Peculiarities of the transient processes in cascades for separation of isotope mixtures with various numbers of components

A Yu Smirnov[1], A K Bonarev[1], G A Sulaberidze[1], V D Borisevich[1], S Zeng[2], D Jiang[2] and Y N Zhang[2]

[1] National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Kashirskoe highway 31, Moscow, 115409, Russia
[2] Tsinghua University, Engineering Physics Department, Beijing, P.R.China

E-mail: AYSmirnov@mephi.ru

Abstract. The peculiarities of the transient processes in separation of isotope mixtures with various numbers of components are considered. A model *Q*-cascade without its depletion part is taken as an object of the research. It is shown that before entering the stationary mode of operation in the cascade the concentrations of the intermediate multicomponent mixture components in the product flow demonstrate the nonmonotonic behavior when they pass through the maximum and the minimum before the subsequent achievement of their steady-state values. It is revealed during calculations that the "abnormal" behavior of the end concentrations is primarily caused by the initial composition of a separated mixture.

1. Introduction

In recent decades, there has been substantial growth in demand for the isotopically modified products. This is the result of intensive development of science, technology, and medicine which requires the use of high purity isotopes of various chemical elements. The most methods of isotope separation are characterized by the use of multistage separation installations (cascades), consisting of series-connected separation stages, which contain parallel connected separating elements (for example, gas centrifuges).

For a theoretical description of isotope separation processes in cascades it was developed various models of molecular-selective mass transfer in multistage installations as well as related methods to calculate their parameters [1-9]. An important role in theory of cascades for isotope separate plays the so-called model cascades [1, 2, 4, 8-12] that allow to simplify the analysis of the mass transfer laws in a cascade and corresponding computational procedures for calculation and optimization of its parameters. At the same the results obtained correspond to the separation process in the cascades used in practice. One of the widely used for this purpose model cascade is a *Q*-cascade [4, 11-18].

The duration of transient processes largely determines the efficiency of the cascade work, is one of its important characteristics. The main way to solve the problem of describing the non-stationary mass transfer in cascades for the separation of multicomponent mixtures is the numerical calculation of parameters for the symmetrically countercurrent cascade [19-23]. In the recent paper [24] Recently the

mathematical model of a Q-cascade without its depleted part has been offer to study the physical laws of the transient processes in cascades for separation of multicomponent mixtures.

For the first time it was obtained the solution allowed to calculate analytically the values of the partial flows and concentrations for all components of a separated mixture at any time at any stage of the cascade. It was found that before entering a steady state operation mode of the cascade, the concentrations of some components with intermediate mass numbers in the product flow of the cascade demonstrate nonmonotonic behavior, passing through the maximum and minimum values before reaching their steady-state values. It was found in the conducted computational experiments that such the "abnormal" behavior is determined by the initial composition of a separated mixture, i.e. by the relations between the concentrations of its various components.

The present work is an extension of the research which results are published in [24]. It is devoted to an in-depth analysis of the physical reasons of the anomalous behavior of intermediate component concentrations when a cascade entering its stationary mode.

2. Mathematical model

As the object to study we use a Q-cascade without a depletion part which mathematical model is described in detail in [24]. We consider that this Q-cascade is connected to the infinite volume reservoir (V) filled with a mixture of the component concentrations C_i^F where i is a number of an arbitrary component of a mixture consisting of the m components (see figure 1). The product flow P is withdrawn from the opposite end of the cascade, where the light components of the mixture are enriched (so-called the "light" end of a cascade). We believe that at the initial time moment the cascade is also completely filled the homogeneous mixture whose composition corresponds to that in the reservoir. The length of the cascade (the number of stages in it) is denoted as S_P. The stage number is counted from the stage where the product flow P is withdrawn to the stage where a feed flow enters the cascade.
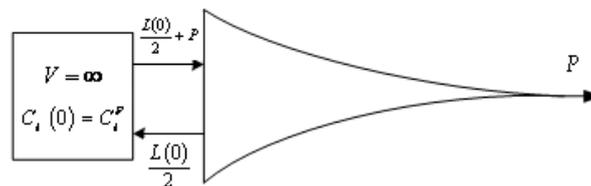

Figure 1. Schematic drawing of a Q-cascade with the infinite reservoir at its heavy end.

