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Original Article

A method for purifying reprocessed uranium from even isotopes under conditions of multiple recycle

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ABSTRACT

We proposed a modification of a double cascade scheme to enrich reprocessed uranium. Such a cascade scheme represents a combination of one cascade with "broadening" of the flow and an ordinary three-flow cascade. A calculation and optimization method has been developed for the proposed scheme according to various efficiency criteria.

It is shown that the proposed scheme makes it possible to obtain low-enriched uranium of commercial quality using reprocessed uranium of different initial compositions. For example, the enrichment of reprocessed uranium, which had gone through five consequent recycles, was considered. The proposed scheme allowed to enrich it with simultaneous fulfillment of restrictions on isotopes ^{232}U, ^{234}U, and ^{236}U. Such results indicate the scheme's applicability under conditions of multiple recycling of uranium in reactor fuel.

Computational experiments have shown that in the proposed modification, a noticeable saving of natural uranium in the cycle (~18%) can be achieved, provided that the additional consumption of separative work does not exceed 10%, compared with the case of enrichment of natural uranium to obtain LEU of equivalent quality.

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1. Introduction

1.1. Background

Light water reactors on thermal neutrons make up most power reactors worldwide. In this regard, there is a problem of handling their spent nuclear fuel (SNF) due to its continuous accumulation [1,2]. According to some estimates, the annual increase in SNF in the world is ~11 thousand tons, and the total mass of the accumulated fuel is ~400 thousand tons [3].

The bulk of spent nuclear fuel (excluding structural materials) is uranium, which has a higher concentration of ^{235}U than natural uranium (NatU). The involvement of such reprocessed uranium (RepU) in nuclear fuel production can reduce the volume of spent nuclear fuel disposal, reduce the costs of separative work in the fuel cycle, and partially replace natural uranium as a raw material [2,4,5]. The latter helps to save some natural uranium which is an important part of rational usage of non-renewable resources.

The RepU separated from SNF can be used in various ways in the production of nuclear fuel: a) separately from plutonium (regenerated uranium fuel - RUF), b) as part of mixed types of fuel (MOX, REMIX) [6,7]. Most options for both types of fuel require pre-enriched reprocessed uranium.

However, use of RepU for producing fresh fuel has some serious drawbacks:

- Nuclear reprocessing technology for separating and recovering fissionable materials (uranium and plutonium) is quite expensive and complicated. Moreover, it brings additional risks for nuclear safety;
- Re-enrichment process and handling of uranium hexafluoride made of RepU have many difficulties associated with artificial uranium isotopes (first of all, ^{232}U, ^{236}U), which appears in fuel during its burning in the reactor core.

Let's consider the latter item.

Abbreviations: NatU, Natural uranium; LEU, Low-enriched uranium; RepU, Reprocessed uranium; RUF, Regenerated uranium fuel; SNF, Spent nuclear fuel; SWU, Separative work unit.

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Enriching the reprocessed uranium can be solved by means of gas centrifuge cascades used to enrich natural uranium. However, its solution is complicated due to the presence of artificial isotopes ^{232}U and ^{236}U in the RepU and a higher content of ^{234}U in relation to the natural mixture.

In accordance with the technical conditions in force, for example, international specifications for low-enriched uranium, the permissible concentrations of ^{232}U and ^{234}U isotopes in commercial LEU are limited to certain values [8,9]. The presence of ^{232}U significantly increases the radiation background of the material, which creates problems during the manufacturing of fresh fuel assemblies and the subsequent handling of these fresh fuel assemblies at nuclear power plants. Main contributors to the radiation background are decay products of ^{232}U (^{228}Th, ^{212}Pb, ^{208}Tl, others) which include sources of hard γ-rays with energies of up to 2.6 MeV [8]. So, to cope with such problems it is necessary to purify RepU hexafluoride from impurities and fission products. Prevention consequences of further appearance of fission products limits the range of time needed to enrich RepU and fabricate fuel assemblies. Also, these factors lead to necessity to fulfill some protective measures for the personnel of enrichment plants as well as using shielded gloveboxes to protect workers of fuel fabrication plants [5].

The presence of the ^{236}U isotope in the RepU, which is an active absorber of thermal neutrons, impairs the multiplying characteristics of the fuel and, therefore, requires an increase in the ^{235}U enrichment to restore them [4]. As a consequence, this presence leads to additional costs of separative work [4].

Not only restrictions on the concentration of even isotopes can be implemented. In some cases, it is necessary to return to the production of low-enriched uranium (LEU) the entire mass of the RepU from SNF. Such a condition means closing its fuel cycle in terms of the uranium component for a single reactor. It means that the RepU must be consumed entirely during fuel fabrication for the subsequent loading of the same reactor [6,10]. Such a condition, first, is stipulated for legal reasons. If using the whole amount of RepU to produce fresh nuclear fuel, it is possible to prevent the accumulation of RepU in warehouses and ensure the efficient return of ^{235}U from spent nuclear fuel to the cycle. Additionally, it simplifies control of the fissile material balance. Further, such a condition will be called the “full return” of the reprocessed uranium.

Another factor strengthens the mentioned above troubles. The problem is that if using uranium in several consequent recycles a degradation of its isotopic composition takes place. This means that concentrations of even isotopes grow significantly [4,11]. It is important to take this factor into consideration while suggesting how to enrich the RepU. Therefore, the enrichment method should provide a solution to the problem in a relatively wide range of initial concentrations of even isotopes in the regenerate.

Thus, using of RepU along with the profits brings some challenges to nuclear industry. All these issues should be taken into account during technical and economical assessment of possible variants to close nuclear fuel cycle of light water reactors.

In this paper we focused on the issues connected with the RepU enrichment in cascades of gas centrifuges. The problem of RepU enrichment from the viewpoint of separation technologies is more complicated than the enrichment of natural uranium. The separation task can be described as follows. It is necessary to produce a given mass of commercial LEU under the following conditions:

- the concentration of the ^{235}U isotope in the resulting LEU corresponds to a given value;
- the concentration of ^{232}U in LEU should not be higher than allowable limits;
- the ratio of concentrations of ^{234}U and ^{235}U in LEU should be lower than a given value;
- an additional enrichment of the ^{235}U isotope must compensate the parasitic absorption of neutrons by the ^{236}U isotope.
- the “full-return” condition.

