Comparison of the efficiency of square cascades with an additional product flow and double cascades to concentrate intermediate isotopes

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Abstract. Original techniques were proposed for calculating and optimizing double cascades (DC) built of square cascades and a square cascade with an additional product flow (SCAP). Both cascade schemes (DC and SCAP) were tested in solving a task of simultaneous enrichment of three components of a model mixture to a pre-defined level. A comparison of the DC and SCAP efficiency was made according to the minimum of total cascade flow. The results have shown that the double cascade is more efficient in terms of the chosen criterion.

1. Introduction
At the moment, stable isotopes demand is growing in various fields, from basic research to medicine and nuclear energy.

Considering the fact that many chemical elements have three or more isotopes, it is common for different isotopes of a particular chemical element to be used in various applications. For example, the $^{180}$W isotope is in demand for producing the $^{181}$W radioisotope, which, in turn, is used to manufacture sources for X-ray fluorescence analysis of ores and for completing geophysical instruments. At the same time, $^{182}$W is also used in the metallurgical industry for materials research using nuclear gamma resonance. Additionally, isotope $^{186}$W is in demand in nuclear medicine, which is used to obtain the $^{188}$W radioisotope, the generator of the $^{188}$Re. Other isotopes of the natural mixture of tungsten are also used in various industries [1]. In addition to the above examples, we can also mention mixtures of isotopes of lead [2–5], cadmium, molybdenum, and some others [1, 6–8].

One of the methods broadly used for the separation of stable isotopes is the gas centrifuge method. The production of isotopes of more than twenty chemical elements is possible using cascades of gas centrifuges [1]. To make the production more efficient, it is important to optimize the separation process, including transient processes [9, 10] and separation of non-typical for this method isotopes such as carbon isotopes [11].

In most cases, to separate stable isotopes, one uses the so-called square cascades (SCs). Such a cascade has equal internal feed flows through all stages, making them convenient for practical implementation. It also allows a manufacturer to use an SC to separate various mixtures without significant restructuring of the cascade.

One of the critical problems in separating multicomponent mixtures of stable isotopes is extracting the isotopes of intermediate mass numbers. However, obtaining such isotopes with high concentrations in the outgoing flows of an ordinary triple-flow cascade is possible only to a certain degree. This
limitation can be estimated analytically based on the initial isotopic composition of the mixture to be enriched [12, 13].

Though, there are several approaches to achieve concentrations of intermediate components above the limitation. The first is using the additional product flow in a cascade [14]. The idea of the method is based on the fact that concentrations of intermediate components can reach maximum values at internal stages of the cascade. Therefore, the additional withdrawal from the stage near such a maximum, provides isotopic composition with the concentration of the intermediate component, which is greater than the theoretical limit for three-flow cascade. In some cases, it is possible to obtain relatively high concentrations of two intermediate components simultaneously in a cascade with two additional product flows [15–17].

An alternative way to reach peak concentrations of intermediate components is the use of connected cascades. One of the outgoing flows from the first cascade goes into the second one. Such double-cascades (DCs) make it possible to redistribute the initial mixture between more than two outgoing flows, thereby creating conditions under which the intermediate target isotope will be the heaviest or the lightest in one of the outgoing flows of such a cascade scheme. This step-by-step growth of the target intermediate component’s concentration can significantly increase its final concentration at the double cascade’s output compared to a single triple-flow cascade. At the same time, there are practically no publications devoted to optimizing consequent connections of two or more cascades. The optimization techniques for single cascades with additional outgoing flows for the concentration of intermediate isotopes are mainly limited to the case of a Q-cascade [18]. The question is which one is more efficient: single cascade with additional outgoing flows or double cascade?

Considering the above, the main goals of this work are:

1. Development of a method for calculating and optimizing double cascades built from SCs to concentrate one intermediate isotope and two utmost in mass components (the lightest and the heaviest).
2. Development of a method for calculating and optimizing an SC with an additional product flow (SCAP) for the simultaneous concentration of three components.
3. Comparison of the efficiency of the SCAP and the DC to concentrate the intermediate target isotope.