The partial flow of the ith component in the non-stationary regime of the Q-cascade under consideration is presented as follows [24]:

$$G_i(y_i, \tau_i) = 2P \cdot C_i^F \frac{1 - \exp\left(y_i - y_i^P\right)}{1 - \exp\left(-y_i^P\right)} \sum_{j=1}^{m} \frac{C_j^P}{Q_j}\left[1 - \exp\left(-y_i^P\right)\right] +$$

$$+ \sum_{l=1}^{\infty} A_{l,i} \sin\left(\frac{\pi l y_i}{y_i^P}\right) \exp\left[\frac{1}{2} y_i - \left(\frac{1}{4} + \left(\frac{\pi l}{y_i^P}\right)^2\right)\tau_i\right]$$

(1)

where $y_i = Q_i s$, $\tau_i = \dfrac{2Q_i^2}{\omega}t$ are the new variables; $y_i^P = Q_i S_P$; ω is the residence time of a separated mixture at a cascade stage which value is considered for all cascade stages; Q_i are the constants connected by the condition $Q_i - Q_j = \varepsilon_{ij}$, which for the molecular kinetic separation methods are transformed to the following relationship $Q_i = \varepsilon_0(M - M_i)$ [4, 12]; M_i is a mass number of the

component with the i number; M is a parameter which allows to calculate the Q_i constants for all components of the separated mixture; ε_{ij} are the relative enrichment factors for the pair of components with the i and j numbers; ε_0 is the enrichment factor for the unit mass difference. The $A_{l,i}$ are expressed as follows

$$
A_{l,i} = \frac{4P \cdot C_i^F}{S_P} \sum_{j=1}^{m} \frac{C_j^P}{Q_j} \left\{ \left[1 - \frac{1 - \exp\left(-Q_j S_P\right)}{1 - \exp\left(-Q_i S_P\right)} \right] \frac{S_P \big/ \pi l \left[1 - \exp\left(-1/2 Q_i S_P\right) \cos \pi l \right]}{1 + \left(Q_i S_P \big/ 2\pi l \right)^2} - \right.
$$
$$
- \exp\left(-Q_j S_P\right) \frac{S_P \big/ \pi l \left[1 - \exp\left(\left(Q_j - 1/2 Q_i\right) S_P\right) \cos \pi l \right]}{1 + \left(\left(Q_j - 1/2 Q_i\right) S_P \big/ \pi l \right)^2} + \tag{2}
$$
$$
\left. + \exp\left(-Q_i S_P\right) \frac{1 - \exp\left(-Q_j S_P\right)}{1 - \exp\left(-Q_i S_P\right)} \frac{S_P \big/ \pi l \left[1 - \exp\left(1/2 Q_i S_P\right) \cos \pi l \right]}{1 + \left(Q_i S_P \big/ 2\pi l\right)^2} \right\}
$$

The values of the component concentrations at the product end of the cascade in the stationary mode of its exploitation can be calculated by the following formula:

$$
C_i^P = \frac{Q_i C_i^F}{1 - \exp\left(-y_i^P\right)} \bigg/ \sum_{j=1}^{m} \frac{Q_j C_j^F}{1 - \exp\left(-y_j^P\right)} \tag{3}
$$

Thus, after specifying the number of stages in the cascade S_P, the initial composition of a separated mixture as well as the values of the parameters M by the formulas (1) - (3) one can carry out the analytical calculation of the non-stationary concentration distributions over cascade length at various moments of time.

It should be emphasized that despite of the fact that the mathematical model of a Q-cascade is developed for the case of the so-called "low enrichment", the results obtained using this model, valid also in the case of arbitrary enrichments at cascade stages [12, 24].

3. Results and discussion

The model of non-stationary mass transfer in a Q-cascade briefly described above was used to study the basic laws of transient processes in multistage installations for separation of multicomponent isotope mixtures. The calculations were performed for isotopic mixtures with the various numbers of components (from 3 to 5). The isotopic compositions of examined mixtures are presented in tables 1-3. The details of the algorithms to calculate the parameters of the transient processes in a Q-cascade may be found in [24].

