Original Article

# Restoration of the isotopic composition of reprocessed uranium hexafluoride using cascade with additional product

Valerii Palkin, Eugenii Maslyukov[*]

Department of Technical Physics, Ural Federal University, Mira 19, Ekaterinburg, 620002, Russia

ABSTRACT

In reprocessed uranium, derived from an impoverished fuel of light-water moderated reactors, there are isotopes of $^{232, 234, 236}$U, which make its recycling remarkably difficult. A method of concentration of $^{235}$U target isotope in cascade's additional product was proposed to recover the isotopic composition of reprocessed uranium. A general calculation procedure is presented and a parameters' optimization of multi-flow cascades with additional products. For the first time a numeric model of a cascade that uses the cuts of partial flows of stages with relatively high separation factors was applied in this procedure. A novel computing experiment is carried out on separation of reprocessed uranium hexafluoride with providing a high concentration of $^{235}$U in cascade's additional product with subsequent dilution. The parameters of cascades' stages are determined so as to allow reducing the $^{232, 234, 236}$U isotope content up to the acceptable. It was demonstrated that the dilution of selected products by the natural waste makes it possible to receive a low enriched uranium hexafluoride that meets the ASTM C996-15 specification for commercial grade.

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## 1. Introduction

In reprocessed uranium, derived from an impoverished fuel of the light-water moderated reactors, the concentration of the $^{235}$U base fissile substance is usually higher than in natural uranium [1]. This fact makes it attractive for a rerun and nuclear fuel fabrication. However, there are number of complications that makes usage of the reprocessed uranium difficult. The most noticeable item is the presence of $^{232, 234, 236}$U isotopes in the reprocessed uranium. Due to the necessity to fulfill conditions of the radiological safety for fuel fabrication, the narrow requirements are imposed upon the content of $^{232, 234}$U in the low enriched uranium (concentration of $^{235}$U is less than 5%). High concentration of $^{236}$U in fuel leads to increase of the neutron parasitic capture and, as a consequence, to the necessity of increase of $^{235}$U concentration. It is necessary to clean the reprocessed uranium by $^{232, 234, 236}$U impoverishment for a reproduction of a reactor fuel. For this purpose, different methods of uranium hexafluoride separation in cascades and dilution procedures were worked out. They comprise an enrichment of the reprocessed uranium in a double cascade up to 97% of $^{235}$U [2], a

usage of natural and waste uranium during an additional feed flow of cascades [3,4] and a low enriched product as a diluting agent [5,6]. These methods allow partially decrease the concentrations of $^{232, 234, 236}$U but have some drawbacks. In the majority of methods these isotopes' content fails to meet the requirements to the commercial grade of low enriched uranium hexafluoride, which is stated in the ASTM C996-15. Moreover, a loss of the valuable natural raw materials and additional losses for the separation work happen there.

In order to impoverish the $^{232, 234, 236}$U the present paper proposes a new method of concentrating $^{235}$U target isotope with use of an ordinary cascade with additional product and successively diluting the resulting product with the waist natural uranium. Concentrating is followed by dilution of the resulting product by the waste uranium. The present approach provides the low content of $^{232, 234}$U while a production of high concentration of $^{235}$U in the main product is followed by a significant rise of these concentrations. An efficiency of an identical method of concentration of stable isotopes of medium mass is shown on different models: on the MARC-cascades with a feed flow which differs through the stages [7,8]; on the model Q-cascades with a continuous flow profile and its extension in the additional product point [9–11]; and on the rectangular-sectional cascades with a common flow in some separate sections of stages [12]. It should be noted that MARC-

cascade is matched abundance ratio cascade with equal relative concentrations of two key components. Q-cascade is a model cascade with continuous profile and relatively small separation factors that is used for multicomponent isotope separation. Rectangular-sectional cascade consists of several sections. Each of them contains stages with equal feed flow.

The purpose of the present paper is applying of this approach to the cascades with bigger stage separation factors, which correspond to the gas centrifuges. A multi-flow cascade [13] was chosen as the study object. This model is applied for the first time for concentrating uranium hexafluoride basing on adjustment of parameters of each stage through the partial flows cuts. The properties analysis of the cascades with an additional product was made by the computing experiment separating the mixture of reprocessed uranium hexafluoride which is derived from the impoverished fuel of light water reactor. It should be noted that present paper is considering only isotope separation issue and omitting other complications of reusing the reprocessed uranium.

### 1.1. Parameters and equations of multi-flow cascade

The model of multi-flow symmetrical cascade of $n$ stages comprises a supply of external feed flow to each stage and taking therefrom additional product flows of cascade (Fig. 1). The following notations of the external flows of the cascade were agreed: $W$ − waste, $F_1$, $F_2$, ..., $F_n$ − feed, $P$ − main product flow, $E_1$, $E_2$, ..., $E_{n-1}$ and $G_2$, $G_3$, ..., $G_n$ − additional products from product and waste flows of stages consequently. The lightest isotope is enriched in the main product flow of the cascade and the heaviest one is enriched in the waste flow. The same parameters have the product and waste flows of the stages. The scheme allows to calculate cascades with any number of external flows. To do this all the really missing flows have to be set equal to zero.

