety during the manufacture of fuel elements (conditioned by ^{232}U presence), and preservation the neutron-physical characteristics of the fuel (owing to ^{236}U). Such limitations stem from ^{232}U and ^{236}U properties: ^{232}U is particularly dangerous contaminant because of intense gamma radiation emitted by Thallium (a daughter of ^{232}U , produced relatively quickly); ^{236}U acts as a parasitic neutron absorber that impedes the chain reaction (this means that a higher level of ^{235}U enrichment is required in the product) [4].

Once again, this study examines employing reprocessed uranium (RepU) – material, which is free of impurities and consists of only uranium isotopes. So, the scope of this paper was limited to describing the technical effects of the presence of the various isotopes of uranium in RepU. And this issue is considered mainly from an enrichment process point of view: how to deal with and overcome the harmful influence of $^{232,234,236}\text{U}$ [7]. In other words, the main purpose of the work was to illustrate the role of separation technologies, in particular, the applicability of centrifuge cascades to re-enrich reprocessed uranium efficiently. This needs to be done in order to sort out the general problem of closing the fuel cycle of light water reactors with uranium as the main component of nuclear fuel.

The impact of $^{232,234,236}\text{U}$ on re-enrichment process has been estimated using two key parameters: consumption of natural uranium and separative work requirements. It should be noted, that calculations were done while meeting the ASTM limitations on the presence of $^{232,234}\text{U}$ in enriched product [5]. In the same manner, additional enrichment in order to compensate the effect of ^{236}U should be done [8].

It is assumed that the spent fuel from a reactor pass is reprocessed into oxide, converted to UF_6 , and used as enrichment feed along with sufficient natural-assay UF_6 feed to produce the required enrichment product to fuel the next reactor pass. In our case, this process is repeated over and over again (here, calculations were made for four consequent reactor passes). The whole process depicted in Fig.1, here, for the first reactor pass, the enriched uranium for the reactor fuel was produced in centrifuge enrichment plant using all-natural-uranium feed. Beginning with the first cycle on NatU, spent fuel from the reactor core goes to cooling and reprocessing. Then, starting with cycle 2, reprocessed fuel undergoes re-enrichment along with natural uranium in a cascade. Afterwards, LEU is used for fuel assembly manufacturing, these bundles are inserted into the reactor core. At the end of operating period, used fuel is placed in cooling ponds. After that, as the last step of each cycle, SNF is reprocessed and could be used over again.

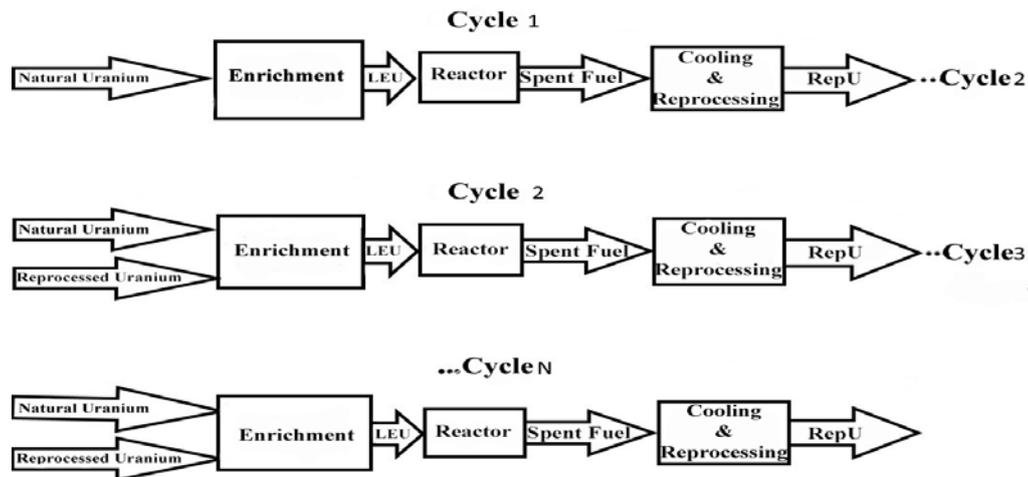


Figure 1. Multiple recycling of LWR uranium.

It should be pointed out, that in case of multiple recycling, we will face steady accumulation of undesirable isotopes $^{232,234,236}\text{U}$ [3]. Unfortunately, such consistent RepU re-enrichment inevitably leads to deterioration of fuel isotopic composition: build-up of minor isotopes significantly worsens the quality of reused material, because more NatU is needed to dilute these pollutants.