2. Instruments and Methods

2.1 Background

Let’s consider the simplest version of a symmetrical counter-current cascade (figure 1). It is broadly used in the practice of isotope separation in centrifuge cascades [13]. Each of squares in the figure 1 represents one cascade stage (parallel connection of gas centrifuges). The arrows in the figure 1 represent flows between stages and external flows (P, W, F). The meaning for each flow will be clarified below.

For such a cascade type, it is typical, when at the inlet to the s-th stage, the flows from the previous (s - 1)-th stage and from the next (s + 1)-th stage are mixed. The feed flow F with the concentration $C_i^F$ is fed to the input of the stage with the number $s = f$. The cascade stages are numbered sequentially from left ($s = 1$) to right ($s = N$). From the right end of the cascade, a flow enriched in light isotopes (‘product’ – P) is withdrawn, and from the left end, a flow enriched in heavy isotopes (‘waste’ – W). It is noteworthy that in the case of multi-component mixtures, separation concepts ‘product’ and ‘waste’ are quite flexible because both of the flows might be valuable in reality. That is why in the text below, we will refer to them just as flows P and W. Concentrations of components in flows P and W are $C_i^P$, $C_i^W$, respectively.

Flows F, P, W with corresponding concentrations $C_i^F$, $C_i^P$, $C_i^W$ ($i = \bar{1, m}$, $m$ is a number of components in a mixture under separation) together are considered as external cascade parameters. Every stage has three flows, which are denoted as $L_s$, $L_s^L$, $L_s^R$ ($s = 1, 2, \ldots N$) and objectify stage feed, ‘product’ (enriched in light components) and ‘waste’ (enriched in heavy components) flows, respectively. In case of square cascade $L_s = \text{const} = L$. It is necessary to make so-called ‘swirls’ of flows at the first and last stages of the cascade to fulfill the requirement of constant value for $L_s$. Making ‘swirls’ of flows means a partial return of outgoing flows from those stages to their inputs. Each of the flows $L_s$, $L_s^L$, $L_s^R$ on each stage
characterized by a set of concentrations $C_{i,s}$, $C_{i,s}'$, $C_{i,s}''$. The stage cut $\theta_s = L_s'/L_s$, separation coefficients $\alpha_{i,j,s} = \left(\frac{C_{i,s}}{C_{j,s}}\right)^{-1}$, $\beta_{i,j,s} = \left(\frac{C_{i,s}}{C_{j,s}}\right)^{-1}$, $q_{i,j,s} = \left(\frac{C_{i,s}}{C_{j,s}}\right)^{-1}$ complement the set of stage parameters.

![Figure 1. Schematic drawing of a symmetrical counter-current cascade.](image)

In the absence of losses of the working substance in the stages of the cascade and in the stationary mode of operation, the external parameters must satisfy the material balances [13]:

$$\begin{align*}
F &= P + W, \\
FC_i^F &= PC_i^P + WC_i^W, \quad i = 1, m.
\end{align*}$$

(1)

(2)

Equivalent equations can be written for each cascade stage [13]:

$$\begin{align*}
L_s' + L_s'' &= L_s, \\
L_i'C_{i,s}' + L_i'C_{i,s}'' &= L_i'C_{i,s}, \quad i = 1, m.
\end{align*}$$

(3)

(4)

External and internal parameters of the cascade are related by boundary conditions

$$\begin{align*}
L_1' &= (1 - \theta_1)L_1 = W, \\
L_N &= \theta_NL_N = P, \\
C_N &= C_N^P, \quad i = 1, m, \\
C_i' &= C_i^W, \quad i = 1, m.
\end{align*}$$

(5)

(6)

(7)

(8)

Let us describe the idea of the simultaneous concentrating of isotopes when connecting ordinary cascades into a DC. Let each of the cascades numbered $j$ be an ordinary SC with three external flows: the feed flow $F_j$, the flow of the light fraction $P_j$, and the flow of the heavy fraction $W_j$. The concentration of the $i$-th component in the corresponding flow is denoted by $C_{i,F_j}$, $C_{i,P_j}$, $C_{i,W_j}$. The above parameters are external parameters of the cascade. The following values are introduced for each cascade: $N_j$ is the number of stages of the cascade, $f_j$ is the number of the feed stage.

