A method to enrich reprocessed uranium with various initial contents of even-numbered isotopes

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Andrei Smirnov, Vladislav Gusev, Georgy Sulaberidze, and Vladimir Nevinitsa

A Method to Enrich Reprocessed Uranium with Various Initial Contents of Even-numbered Isotopes

Andrei Smirnov[1,a)], Vladislav Gusev[1,b)], Georgy Sulaberidze[1] and Vladimir Nevinitsa[2]

[1]*National Research Nuclear University "MEPhI", 31 Kashirskoe road, 115409, Moscow, Russia.*
[2]*National Research Centre Kurchatov Institute, 1 Akademika Kurchatova sq., Moscow, 123182, Russia*

[a)]Corresponding author: a.y.smirnoff@rambler.ru
[b)]gusevvlad13@mail.ru

Abstract. Nuclear industry needs effective recovering of fissile material from used nuclear fuel, as uranium accounting more than 90% of the spent nuclear fuel (SNF) volume. Use of reprocessed uranium (RepU) is associated with the difficulties due to the presence of ^{232,234,236}U isotopes in its composition. The content of these isotopes in fresh fuel is limited in accordance with specifications for low-enriched uranium (LEU). In this regard, in order to make a product of the required quality, it is necessary to modify the regular cascade scheme for enriching natural uranium and/or partially dilute the RepU with raw materials not containing ^{232,234,236}U (for example, natural uranium or depleted uranium). To solve such problems, a number of cascade schemes have been proposed for the last decades. However, there is still no answer what kind of scheme is preferable. In addition, most of them are unsuitable for full reuse of uranium extracted from spent fuel. Within the framework of the present paper, a double cascade scheme is proposed that allows a full use of reprocessed uranium (of any composition, including "dirty" one) in fuel production in compliance with restrictions on even-numbered isotopes. The "quasi-ideal" cascade, widely used in modeling separation processes in cascades for the separation of multicomponent mixtures, was chosen as the object of this theoretical study. The physical regularities of mass transfer in the proposed cascade scheme are analyzed. The interdependencies of cascade parameters are studied. It is shown that this scheme can be effectively employed to enrich the RepU of "dirty" composition, which are typical for SNF after several irradiation cycles.

INTRODUCTION

The issue of using uranium and plutonium repeatedly in light-water reactors (LWRs) has become even more important due to the current transition of the Russian nuclear industry (and some other global participants) to the closed nuclear fuel cycle. One of the reasons for the emergence of this trend is the continuous and inevitable accumulation of spent nuclear fuel (SNF) from LWRs operating worldwide [1, 2]. For the Russian nuclear industry, the problem of fuel treatment is even more pressing due to the necessity to provide nuclear fuel for overseas nuclear power plants and then to bring it back for the following processing. An additional factor causing interest in nuclear materials recycling is the anticipated shortage of natural uranium by the end of the century.

Obviously, as the uranium is the predominant material (more than 90%) in the SNF, it should be considered as the main stock for the subsequent fuel reproduction, so, the reprocessed uranium can partly 'replace' the natural one, acting as an alternative feedstock for LWR fuel production. It is certainly helpful in solving at least two problems: (1) the natural uranium resource scarcity, (2) the growing accumulation of spent nuclear fuel in interim storages. In addition, such an approach drastically reduces the volume of high-level wastes (HLW), therefore eases the burden of its final disposal – one of the most complicated problem in nuclear industry [3, 4].

Various studies on the issues of uranium multiple recycling in light-water reactors show that the major obstacle on the way to achieve widespread uranium reusing is the presence of even-numbered isotopes ^{232}U, ^{234}U и ^{236}U in reclaimed uranium, let alone the high cost of the reprocessing itself [5-8]. The content of these isotopes in low-enriched uranium LEU is subject to strict requirements related to the need to fulfill radiation safety conditions when fabricating nuclear fuel and preserving its neutron-physical characteristics in the reactor

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core [8]. In this way, firstly, the concentration of ^{232}U isotope in final product is strictly limited by $2 \cdot 10^{-7}$ %, or sometimes $5 \cdot 10^{-7}$ % (percent by weight) and the presence of ^{236}U leads to the necessity to increase the ^{235}U concentration to compensate the first one [8].