Let $m$ − number of components of isotope mixture. Let's denote concentrations (molar fractions) of $j$-component of isotope mixture in flows: $C_j^W$ − waste, $C_{j,1}^F$, $C_{j,2}^F$, ..., $C_{j,n}^F$ − feed, $C_j^P$ − main product, $C_{j,1}^E$, $C_{j,2}^E$, ..., $C_{j,n-1}^E$ and $C_{j,2}^G$, $C_{j,3}^G$, ..., $C_{j,n}^G$ − additional products, $j = \overline{1,m}$. Then, if there are no losses, the equations of mass balance and each component balance are valid

$$\sum_{i=1}^{n} F_i = P + W + \sum_{i=1}^{n-1} E_i + \sum_{i=2}^{n} G_i;$$

$$\sum_{i=1}^{n} F_i C_{j,i}^F = P C_j^P + W C_j^W + \sum_{i=1}^{n-1} E_i C_{j,i}^E + \sum_{i=2}^{n} G_i C_{j,i}^G, \ j = \overline{1,m}.$$

Component-wise sum of concentrations for any flow equals to one. So from the reduced equations there are $m$ independent parameters of the cascade.
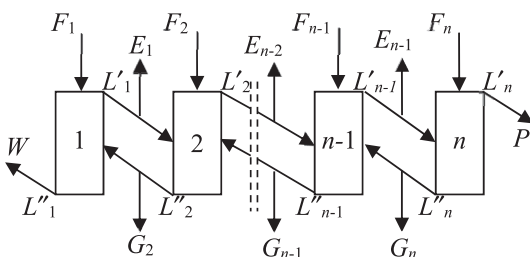


**Fig. 1.** Stage connection model in the multi-flow symmetrical cascade: 1, 2, n-1, n − stage number.

Internal parameters of cascade include feed, product and waste flows of stages, concentrations of components in these flows. They are connected by the equations of mass and components balance, equations of flows' connections and by the separation characteristics. Balance equations for the stages are

$$L_i = L_i' + L_i''; \ L_i C_{j,i} = L_i' C_{j,i}' + L_i'' C_{j,i}'', \ i = \overline{1,n}, \ j = \overline{1,m},$$

where $L_i, L_i', L_i''$ − feed, product and waste flows of $i$-stage, $C_{j,i}, C_{j,i}', C_{j,i}''$ − concentrations of $j$-component in the corresponding flows. In the given scheme each stage with number $i = \overline{2,n-1}$ is fed by the flow $L_{i-1}' - E_{i-1}$ of the previous stage and flow $L_{i+1}'' - G_{i+1}$ of the following one. At the cascade edges flow $L_2'' - G_2$ feeds the first stage, and flow $L_{n-1}' - E_{n-1}$ feeds the last stage. Moreover every stage can have the external feed flow $F_i$. Thereby the equations of flow stages connections are expressed as balance correlations

$$L_1 = L_2'' - G_2 + F_1; \ L_n = L_{n-1}' - E_{n-1} + F_n; \ L_i$$
$$= L_{i-1}' - E_{i-1} + L_{i+1}'' - G_{i+1} + F_i;$$

$$L_1 C_{j,1} = L_2'' C_{j,2}'' - G_2 C_{j,2}^G + F_1 C_{j,1}^F; \ L_n C_{j,n}$$
$$= L_{n-1}' C_{j,n-1}' - E_{n-1} C_{j,n-1}^E + F_n C_{j,n}^F;$$

$$L_i C_{j,i} = L_{i-1}' C_{j,i-1}' - E_{i-1} C_{j,i-1}^E + L_{i+1}'' C_{j,i+1}'' - G_{i+1} C_{j,i+1}^G + F_i C_{j,i}^F,$$
$$i = \overline{2,n-1}, \ j = \overline{1,m}.$$

The separation characteristics are set as separation factors $q_{j,i}$, $j = \overline{1, \ m}$, $i = \overline{1, \ n}$. Let's number the components in the order of increasing of their mass number (molar or atomic weight). Then, taking the heaviest isotope as base component

$$q_{j,i} = \frac{C_{j,i}' C_{m,i}''}{C_{j,i}'' C_{m,i}'}.$$

In molecular-kinetic method of separation the separation factors $q_{j,i}$ are expressed through $\Delta M_{jm} = M_m - M_j$ using the formulas $q_{j,i} = (q_{0,i})^{\Delta M_{jm}}$, where $q_{0,i}$ − separation factor per unit of mass number difference [14].