The requirements listed above are only necessary conditions, and not sufficient ones. We have considered them in detail because they are important in modeling the process of enrichment of RepU in gas centrifuge cascades. Simultaneously, it is important to mention that the reuse of RepU for the production of fuel for thermal neutron reactors requires more extensive changes in the entire fuel production chain, from adjusting technologies and means of protection at separation plants and fuel fabrication plants to even changing legislative acts. For example, if the concentration of ^{235}U exceeds 5%, which is necessary to compensate for the impact of ^{236}U, new licensing of enrichment plants may be required [5]. Obviously, when deciding on the reuse of reprocessed uranium, all of these factors must be taken into account to correctly calculate the unit costs in such a fuel cycle. Only on the basis of such economic assessments it is possible to decide on the expediency of implementing various nuclear fuel cycle options.

1.2. Existing methods to solve the problem

In most cases, it is impossible to solve the described problem in an ordinary or three-flow cascade (incoming flow: external feed, and outgoing flows: product and waste) used for enriching natural uranium [10].

Several methods to enrich the reprocessed uranium have been proposed so far [10–27]. Many of them are based on various options for diluting the RepU with materials that do not contain even isotopes. Some other methods allow to separate the fractions containing ^{235}U and ^{232}U partially. Mostly they are based on modifications of double cascades [14–17,19]. Significant disadvantages of diluting methods and double cascades are the impossibility of fulfilling the condition of “full return” along with all requirements on concentrations of ^{232}U, ^{234}U, ^{236}U isotopes.

There are a few methods for enriching the RepU, which makes it possible completely to solve the problem. We call them “combined schemes” since they represent a combination of the “dilution” of the RepU and its purification [10,26,27]. These methods are based on the use of multi-cascade schemes, consisting of two [26] or three cascades [10,27] and a separate cascade for enriching natural uranium to obtain low-enriched uranium-diluent.

The disadvantages of such schemes are either the presence of a waste fraction representing a radiation hazard [26] or a noticeable increase in the cost of separating work in the fuel cycle [27].

A method is based on the use of the so-called cascades with “flow broadening” [20]. Originally such cascades were proposed to concentrate isotopes of intermediate mass numbers in the separation of multicomponent mixtures [28]. However, the problem of this method is the failure to meet the condition of “full return”, as well as the presence of highly enriched byproduct waste.

Considering the above, an urgent task for theoretical research and subsequent practical application is the development of a cascade scheme that would simultaneously ensure the saving of natural raw materials and the separative work when enriching reprocessed uranium with an arbitrary initial composition.

In this paper, we propose a modification of a double cascade scheme for enriching reprocessed uranium, which is a combination of a cascade with “broadening” of the flow and a three-flow cascade for enriching natural uranium. The advantages of the proposed scheme in relation to the known modification of double cascades are shown.

2. Description of the cascade scheme and methods for calculating its parameters

2.1. Description of the scheme

The cascade scheme proposed in work is described below.

The proposed approach uses the principle of separating intermediate isotopes from multicomponent mixtures of stable isotopes in cascades with additional product flows [28,29].

Considering that the ^{235}U isotope is intermediate in mass number in the mixture of reprocessed uranium, this method can also be used to concentrate this isotope during the enrichment of RepU.

The main idea of cascades with "broadening" of the flow can be explained as follows.

It is possible to achieve a relatively high concentration of the target intermediate component on the internal stages by choosing certainly the form of the distribution function of the feed flow over the stages of the cascade.

If to withdraw an additional product flow at a stage in the region of the maximum concentration of the target intermediate component within the cascade, it is possible to obtain a fraction with the maximum content of this isotope compared to it than in the withdrawal at the end of the cascade [28].

In this work, a modification of the cascade scheme is proposed, which allows solving the problem of enrichment of RepU with the satisfaction of all the requirements described above.

The proposed modification includes two cascades: the first has a "broadening" of the flow and an additional withdrawal, in which a mixture enriched in ^{235}U is obtained, the second cascade produces a low-enriched uranium-diluent (LEU-diluent) for the final correction of the isotopic composition of the obtained commercial LEU, as well as its given mass.

Let's consider in more detail the proposed scheme.

The flow of RepU F_{rep} is fed to the input of the first cascade (cascade 1 in Fig. 1).

There are three outgoing flows from the first cascade: waste flow W_1, additional product flow E, and end product flow P_1.

In P_1, the maximum concentration of ^{235}U is achieved (up to several tens of percent).

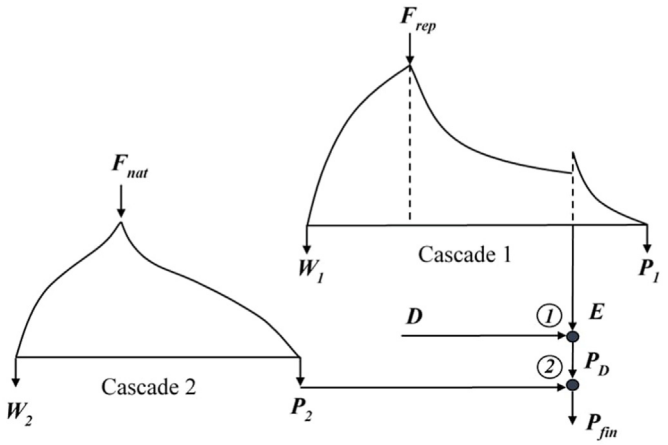

Fig. 1. The modification of a double cascade, consisting of a cascade with "broadening" and an ordinary cascade to produce LEU-diluent from natural uranium. Abbreviations: F_{rep} is a flow of initial RepU; P_1 is a flow enriched with light components in the cascade 1; W_1 is a waste flow of the cascade 1; E is a flow of additional product of the cascade 1; D is a depleted uranium; P_D is a flow of enriched RepU diluted with depleted uranium; P_{fin} is commercial LEU; F_{nat} is natural uranium, which is fed to the second cascade to produce LEU-diluent; P_2 is a flow of the "light" fraction of the auxiliary cascade; W_1 is a waste flow of the auxiliary cascade.

