Optimization of a system of square cascades for efficient concentration of intermediate isotopes

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Abstract. The problem of obtaining relatively high concentrations of stable isotopes of intermediate mass numbers is considered. The object of research is a connection of three ordinary square cascades. The possibility of simultaneous concentration of isotopes of intermediate masses in different output flows of cascades is shown. The dependencies of the parameters of the cascade schemes under consideration are investigated. The method of optimization of such system of square cascades according to the criterion of minimum relative total flow at given concentrations of target components is proposed. Calculations were made on the example of isotope mixtures with the number of components 4 and 5.

1. Introduction

Currently, stable isotopes of various chemical elements are becoming more and more in demand in various fields, starting from basic research to medicine and nuclear energy. Gas centrifuge method is actively developing [1] [2] and it is the main industrial method for enriching natural uranium [3][4]. In addition, production of about a hundred stable isotopes mastered using the gas centrifuge method [4] [5] [6] [7] [8]. Considering that a significant part of chemical elements has 3 or more isotopes, situations are not uncommon, in which several isotopes of separated multicomponent mixtures are used in various applications. For example, $^{180}$W is in demand for obtaining the radioisotope $^{181}$W, which is used to analyze ores and complete geophysical instruments; at the same time, $^{182}$W is also used in the metallurgical industry to study materials by nuclear gamma resonance [5].

Another example is lead isotope $^{206}$Pb which is used to produce the medical radioisotope $^{205}$Bi, while $^{207}$Pb is also used in control and protection systems in reactors. Moreover, in recent years, the possibility of expanding the use of these isotopes in nuclear energy, as well as the use of another lead isotope - $^{208}$Pb - has been actively discussed [9] [10] [11] [12]. Another example of the simultaneous use of different isotopes of the same chemical element is a mixture of molybdenum isotopes, several of which are used in basic research and medicine [5].

One of the critical problems in the separation of multicomponent mixtures of stable isotopes is the concentration of isotopes of intermediate mass numbers. The problem is that the enrichment of these isotopes in an ordinary cascade is difficult due to their “competition” with the isotopes of the utmost mass numbers (the lightest the heaviest), as a result, the concentrations of the intermediate components attainable at the outgoing flows of an ordinary (triple-flow) cascade are limited by some certain values [13].

There are several ways to solve this problem. The first way is to use multi-flow cascades [14] [15] [16]. Another way to solve this problem is to use multi-cascade schemes [17], in which the external outgoing flows of one cascade are used as external feed flows for other cascades. This allows to divide
the initial mixture into more than 2 outgoing flows, thereby creating the conditions under which the

target intermediate isotope will actually utmost mass number in one of the outgoing flows of such a
scheme. This consistent concentration of the target intermediate components can significantly increase
the achieved concentration in the outgoing flows of the cascade scheme. However, such schemes, as a
rule, are aimed at concentrating one of the isotopes of the mixture under separation. At the same time,
when such schemes are implemented, side fractions will inevitably arise in which some of the remaining
isotopes, including intermediate ones, of the separated mixture are enriched. Considering that there are
examples of isotopic mixtures, several isotopes of which are simultaneously in demand in various
applications, it seems appropriate to assess the possibility of using such multi-cascade schemes for the
simultaneous concentration of several isotopes of intermediate mass numbers. Along with the
concentration of intermediate isotopes in such cascade schemes, it is possible to organize the production
of relatively high concentrations of components with extreme mass numbers, which can be no less in
demand.

This work is devoted to the analysis of the possibility of simultaneous concentration of at least four
isotopes to obtain their relatively high concentrations in a system of three cascades. As a single cascade
for consideration, a cascade with a constant stage feed flows or a square cascade (SC) was chosen,
because this type of cascade is often used in the practice of separation of stable isotopes.

An effective technique for optimizing a system of three SCs for the simultaneous concentration of
four components in a mixture under separation is proposed.