The presence of such restrictions complicates the enrichment of the reprocessed uranium and, in most situations, requires modification of the standard enrichment cascade [9-18]. The key problem here is that the enrichment of reprocessed uranium is not just a task of enriching a multicomponent mixture of uranium isotopes, but the task of fulfilling all the requirements on the concentrations of 'undesirable' isotopes ($^{232,234,236}U$), while reaching the necessary concentration of the fissile target component - ^{235}U.

The situation could be complicated further by the following fact: the uranium recycling could (and it is a reasonable from the control measures point of view) lay down the additional condition – to spend the reprocessed uranium per unit of production at a prerequisite level. For instance, chemical reprocessing of 1 kg of SNF, yield ~0.9 kg of uranium, which we suppose to make use of to produce 1 kg of fresh fuel. Only in this case it is be possible to talk about the real closure of the nuclear fuel cycle (NFC) for the uranium component of the fuel.

Calculations show that in most schemes (even modified ones), it is not possible to withstand the conditions described above [19]. However, in some cases, the compliance with the prescribed ratio between the mass of the reprocessed uranium to be enriched and the mass of the final product (LEU) is a must owing to nuclear fuel export control over the fissile material flow.

All in all, we try to produce reactor-grade low-enriched uranium (LEU) that meets all the $^{232,234,236}U$ presence requirements, while spending the reprocessed uranium per unit of production at a prerequisite level.

This work aims to develop a cascade scheme for reprocessed uranium enrichment, which would allow soling the above-formulated problem for any given composition of this feedstock. By "any given composition" we mean that it can correspond to the repeatedly recycled uranium characterized by relatively high concentrations of isotopes ^{232}U и ^{234}U, many times higher than permissible limits.

CASCADE SCHEMES: A BRAND-NEW CONFIGURATION AND ITS PRECURSORS

Analysis of Schemes for Reprocessed Uranium Enrichment

To date, there are approximately 20 different cascade schemes that could enrich the reprocessed uranium of different initial composition with simultaneous fulfillment of the conditions on even-numbered isotopes [9–23]. Summarizing, these schemes can be divided into three categories: 1) schemes based on the use of a triple-flow ('ordinary') cascade; 2) schemes based on the use of cascades with additional external flows (inputs or outputs); 3) double cascade schemes (combined ordinary cascades). These schemes are illustrated in Fig. 1-4.

The analysis shows that most of the proposed schemes either cannot, in principle, solve the problem formulated above, or are able to solve it only for the case when the concentrations of even-numbered isotopes are below the permissible limits (such compositions are typical for the first recycle, when the materials were used only once).

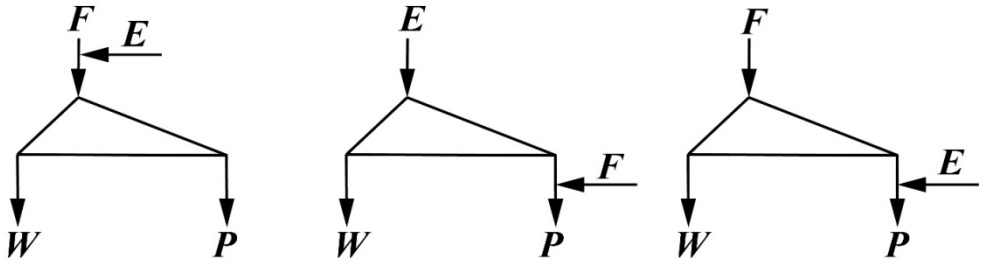
Let us briefly outline these basic options.