2. Main part

2.1 Brief review of cascade schemes for RepU re-enrichment

The industrial cascade for uranium enrichment is an expensive large-scale hi-tech structure. In this connection, it is particularly important to design such installations properly. That is why for more than seventy years along with the improvement of separation technologies, the theory of binary and multicomponent isotopic mixtures separation in cascades is developing. Mathematical modeling of enrichment cascades allows to make an adequate analysis of mass transfer processes in order to find their best parameters for solving the separation problems. Below we present an analysis of modern approaches to the solution of the RepU re-enrichment problem, based on the gas centrifuge separation technology and the achievements of cascade theory for the separation of multicomponent mixtures.

Lots of them are based on ‘ordinary’ (triple-flow) cascade, which could be applied, for instance, in following ways (Fig. 2) [8]:

- to enrich the mixture of natural (F) and reprocessed uranium (E).
- to produce enriched product from RepU (E) and then to dilute it with NatU (F) or with other uranium mixture.
- to over-enrich NatU (F) a bit, than it needed for reactor-grade fuel, and then to dilute with RepU (E).

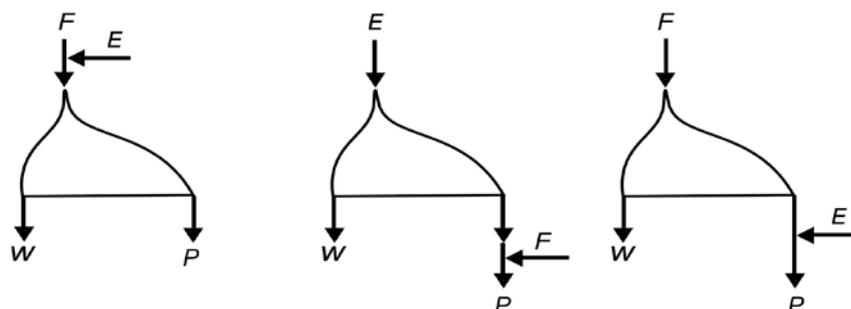


Figure 2. Schemes for RepU re-enrichment based on ordinary cascade.

The main drawback of all these schemes is losses of separative work, due to mixing flows with different concentration of ^{235}U .

Here, RepU needs to be somehow diluted by mixtures without harmful $^{232,236}\text{U}$ isotopes (usually NatU, but also it can be DepU). And as we can see on these figures, commercial LEU could be produced by watering down RepU at any outer point [8]. In this case, the ratio between the RepU flow and the diluent is determined by the limit of ^{232}U content in product (P). At the same time, the concentration of ^{235}U should not be lower than required for LEU with specific properties.

The main advantage of such schemes is simplicity of implementation, because there is no need for a nonstandard modification of the cascade. Moreover, we already have necessary technologies for mixing gaseous uranium with different isotope content.

The main drawback here is losses of separative work due to mixing of flows with different isotopic concentration.

Apart from these simple methods there are other variants based on the use of multi-flow cascades with additional feeds. The advantage of using such cascades, comparing to the previous ordinary schemes, is that the RepU is directed into the cascade through a distinct feed point without losses of separative work [8-10]. However, in such schemes, a noticeable decrease in the content of $^{232,234,236}\text{U}$ isotopes in LEU is achieved, first of all, as in previous cases, due to dilution of the RepU with natural raw materials within the cascade.

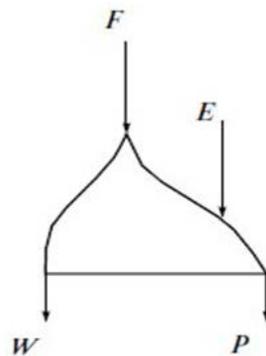


Figure 3. Cascade scheme with feeds of RepU (E) and NatU (F).

In some cases, multi-flow cascades may have an additional external flow – additional product [8, 11]. Such a scheme of the cascade for purification of the RepU from minor isotopes is presented below (Fig. 4). Here, in (P_2) flow, a LEU is obtained, and in the intermediate (P_1) flow, a semi-product with a reduced content of minor isotopes is produced. The ^{235}U concentration in this "purified" product is about 0.8-1.1 of its value in the "purified" RepU and is regulated both by the ratio of the incoming fluxes (RepU to NatU) and by the relocation of intermediate outlet point (P_1) or additional feed point (F). As a main disadvantage, this configuration is subject to radiation contamination, and the cleaning effect is caused, primarily, by the lesser stream of purified material (P_1). But in this scheme there are practically no loss of separative work and no extra cascade is required [11].