2. **Instruments and Methods**

![Figure 1. Schematic drawing of a system of three square cascades.](image)

We present the idea of the simultaneous concentration of several isotopes in a triple cascade scheme
composed of SC. A schematic representation of such a cascade system is shown in Figure 1. Each of
the cascades is a square cascade having an external flow of feed $F_j$, product $P_j$ and waste $W_j$ ($j = 1, 3$).
By “product” we mean the end of the cascade at which enrichment with the lightest component takes
place, under the “waste” - in which the heaviest is enriched. The concentration of the $i$-th component
in the corresponding flow is denoted by $C_{Fj}$, $C_{Pj}$, $C_{Wj}$ ($i = 1, ..., m$, where $m$ – is the number of
components of the mixture being separated, \( j = 1, 3 \). The parameters listed above are called the external parameters of the cascade. Its internal parameters are: stage feed flow (productivity) \( \dot{L}_s \) of stage with number \( s \) with concentrations \( C_{i,s} (i = 1, m) \); the stage outgoing flows leaving the stage enriched and depleted in light components, \( L'_s, L'_s \), respectively, with concentrations \( C'_{i,s} \) and \( C_{i,s} (i = 1, m) \). In the case of a square cascade \( L_s = \text{const} = L \). The cascade configuration is determined by the values of \( N_j \) and \( f_j \) – the number of stages and the number of the external feed supply stage in the \( j \)-th cascade.

The idea of the presented scheme is as follows. The initial separated mixture enters the first cascade, in which it is divided into 2 outgoing flows: in the first, the light part of the mass spectrum of isotopes of the initial mixture is enriched, in the second, heavy isotopes are enriched. Further, both outgoing flows are supplied as external feed flows to the second and third cascades. Moreover, in each of them, there is also a division of each of the fractions (light and heavy) into 2 groups. As a result, if a four-component mixture is fed to the cascade scheme, then in the limiting case it is possible to concentrate each of the components of the mixture in one of the outgoing flows. If mixtures with more than four components are fed to such a scheme, it is possible to higher concentrations of four isotopes (two utmost and two intermediate).

To assess the capabilities of such a scheme, it is necessary to develop an effective methodology for calculating and optimizing its parameters according to specified efficiency criteria. In the case of considering a cascade with separation coefficients that are constant over cascade stages, the value of the relative total flow of the scheme can be used as such a criterion. Below is a description of the proposed methodology.

3. Methodology for calculating and optimizing the parameters of the triple cascade.

We formulate a mathematical formulation for the problem under consideration. As the constituent elements of the system, we have three rectangular cascades. As the constituent elements of the system, we have three rectangular cascades.

For the system under consideration, the following parameters are given:

- concentrations of components in the feed mixture \( C^F \);  
- he required concentrations of the four target isotopes in the outgoing flows of the scheme \( C^P_{n_2}, C^P_{k_2}, C^P_{n_3}, C^P_{k_3} \);  
- parameters of a single separating element. In the case of constant stepwise separation coefficients, the separation coefficient per unit difference of mass numbers \( q_{P_0} \).

In the calculation process, it is necessary to determine all the other parameters of the considered scheme, namely:

1) the values of the other external parameters: the concentrations of the other components in the external outgoing flows, the ratio between the values of the external outgoing and ingoing flows.

2) Internal parameters of the scheme: concentrations in the withdrawals of the first cascade, internal parameters and configurations of all square cascades in the system \( \left( \frac{L}{L} \right)_j, \left( \frac{W}{L} \right)_j, N_j, f_j, j = 1, 3 \)

In this case, the found cascade configuration must meet the minimum of the specified performance criterion. The following parameters act as variable parameters when solving such a problem:

- flow relations in all three cascades \( \left( \frac{L}{L} \right)_j, \left( \frac{W}{L} \right)_j, j = 1, 3 \)
- lengths of all cascades \( N_j \) and feed supply stage numbers \( f_j, j = 1, 3 \)
- concentrations of components in the outgoing flows of the first cascade.

For each set of the above-mentioned variable parameters, a sequential calculation of all SCs of the configuration specified in this step is performed. A similar calculation can be carried out by one of the known methods. In the framework of the work, was used a method based on the approximation of the separation factor of the cascade [18].
The formulated problem is the optimization problem on the space of variables of mixed type, since the variables \( N_j \) and \( f_j \) change discretely, while the desired flow ratios \( \left( \frac{W}{L} \right)_j \) - continuously. The proposed method for solving the problem is described below.