Ordinary Cascades

The ordinary cascade modifications for reprocessed uranium enrichment are usually associated with a diluting agent that does not contain even-numbered isotopes $^{232,236}U$ (natural uranium is used as such diluent mostly). For example, the mixing happens at the entrance to the cascade, or at the exit (Fig. 1) [5, 21]. However, a more efficient method is to direct diluent as the additional feed to the intermediate section of the cascade [5, 12, 21], because in this case it is possible to minimize the separative work losses when mixing streams with different isotope concentrations ^{235}U (Fig. 2). As a diluent, besides natural uranium, a LEU or a depleted uranium, as they are also do not contain artificial isotopes $^{232,236}U$, could be used as well. Their combination (Fig. 3) is also possible [14].

However, the main idea of the schemes presented in Fig. 1-3 is that by using diluents it is possible to reduce the relative concentration of isotopes ^{232}U and ^{235}U in the cumulative feed of the cascade, thereby staying within the allowable range of ^{232}U and other undesirable isotopes in product. Moreover, in these schemes, there are no parameters that allow reducing the relative concentrations of ^{232}U to ^{235}U across the cascade stages during the centrifugation process. This leads to the fact that enrichment of the reprocessed uranium with simultaneous fulfillment of: (1) restrictions on the concentrations of even-numbered isotopes, and (2) condition of the complete return of the material to the nuclear fuel cycle; is possible only if the ^{232}U concentration in the initial

feedstock of reprocessed uranium is relatively small and only slightly exceeds the allowable limits for the final LEU product.



(a) (b) (c)

FIGURE 1. The ordinary cascade modifications for reprocessed uranium enrichment: *F* – natural uranium; *E* – reprocessed uranium; *P* – product; *W* – waste.

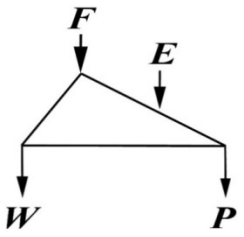


FIGURE 2. Cascade with additional feed: *F* – natural uranium; *E* – reprocessed uranium; *P* – product; *W* – waste.

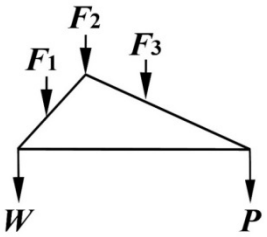


FIGURE 3. Cascade with two additional feeds: F_1 – depleted uranium; F_2 – natural uranium; F_3 – reprocessed uranium; *P* – product; *W* – waste.

The research on multiple uranium recycling shows that the concentration of ^{232}U in the initial material, exceeds the accepted limits for LEU, usually starting from the second recycling [4]. This circumstance makes it impossible to return all the reprocessed uranium to fuel cycle (Fig. 1-3).

Double Cascades for Reprocessed Uranium Enrichment

A solution to the problem of even-numbered isotopes in the reprocessed uranium may be based on the use of a consequent connection of ordinary cascades — double cascades [9, 16-18, 22, 23]. The main idea of using such a cascade scheme is that due to the higher number of flows, and, consequently, a larger number of parameters to control the process, it is possible to achieve to concentrate ^{232}U and ^{235}U in different outgoing flows.

Fig. 4 depicts one of the possible double cascade configurations for reprocessed uranium enrichment. The purification and enrichment are achieved in such cascades in the following way: the first cascade at the P_1 enriches ^{235}U with a simultaneous increase in the concentrations of isotopes ^{232}U and ^{234}U; then, this flow feeds the second cascade, where this intermediate product is divided into a light (^{232}U, ^{233}U, ^{234}U) and heavy fractions (^{235}U, ^{236}U). The last one is responsible for LEU production. And the content of ^{236}U in it meets the conditions of parasitic absorption compensation.

The obvious advantage of this scheme is that there is no need for natural uranium. However, significant drawbacks are:

– radiation contamination of both cascades;

– intermediate product - highly enriched uranium (in the P_2) where ^{235}U concentration could exceed 20% level [19], which complicates the situation with the international standards for the treatment of fissile materials;

–this scheme could not provide the complete return of the material into the NFC, because it consumes more than 1 kg of reprocessed uranium per unit of LEU product.