Taking into consideration the limiting conditions which link the external and internal parameters

$$W = L_1''; \ P = L_n'; \ C_j^W = C_{j,1}''; \ C_j^P = C_{j,n}';$$

$$C_{j,i}^E = C_{j,i}'; \ C_{j,k}^G = C_{j,k}'', \ j = \overline{1,m}, \ i = \overline{1,n-1}, \ k = \overline{2,n},$$

the number of the independent parameters of multi-flow cascade with known number of stages and defined separation factors, equals to $n(m+3) - 2$.

### 1.2. Calculation and optimization of the cascade

Let given: $F_1$, $F_2$, ..., $F_n$ − external feed flows, $E_1$, $E_2$, ..., $E_{n-1}$ and $G_2$, $G_3$, ..., $G_n$ − additional products, $C_{j,1}^F$, $C_{j,2}^F$, ..., $C_{j,n}^F$ − components concentrations in the feed flows of the cascade, $j = \overline{1,m}$. There are $n$ independent parameters remain, which can be chosen in passing to partial flows of components of mixture. For each $j$-th component of $i$-th stage the partial flows equal to

$L_{j,i} = L_i C_{j,i}; \; L'_{j,i} = L'_i C'_{j,i}; \; L''_{j,i} = L''_i C''_{j,i}.$

It follows that the partial flows cuts meet the correlations [15].

$$\varphi_{j,i} = L'_{j,i} / L_{j,i} = \sigma_i q_{j,i} / \left( 1 + \sigma_i q_{j,i} \right),$$

where $\sigma_i$ − parameter, defining a relation of waste and product enrichment factors of $i$-th stage. Parameters $\sigma_i$, $i = \overline{1,n}$ do not depend on the concerned component and can be used when calculating the cascade as $n$ independent variables.

Keeping in mind balances of partial flows of components in the cascade's sections, analogically to Ref. [16], it can be shown that

$$L''_{j,i} - L'_{j,i-1} = W_{j,i}, \; j = \overline{1,m}, \quad i = \overline{1,n},$$

where $W_{j,i}$ − transit flow of $j$-th component in the section before the $i$-th stage, that equals to

$$W_{j,i} = W C^W_j - \sum_{l=1}^{i-1} F_l C^F_{j,l} + \sum_{l=1}^{i-2} E_l C^E_{j,l} + \sum_{l=2}^{i} G_l C^G_{j,l}.$$

In these correlations there are $L_{j,0} = 0$, and sums equal to zero if upper summation indexes are smaller than the lower ones. Evaluating the partial flows through the cuts, in other words $L_{j,i} = \frac{1}{1-\varphi_{j,i}}(W_{j,i} + \varphi_{j,i-1} L_{j,i-1})$, we can get the general formula

$$L_{j,i} = \frac{1}{\varphi_{j,i}} \sum_{l=1}^{i} W_{j,l} \prod_{s=l}^{i} \frac{\varphi_{j,s}}{1 - \varphi_{j,s}}, \; j = \overline{1,m}, \quad i = \overline{1,n}.$$

At the product end of the cascade for each $j$-th component the present condition should be met

$$L'_{j,n} = L_{j,n} \varphi_{j,n} = \sum_{i=1}^{n} F_i C^F_{j,i} - \sum_{i=1}^{n-1} E_i C^E_{j,i} - \sum_{i=2}^{n} G_i C^G_{j,i} - W_j,$$

where $W_j = W C^W_j$ − partial flow of $j$-th component in the waste flow of the cascade. The formula on the right-hand side equal to the partial flow $P_j = P C^P_j$ in product. From here it follows that on rearrangement we deduce

$$W_j = \sum_{i=1}^{n} \left( F_i C^F_{j,i} - E_{i-1} C^E_{j,i-1} - G_{i+1} C^G_{j,i+1} \right) A_{j,i} / B_j,$$

where $E_0 = G_{n+1} = 0$ and the $A_{j,i}$ and $B_j$ coefficients equal to

$$A_{j,i} = 1 + \sum_{l=i+1}^{n} \prod_{s=l}^{n} \frac{\varphi_{j,s}}{1 - \varphi_{j,s}}; \; B_j = 1 + \sum_{l=1}^{n} \prod_{s=l}^{n} \frac{\varphi_{j,s}}{1 - \varphi_{j,s}}.$$

This formula allows to calculate the partial flows $W_j$, $j = \overline{1,m}$ using the selected parameters of the stages $\sigma_i$, $i = \overline{1,n}$. The peculiarity of that formula is that the components concentration in the additional product flows of the cascade is not independent and is found during the process of calculation. Consequently it makes sense to use the iterative procedure of concentrations matching $C^E_{j,1}$, $C^E_{j,2},...,C^E_{j,n-1}, C^G_{j,2}, C^G_{j,3},...,C^G_{j,n}, j = \overline{1,m}$. So the first calculation can be done without additional product flows and the received values of components concentration in the waste and product flows can be accepted as the initial approximations.