The coupling of two SCs is possible in two ways (figure 2). There will be four external flows in a double cascade with any such connection – one incoming (feed flow of the first cascade) and three outgoing ones. It is taken into account that one of the outgoing flows of the first cascade is fed to the input of the second. Therefore, from the point of view of the entire double cascade, this flow cannot be considered as an external one.

It is possible to develop a universal (independent of the particular variant) method for calculating and optimizing DC’s parameters according to a given efficiency criterion. In the case of a cascade with constant separation coefficients in stages, the value of the cascade total flow can be used as such a criterion. The minimum of this value will mean the minimum number of used separation elements (gas centrifuges) [19].
In this paper, we propose a technique for optimizing both variants of the DC based on modern optimization methods of finding the global extremum in multi-dimensional space. The optimization problem and the method are described below.

2.2 Method to optimize a double cascade built from SCs

Let’s formulate the problem statement.

Given values: concentration of components in the feed mixture $C_i^F$; the required concentrations of three isotopes with numbers $n_k$ in the outgoing flows of the scheme $T_k - C_i^{T_k}$, where $k = 1, 3$, $T_k$ can be equal to $P_1$ (or $W_1$), $P_2$, $W_2$; parameters of a single separating element for the cascade with constant separation coefficients in stages – the separation coefficient per unit mass difference $q_0$.

It is necessary to determine the following parameters during calculation:

- The values of the remaining external parameters of the cascade scheme: the concentration of non-target components for the $T_k$ flow, ratios of external flows for each cascade.
- The values of the outgoing flows of the first cascade that are fed to the second cascade (for example, for the variant I shown in figure 1 – the value of the flow $P_1$, for the variant II – the value $W_1$), as well as the concentration of all components in them.

The found parameters should ensure the achievement of the required concentrations in the outgoing flows of the double cascade and correspond to the minimum of the chosen efficiency criterion.

As an optimization criterion for the double cascade, the total flow of both cascades normalized to the sum of the outgoing flows was chosen:

$$\psi = \frac{\sum_{k=1}^{N_1} L_{1_k} + \sum_{k=1}^{N_2} L_{2_k}}{\sum_{k=1}^{N_1} T_k} = \frac{N_1 L_1 + N_2 L_2}{\sum_{k=1}^{N_1} T_k}.$$

(9)

The parameters listed below are used as variables in solving the problem:

- The flow ratios in each of the two cascades are $(P/L)_j$, $(W/L)_j$ ($j = 1, 2$).
- The length of each cascade is $N_j$, the index of the feed stage is $f_j$ ($j = 1, 2$).

At each iteration of the search for the required values of $N_j$ and $f_j$ it is necessary to carry out a stage-by-stage calculation of all parameters of both cascades. This procedure can be implemented by one of the known methods of single cascade calculation. In this paper, we used the method of approximation of the separation factor [20] due to its fast convergence and stability for the choice of initial approximations.

The optimization problem described above is a numerical optimization problem on the space of variables of mixed type since the values of the flow ratios $(P/L)_j$, $(W/L)_j$ are real, and the values $N_j$ and $f_j$ are integers. Below is the proposed method for solving the formulated problem.
It is logical to divide the problem being solved into two steps. The first is to determine the values of the flow ratios \((P/L)_s\), \((W/L)_s\) at fixed \(N_l\) and \(f_i\), corresponding to the required concentrations of the target components in the outgoing flows of cascades. The second is the search for \(N_l\) and \(f_i\) that ensures that the minimum of the given optimization criterion is met.