The last flaw means that it is impossible to solve the enrichment problem for reprocessed uranium in the general statement described above. In other words, double cascade scheme is not able to provide the LEU for a reactor using only reprocessed uranium obtained from the SNF of the same reactor. Therefore, to ensure the required mass of LEU in the reactor load, it is necessary to use the supplement, for example, fuel from natural uranium. This circumstance eliminates the main advantage of the double cascade – no consumption of natural

uranium. Since the use of such scheme cannot produce a fuel for a single reactor loading without natural uranium supplement, which significantly reduces its savings, thereby putting this cascade scheme in one row with the modifications of ordinary cascade.

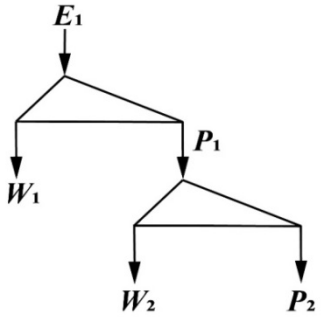


FIGURE 4. Double cascade for reprocessed uranium enrichment: E_1 – reprocessed uranium; P_1 – product flow of the first cascade, directing as a feed to the second cascade; P_2 – product flow of the second cascade; W_1 – waste flow of the first cascade; W_2 – heavy fraction flow of the second cascade – the final LEU product.

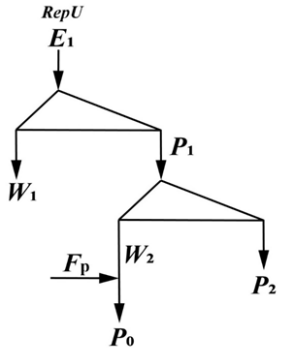


FIGURE 5. Double cascade for reprocessed uranium enrichment: E_1 – reprocessed uranium; P_1 – product flow of the first cascade, directing as a feed to the second cascade; P_2 – product flow of the second cascade; W_1 – waste flow of the first cascade; W_2 – heavy fraction flow of the second cascade; F_P – LEU-diluent; P_0 – final product (commercial reactor-grade LEU).

Considering this, a modification of the double cascade has been proposed recently. The new scheme makes it possible to enrich the reprocessed uranium while fulfilling the restrictions on even-numbered isotopes and the condition of complete return (Fig. 5) [19].

Such scheme is similar to the double cascade considered above. The key difference is that W_2 is not the final product. The final step in obtaining a commercial product is to dilute the flow W_2 by a material that do not contain artificial isotopes of uranium to fulfill the restrictions on them. At the same time, in order to fulfill the condition of complete return of the reprocessed uranium to the NFC, the mixing ratio is actually predetermined, since at this point the proportion of flows W_2 to E_1 is known and the value of the additional diluent flow is chosen so as to ensure the specified consumption of the reprocessed uranium per unit of the final LEU product [19].

In this case, the only parameter that allows you to influence the concentration of ^{235}U in the product is the concentration of this isotope in the diluent (F_p). To obtain a product that meets the specified conditions for all the isotopes, this concentration must be strictly defined. Calculations show that low-enriched uranium should be used as a diluent. This diluent can be obtained, for example, by mixing LEU with a concentration of 5% and depleted uranium [19].

According to [19] the scheme shown in Fig. 5 allows to enrich the reprocessed uranium even after five consecutive recycles in the fuel of the VVER-1000 reactors ('dirty' uranium composition) when a ratio of the consumption of the reprocessed uranium to product is equal to 0.93, while satisfying the restriction on the concentration of isotope ^{232}U - $5 \cdot 10^{-7}$%.

Thus, at least formally, such scheme solves the problem. However, it is obvious that there is a problem (as of other double cascades) of handling the light fraction of the second cascade (P_2).