After a determination of the partial flows of components in the waste flow of cascade there can be calculated $L_{j,i}, j = \overline{1,m}, \quad i = \overline{1,n}$

flows and other parameters: $P_j = \sum_{i=1}^{n} F_i C^F_{j,i} - \sum_{i=1}^{n-1} E_i C^E_{j,i} - \sum_{i=2}^{n} G_i C^G_{j,i} - W_j; \; W = \sum_{j=1}^{m} W_j; \; P = \sum_{j=1}^{m} P_j; \; C^W_j = W_j/W; \; C^P_j = P_j/P; \; L_i = \sum_{j=1}^{m} L_{j,i};$ $C_{j,i} = L_{j,i}/L_i$ and etc. It should be pointed out that at the equal parameters $\sigma$ through the stages it is obtained a quasi-ideal cascade. It has a random number of feed flows and additional products. In particular case of one feed flow a *MARC*-cascade with additional product flows is calculated. Whereby

$$\sigma = 1/\sqrt{q},$$

where $q$ − is the separation factor, determined from the mass numbers of the key components. But if vary parameters of stages $\sigma_i$, $i = \overline{1,n}$, an optimization of the cascade can be made in accordance with the accepted criterion. It should be noted that the method described above can be applied for the cascade with relatively high separation factors.

To verify the calculation and optimization procedure the computing experiment have been carried out. A double cascade scheme [17] for molybdenum separation was taken as a reference. First cascade has 71 stage; feed stage number is 13. Second cascade has 39 stages; feed stage number is 36. Waste of the first cascade is the feed of the second one. Denote $\sigma_C$ − thermal neutron cross section, $\Sigma L$ − total feed flow, $P_0$ − the sum of the first cascade product flow and second cascade waste. The computing experiment presented on Table 1 shows the model correctness: results are practically match each other.

The conservation of isotopes was controlled with an accuracy of $10^{-10}\%$ on each calculation stage.

### 1.3. Calculation data and argument

At first, the computing experiment was performed on the separation of uranium hexafluoride in cascades with one feed flow, one main product flow, one additional product flow and one waste flow. Components concentrations in feed flow corresponded to the reprocessed uranium of the first cycle from an impoverished fuel of light water reactor VVER-1000 [18]: $^{232}$U $2.9803 \cdot 10^{-7}\%$, $^{233}$U $5.8144 \cdot 10^{-7}\%$, $^{234}$U $1.9106 \cdot 10^{-2}\%$, $^{235}$U $0.90076\%$, $^{236}$U $0.57311\%$, $^{238}$U − the rest. Cascades flows were calculated according to the initial mass of uranium hexafluoride in feed 100 ton, used within one working year. In addition, the total feed flow of stages characterizes the number of used separation elements. Additional product flow was calculated from the waste flow of cascade stage

**Table 1**
Comparison of calculation methods on double cascade for molybdenum separation.

| Parameter | Numeric model | Reference model [15] |
|---|---|---|
| First cascade | | |
| Feed flow, ton/y | 10.0 | No data |
| Product, ton/y | 1.184 | |
| Waste, ton/y | 8.816 | |
| $\Sigma L$, ton/y | 2750.137 | |
| $\sigma_C$, barn | 0.122 | |
| Second cascade | | |
| Feed flow, ton/y | 8.816 | No data |
| Product, ton/y | 7.985 | |
| Waste, ton/y | 0.831 | |
| $\Sigma L$, ton/y | 878.781 | |
| $\sigma_C$, barn | 0.287 | |
| Total | | |
| $\Sigma L/P_0$ | 1800.95 | 1807.68 |
| $F/P_0$ | 4.96 | 4.98 |
| $\sigma_C$, barn | 0.190 | 0.190 |

**Table 2**
Manufacturing of the low enriched reprocessed uranium hexafluoride in the optimal cascade at the total feed flow $\sum L = 8621.9$ ton/y.

| Parameter | UF$_6$ mass, ton/y | $^{232}$U, % | $^{233}$U, % | $^{234}$U, % | $^{235}$U, % | $^{236}$U, % |
|---|---|---|---|---|---|---|
| Feed flow | 100 | $2.98 \cdot 10^{-7}$ | $5.81 \cdot 10^{-7}$ | $1.91 \cdot 10^{-2}$ | 0.901 | 0.573 |
| Product | 17.66 | $1.68 \cdot 10^{-6}$ | $3.24 \cdot 10^{-6}$ | $1.03 \cdot 10^{-1}$ | 4.4 | 2.145 |
| Waste | 82.34 | $1.56 \cdot 10^{-9}$ | $1.01 \cdot 10^{-8}$ | $1.07 \cdot 10^{-3}$ | 0.15 | 0.236 |

with the highest concentration of $^{235}$U. The stage number was chosen by sample calculation of $^{235}$U concentration changes in triple-flow cascade without an additional product flow. Separation factors of stages were defined from the accepted one per unit of difference of mass numbers $q_0 = 1.119$. The calculations were made using the dilution of concentrated additional product by the waste natural uranium with concentration of $^{235}$U 0.15% and 0.3%.