For each new value of \(N_l\) and \(f_i\), the values of the flow ratios \((P/L)_s\), \((W/L)_s\) also need to be recalculated. Thus, varying the integer variables \(N_l\) and \(f_i\) will act as an ‘outer loop’ over the first part of the problem.

At the ‘inner loop’ of the problem, the function \(\Delta_1\) is minimized:

\[
\Delta_1\left(\left(\frac{P}{L}\right)_j, \left(\frac{W}{L}\right)_j\right) = \sum_{k=1}^{3} \left( C_{n_k}^{T_k} - C_{n_k}^{T_k \text{ given}} \right)^2,
\]

where \(C_{n_k}^{T_k}\) are calculated concentrations of mixture components in outgoing flows; \(C_{n_k}^{T_k \text{ given}}\) are given values of mixture components in outgoing flows; \(k = 1,3\).

The advantage of solving the first subproblem in this form is that the set of variable parameters does not depend on how the ordinary SCs connect in a double cascade. When the type of connection changes, only the indices in the function under minimization change. The minimization problem described above is the numerical optimization of the function that depends on four continuous variables. Various methods have been proposed so far to solve problems of this type. In this paper, we choose the Levenberg-Marquardt algorithm [21]. This method has fast convergence and the possibility of working with limited intervals for variables.

The second part of the problem is the minimization of the function \(\Psi\), depending on four integer variables \(N_l, f_i\). Mathematically it is a constrained combinatorial optimization problem. The combinatorial optimization problem in the general case is NP-complete. Thus, finding the exact value of the minimum is possible only by a complete enumeration of all admissible values of the variables [22]. The total number of options for the search grows geometrically even with a slight increase in the range for the examination. In such a situation, it is possible to use approximate methods for finding the extremum of a function, based, in particular, on a random search. Metaheuristic (sometimes called ‘bionic’) algorithms, which are essentially a subclass of random search algorithms, are also widely used. In this work, to solve the problem, one of such methods was used, namely, the bee colony algorithm (‘ABC’-algorithm) [23].

It is necessary to have correctly selected initial approximations for this algorithm to work efficiently. It is hard to find them from any simple considerations or using simple analytical calculations due to the significant uncertainty in values of cascade parameters. In this regard, to search for initial approximations, it is reasonable to use the parameters of the optimal schemes of model cascades. For this purpose, the corresponding double cascades built of Q-cascades were chosen [24]. The optimization of the parameters of Q-cascades is less complicated compared to SC, including the case of optimization of a system of Q-cascades [18, 25, 26].

2.3 Calculation and optimization of a SC with an additional product flow to simultaneously concentrate three components of an isotopic mixture

Consider the SC with one additional product flow, shown in figure 3.

The idea of using such a cascade is based on the fact that the concentration distributions of components with intermediate masses can reach maximum values at the inner stages of the cascade. Therefore, it is possible to obtain the concentration of the intermediate isotope significantly higher than limiting value, if to turn on an additional withdrawal (the flow \(E\) in figure 3 with concentrations \(C_{n_k}^{E}\)) near the stage, where the maximum is reached [16]. In addition, one can get relatively high concentrations of the lightest and heaviest isotope in the flows \(P\) and \(W\).

Let us formulate a mathematical statement of the optimization problem for the SCAP. Suppose that the following parameters are to be set for the optimized cascade: concentrations of components in the feed mixture \(C_{n_k}^{E}\); required concentrations of three isotopes: 1) number \(n\) in the flow \(P - C_{n_k}^{P}\); 2) number \(k\) in the flow \(W - C_{n_k}^{W}\); 3) number \(I\) in the flow \(E - C_{n_k}^{E}\).
It is required to determine the following parameters during the optimization:

- Concentration of non-target isotopes for each of the outgoing flows.
- The ratios of the outgoing and incoming flows.
- The feed flow distribution over cascade stages.
- Distribution of component concentrations over cascade stages.
- etc.