In the analysis of the results of our computational experiments we based on the assumption that the process of reaching the stationary mode of operation of a cascade can be conditionally divided into 2 parts [25, 26]. At first one the change in concentration of the mixture components occurs independently. At the second part there is the mutual influence of the components on each other, associated with the process of their accumulation on the cascade stages. If one would like to consider the specifics of the initial part of the process in detail, it can be assumed that at this stage the dynamics of concentration change is determined by the below system of partial differential equations, which describes the unsteady separation process in the case of the "weak" enrichment [25-26]:

$$
\frac{\partial C_i}{\partial \tau} = \frac{\partial^2 C_i}{\partial y^2} - \frac{\partial C_i}{\partial y} \sum_{j=1}^{m} \left(M_j - M_i\right) \cdot C_j - C_i \sum_{j=1}^{m} \left(M_j - M_i\right) \frac{\partial C_j}{\partial y} - \frac{2T}{\varepsilon_0 \cdot L} \frac{\partial C_i}{\partial y} \tag{4}
$$

$$y = \varepsilon_0 s, \quad \tau = \frac{\varepsilon_0^2 t}{\omega}, \quad \omega = \frac{H(s)}{L(s)} = const \ .$$

where $H(s), T(s), L(s)$ are the stage holdup (the amount of a working substance that is held by a separation stage of a cascade), the mass transfer and the flow of a working substance at the entrance of the cascade stage with the s number, respectively.

The nonlinear term in equation (4) can be replaced by the following linear approximation

$$C_i \sum_{j=1}^{m} (M_j - M_i) \cdot C_j \approx b_i \cdot C_i \tag{5}$$

where $b_i = \overline{M} - M_i$; $\overline{M} = \sum_{j=1}^{m} M_j C_j^F$ is the average mass number of the initial mixture of isotopes.

Three multi-isotope mixtures were investigated in the computing experiments (see tables 1-3). The first one was the 5-component natural mixture of the tungsten isotopes. The intermediate component with the mass number 184 was chosen as a target component; the separation factor for the unit mass difference was equal to $q_0 = 1.16$ [9]; the parameter M was equal to be 184.5, which corresponds to the preferential enrichment of the key isotope in the flow of the light fractions. In addition, two model 3 and 4-component mixtures which compositions were chosen so that the content of the intermediate components was comparable with that for the utmost components were investigated. This suggests that the impact of these components on the mass transfer laws in the transient regimes is comparable to the effect of utmost isotope masses.

The mass numbers and concentrations of isotopes, as well as the number of the target component for the model mixtures were chosen so that the b_i parameter for the key isotope was close to zero. As is shown in [25, 26], it is crucial for the emergence of the "abnormal" behavior of a component. The M parameter for each of the model mixtures corresponded to enrichment the targeted "intermediate" isotopes at the light end of the cascade. The separation cascade parameters for each case under consideration are listed in table 4.

The dependences of the component concentrations for the intermediate masses in the considered multi-component mixtures on the relative time of the cascade work are shown in figures 2-6. As the relative time we understand the ratio of the current time of the cascade work (t) to the residence time of a separated mixture at a cascade stage (ω=const). The quantity t/ω was varied in the limits from 0 to a value which is corresponded to the steady state mode of a cascade exploitation.

The analysis of the computational experiment results allowed confirming the following fact. The concentrations of some intermediate components in the transient mode in the product flow from the cascade successively pass through the maximum and minimum values that resemble a kind of a swing and then reaching their steady-state values. The results are qualitatively consistent with that of [25, 26] in which the problem of the non-stationary mass transfer in cascades for the separation of multicomponent isotope mixtures is solved numerically.

As it follows from the analysis of the initial compositions of the separated mixtures considered in this research and the features of the curves in figures 2-6, the "abnormal" change of concentration takes place for those components of the mixture, in which the absolute value of the b_n coefficient is the smallest one or close to zero.

From the physical point of view, such a non-monotonic behavior of the component concentrations in the separated mixture can be explained by the proximity of an average mass number and the mass of the component. It means that this component has almost no the strongly marked direction of enrichment in a cascade. In other words, the concentration gradient of this isotope is the alternating one within a cascade, including the area next to its product end. This fact is illustrated by the curves in figure 7, which demonstrate the dependence of the b_n parameter for the [183]W isotope. It can be seen that the magnitude of b_n in the transient process takes both positive and negative values. When

comparing the curves in figures 5 and 7 one can notice that the area of the "abnormal" behavior corresponds to that, in which the value of b_n changes its sign. In addition, for the components ^{182,184}W the b_n parameter had the permanent signs throughout all transition process. Note that such kind of instability in the enrichment of the intermediate components can be observed in inside cascade either in the transient [21] or in the steady regime of a cascade work [27].