Despite the fact that P_2 makes up only a few percent of the main product flow, it cannot be considered as a waste that could be sent for the long-term storage, because this is a highly active radioactive product due to a significant (1-2 orders of magnitude) increase in ^{232}U concentration comparing to the initial mixture. Worth noting that P_2 contains up to 20% of the isotope ^{235}U. These factors make such material: (1) dangerous due to the relatively high level of radioactivity, (2) sensitive to proliferation of nuclear fissile materials.

Some patents analyze the possible fate of this troublesome material. For example, [23] proposed to use depleted uranium as diluent to decrease the ^{235}U concentration until it reaches 1% and then store or use it for fuel production. So, we get additional material, though not a regular waste, but sort of intermediate product that needs to be used for the sake of complete recycling of uranium. Basically, only by making use of P_2, we ensure the full closure of the fuel cycle on the uranium component.

The Idea of Triple Cascade for Reprocessed Uranium Enrichment

Considering the drawbacks of the previously discussed cascade schemes, another modification of the double cascade is presented on Fig. 5. Here the "troublesome" flow (P_2) is mixed with depleted uranium and sent to the

subsequent cascade (Fig. 6). The mixing ratio is determined in such a way that with the enrichment of the mixture 'P_2 + *DepU*' it is possible to produce LEU of commercial quality, i.e. with even-numbered isotopes constraints fulfilled. This scheme could be named as a triple cascade. Its parameters should be selected in such a way that the final product consists of two LEU flows (P_3 and P_4 on Fig. 6). By varying the concentration at the outputs of the first two cascades, as well as in the flow of LEU diluent (F_P) it is possible to provide the specified mass ratio of the final product and the initial reprocessed uranium, thereby fully solving the problem of highly active waste. Analysis of this scheme allows us to state that in addition to the final product (commercial LEU – P_0) this scheme produces only regular waste (conventional depleted uranium) on W_1 and W_4 (for these byproducts the procedures are known and technologically proven).

RESULTS AND DISCUSSION

For the proposed scheme of reprocessed uranium enrichment, a series of computational experiments has been conducted to verify its efficiency. It was done for the reprocessed uranium that passed through several recycles in the fuel of the VVER-1000 reactor (Table 1) [20]. Separated mixture – uranium hexafluoride, which is the working substance in the enrichment of uranium by the gas centrifuge [24-27]. Inputs:

– ^{235}U enrichment in LEU – 4.95 %;
– Separation factor for $^{235}UF_6$ to $^{238}UF_6$ is 1.2 [19];
– The concentration of ^{232}U in LEU is limited by $5 \cdot 10^{-7}$% [19];
– Loss of reactivity due to the presence ^{236}U compensated by additional enrichment of ^{235}U, reactivity compensation coefficient equals 0.29 [5];
– 'Consumption' of reprocessed uranium per unit of the product was considered equal to 0.93 kg per 1 kg of LEU [19].
– Natural uranium savings in the scheme should not be less than 35%.

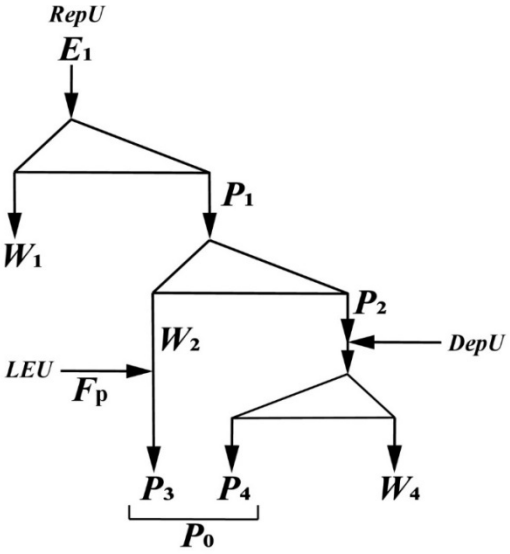


FIGURE 6. Triple cascade for reprocessed uranium enrichment: E_1 – reprocessed uranium; P_1 – product flow of the first cascade, directing as a feed to the second cascade; P_2 – product flow of the second cascade; W_1 – waste flow of the first cascade; W_2 – heavy fraction flow of the second cascade; F_P – LEU-diluent; P_0 – final product (commercial reactor-grade LEU); P_3 – LEU_1; P_4 – LEU_2; W_4 – waste flow of the third cascade; *DepU* – depleted uranium.