The found parameters should ensure the achievement of the required concentrations in the outgoing flows and also correspond to the minimum of the chosen optimization criterion.

As an optimization criterion, the total flow normalized to the sum of the outgoing flows was chosen:

$$\psi = \frac{\sum_{s=1}^{N} L_s}{P + W + E} = \frac{NL}{P + W + E}.$$  \hspace{1cm} (11)

The parameters listed below act as variables in solving the problem: 1) the ratios $P/L, W/L, E/P$; 2) the values of $N, f,$ and $S_E$ (the number of the stage, from which the flow $E$ is withdrawn).

At each iteration of the search for the required configuration for a given set of variable parameters, it is necessary to carry out a sequential calculation of the cascade parameters (verification calculation). As in the case of the double cascade scheme, the method [20] was used. This method was tested on SCs with additional outgoing flows before [16].

The optimization problem described above is also a problem of numerical optimization in the space of variables of mixed type. The values of the flow ratios $P/L, W/L, E/P$ are real, and the number of stages in the cascade $N$, the number of the feed flow injection $f$, the number of the stage, from which the additional product flow is withdrawn $S_E$ are integers.

By analogy with the double cascade, it is logical to divide the problem into two parts. The first is to determine the flow ratios $P/L, W/L, E/P$ at fixed $N, f,$ and $S_E$, corresponding to the required concentrations of the target components in the outgoing flows. The second is the search for $N, f,$ and $S_E$ that ensures that the minimum of a given optimization criterion is met.

The first part of the problem can be solved as the problem of minimizing the function $\Delta_2$:

$$\Delta_2 \left( \frac{P}{L}, \frac{W}{L}, \frac{E}{P} \right) = \left( C_{n, \text{calc}}^P - C_{n, \text{given}}^P \right)^2 + \left( C_{k, \text{calc}}^W - C_{k, \text{given}}^W \right)^2 + \left( C_{l, \text{calc}}^E - C_{l, \text{given}}^E \right)^2,$$  \hspace{1cm} (12)

where $C_{n, \text{calc}}^P, C_{k, \text{calc}}^W, C_{l, \text{calc}}^E$ are calculated concentrations in corresponding outgoing flows of the cascade; $C_{n, \text{given}}^P, C_{k, \text{given}}^W, C_{l, \text{given}}^E$ are required values for concentrations of components $n, l, k$ in cascade outgoing flows.
The problem to minimize function \( \Delta_2 \) is a numerical optimization problem for a function that depends on three variables of real type. This problem can be easily solved using the Levenberg-Marquardt algorithm.

The second part of the problem is the minimization of the function \( \Phi \) depending on three integer variables \( N, f, \) and \( S_E \). This is a constrained combinatorial optimization problem. In the framework of the problem under consideration, the method of full enumeration of the values of \( N, f \) and \( S_E \) with constraints was used owing to the relatively small number of possible combinations of the parameters \( N, f, \) and \( S_E \), as well as their interrelation \( N > f, N > S_E, S_E > f \). Such an approach guarantees finding a minimum in a given area.

3. Results and Discussion

Within the framework of this work, the comparison of the parameters of the optimal DCs and SCAP was carried out. As an example, we considered a three-component model mixture. The mass numbers and concentrations of the components are presented in Table 1. They were selected to correspond to the average values for the spectrum of isotopic mixtures separated by the gas centrifuge method. The concentrations of the components are set in such a way to ensure the limiting values of the concentration of the intermediate component in the triple-flow cascade at a level not higher than 30\%. A simple estimation shows that for the component with the mass number equal to 201, the limiting concentration in the light fraction is 29.4\%, and in the heavy fraction is 13.2\%. Thus, the composition of the model mixture illustrates one of the typical situations in the separation of multicomponent mixtures, when the maximum attainable concentrations of intermediate isotopes are not high enough.