At the first step of enrichment, in cascade 1, flow E is obtained, which is included in the stage corresponding to the maximum distribution of the ^{235}U isotope concentration. Next, flow E is mixed with depleted uranium (flow D) to obtain low-enriched (P_D) uranium.

The use of depleted uranium allows one to reduce the content of even isotopes, as well as to correct the concentration of ^{235}U without using natural uranium.

At the second stage, to obtain a commercial product, it is necessary to mix the resulting mixture with low-enriched uranium-diluent, which is obtained by enriching natural uranium in a separate three-flow cascade.

The ^{235}U concentration is chosen equal to or close to the ^{235}U concentration in P_D flow and in the final P_{fin} flow to minimize the loss of separative work.

2.2. Governing equations for a Q-cascade with flow "broadening"

The Q-cascade model with "broadening" of the flow was used to simulate the enrichment of reprocessed uranium in such a scheme [28].

The Q-cascade is essentially a special case of symmetrical counter-current cascade since it has constant separation coefficients in stages.

Simultaneously, while maintaining the adequacy of the processes of mass transfer in cascades for separating multicomponent mixtures, this model makes it possible to simplify the procedures for calculating and optimizing their parameters [30–33]. This feature is the reason for the widespread use of the Q-cascade in modeling the separation of multicomponent mixtures.

Thus, this model is a convenient tool for studying the physical laws of mass transfer in such installations and evaluating their optimal parameters.

Let us briefly outline the basic equations of the mathematical model of the Q-cascade with the "broadening" of the flow [28,29].

Consider a cascade with one external feed flow F with concentrations C_i^F, and three outgoing flows: end withdrawal P (enriched with light components of the mixture) with concentrations C_i^P, additional withdrawal E with concentrations C_i^E, and a flow W with enriched heavy components and concentrations C_i^W (Fig. 2). The lengths (number of stages) of the sections of the cascade between the end and intermediate withdrawals will be denoted S_E, between

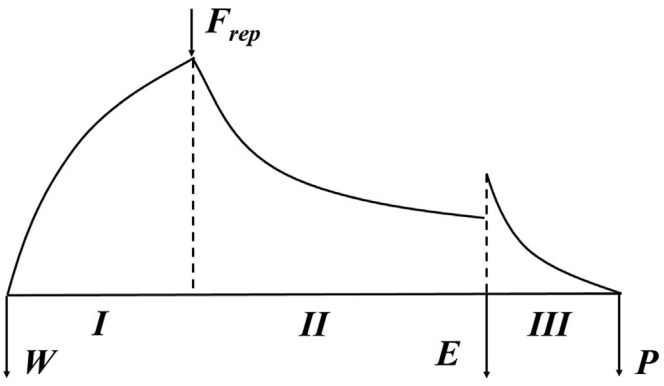

Fig. 2. The schematic drawing of the cascade with "broadening" of the flow and an additional product flow (E). The figure represents the qualitive form of the function for stage feed flow along the cascade length.

the flows W and $F - S_W$, between the flows P and $F - S_P$. From flow W to flow P the cascade has three sections which are numbered in Fig. 2. The number of components in a mixture under separation is m.

External flows and concentrations of components in them in a stationary mode of operation and the absence of losses of working substance should satisfy balance equations:

$$F - P - E - W = 0 \tag{1}$$

$$FC_i^F - PC_i^P - EC_i^E - WC_i^W = 0,\ i = \overline{1,\ m} \tag{2}$$

The internal parameters of the cascade include the feed flows of the stages and the concentration of components in the flows entering/leaving the cascade. The following equations connect concentrations of components and lengths of individual sections of such a cascade [28]:

$$KL_E C_i^E = 2PC_i^P \cdot \frac{1 - e^{-Q_i^{II} S_E}}{Q_i^{II}}, \tag{3}$$

where L_E is a flow at the point where the additional product flow is, K is a parameter which defines the value of flow "jump." $Q_{i\text{II}}$ is a constant value which meaning will be explain further as well as $_{Q\text{II}}$.

$$\begin{aligned} L_f C_i^f = {} & 2PC_i^P \left[\frac{1 - e^{-Q_i^{II} S_E}}{K \cdot Q_i^{II} \cdot e^{Q_i^I (S_P - S_E)}} + \frac{1 - e^{-Q_i^I (S_P - S_E)}}{Q_i^I} \right] \\ & + 2EC_i^E \cdot \frac{1 - e^{-Q_i^I (S_P - S_E)}}{Q_i^I}, \end{aligned} \tag{4}$$

$$L_f C_i^f = 2WC_i^W \cdot \frac{e^{-Q_i^I S_W} - 1}{Q_i^I}, \tag{5}$$

where L_f and C_i^f are a flow and concentrations of components at the point where the feed flow is entered the cascade.

Solving equations (3), (5), and (6) together, after transformations, we obtain the following solutions.

For sections II and III:

$$\frac{P}{F} = \sum_{j=1}^{m} \left\{ \frac{C_j^F \cdot \frac{e^{Q_j^I S_W} - 1}{Q_j^I}}{\frac{1 - e^{-Q_j^{II} S_E}}{K \cdot Q_j^{II} \cdot e^{Q_j^I (S_P - S_E)}} + \left[1 + \frac{E}{P} \frac{C_j^E}{C_j^P} \right] \frac{e^{Q_j^I S_W} - e^{-Q_j^I (S_P - S_E)}}{Q_j^I}} \right\} \tag{6}$$

$$C_j^P = \frac{F}{P} \cdot \frac{C_j^F \cdot \frac{e^{Q_j^I S_W} - 1}{Q_j^I}}{\frac{1 - e^{-Q_j^{II} S_E}}{K \cdot Q_j^{II} \cdot e^{Q_j^I (S_P - S_E)}} + \left[1 + \frac{E}{P} \frac{C_j^E}{C_j^P} \right] \frac{e^{Q_j^I S_W} - e^{-Q_j^I (S_P - S_E)}}{Q_j^I}}, \tag{7}$$