Table 2 shows the calculations of enrichment of reprocessed uranium up to the concentration 4.4% of $^{235}$U in cascade with the minimal total feed flow. Number of cascade stages is $n = 20$, stage number of feed supply is $f = 11$. The received concentrations of $^{232, 234, 236}$U in the product flow meet the ASTM C996-15 specification for the low enriched reprocessed uranium with additional arrangement between the parties. But they are essentially more than the defined in ASTM C996-15 limits for the commercial uranium ($^{232}$U not more than $1 \cdot 10^{-8}$%, $^{234}$U–11 000 mg/g$^{235}$U, $^{236}$U–0,025%). Particularly this is the case of the $^{232, 236}$U isotopes, of which content in the selected product equals to $1.68 \cdot 10^{-6}$ and 2.145% respectively. And that leads to different restrictions. In particular, because of high concentration of $^{232}$U, at nuclear fuel fabrication a radiation situation becomes significantly worse. That concentration of $^{236}$U expects an increase in content of $^{235}$U in the product by 0.4–0.6%. It is calculated with the compensation

coefficient of 0.2–0.3 ($^{236}$U/$^{235}$U concentration ratio) in order to compensate for $^{236}$U parasitic captures. So the real concentration of $^{235}$U in the product without regard to $^{236}$U neutron parasitic capture is 3.8–4.0%.

Improvement of isotopic composition of reprocessed uranium hexafluoride is possible when a $^{235}$U concentration of 97% and more is provided in the additional product flow. However, the accomplishment of such a content of $^{235}$U does not guarantee low concentration of each of $^{232, 234, 236}$U isotopes. These concentrations can redistribute depending upon the place of additional product flow. In this context the parameters $\sigma_i$ were chosen on the separated sections of the cascade with content analysis of the mentioned isotopes. Referring to this reason the total feed flow of stages was not minimized at given concentration of $^{235}$U. The flow was taken into account during comparison of different options of cascade.

Table 3 shows the calculations of $^{235}$U concentrations in the cascade of 160 stages when the feed was given to the 40-th stage. Parameters $\sigma_i$ were set equal to 0.752 for the section of stages from 1-st up to 80-th and 0.65 — from 81-st up to 160-th. This conforms to a direction of enrichment of $^{235}$U and lighter isotopes in the first section towards the product to the 80-th stage. In the second section the direction is changed — $^{235}$U and more heavy isotopes are enriched towards the waste. The flow of the additional product was chosen close to the maximum possible. As a result, a concentration of $^{235}$U 96.9% was received in additional product of the 82-nd stage. The total feed flow is 31.2 thousand ton/y that is 3.6 times higher than the flow showed in Table 2. This is resulted from the enrichment up to the high concentration of $^{235}$U, as well as from its bigger extract to the products owing to the lower concentration of waste. Concentrations of $^{232, 234, 236}$U in the resulting product, received after dilution, are significantly lower than that given in Table 2. Concentration of $^{232}$U is less by an order of magnitude, $^{236}$U is around 30 times lower. The product mass 18.2–18.9 ton is close to

**Table 3**
Concentration of $^{235}$U in additional product flow of the cascade ($n = 160, f = 40$) at the total feed flow $\sum L = 31192.3$ ton/y.

| Parameter | UF$_6$ mass, ton/y | $^{232}$U, % | $^{233}$U, % | $^{234}$U, % | $^{235}$U, % | $^{236}$U, % |
|---|---|---|---|---|---|---|
| Cascade: | | | | | | |
| Feed flow | 100 | $2.98 \cdot 10^{-7}$ | $5.81 \cdot 10^{-7}$ | $1.91 \cdot 10^{-2}$ | 0.901 | 0.573 |
| Product $P$ | 0.009 | $2.89 \cdot 10^{-3}$ | $5.08 \cdot 10^{-3}$ | 86.646 | 13.346 | $6.86 \cdot 10^{-5}$ |
| Product $G_{82}$ | 0.8 | $3.99 \cdot 10^{-6}$ | $1.39 \cdot 10^{-5}$ | 1.382 | 96.914 | 1.696 |
| Waste | 99.191 | $5.50 \cdot 10^{-14}$ | $9.49 \cdot 10^{-12}$ | $2.78 \cdot 10^{-5}$ | 0.125 | 0.564 |
| Dilution No. 1: | | | | | | |
| Diluting agent | 17.41 | | | $5 \cdot 10^{-4}$ | 0.15 | |
| Product | 18.21 | $1.75 \cdot 10^{-7}$ | $6.11 \cdot 10^{-7}$ | $6.12 \cdot 10^{-2}$ | 4.4 | $7.45 \cdot 10^{-2}$ |
| Dilution No. 2: | | | | | | |
| Diluting agent | 18.05 | | | $1.5 \cdot 10^{-3}$ | 0.3 | |
| Product | 18.85 | $1.69 \cdot 10^{-7}$ | $5.90 \cdot 10^{-7}$ | $6.01 \cdot 10^{-2}$ | 4.4 | $7.20 \cdot 10^{-2}$ |