Given the specifics of the problem, it was proposed to break its solution into two steps: the first is the determination of the flow ratios \( \left( \frac{W}{L} \right)_j \), \( \left( \frac{W}{L} \right)_j \) for fixed \( N_j \) and \( f_j \), the second is the search for combinations of variables \( N_j \) and \( f_j \), that provide the best possible configuration of the cascade scheme by the selected efficiency criterion. In the case of equal stage separation coefficients on every stage, it is reasonable to use relative total flow as an efficiency criterion [19]. Since, as noted above, it is critical to obtain high concentrations of isotopes with intermediate mass numbers, the function \( \psi \) in the following form was chosen as an efficiency criterion:

\[
\psi = \sum L \left( \frac{P_3}{P_3 + W_2} \right)
\]  

(1)

The task described above is a combinatorial optimization problem, since in this case \( N_j \) and \( f_j \) can only take integer values. The problems of combinatorial optimization are \( NP \)-complete in the general case, which means that finding the exact solution is possible only by exhaustive search of all possible combinations [20]. For the task described above, the number of options for exhaustive search can be calculated by the formula:

\[
\prod_{j=1}^{3} \dim\{N_j\} \ast \dim\{f_j\}
\]  

(2)

where \( \dim\{N_j\}, \dim\{f_j\} \) are dimensions of the sets of possible values to enumerate.

The formula shows that the number of options grows geometrically, even with a small change in the range of variation of each variable. A preliminary analysis shows that implementing direct search to solve the problem is not practical, since the time to obtain a result using a personal computer will be unreasonably long. In such a situation, it is possible to use approximate methods to find the extremum of a function, based, in particular, on random search. The efficiency of a random search is explained by its good “performance” with great complexity and multi-extremity of the objective function. An important fact is that random search algorithms are universal and simple to implement. Unlike any deterministic algorithm, random search allows you to optimize any function, albeit with different efficiency.

The most general iterative formula by random search has the form:

\[
X_{k+1} = X_k + \xi_k
\]  

(3)

where \( \xi_k \) is an \( n \)-dimensional random variable, \( X_k, X_{k+1} \) the value of the variable vector at the current and next iteration, respectively. The probability distributions of this random variable, their changes in various steps of the method, determine the search method.

The class of algorithms is widely used, which is essentially a subclass of random search algorithms - metaheuristic (sometimes called "bionic") algorithms. The advantage of these methods is their ability to solve complex problems without first analyzing the search area. It can be simplified to consider metaheuristic methods as algorithms that implement a direct random search for possible solutions to the problem, optimal or close to optimal, until a certain condition is met or a given number of iterations is reached. In this work, to solve the problem, we used the bee colony algorithm ("ABC" algorithm) [21].

The algorithm, according to its name, is based on simulating the behavior of a colony of honey bees when collecting nectar in nature. The main purpose of the bee colony in nature is to explore the space around the hive in order to search for nectar with its subsequent ingathering. For this, there are various types of bees in the colony: scout bees and working forager bees. Scouts investigate the surrounding
hive space and provide information on promising places in which the largest amount of nectar was
discovered. This optimization method is described in detail in [21].

One of the reasons why the described optimization method was chosen is that there are no restrictions
on the type of variables used, which allows their combination (integer, real). However, for the effective
implementation of this method, it is necessary to have correctly selected initial approximations. Due to
the great uncertainty of the parameters of the triple scheme, it is not possible to find initial
approximations for its parameters of any simple considerations or using simple analytical calculations.
In this regard, to search for initial approximations, a scheme of three model cascades was used, which
are widely used in the theory of separation of multicomponent mixtures for estimation calculations. As
part of the work, a model cascade was selected that has one of the simplest mathematical models - the
Q-cascade.

The mathematical model of the Q-cascade is based on a system of differential equations describing
the molecular-selective mass transfer in the cascade for the separation of multicomponent mixtures [22]
in the case of small enrichments per stage. However, in the scientific papers that appeared in the last 10
years, the equivalence of the Q-cascade to another model cascade — the “quasi-ideal” cascade, as well
as the fact that both models are in fact particular cases of a symmetric countercurrent cascade with
separation coefficients that are constant over the steps [23], is shown. The results of these studies have
opened up the possibility of using Q-cascades, including for the case of isotope separation by the gas
centrifuge method.

The work has developed a methodology for calculating and optimizing the system of ordinary Q-
cascades. This system is inherently similar to the SCs scheme shown in Figure 1, however, the
calculation and optimization of such a scheme is much less complicated than the calculation and
optimization of a SC scheme; when calculating the Q-cascade, it is necessary to vary a much smaller
number of parameters, and there is also no need for the step-by-step calculation of its internal
parameters. At the same time the regularities of mass transfer of components in such a model are
preserved and correspond to a symmetric countercurrent cascade for the separation of multicomponent
mixtures. In the simplest case, the Q-cascade that provides the desired concentrations of the target
isotope in the outgoing flows can be optimized by the criterion of the minimum relative total flow
depending on only one parameter — the so-called M value, which actually represents the average mass
number of the mixture being separated [24]. That is why Q-cascades are widely used in theoretical
works devoted to the optimization of cascade schemes for the separation of multicomponent mixtures
[25]. Moreover, Q-cascades, like other model cascades, can be considered as “good” initial
approximations in the calculation and optimization of symmetrically countercurrent cascades of a square
or squared off profile [26].