For section I:

$$\frac{W}{F} = \sum_{j=1}^{m} \left\{ \frac{C_j^F \cdot \left\{ \frac{1 - e^{-Q_j^{II} S_E}}{K \cdot Q_j^{II} \cdot e^{Q_j^I (S_P - S_E)}} + \left[1 + \frac{E}{P} \frac{C_j^E}{C_j^P} \right] \frac{1 - e^{-Q_j^I (S_P - S_E)}}{Q_j^I} \right\}}{\frac{1 - e^{-Q_j^{II} S_E}}{K \cdot Q_j^{II} \cdot e^{Q_j^I (S_P - S_E)}} + \left[1 + \frac{E}{P} \frac{C_j^E}{C_j^P} \right] \frac{e^{Q_j^I S_W} - e^{-Q_j^I (S_P - S_E)}}{Q_j^I}} \right\}, \tag{8}$$

$$C_j^W = \frac{F}{W} \cdot \frac{C_j^F \cdot \left\{ \frac{1 - e^{-Q_j^{II} S_E}}{K \cdot Q_j^{II} \cdot e^{Q_j^I (S_P - S_E)}} + \left[1 + \frac{E}{P} \frac{C_j^E}{C_j^P} \right] \frac{1 - e^{-Q_j^I (S_P - S_E)}}{Q_j^I} \right\}}{\frac{1 - e^{-Q_j^{II} S_E}}{K \cdot Q_j^{II} \cdot e^{Q_j^I (S_P - S_E)}} + \left[1 + \frac{E}{P} \frac{C_j^E}{C_j^P} \right] \frac{e^{Q_j^I S_W} - e^{-Q_j^I (S_P - S_E)}}{Q_j^I}}, \tag{9}$$

From expression (10) it is easy to obtain the following relations:

$$C_j^W = \frac{C_j^P \cdot \frac{1 - e^{-Q_j^{II} S_E}}{Q_j^{II}}}{\sum_{i=1}^{m} \left(C_i^P \cdot \frac{1 - e^{-Q_i^{II} S_E}}{Q_i^{II}} \right)} \tag{10}$$

$$\frac{C_j^E}{C_j^P} = \frac{C_n^E}{C_n^P} \cdot \frac{Q_n^{II}}{Q_i^{II}} \cdot \frac{1 - e^{-Q_j^{II} S_E}}{1 - e^{-Q_n^{II} S_E}} \tag{11}$$

Constants Q_i can be defined as follows [28]:

$$Q_i = \varepsilon_0 (M - M_i), \tag{12}$$

where M is a special parameter. The choice of particular value for M in a range $M_1 \le M \le M_m$ allows to determine all values for Q_i and a particular form of the function $L(s)$.

We will assume that the flow of the mixture to be separated is continuous along the length of the cascade, except for the points at which additional product flow and external feed are performed.

Specifying the value of M determines the nature of the distribution $L(s)$ and the ratio of flows P and W. In its turn it determines conditions under which components with mass numbers $M_i < M$ are enriched together with the lightest component to the "light" end of the cascade, and components with $M_i > M$, respectively, to the heavy one.

The principle of operation of a Q-cascade with "broadening" is as follows: if to set different values for the value of M in various sections of the cascade, so that in one of them – $M_n < M$ (n is the number of the target (desirable) component), and in another section – $M_n > M$, then it can be assumed that the distribution of the flow $L(s)$ corresponding to such a setting of the parameter M will be such that the n-th component in two neighbor sections will begin to enrich in opposite directions. As a result, one should expect an increase in the concentration of the n-th component in that region.

Let the parameter M has a value M^I with the corresponding constants Q_i^I ($i = \overline{1,\ m}$) in the sections between flows W, and E. In the section between flows E and P M takes on a value M^{II} with the corresponding constants Q_i^{II} ($i = \overline{1,\ m}$).

2.3. Optimization of the proposed scheme

Taking into account the general formulation of the problem to enrich reprocessed uranium described in Introduction, we will formulate the optimization problem for the proposed cascade scheme.

The following external parameters are specified:

- C_{235}^E is a concentration of ^{235}U in the flow E;
- C_{235}^{Pfin} is a concentration of ^{235}U in the final product – commercial LEU;
- C_i^{Frep} are concentrations of components (i = 1,2, …m) in the initial RepU;
- C_i^D are concentrations of components (i = 1,2, …m) in depleted uranium;

- C_i^{Fnat} are concentrations of components (i = 1,2, …m) in natural uranium;
- restrictions on the concentrations of even isotopes;
- the form of the function for calculating the value of the additional ^{235}U enrichment compensating for the effect of ^{236}U.

During calculation process, it is necessary to determine: lengths of sections of the cascades, the distribution function of the feed flows of the stages in each cascade.

As decision variables we used values $\epsilon_0 S_W$, $\epsilon_0 S_P$, $\epsilon_0 S_E$.

These values were varied to minimize residuals of concentrations of C_{235}^E, C_{235}^P and C_{235}^W.

When the values $\epsilon_0 S_W$, $\epsilon_0 S_P$, $\epsilon_0 S_E$ are found, the calculation of other parameters of the Q-cascade with "broadening" begins with use of relations (3)–(17).

After that, it is easy to calculate the ratio between flows E and D.

It helps to find the isotopic composition of the final product obtained by mixing the diluted RepU ($E + D$) with an LEU-diluent produced from natural uranium.

It is necessary to implement an algorithm that will consider the condition of compensation ^{236}U in the flow P_{fin} to solve the problem correctly (see Fig. 1).

That condition affects isotopic composition in the final product.

Since all diluents (depleted uranium and LEU) have a fixed concentration composition, only the ratio between mixed flows will affect the composition of the final product.

Finding the appropriate value for the mentioned above ratio minimizes the discrepancy in the ^{235}U concentration in the final product, considering the specific type of function for calculating the compensating additive for the ^{235}U concentration.

After determining the values of the flows of diluents, the remaining parameters of the cascade are calculated (integral characteristics of the cascade, the ratio of flows, etc.).

The Q-cascade model was also used to calculate the cascade parameters of the cascade producing the LEU-diluent.