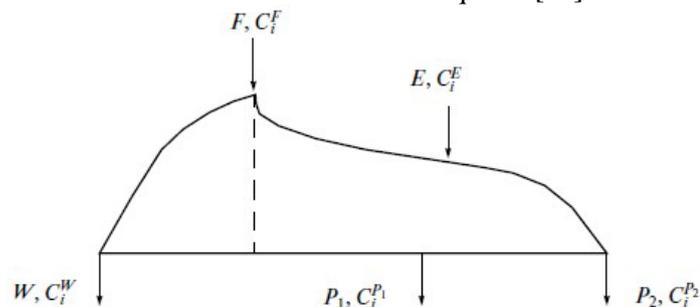


Figure 4. Cascade scheme with two feeds (NatU and RepU) and additional purified product (P_1).

Extensive NatU use for dilution of RepU is a common drawback of abovementioned cascade schemes (Fig. 2-4), because it is necessary, first of all, to decrease ^{232}U content in product. So, it is not helpful enough for NatU saving (no more than 15-18% economy is achievable, according to preliminary estimations).

To overcome this shortcoming, depleted uranium (DepU, byproduct of the enrichment process, so-called ‘tails’) could be employed, as alternative to NatU diluent, although this material often treated as ‘waste’. The main advantage of its use is possibility of significant natural uranium saving. But such approach could also be helpful in reducing its own stored amounts.

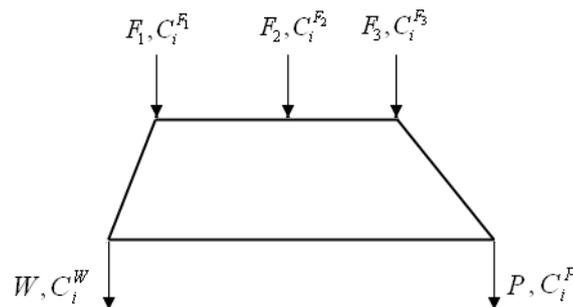


Figure 5. The schematic drawing of the cascade with three feeds.

In this paper, we consider a scheme, where DepU and NatU are used together as the cascade feeds to water down harmful $^{232,234,236}\text{U}$ isotopes in RepU (Fig. 5) [12]. F_1 , F_2 and F_3 are flows with concentrations $C_i^{F_1}$, $C_i^{F_2}$, $C_i^{F_3}$ into feed points of DepU, NatU and RepU respectively. This cascade has product (P) and waste (W) outlets with a concentrations C_i^P , C_i^W (from now on, the concentration is expressed as a percent by mass; $i = \overline{1, m}$, i - index number of each isotope; m - the total number of components in mixture). Detailed description of mathematical model is given in [11].

This feature makes this scheme promising, as it combines the ratio of two diluents (DepU and NatU) to achieve proper natural uranium or separative work saving, which allows to "tune" the cascade for specific isotopic compositions of feed mixtures and current prices of NatU and separative work unit (SWU).

Yet there are other options, such as double cascade, which is formed by two ‘ordinary’ cascades through serial connection. It helps to re-enrich reprocessed uranium even without diluting it with raw materials, but such scheme usually considered to be ‘dirty’ because of polluting enrichment devices, as its main drawback [8]. Another shortcoming of this option is mounting concentration of fissile ^{235}U in a by-product, which could even reach the level of highly enriched uranium [8], and this is inappropriate for meeting the terms of non-proliferation treaty designed to control spreading of fissile materials. Finishing the brief review of the proposed methods for RepU re-enrichment, it should be noted that there is still no answer to the question which of the schemes is inherently better and more effective. And the final answer could not be given, because the advantages of every cascade is inseparable from the industry aims and fuel cycle short-term goals (connected with current market prices) or long-term strategies (which are basically based on specific national policy). So, for instance, in the first case, we could pick the right cascade, focusing on the immediate task after carrying out detailed technological and economic evaluations of the effectiveness of the schemes considered. And, in an ideal world, the outcome of the choice would always be closely connected with path that industry pursue, and the choice itself would be obvious due to comprehensive scenario analysis.

2.2 Multiple recycling

Let's look closely at the method employed in this research. The simulation of multiple recycling of LWR uranium was carried out in this study. Here, we used complex approach based on coordinated simulations of neutron-physical processes (during reactor operation and spent fuel storing in cooling pools at the reactor site) and molecular-selective transfer [13]. In this way, neutronic calculations were

made at NRC KI for VVER-1000 fuel burnup of 46.9 GWd/tU and sequential 5 years cooling period for each cycle (Fig. 1). NRNU MEPhI, in its turn, performed computer simulation of molecular-selective transfer in studied cascades for each cycle.