For modeling of the enrichment of the reprocessed uranium in the proposed scheme, a model of a "quasi-ideal" cascade with non-mixing by relative concentrations of the selected pair of components (Matched Abundance Ratio Cascade) was used [28, 29]. As a result, the overall cascade design was calculated in order to derive all the necessary parameters (numbers of stages and lengths of sections) and integral characteristics.

As a baseline we chose the following schemes: 1) three-flow ordinary cascade for natural uranium enrichment; 2) the simplest modification of a three-flow ordinary cascade to enrich the reprocessed uranium, diluted by natural uranium (left scheme) on Fig. 1). Both reference schemes provided LEU of the same commercial quality.

TABLE 1. Reprocessed uranium isotopic composition.

Index of component	Mass number	Concentration, %
1	232	$1.03 \cdot 10^{-6}$
2	233	$1.30 \cdot 10^{-6}$
3	234	$3.91 \cdot 10^{-2}$
4	235	1.07
5	236	1.45
6	238	The remains

The key integral characteristics for comparison were the values of natural uranium savings and the number of centrifuges in the cascades.

When calculating the parameters of the cascade, we dealt with the system of nonlinear equations, formed by closure errors for given concentrations of components in the outgoing flows. The solution of this system was carried out using numerical methods. In this case, the relative error in finding the unknown parameters of the cascade was 10^{-7}. Table 2 shows the isotopic composition of commercial-level LEU obtained in proposed triple-cascade. Fig. 7 presents the magnitude of the natural uranium savings, the consumption of reprocessed uranium per unit of product and the number of centrifuges for proposed triple-cascade scheme, modified ordinary cascade, and basic cascade, which enriches natural uranium. The number of centrifuges for all variants is normalized to that in the cascade for the enrichment of only natural uranium.

TABLE 2. Isotopic composition of LEU product ($P_3 + P_4$) of triple-cascade scheme.

Mass number	232	233	234	235	236	238
$C_i^{P_0}$, %	$5.00 \cdot 10^{-7}$	$6.88 \cdot 10^{-7}$	$5.31 \cdot 10^{-2}$	5.11	0.57	The rest

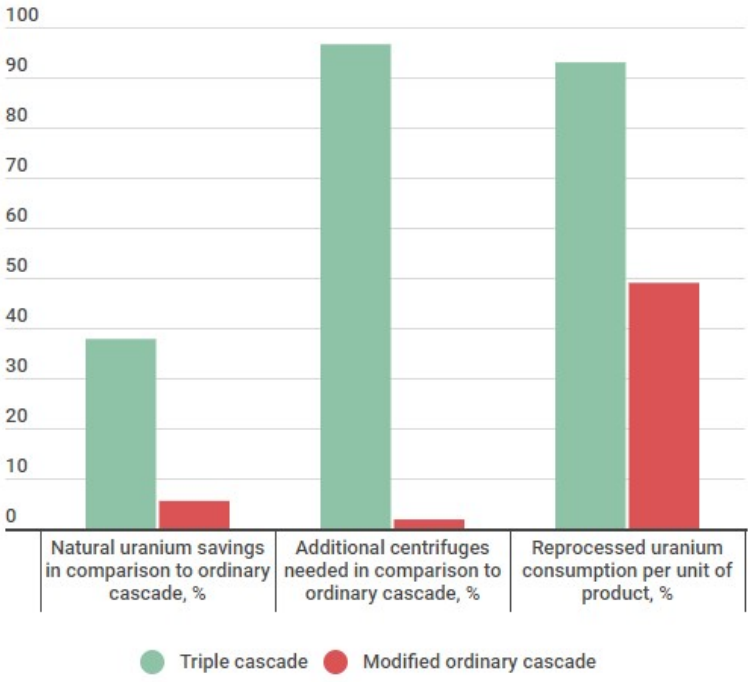


FIGURE 7. Integral parameters of evaluated cascade schemes.