Analysis of the above formulation of the problem shows many combinations of the parameters C_{235}^E, C_{235}^P and C_{235}^W to solve it.

Additionally, parameters M_1, M_2, and E/P give additional "degrees of freedom" to the system.

Thus, it is reasonable to optimize the scheme according to some efficiency criterion.

In the general case, when solving the optimization problem of the proposed scheme, it is necessary to find such a set of parameters M_1, M_2, C_n^E, C_n^P, C_n^W and E/P, at which the condition is valid:

$\Psi = \Psi\ (M_1, M_2, C_n^E, C_n^P, C_n^W, E/P) \rightarrow$ min (max).where Ψ is the target function (efficiency criterion).

The following values were considered as efficiency criteria: $\delta\left(\frac{F}{P}\right) = F_1(M_1, M_2, C_n^E, C_n^P, C_n^W, E/P)$ is a savings of natural uranium in the production of commercial LEU in comparison with the open fuel cycle; $\delta\left(\frac{SWU}{P}\right) = F_2(M_1, M_2, C_n^E, C_n^P, C_n^W, E/P)$ is an overexpenditure of the separative work when obtaining commercial LEU in relation to the open fuel cycle. Simultaneously, special attention was paid to the first criterion, since saving natural uranium is one of the main goals of re-enrichment of reprocessed uranium.

We should emphasize that the concepts of "separative work" and "separative work unit (SWU)" were first introduced for a binary mixture of natural uranium. For a multicomponent mixture of RepU, the concept of separative work is still under discussion [34]. Therefore, in the results below, we meant a conditional value directly proportional to the number of gas centrifuges in the cascade as separative work.

Below are the formulas for calculating the values of the chosen efficiency criteria:

$$\delta\left(\frac{F}{P}\right) = \frac{F_{nat} - F^*}{F_{nat}} \cdot 100\% \tag{13}$$

$$\delta\left(\frac{SWU}{P}\right) = \frac{\sum L_{nat} - \sum L^*}{\sum L_{nat}} \cdot\ 100\%, \tag{14}$$

where F_{nat} and $\sum L_{nat}$ are natural uranium consumption and the total flow in three-flow cascade for natural uranium enrichment to obtain the same amount of commercial LEU of the same quality, F^* and $\sum L^*$ are natural uranium consumption and the total flow in the proposed cascade scheme.

Another essential integral characteristic of the cascade is the recovery rate of ^{235}U – $\delta\left(\frac{M_P}{M_F}\right)$:

$$\delta\left(\frac{M_P}{M_F}\right) = \frac{\sum_i M_{Pi235}}{\sum_i M_{Fi235}} \cdot\ 100\%. \tag{15}$$

In formula (15), the summation is performed over the ^{235}U masses contained in all "useful" withdrawals and feed flows (F_{nat}, F_{rep}), respectively. It should be noted that in a cascade with "broadening", the flow E is selected and flow P_2 for cascade 2 (see Fig. 1). The flow P in this scheme acts as the second waste flow. Thus, the recovery in the cascade with "broadening" $\delta\left(\frac{M_P}{M_F}\right)_{ext} = \frac{M_{P235}}{M_{F235}} \cdot 100\%$. Obviously, relative mass losses of ^{235}U can be calculated as follows:

$$m_{235,\ loss} = 100\% - \delta\left(\frac{M_P}{M_F}\right). \tag{16}$$

The problem formulated above is a conditional optimization problem for the function Ψ in a subdomain of a 6-dimensional space of continuous variables. The "particle swarm" method is appropriate for solving the problem [35]. This method is metaheuristic and does not require the calculation of the objective function gradients. Optimization methods similar to the one indicated are finding widespread use in various applications, including the optimization of cascade installations for isotope separation [36–38]. The advantage of the method is that it avoids getting stuck in the local extrema of the objective function. The user limits the number of iterations, and the implementation of the technique allows one to consider the constraints on the variables of the optimization problem.

A program code in Python 3 was developed using "SciPy" and "pyswarm" libraries to implement the optimization technique. Fig. 3 shows the block diagram of the proposed algorithm.

3. Results and discussion

Let's consider an example of how the proposed scheme works. The task was to enrich reprocessed uranium that has undergone five irradiation recycles. Table 1 presents an isotopic composition of RepU under consideration [28]. The considered composition is characterized by a relatively high content of even isotopes. Consequently, according to the results obtained, it will be possible to judge the applicability of the proposed scheme under conditions of multiple recycling. In this case, it is necessary to satisfy the following requirements for the product received:

Concentration of ^{235}U in commercial LEU – 4.7%.

Concentration of ^{235}U in depleted uranium – 0.15%.

Concentration of ^{235}U in LEU-diluent – 4.7–5.0%.

The limiting concentration of ^{232}U in commercial LEU – not higher than $5 \cdot 10^{-7}$%.