**Table 4**
Concentration of $^{235}$U in the additional product flow of the cascade ($n = 130, f = 25$) at the total feed flow $\sum L = 30990.4$ ton/y.

| Parameter | UF$_6$ mass, ton/y | $^{232}$U, % | $^{233}$U, % | $^{234}$U, % | $^{235}$U, % | $^{236}$U, % |
|---|---|---|---|---|---|---|
| Cascade: | | | | | | |
| Feed flow | 100 | $2.98 \cdot 10^{-7}$ | $5.81 \cdot 10^{-7}$ | $1.91 \cdot 10^{-2}$ | 0.901 | 0.573 |
| Product $P$ | 0.012 | $2.32 \cdot 10^{-3}$ | $4.35 \cdot 10^{-3}$ | 72.283 | 27.709 | $1.11 \cdot 10^{-3}$ |
| Product $G_{73}$ | 0.85 | $1.40 \cdot 10^{-6}$ | $5.21 \cdot 10^{-6}$ | 1.193 | 97.202 | 1.603 |
| Waste | 99.138 | $2.04 \cdot 10^{-12}$ | $6.77 \cdot 10^{-11}$ | $4.11 \cdot 10^{-5}$ | $7.17 \cdot 10^{-2}$ | 0.564 |
| Dilution No. 1: | | | | | | |
| Diluting agent | 18.56 | | | $5 \cdot 10^{-4}$ | 0.15 | |
| Product | 19.41 | $6.11 \cdot 10^{-8}$ | $2.28 \cdot 10^{-7}$ | $5.28 \cdot 10^{-2}$ | 4.4 | $7.02 \cdot 10^{-2}$ |
| Dilution No. 2: | | | | | | |
| Diluting agent | 19.24 | | | $1.5 \cdot 10^{-3}$ | 0.3 | |
| Product | 20.09 | $5.91 \cdot 10^{-8}$ | $2.20 \cdot 10^{-7}$ | $5.20 \cdot 10^{-2}$ | 4.4 | $6.78 \cdot 10^{-2}$ |

**Table 5**
Concentration of $^{235}U$ in the additional product flow of the cascade ($n = 130, f = 25$) at the total feed flow $\sum L = 54863.2$ ton/y.

| Parameter | UF$_6$ mass, ton/y | $^{232}U$, % | $^{233}U$, % | $^{234}U$, % | $^{235}U$, % | $^{236}U$, % |
|---|---|---|---|---|---|---|
| Cascade: | | | | | | |
| Feed flow | 100 | $2.98 \cdot 10^{-7}$ | $5.81 \cdot 10^{-7}$ | $1.91 \cdot 10^{-2}$ | 0.901 | 0.573 |
| Product $P$ | 0.040 | $7.40 \cdot 10^{-4}$ | $1.43 \cdot 10^{-3}$ | 38.797 | 61.200 | $1.18 \cdot 10^{-3}$ |
| Product $G_{77}$ | 0.8 | $2.17 \cdot 10^{-7}$ | $1.04 \cdot 10^{-6}$ | 0.443 | 99.055 | 0.501 |
| Waste | 99.160 | $1.99 \cdot 10^{-12}$ | $6.43 \cdot 10^{-11}$ | $3.54 \cdot 10^{-5}$ | $8.45 \cdot 10^{-2}$ | 0.574 |
| Dilution No. 1: | | | | | | |
| Diluting agent | 17.82 | | | $5 \cdot 10^{-4}$ | 0.15 | |
| Product | 18.62 | $9.31 \cdot 10^{-9}$ | $4.47 \cdot 10^{-8}$ | $1.95 \cdot 10^{-2}$ | 4.4 | $2.15 \cdot 10^{-2}$ |
| Dilution No. 2: | | | | | | |
| Diluting agent | 18.47 | | | $1.5 \cdot 10^{-3}$ | 0.3 | |
| Product | 19.27 | $9.00 \cdot 10^{-9}$ | $4.32 \cdot 10^{-8}$ | $1.98 \cdot 10^{-2}$ | 4.4 | $2.08 \cdot 10^{-2}$ |

the mass of low enriched reprocessed uranium, derived by the direct process without concentration, overpassing it by 0.5–1.2 ton.