Two of the above-mentioned cascade schemes (with two feeds (Fig. 3) and three feeds (Fig. 5)) were taken as reference objects (for convenience, denote them as scheme #1 and scheme #2, respectively). The basic ordinary cascade will serve as a benchmark for making comparisons using fundamental indicators: consumption of natural uranium, separative work requirements. To represent proposed methodology when considering the issue of RepU multiple recycling in VVER, these two schemes were picked for the following reasons: (1) the first configuration being the best option for RepU re-enrichment, comparing with cascades based on three-flows schemes [8, 9], could facilitate comparison between RepU employment and basic open fuel cycle; (2) the second scheme could show the ability of isotope separation technologies to attain significant saving of NatU. So, primarily, the schemes serve to illustrate the potential of gas centrifuge enrichment plants for increase of NatU saving.

The external parameters of the cascade were specified: the required concentrations of the target component ^{235}U in the waste and product flows, the ^{236}U penalty, and the maximum acceptable ^{232}U concentration in the product.

R-cascade has been considered as a calculation model for both schemes [14]. It is a special case of so-called "quasi-ideal" cascade – cascade, where condition of no mixing is met for chosen pair of components (in our cases: ^{235}U and ^{238}U isotopes). In the molecular-selective transfer calculations the following parameters were given: concentration of ^{235}U in product was 4.6% (not taking into account required additional enrichment in order to compensate the effect of ^{236}U); the ^{236}U compensation factor was 0.29; concentration of ^{235}U in waste end of the cascade was 0.1%. The concentration of ^{232}U in the product was limited by value $5 \cdot 10^{-7}$ %; and ratio of ^{234}U to ^{235}U must be lower than 0.02 [5]. DepU with 0.2% of ^{235}U (in scheme #2) and NatU (in both configurations) acted as diluents. Let us remind you that feeds of different compositions are directed into separated feed points of cascade. The locations of such inlets are placed where ^{235}U concentrations of external and internal flows coincide (external flows are feeds of the cascade, when internal flows are already circulating inside a set of gas centrifuges, in other words, cascade).

For instance, the calculations for cascade with three feeds were performed for the problem statement as follows: the concentrations and relations F_2/F_3 , F_1/F_3 are given (n – the index number of the target component). Through calculations is necessary to compute remaining parameters of the cascade, including the concentrations of other components in outgoing flows and the number of stages in the cascade, etc. The problem is interesting from a practical point of view, because in this case clearly defined product quality allows comparing the effectiveness of different ways to produce commercial reactor-grade LEU.

As a benchmark for summarizing the results, the ordinary *R*-cascade is considered [14].

2.3 Results and discussion

Evolution of the isotopic composition during multiple recycling is shown in tables 1-2.

Table 1. Scheme #1 calculation results for a sequence of U recycles, taking into account ^{236}U compensation and ^{232}U limit

Cycle	Before enrichment				After enrichment			
	^{232}U , %	^{234}U , %	^{235}U , %	^{236}U , %	^{232}U , %	^{234}U , %	^{235}U , %	^{236}U , %
Recycle								
1	2.44e-7	2.26e-2	1.16	6.38e-1	2.28e-7	5.18e-2	4.73	4.61e-1
2	5.25e-7	3.17e-2	1.26	1.07	4.92e-7	6.03e-2	4.83	7.84e-1
3	7.64e-7	3.75e-2	1.32	1.38	5.0e-07	5.75e-2	4.81	7.16e-1
4	7.40e-7	3.58e-2	1.31	1.31	5.0e-7	5.70e-2	4.80	7.03e-1

For cascade with two feeds starting from the third recycle, total employment of RepU becomes impossible, due to the deterioration of the isotopic composition of uranium and the presence of limitations on the isotopes $^{232,236}\text{U}$.