The ratio of concentrations ^{234}U and ^{235}U – not higher than

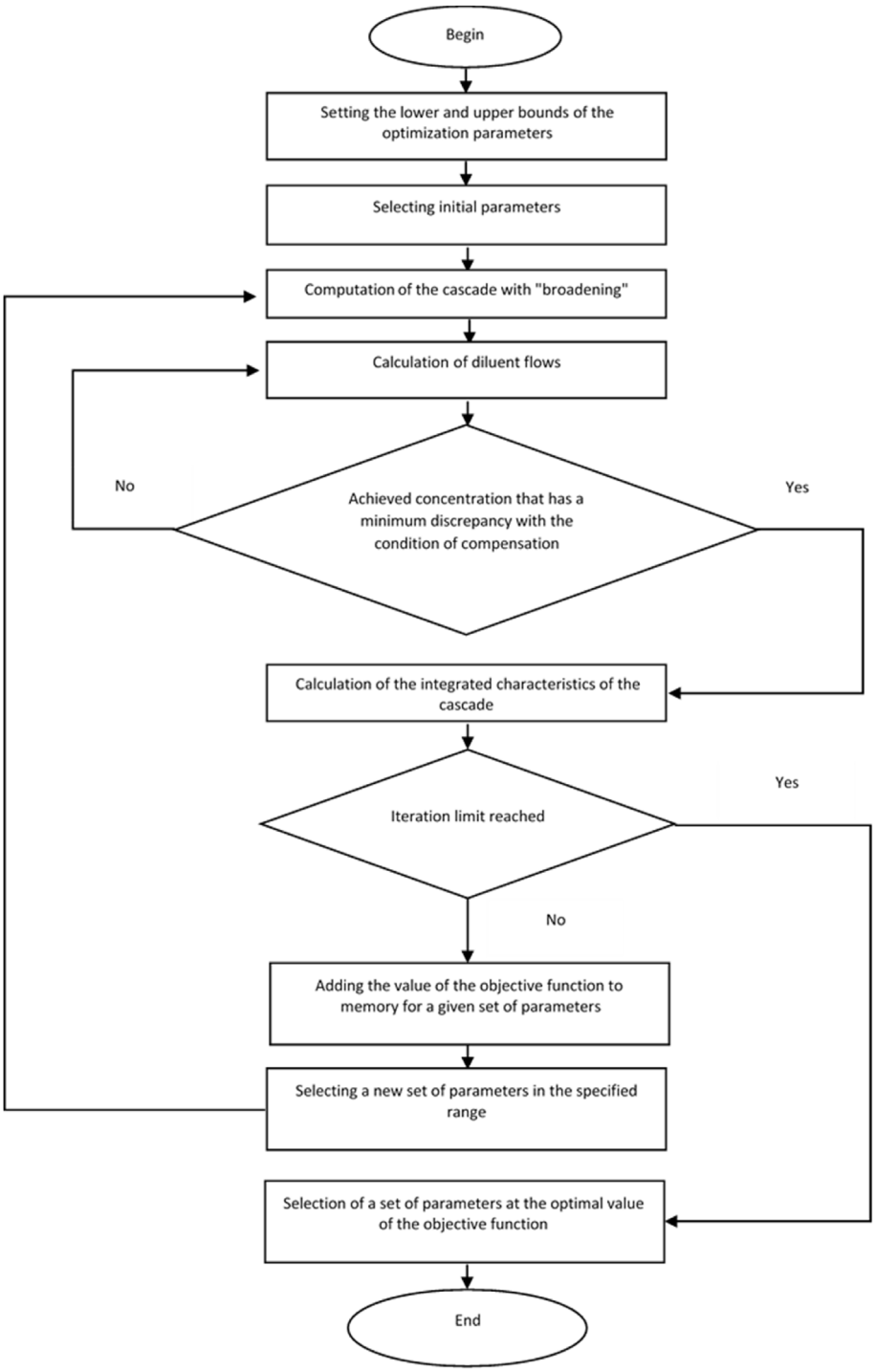

Fig. 3. A block diagram of the optimization algorithm for the proposed version of the cascade enrichment of reprocessed uranium.

Table 1
The isotopic composition of RepU.

Component	^{232}U	^{233}U	^{234}U	^{235}U	^{236}U	^{238}U
Concentration, wt. %	$1.03 \cdot 10^{-6}$	$1.30 \cdot 10^{-6}$	0.0391	1.07	1.45	The rest

0.02;

The ratio (initial RepU)/(commercial LEU) – 0.93 [28].

The compensation condition for ^{236}U in the resulting composition must also be met.

Uranium hexafluoride UF_6 was considered as a working substance [39, 40].

The proposed cascade scheme was compared with the modified double cascade described in Ref. [26]. That cascade was chosen for comparison because it can also solve the set problem under conditions of multiple recycle of uranium. Additionally, parameters of an ordinary cascade for enrichment of natural uranium were also taken into account. Consideration of the ordinary cascade helps to estimate the efficiency of reprocessed uranium recycling compared

Table 2
Relative integral characteristics of the proposed cascade and the scheme of work [26] compared with the case of a three-flow cascade for enrichment of natural uranium in an open fuel cycle.

Cascade type	Saving natural uranium ($\delta(\frac{F}{P})$), %	Overrun separative work ($\delta(\frac{SWU}{P})$), %	Recovery of ^{235}U ($\delta(\frac{M_P}{M_F})$), %
Proposed cascade	18.1	10.0	88.9
Modified double cascade	8.21	12.75	85.94

Table 3
Concentrations of the components of the mixture to be separated in the outgoing flows of the scheme (wt. %).

Flow	^{232}U	^{233}U	^{234}U	^{235}U	^{236}U
P_1	0.15	0.12	96.58	3.15	$6\bullet10^{-4}$
W_1	$1.37\bullet10^{-11}$	$3.43\bullet10^{-10}$	$2.06\bullet10^{-4}$	0.12	1.24
E	$1.85\bullet10^{-5}$	$5.28\bullet10^{-5}$	3.15	78.34	18.15
P_D	$1.16\bullet10^{-6}$	$3.29\bullet10^{-6}$	0.19	5.02	1.13
P_{fin}	$2.11\bullet10^{-7}$	$5.98\bullet10^{-7}$	0.07	4.75	0.2

to an open cycle using natural uranium. The proposed scheme was optimized according to the minimum consumption of natural uranium.

The results of the comparison of the cascade schemes are shown in Table 2. The values of economy of natural uranium and losses of separative work as well as the recovery coefficient of ^{235}U were calculated using formulas (13)–(15). As follows from the analysis of the data obtained, the proposed cascade is more efficient, both from the viewpoint of saving natural uranium, and from the perspective of the costs of separative work, as well as the degree of recovery of the ^{235}U isotope during the enrichment of the regenerate. The latter circumstance suggests that the proposed scheme more effectively solves the problem of returning the target isotope ^{235}U to the cycle, thereby increasing the efficiency of using the resource of reprocessed uranium in the fuel cycle.

Table 3 and Fig. 4 show the concentrations of the components of the mixture to be separated in the cascades' outgoing flows and illustrate the movement of material flows in the scheme. The data in Fig. 4 are calculated for 1 kg of marketable product ($P_{fin} = 1$ kg). The concentration of the ^{235}U in the diluent (P_2) was equal to 4.7%.