Splitting the cascade into three sections on the parameter $\sigma_i$ could increase the flow of the additional product. Table 4 shows calculations of cascade made of 130 stages when the feed flow is given to the 25-th stage. The first section consists of stages from 1-st up to 30-th, second — from 31-st up to 80-th, third — from 81-st up to 130-th. Parameter $\sigma_i$ through the sections was: 0.822; 0.74 and 0.61. This selection of sections was defined by creation of concentrations maximums: $^{236}U$ in the depletion part of the cascade, $^{235}U$ — in the enrichment one. In the first section the enrichment of $^{236}U$ and lighter isotopes was in the enrichment part. In the third section $^{235}U$ and more heavy isotopes were concentrated in the depletion part. In the second section the directions of $^{235}U$ and $^{236}U$ isotopes enrichment were opposite: first concentrated in the enrichment part, the second — in the depletion one. This setting of $\sigma_i$ provided the maximum concentration of $^{236}U$ in the 40-th stage waste equal to 22.0%. Concentration of $^{235}U$ in the additional product of the 73-rd stage amounted to 97.2%. After its dilution the mass of low enriched reprocessed uranium with the concentration of $^{235}U$ 4.4% amounted to 19.4–20.1 ton. The increase of mass if compared with the above example can be explained by the lower concentration of $^{235}U$ (0.07%) in the waste of cascade and its bigger extract into the products consequently. As to the concentrations of $^{232, 234, 236}U$ in the resulting products, they are significantly better than shown in Table 3.

The reduction of the flow of additional product when using three sections of cascade allows to improve the concentrations of $^{232, 234, 236}U$ in the resulting product of reprocessed uranium. This is followed by an increase of concentration of $^{235}U$ and the total feed flow. So in the previous cascade the boundary of the first and the second sections can be changed to the 40-th stage and the
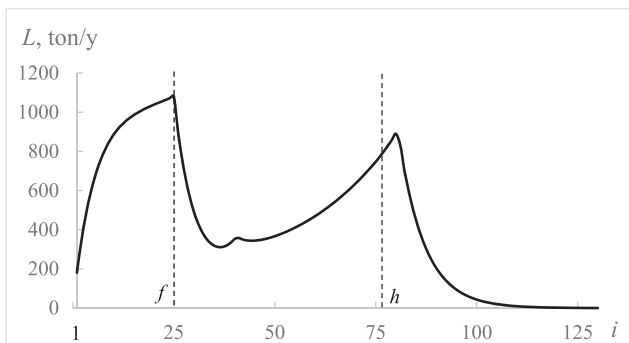
parameter $\sigma_i$ can be set through the sections equal to 0.82; 0.74 and 0.62. Then, by reducing additional product mass down to 0.83 ton, the concentration of $^{235}U$ in it is equal to 98.6%, and the total feed flow of the cascade equals to 40.4 thousand ton/y. After dilution by the waste uranium hexafluoride with the content of $^{235}U$ 0.3% and $^{234}U$ 0.0015% the concentrations in 19.9 ton of the diluted product equal to, %: $^{232}U$ $2.15 \cdot 10^{-8}$, $^{233}U$ $9.94 \cdot 10^{-8}$, $^{234}U$ $2.77 \cdot 10^{-2}$, $^{235}U$ 4.4, $^{236}U$ $3.31 \cdot 10^{-2}$. The mass content of $^{234}U$ is fully correspond to the requirements of the ASTM C996-15 specification for the
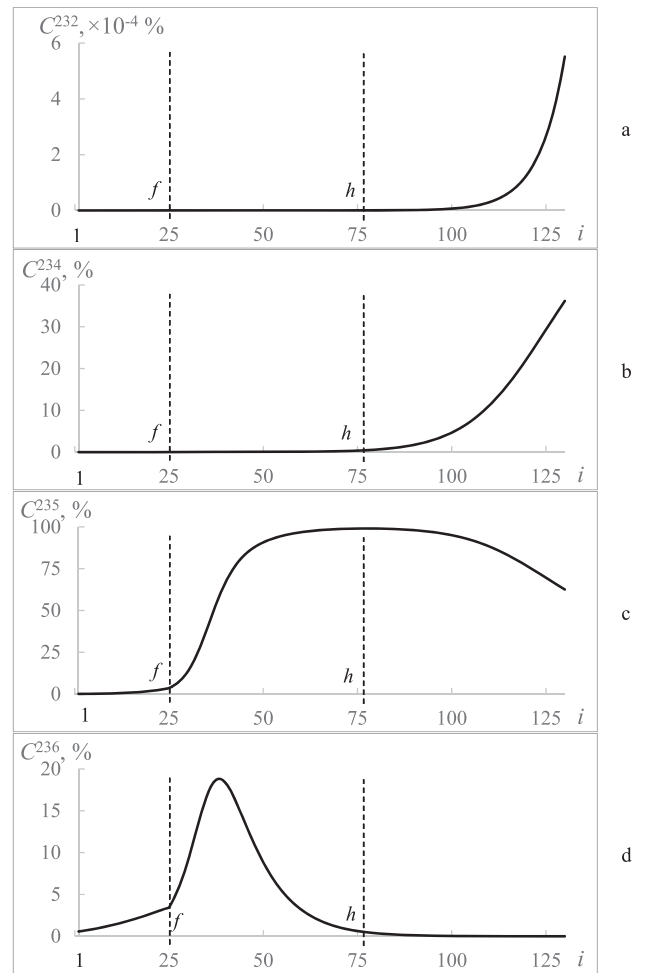
**Fig. 3.** $^{232}U$ (a), $^{234}U$ (b), $^{235}U$ (c) and $^{236}U$ (d) concentration changes in the waste of cascade stages.