Calculations performed for the 3 and 4-component mixtures have led to the similar results. At the initial stage of the transient process there were observed the maxima and minima ("a swing") for the end concentrations of the intermediate components, for which the value of the b_i parameter was close to zero (see figures 2 and 3).

Thus, the "abnormal" change of the concentrations of the isotope with the intermediate masses in the product flow from a cascade is mostly due to the initial composition of the separated mixture.

Table 1. Composition of the three-component model mixture

Number of component i	Mass number of isotope	c_{iF},%
1	100	20.0
2	101	40.0
3	102	40.0

Table 2. Composition of the four-component model mixture

Number of component i	Mass number of isotope	c_{iF},%
1	298	10.0
2	299	30.0
3	300	30.0
4	301	30.0

Table 3. Composition of the natural mixture of tungsten isotopes

Number of component i	Mass number of isotope	c_{iF},%
1	180	0.12
2	182	26.50
3	183	14.31
4	184	30.64
5	186	28.43

Table 4. Parameters of a Q-cascades for separation of multicomponent mixture

	Model 3-component mixture	Model 4-component mixture	Natural abundance of the 5-component mixture of the tungsten isotopes
Cascade length, S_p	18.5	18.5	52.5
Parameter M	101.5	299.5	184.5
Separation factor, q_0	2.00	2.00	1.16
Number of a target component, n	2	2	4
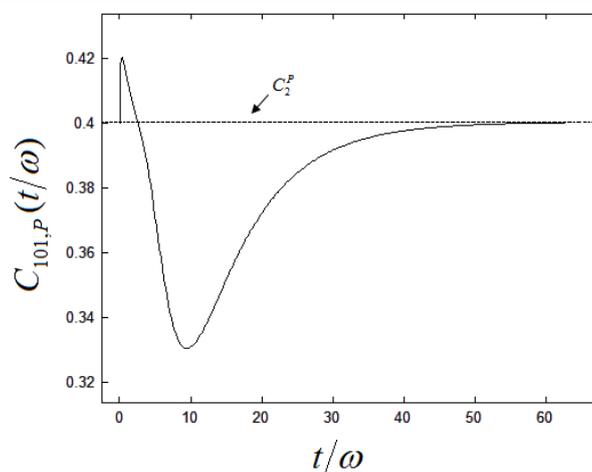
Figure 2. Variation of the ^{101}X component concentration of the model 3-component mixture in the product flow of the cascade on the relative time of its work.
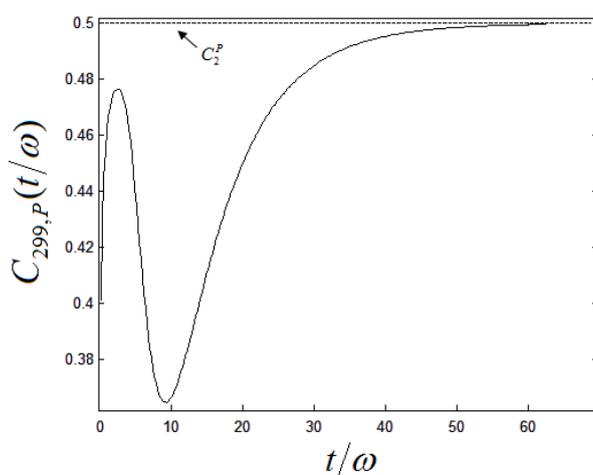
Figure 3. Variation of the ^{299}X component concentration of the model 4-component mixture in the product flow of the cascade on the relative time of its work.
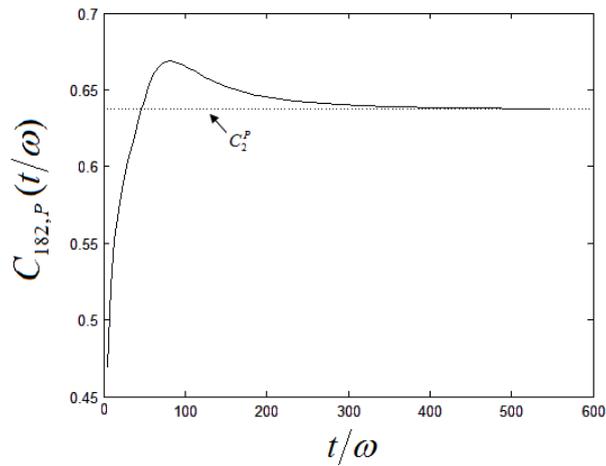
Figure 4. Variation of the ^{182}W component concentration of the tungsten isotopes of a natural abundance in the product flow of the cascade on the relative time of its work.
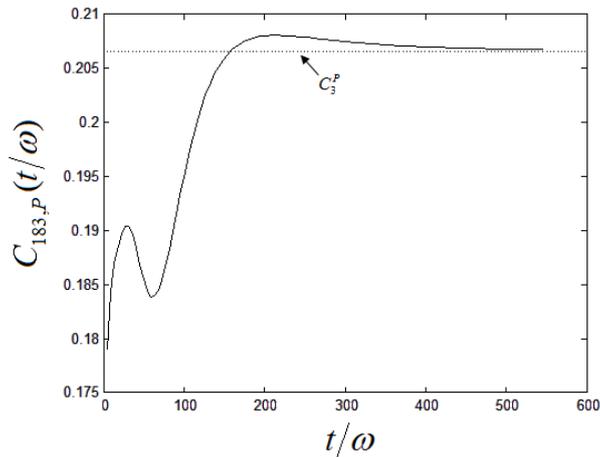