It should be noted that the concentration of ^{235}U in flow E exceeds 20%. This increases the risks of nuclear proliferation. On the other hand, this concentration is reached within the scheme, and the enriched material must be diluted immediately after enrichment. Moreover, in the proposed cascade scheme, the cascade which works with RepU is not connected in any way with the natural uranium enrichment cascade and, therefore, may be located in a separate site, for which enhanced security measures can be applied, just as with regard to radiation safety. These issues require special attention in the future, but nevertheless are not insuperable.

Additionally, the influence of the ^{235}U concentration in the diluent produced at stage 2 in the proposed scheme on its integral characteristics was studied (Figs. 5–7). For this purpose, the concentration of the ^{235}U isotope in the diluent was varied within a range from 4 to 5%. For each value of concentration of the ^{235}U isotope in the diluent integral characteristics of the cascade scheme were calculated using formulas (13), (14), and (16).

As follows from the analysis of curves in Figs. 5–7, an increase in the ^{235}U concentration in the LEU diluent noticeably (approximately three times) increases the saving of natural uranium.

It reduces separative work losses only slightly (by several tenths of a percent), increasing the ^{235}U losses in the cascade scheme.

Note that the chosen value of the concentration in the waste flow corresponds to a specific case and the reasonable question arises of how much the results will change if the concentration in the waste flow changes? To answer this question, other numerical experiments were carried out to reveal the influence of concentration of ^{235}U in waste flows of the cascade 1 and 2 on key characteristics of the cascade scheme. We considered three values for that concentration, namely, 0.1%, 0.2%, and 0.3%. Such values cover the reasonable range for the concentration of ^{235}U in waste flow if the gas centrifuge technology is used for uranium enrichment.

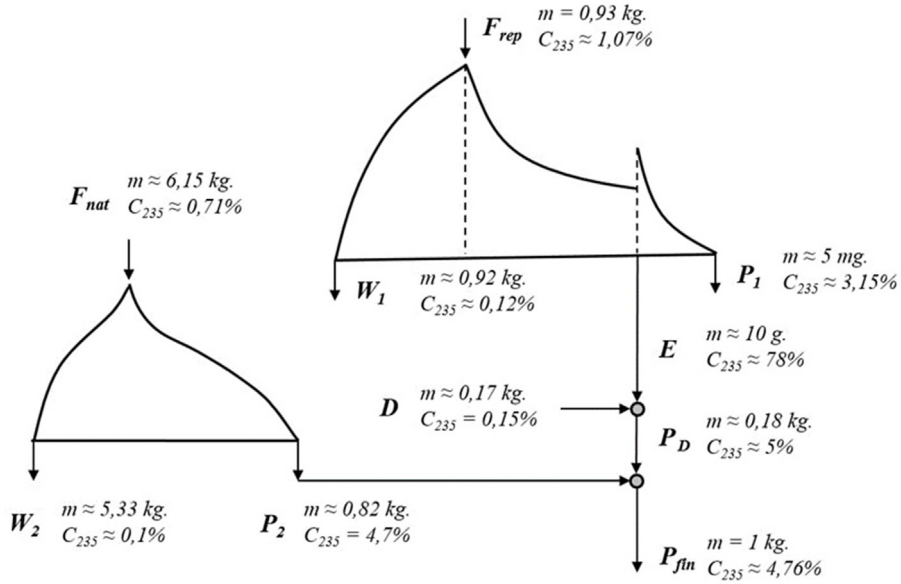

Fig. 4. An illustrative example of the movement of material flows in the proposed scheme.

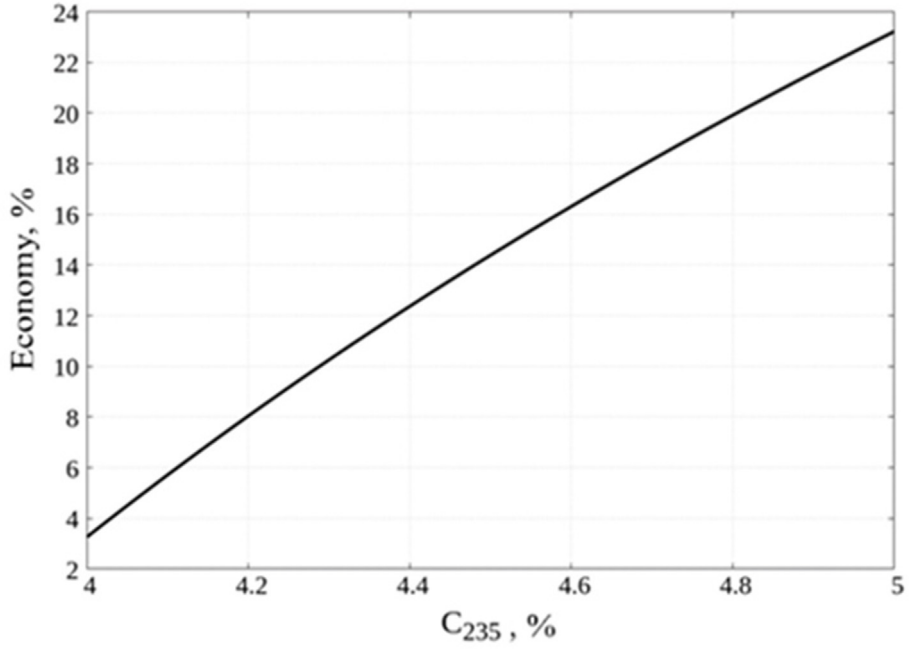

Fig. 5. The economy of natural uranium, depending on the concentration of ^{235}U in the flow of the LEU-diluent.