In connection with the foregoing, in order to search for realistic initial approximations for the
parameters of the SC scheme, the scheme was initially calculated and optimized from three Q-cascades,
which provided the same concentrations in the outgoing flows.

For this system, the task of calculating the optimal parameters can be formulated as follows. For the
given parameters of the initial mixture (mass numbers \( M_1 \) component concentrations in the feed \( C^F \) it is
necessary to determine the optimal concentration values for a pair of different components in the
outflows of the first stage \( (C_{n_1}^{p_1}, C_{k_1}^{w_1}) \), values \( M_1, M_2, M_3 \) of all three cascades are such that in the
outgoing concentration of the selected components in the outgoing flows of the second and third
cascades coincide with the set values \( C_{n_2}^{p_2}, C_{k_2}^{w_2}, C_{n_3}^{p_3}, C_{k_3}^{w_3} \)

The optimization procedure for such a system of Q-cascades consists of the following steps. First,
initial approximations are set for a set of variable parameters: \( C_{n_1}^{p_1}, C_{k_1}^{w_1}, M_1, M_2, M_3 \). Having set the
indicated values, it is possible to first find the unknown lengths of sections of the first cascade \( (S_p, S_w) \),
numerically solving the system of equations for the concentration residuals, \( C_{n_1}^{p_1}, C_{k_1}^{w_1} \), for example, by
the Newton method. After that, the concentrations of the remaining components in each of the outgoing
streams of the first stage are calculated, as well as the ratios of the values of its outgoing flows. Knowing
them, as well as setting the initial values for the quantities $M_2$ and $M_3$, it is possible to implement the design calculation procedures for the second and third cascades. Next, it is possible to calculate the value of the selected criterion for this step (hereinafter - $\psi$). After that, a vector of initial approximations was formed for calculating the SC scheme.

The problem described above is a multidimensional optimization problem with constraints. At the same time, it should be noted that the use of deterministic methods for finding extrema, such as gradient descent [27] or the Nelder-Mead method [28], is problematic for solving this problem, since the methods of this family tend to converge to local extrema, which in our statement optimization tasks are artificial. To solve the problem, it is advisable to use a method related to the statistical family of methods for searching for extrema. In the framework of this work, the method of differential evolution was chosen [29].

4. Results and discussion

The developed methodology for calculating and optimizing a system of three SCs was used for calculations on the separation of several isotopic mixtures with a different number of components. As an example, we consider a four-component mixture of lead isotopes that best fits the main objective of the scheme — the concentration of the four isotopes of a mixture under separation. The concentrations of the components in the natural isotopic mixture of lead are shown in Table 1. The following concentrations of isotopes in the outgoing streams were specified for this mixture: $C_{p1}^{W1} = 99\%$, $C_{p2}^{W2} = 99\%$, $C_{p3}^{W3} = 99\%$, $C_{p4}^{W4} = 99\%$.

The value of the coefficient of separation into the unit difference of mass numbers is set equal to $q_0 = 1.06$. As a working substance we considered Pb(CH$_3$)$_4$ [9].

Table 1 shows the concentrations of all components in the flows of an optimized scheme of three SCs. As can be seen from the analysis of the data in table 1, the proposed scheme really allows to actually completely separate the four-component mixture into flows in which each of the components is enriched.

| $i$ | $M_i$ | $C_{i}^{W1},\%$ | $C_{i}^{P1},\%$ | $C_{i}^{W2},\%$ | $C_{i}^{P2},\%$ | $C_{i}^{W3},\%$ | $C_{i}^{P3},\%$ | $C_{i}^{W4},\%$ | $C_{i}^{P4},\%$ |
|-----|-------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| 1   | 204   | 7.79·10^{-23}   | 4.14·10^{-10}   | 0.152           | 99.0            | 5.48            | 1.0·10^{-13}    | 1.40            |
| 2   | 206   | 3.78·10^{-8}    | 0.644           | 99.0            | 0.999           | 93.65           | 0.22            | 24.10           |
| 3   | 207   | 0.999           | 99.0            | 0.847           | 5.24·10^{-7}    | 0.87            | 29.39           | 22.10           |
| 4   | 208   | 99.0            | 0.356           | 4.97·10^{-10}   | 1.95·10^{-13}   | 1.0·10^{-7}     | 70.39           | 52.40           |