These results show that the proposed scheme solves the task formulated in this paper. A comparison with the reference cascade (ordinary one) shows that even for the chosen 'dirty' composition of the reprocessed uranium, it is possible to save more than a third of natural uranium, which is a lot more than the simple modifications could give [5]. However, such benefits entail the increase in separative work, hence the number of centrifuges, by approximately 97%, comparing to conventional cascade that enriches natural uranium. The scheme also makes it possible to produce a commercial quality LEU by spending a predetermined amount of reprocessed uranium and do it without giving off of any undesirable by-products, except for the standard tails (waste flows)

of the separation cascades. Worth mentioning that the example mentioned is for illustrative purposes only. To put this scheme in practice, first of all, we should optimize it according to the selected efficiency criterion.

As an addition to the results described above, we will calculate the material balances in this scheme on the assumption of producing precisely 21 tons of LEU. Approximately this mass of uranium is required to load the VVER-1200 reactor with fuel rods with an enrichment of 4.95%.

Knowing that E_1 to P_0 ratio equals 0.93, the reprocessed uranium is going to be spent at the rate of 19.53 tons per 21 tons of final LEU product. In our case, the first cascade gives off 17.5 tons of depleted uranium from W_1, yielding ≈2.03 tons of P_1 with concentration of ^{235}U is equal to 9.41%. The latter flow P_1 acts as a feed of the second cascade, which produce a 'purified' mixture W_2 (1.7 tons that has 7.34% of ^{235}U) and contaminated P_2 (≈332 kg) that has 20% of ^{235}U. This P_2 goes to the third cascade, and is diluted there by 3298.88 tons of depleted uranium (with ^{235}U concentration only 0.1%, which is not the best quality). In the third cascade the depleting part consists of just 1 stage, giving out ≈ 3293.1 tons of waste W_4 with 0.093% of ^{235}U. LEU-diluent F_p≈13.2 tons is mixed with 1.7 tons of W_2 forming ≈14.9 tons of P_3. The final product also comprises 6.1 tons of P_4, thus we have $P_3 + P_4 = P_0$=21 tons.

The cascade that produce LEU-diluent F_p (concentration of ^{235}U is equal to 4.9%) in our case consumes ≈103.6 tons of natural uranium, releasing 90.4 tons from its waste flow (0.1% of ^{235}U). As a result, our scheme gives off (90.4+17.5+3293) ≈3401 tons of depleted uranium tails. At the same time, the scheme takes ≈3300 tons of stored depleted uranium. Consequently, the actual outturn of depleted uranium equals ≈100 tons. At the same time, the ordinary cascade gives off ≈146 tons, which means that, as a side effect, we could reduce the accumulation of tails by half.

Of course, it should be noted that the 'mobilization' of a significant amount of depleted uranium from a warehouse will require additional expenses, but on the other hand, the storage of this material is a problem itself, and its use have been the subject of discussion for more than a decade [30-31].

CONCLUSION

To solve the problem of reprocessed uranium complete recycle it is proposed to use a triple-cascade scheme. The calculations of "quasi-ideal" cascade model showed that such approach make it possible to produce a commercial quality LEU by spending a given amount of reprocessed uranium, while meeting the ^{232}U standard specifications (and conditions laid on other even-numbered isotopes). At the same time, the proposed scheme provides more natural uranium savings than the majority of schemes for reprocessed uranium enrichment (37%). The material balances for the production of 21 tons of commercial LEU were estimated. It is shown that it produces a smaller amount of depleted uranium (by about 50%) than the standard cascade for enriching natural uranium to produce an equivalent LEU.

ACKNOWLEDGEMENTS

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