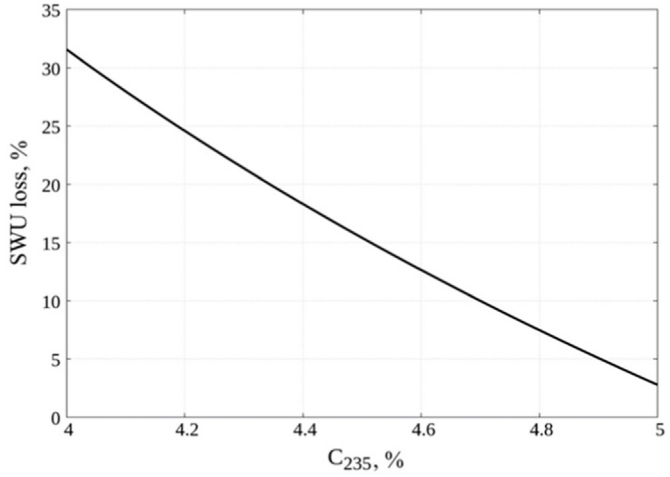

Fig. 6. The overrun of the separative work, depending on the concentration of ^{235}U in the flow of the LEU-diluent.

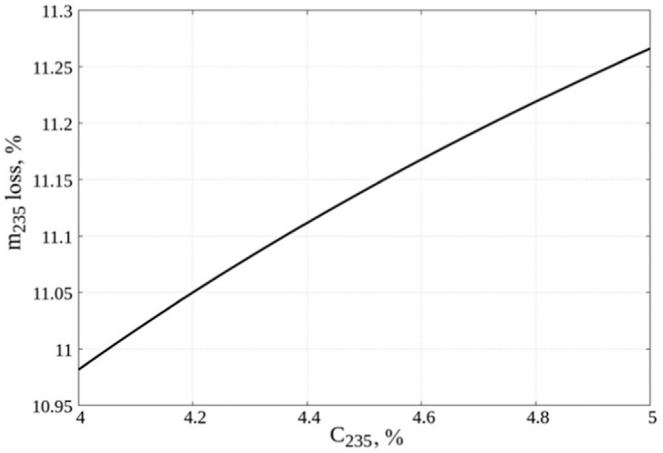

Fig. 7. The relative mass loss of ^{235}U, depending on the concentration of ^{235}U in the flow of the LEU-diluent.

Moreover, for each of those concentrations two cases were considered. First, the concentration of ^{235}U in depleted uranium (flow *D* in Fig. 1) was constant and equal to 0.15% as in illustrative example shown in Fig. 4. Second, the concentration of ^{235}U in flow *D* coincide with the concentration in waste flows. For all the cases the concentration of ^{235}U in the diluent (flow P_2 in Fig. 1) was equal to 4.5% as a "middle point" between utmost values of that concentration in calculations. Concentration of ^{235}U in commercial LEU was the same as in previous examples – 4.7% (before compensation of ^{236}U). When comparing the performance of the cascade scheme with the case of an open fuel cycle on natural uranium, the concentration of ^{235}U in the waste flow during the enrichment of natural uranium is chosen to be the same as for the scheme to enrich RepU.

Relative integral characteristics of the cascade scheme for all the cases are presented in Tables 4 and 5. The analysis of the data shows that with the growth of the concentration of ^{235}U in waste flows the economy of natural uranium decreases which can be explained by the decrease of the recovery of ^{235}U. On the other hand, the relative change of separative work is quite small. The comparison of the results of Tables 4 and 5 shows that the concentration of ^{235}U in depleted uranium (flow *D*) has almost no influence on the integral characteristics of the cascade.

As can be seen from the analysis of the data in Tables 4 and 5, the above conclusions regarding the effectiveness of the proposed cascade scheme will not change qualitatively. It should also be emphasized that the final choice of concentration in the waste flow can only be made on the basis of an economic assessment, considering the cost of SWU, natural uranium, fuel reprocessing, various chemical operations, etc. Such an assessment goes beyond the goals set in the work, so it was not carried out.

Table 4
Relative integral characteristics of the proposed cascade for various concentrations of ^{235}U in waste flows and fixed concentration of ^{235}U in flow *D* compared with the case of a three-flow cascade for enrichment of natural uranium in an open fuel cycle (concentration of ^{235}U in diluent equals to 4.5%).

Concentration of ^{235}U in waste flows, %	Concentration of ^{235}U in flow *D*, %	Saving natural uranium ($\delta(\frac{F}{P})$), %	Overrun separative work ($\delta(\frac{SWU}{P})$), %	Recovery of ^{235}U ($\delta(\frac{M_P}{M_F})$), %
0.1	0.15	12.81	18.56	89.17
0.2	0.15	11.20	18.88	77.55
0.3	0.15	9.50	18.12	65.02

Table 5
Relative integral characteristics of the proposed cascade for various concentrations of ^{235}U in waste flows and flow *D* compared with the case of a three-flow cascade for enrichment of natural uranium in an open fuel cycle (concentration of ^{235}U in diluent equals to 4.5%).

Concentration of ^{235}U in waste flows, %	Concentration of ^{235}U in flow *D*, %	Saving natural uranium ($\delta(\frac{F}{P})$), %	Overrun separative work ($\delta(\frac{SWU}{P})$), %	Recovery of ^{235}U ($\delta(\frac{M_P}{M_F})$), %
0.1	0.1	12.71	18.66	89.16
0.2	0.2	11.29	18.79	77.57
0.3	0.3	9.74	17.88	65.08

4. Conclusion

A modification of a double cascade scheme for enriching reprocessed uranium using a cascade with “broadening” and a three-stream cascade for enriching natural uranium is proposed. For the proposed scheme, a calculation and optimization method was developed according to the following efficiency criteria: saving natural uranium and over-expenditure of separative work in relation to the corresponding characteristics for a standard cascade enriching natural uranium in an open fuel cycle.

It is shown that the proposed scheme makes it possible to obtain low-enriched uranium of commercial quality using regenerated uranium with relatively high concentrations of even numbered isotopes, which indicates the applicability of the scheme under conditions of multiple recycling. Simultaneously, the scheme does not contain unspent regenerated uranium upon receipt of a given amount of a product - commercial LEU.

Computational experiments have shown that in the proposed modification, a noticeable saving of natural uranium in the cycle (up to 18.1%) can be achieved, provided that the additional consumption of the separative work does not exceed 10%, compared with the case of enrichment of natural uranium to obtain LEU of equivalent quality.

The advantage of the proposed scheme is also the involvement in the cycle of depleted uranium accumulated at the moment in significant quantities in the separation plants.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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