Below are the results of applying the proposed scheme for a mixture with the number of components equal to five. An example of such a mixture is a natural mixture of tungsten isotopes (table 2). The following isotope concentrations in the effluent were set for this mixture: $C_{p2}^{W2} = 99\%$, $C_{p3}^{W3} = 95\%$, $C_{p4}^{W4} = 99\%$. The value of the coefficient of separation into a unit mass difference is set equal to $q_0 = 1.16306$. Table 2 shows the concentrations of all components in the outflows of an optimized scheme of three SCs. As can be seen from the analysis of the data in table 2, the proposed scheme also allows in each of the flows to concentrate a separate component of the initial mixture.

| $i$ | $M_i$ | $C_{i}^{W1},\%$ | $C_{i}^{P1},\%$ | $C_{i}^{W2},\%$ | $C_{i}^{P2},\%$ | $C_{i}^{W3},\%$ | $C_{i}^{P3},\%$ | $C_{i}^{W4},\%$ | $C_{i}^{P4},\%$ |
|-----|-------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| 1   | 180   | 1.17·10^{-18}   | 1.52·10^{-8}    | 9.99·10^{-11}   | 0.529           | 0.35            | 1.0·10^{-12}    | 0.14            |
| 2   | 182   | 5.13·10^{-8}    | 0.063           | 1.47            | 99.0            | 66.28           | 1.0·10^{-5}     | 26.41           |
| 3   | 183   | 4.17·10^{-8}    | 4.29            | 95.0            | 0.470           | 32.27           | 2.58            | 14.41           |
| 4   | 184   | 0.999           | 95.0            | 3.52            | 1.17·10^{-6}    | 1.09            | 50.18           | 30.62           |
| 5   | 186   | 99.0            | 0.651           | 4.45·10^{-7}    | 8.55·10^{-24}   | 0.01            | 47.24           | 28.42           |
Also, for each of the mixtures considered, the found SC scheme was compared with the optimal Q-cascade scheme and the conditional shape efficiency was calculated in each case. The efficiency of the form was determined as

$$\eta = \frac{\psi_Q}{\psi_{SC}}$$

The results are shown in table 3.

**Table 3.** Comparison of the optimal scheme of Q-cascades and the optimal scheme of SC

| Mixture | Q-cascade scheme | SC scheme | η, % |
|---------|------------------|-----------|-----|
| W       | 2691.56          | 5322.1    | 50.6|
| Pb      | 29018.4          | 43958.2   | 66.0|

In the table 4 there are founded optimal parameters of SC schemes, described before.

**Table 4.** Optimal parameters of SC schemes

| Parameter | Tungsten case | Lead case |
|-----------|---------------|-----------|
| N₁        | 62            | 318       |
| f₁        | 44            | 171       |
| N₂        | 123           | 143       |
| f₂        | 56            | 40        |
| N₃        | 52            | 307       |
| f₃        | 24            | 129       |
| P₁/L₁     | 0.02096       | 0.00952   |
| W₁/L₁     | 0.03167       | 0.02773   |
| P₂/L₂     | 0.04252       | 0.00127   |
| W₂/L₂     | 0.02155       | 0.02193   |
| P₃/L₃     | 0.03669       | 0.00662   |
| W₃/L₃     | 0.03344       | 0.01624   |

5. **Conclusion**

A scheme of a system of square cascades has been developed for the simultaneous concentration of four isotopes of a multicomponent mixture under separation to obtain relatively high (over 90%) concentrations for each of them.

For the developed cascade scheme, a method for calculating and optimizing its parameters according to the criterion of the minimum total flow normalized to the sum of flows enriched with intermediate isotopes is proposed. An optimization technique is developed based on the use of a metaheuristic algorithm - the “ABC” algorithm. In this case, as initial approximations in the optimization calculation, the parameters of a system of Q-cascades of a similar structure were used, the optimization of which was also carried out by a metaheuristic algorithm - “differential evolution”.

The efficiency of the proposed method for the simultaneous concentration of several isotopes is illustrated by the example of the separation of two isotopic mixtures: lead and tungsten of natural composition. The possibility of a significant excess of the maximum concentrations due to the use of the scheme is shown. In particular, for lead isotopes, it is possible to obtain all four of its isotopes with concentrations of 99%.
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