| Component | Mass number | Initial concentration \((C_i^F, \%)\) (mol. perc.) |
|-----------|-------------|---------------------------------------------|
| 1         | 200         | 24.00                                       |
| 2         | 201         | 10.00                                       |
| 3         | 202         | 66.00                                       |

When modeling the separation of a three-component mixture, the following concentrations of the components in the outgoing flows were specified: \( C_{200}^P = 90\% \), \( C_{201}^E = 60\% \), \( C_{202}^W = 90\% \). It is important that the selected concentration for the intermediate component is approximately twice higher its limiting value in the flow of the light fraction of the ordinary cascade, which is 29.4\%. The value of the separation factor for the unit difference of mass numbers was set equal to 1.2.

Table 2 shows concentrations of components in the withdrawal flows of the optimal SC with an additional product flow.

| Mass number | \( C_i^P, \% \) | \( C_i^E, \% \) | \( C_i^W, \% \) |
|-------------|-----------------|-----------------|-----------------|
| 200         | 24.00           | 90.00           | 35.02           |
| 201         | 10.00           | 10.00           | 60.00           |
| 202         | 66.00           | 0.00            | 4.98            |

For a comparison, table 3 shows the concentrations of the components of the model mixture in the external flows of two variants of double cascade.

Table 4 shows values of relative total flows of all considered cascade schemes to solve the same separation task.
The data in table 4 shows that for the simultaneous enrichment of several components of the model mixture to high concentrations, the double cascade I is more efficient in terms of the chosen criterion.

### Table 3. Concentrations (mol. perc.) of components of the model mixture in outgoing flows of double cascades I and II.

|                        | Double cascade I |         |         | Mass number | Double cascade II |         |         |
|------------------------|------------------|---------|---------|-------------|------------------|---------|---------|
|                        | 200              | 201     | 202     | 200         | 201              | 202     |
| $C_{i1}^F$, %          | 24.00            | 10.00   | 66.00   | 24.00       | 10.00            | 66.00   |
| $C_{i1}^P$, %          | 78.57            | 16.67   | 4.76    | 90.00       | 8.92             | 1.08    |
| $C_{i1}^W$, %          | 2.61             | 7.39    | 90.00   | 2.65        | 10.35            | 87.01   |
| $C_{i2}^P$, %          | 90.00            | 9.33    | 0.67    | 35.42       | 60.00            | 4.58    |
| $C_{i2}^W$, %          | 11.09            | 60.00   | 28.91   | 1.46        | 8.54             | 90.00   |

### Table 4. The values of relative total flows (dim.less) in the optimal SC with an additional product flow and two variants of double cascades.

| Cascade scheme | SCAP | DC I   | DC II  |
|----------------|------|--------|--------|
| $\Phi$         | 539.63| 319.69 | 365.97 |

### 4. Conclusion

A technique for calculating and optimizing double cascades built from SCs is proposed, based on a combination of optimization methods for continuously and discretely varying cascade parameters.

The proposed optimization technique is tested on the example of calculating double cascades for the simultaneous enrichment of all components of a three-component model mixture.

A method is proposed for calculating and optimizing an SCAP. The method allows finding the optimal configuration of the SCAP that could produce highly-enriched isotopes.

A comparison of the efficiency of double cascades and SCAP is carried out on the example of simultaneous enrichment of all components of the three-component model mixture. In this case, the heaviest and the lightest components for all compared cascade schemes were concentrated up to 90%, and the intermediate component up to 60%. In this case the intermediate component's concentration value is approximately twice higher than its attainable concentration in a triple-flow cascade. The results of the research show that both versions of the double cascade are more efficient than the SCAP in terms of chosen efficiency criterion (the total flow per unit of product).

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