Figure 5. Variation of the ^{183}W component concentration of the tungsten isotopes of a natural abundance in the product flow of the cascade on the relative time of its work.
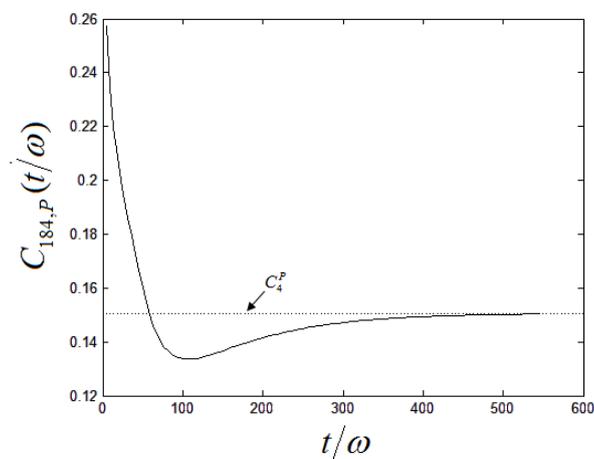

Figure 6. Variation of the ^{184}W component concentration of the tungsten isotopes of a natural abundance in the product flow of the cascade on the relative time of its work.
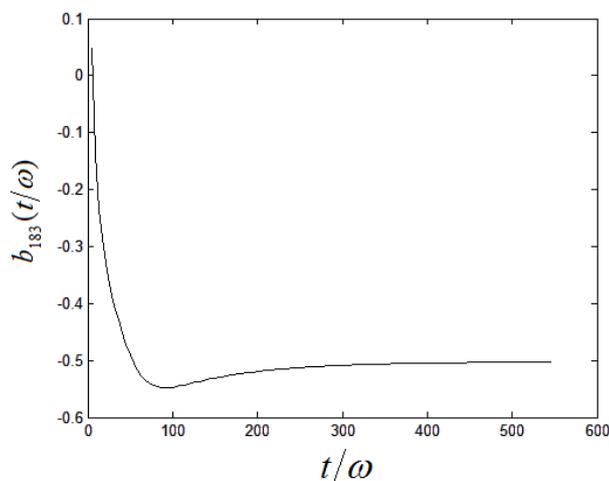
Figure 7. Variation of the b_i parameter for the ^{183}W component concentration of the tungsten isotopes of a natural abundance on the relative time of the cascade work.

4. Conclusion

The results of calculations obtained with help of the developed analytical solution demonstrated the qualitative agreement in regularities of the transient processes in the model cascades and the previously accumulated results of the numerical simulation of the same processes in the symmetric countercurrent cascades for separation of multicomponent isotope mixtures. In particular, it was detected the "abnormal" variation the end concentrations for the intermediate component (passing through the maximum and minimum values before entering the stationary one), which is due both to the initial composition of the separated mixtures, and the parameters of the separation cascade.

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