**Fig. 2.** Feed flow distribution through the stages of the cascade with the additional product flow.

**Table 6**
Expenses for the $UF_6$ separation.

| $C_W$, % | Total flow, ton/y | U equal mass, ton/y | U cost, million USD/y | Separation work, thousands SWU/y | SW cost, million USD/y | Total, million USD/y |
|---|---|---|---|---|---|---|
| 0.15 | 10919 | 92.2 | 7.376 | 103.9 | 4.156 | 11.532 |
| 0.30 | 7701 | 121.4 | 9.712 | 73.5 | 2.940 | 12.652 |

commercial grade of uranium hexafluoride, but the mass content of $^{232, 236}U$ is a little bit higher than necessary.

Full compliance to these requirements is showed in calculation in Table 5. Number of cascade stages is $n = 130$ and stage number of feed supply is $f = 25$. Parameter $\sigma_i$ is set as 0.822 from 1-st up to 40-th stage, 0.74 from 41-st up to 80-th, 0.61 from 81-st up to 130-th. Additional feed is taken from the waste of the stage $h = 77$. The distribution of feed flow through the stages is shown in Fig. 2. Two peaks are characteristic: the first peak is at the point of feed supply in 25-th stage, the second is at the boundary of sections in the 80-th stage, close to the additional product. Characteristically, the flow expansion is similar to that of the $Q$-cascades. In Fig. 3 the diagrams of $^{232, 234, 235, 236}U$ isotope concentration changes in the waste of stages are shown. Maximum concentration of $^{235}U$ is in the stage where after the peak on 38-th stage, the concentration of $^{236}U$ is sufficiently declined and the concentration of $^{234}U$ is not yet increased. The same thing goes for the concentration of $^{232}U$ isotope: it increases towards the product flow with a slight decrease from $3.85 \cdot 10^{-7}\%$ in the 44-th stage to $2.17 \cdot 10^{-7}\%$ in the 77-th stage. This effect is due to the peak-like increase in the feed flow associated with the concentration of the $^{235}U$ isotope in the 77-th stage. It is similar to the influence of the flow expansion noted in Ref. [12]. As a result, the concentrations of lightest components are lower than that in the previous stages. Higher total flow in comparison to previous example shows that this separation program needs more separation elements.

A utilization of the main product containing $^{235}U$ in sufficiently great concentration should be mentioned from the point of view of common use. It is useful to dilute it down to a nuclear-safety mixture with the concentration of $^{235}U < 1\%$, by the waste uranium with the low concentration of the isotope mentioned above. Special mixing station should be used on the final stage of the separation process [2] to exclude military or other non-peaceful application of the product. The mixture is suitable for long-term storage.

To compare the economical productivity of the proposed method two reference cascades for natural uranium separation were calculated: both have product flow 18 ton/y and $^{235}U$ concentration in product equal to 4.4%. First of them has $^{235}U$ concentration 0.15% in the waste flow and separation work is equal to 103.9 thousands SWU. The second one has $^{235}U$ concentration 0.3% in the waste flow and separation work equal to 73.5 thousands SWU. Assuming separation work price around 40 USD/SWU and natural uranium price around 80 USD/kg [19] corresponding expenses are 11.5 million USD/y and 12.7 million USD/y. Calculation results for those cascades are shown in Table 6.

To implement the proposed method in practice the special cascade for the reprocessed uranium purification could be built. This cascade could use gas centrifuge technology. According to total feed flow ratio, number of gas centrifuges there would be 2–7 times more than in the ordinary separation cascade. That means it will pay off when orders for reprocessed uranium are available even taking into account operating and staff costs. Therefore, purification cascade with dilution down to 4.4% could make a profit comparable to expenses for natural uranium enrichment up to the same value (Table 6) and helps to save it.

## 2. Conclusion

A new method is worked out to reuse the reprocessed uranium and save natural uranium. It allows to separate uranium isotopes with use of an ordinary cascade with additional product and successively dilute the resulting product with waste natural uranium. Sufficient separation power makes it possible to meet the requirement of specification ASTM C996-15 for the commercial grade.

### 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.

### Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.net.2020.05.020.

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