Table 2. Scheme #2 calculation results for a sequence of U recycles, taking into account ^{236}U compensation and ^{232}U limit

Cycle	Before enrichment				After enrichment			
	^{232}U , %	^{234}U , %	^{235}U , %	^{236}U , %	^{232}U , %	^{234}U , %	^{235}U , %	^{236}U , %
1	2.44e-7	2.26e-2	1.16	6.38e-1	2.28e-7	4.54e-2	4.67	4.65e-1
2	5.27e-7	2.89e-2	1.26	1.07	4.94e-7	5.25e-2	4.83	7.84e-1
3	7.54e-7	3.31e-2	1.32	1.38	5.0e-7	4.88e-2	4.82	7.27e-1
4	7.45e-7	3.08e-2	1.31	1.32	5.0e-7	4.75e-2	4.81	7.03e-1

As we can see from the following tables, enrichment technologies could equip the nuclear industry with effective instrument for ‘refining’ isotopic composition of reprocessed fuel, even in case of multiple recycling. An increase in the number of consecutive cycles is also possible. And we could achieve even greater results for both schemes if we consider an extension of permissible limit of even-numbered isotopes, first of all ^{232}U . For such purpose ASTM standards need to be re-examined, or maybe fuel production should comply with other international or local specifications. Anyway, the role of ^{232}U in enrichment process needs to be scrutinized over whether it is possible to establish renewed proven limitations for gaseous UF_6 . Restrictions similar in nature were already extended for fuel assembly manufacturing due to robotization of factories, which enables remote fabrication [15].

A synopsis of the results for studied schemes is displayed in the table 3.

Table 3. Deviations for prior results in NatU consumption and separative work (SW) requirements from ordinary cascade parameters (positive values – a potential saving, negative values overspending)

Feeds	NatU+RepU (Scheme #1)		NatU+RepU+DepU (Scheme #2)	
	NatU	SW	NatU	SW
1	19,15	4,47	50,90	-47,09
2	19,02	3,70	51,07	-48,11
3	13,11	2,10	50,90	-58,50
4	13,58	2,28	50,72	-58,22

As we can see from the table 3, the cascade scheme #2, in which DepU is applied for dilution of RepU, is the most effective for all recycles in terms of natural uranium savings. A stable, more than 50% saving of natural uranium per recycle pass is achieved due to 50% separation work increase. In principle, schemes of this type can, theoretically, provide any degree of NatU savings (in this paper, 50% is chosen for illustration). But, when choosing the ratio between the costs of natural uranium and the work of separation, one should also start from economic criteria, and also take into account that this ratio will depend on the parameters of DepU, in particular the residual content of ^{235}U . Worth mentioning, that with this scheme, even greater results could be achieved by employing depleted uranium with higher concentration of fissile ^{235}U , than used in current calculations. Such materials are abundant (world stock is about 1.6 million tons [16]), so detailed inventory of ^{235}U assay in stored casks of depleted UF_6 is needed. Summarizing the advantages of this approach, it can be said, that, in

total, these factors can allow reduction in fuel cycle costs for VVER due to significant decrease in NatU demand and in dynamics of DepU accumulation, as well as a reduction in the SNF that needs to be buried.

It is shown that the schemes considered in the paper may have different applications, depending on the objectives sought. So, the cascade #1 with two feeds allows to simultaneously achieve saving of separative work and natural uranium, in comparison with the regular ordinary cascade, however, the maximum saving of natural uranium could not exceed 19-20%. As for scheme #2 with three feeds, it could provide stable and considerable (>50%) natural uranium saving, by spending more separative work. Thus, the demonstrated possibility of a substantial natural uranium saving might be economically attractive in case of uranium demand growth.

It was noted that with the cycle number there is a tendency to rise in concentrations of $^{232,234,236}\text{U}$ isotopes, in other words, to deterioration of isotope composition. This effect leads to an increase in separative work requirements and, in the absence of an additional diluent (DepU) to higher NatU input [3].

To sum up, implementation of considered 'advanced' schemes could lead to substantial reduction of VVER fuel cycle costs due to significant decrease in NatU demand (and in the accumulation of DepU for scheme with three feeds), as well as SNF that need to be disposed.

Carrying out comprehensive scenario analysis would also be important for more efficient fissile material allocation. It could be helpful in forecasting of RepU final destination.

3. Conclusion

Isotope separation technologies are crucial in efficient uranium multiple recycling. Enrichment advancements are primarily important at pushing the limits of RepU massive usage and limits of natural uranium saving.

If proposed advanced configurations are assembled (which is not that difficult), such benefits, as natural uranium economy and, in some cases, separative work economy, will be received right away.

Finally, technical and economic analysis is necessary for the right decision over cascade scheme. The right choice between proposed methods will play the main role in establishing the most optimal way of solving uranium re-enrichment task, which is vitally important for nuclear fuel cycle closing. Feasibility study also could help with a choice of best strategy and with establishing optimal share of different ways to close nuclear fuel cycles. Holistic approach would certainly be useful, if every entity in nuclear supply chain was willing to work toward shared goals.

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