Peculiar properties of transient processes in cascades with additional product flow

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Abstract. One of the important stages in the campaign for separation of multicomponent isotopic mixtures is bringing the separation cascade to a stationary mode of operation. This process, depending on the applied separation method, can be characterized by a relatively long time with associated consumption of the working substance and operating costs. Therefore, the search for the values of the control parameters of the cascade at which it is possible to reduce the transient process time is practically a significant problem in the theory of isotope separation in cascades. One of the current trends in the modern theory of cascades is the study of physical laws of stationary mass transfer in cascades with additional outgoing flows. Such cascades can find application for solving a number of urgent problems, in particular, obtaining relatively high concentrations of isotopes with intermediate masses. At the same time, the laws of unsteady mass transfer in such cascades remain poorly understood. The paper analyses the key patterns of transient processes in a square cascade with an additional product flow. The interconnections between the time of bringing such a cascade to the stationary mode and the external conditions under which the process of separation of the mixture occurs are revealed.

1. Introduction

In contrast to the case of binary isotope mixture separation, a few target components in multicomponent isotopic mixtures can be. For instance, the isotopes having the intermediate mass numbers among the mass numbers of the separating components are often the subject to concentrate. Moreover, in the case of multicomponent mixture separation in an ordinary (three-stream) cascade, it is impossible to obtain the concentrations of intermediate isotopes above the limit which is determined by the composition of the initial mixture [1]. However, it is often required to obtain a concentration of the intermediate component larger than the limited one [2]. To solve this problem, it was proposed to use the series connection of several cascades (double cascades, etc.) [2], or to apply the cascades with the additional outflows (the product flows), which are switch on at the internal stages of the cascade, where the concentrations of the intermediate components may exceed the theoretical limits for the end outgoing flow [2].

Along with the necessity to study the stationary mass transfer regularities in cascades, it is also important to study the non-stationary mass transfer laws in the process of achievement the stationary target component concentrations in the outgoing flows of the cascade. Reducing the steady state exit time of a cascade effects the unit costs of the final product, caused by exploitation of the separation and auxiliary equipment, as well as the shared separated mixture flow rate.
Most of the published papers on the topic of the transient processes in separation cascades are aimed to study the non-stationary mass transfer processes in the three-stream separation cascades [8]. Special attention is paid to the study of the processes associated with cascade filling with a working substance [8] at unsteady flows and their influence on the further dynamics of achievement the stationary concentrations. In particular, it was shown that taking into account the non-stationary flows of the cascade leads to the fact that the distribution of component concentrations along the length of the cascade differs from that in the “classical” case when non-stationary processes in cascades are considered in the approximation of stationary flows. However, a significant difference is observed only in the initial period of the transient process, after which the distributions of component concentrations for both approximations practically coincide. Also, the influence of the values of the external flows on the concentration distributions and the time to reach the stationary concentrations were analyzed [10]. At the same time, the physical laws of the transient processes in the multi-stream cascades remain are not studied enough.

It follows that the study of non-stationary mass transfer in the multi-stream cascades as well as the research of the relationship between the parameters of such cascades with the attainment of the stationary concentrations in the outflows (external) and the dynamics of changes in the concentration distributions within the cascade is an important problem of the separation theory of the multicomponent isotopic mixtures which has to be solved.

This work aimed to study the physical laws of mass transfer in the cascades with the additional product flow in the process to achieve its stationary mode of operation. The influence of the choice the time point to switch on an additional product flow counting from the beginning of cascade operation as well as a course of an unsteady process in cascade are studied.

2. Theory

A square symmetric cascade (Figure 1) was chosen as the object to study separation of a multicomponent mixture consisting of $K$ components in the $N$ stages cascade of gas centrifuges. The direction of enrichment of the lightest component in such cascade is defined as the direction of enrichment while the opposite one is the depletion direction for this component. The main equations in a stationary state in a cascade are the equations of the material balance of the stage (1)

$$
\theta_n L_n - (1 - \theta_n) L_{n-1} = P^*_n
$$

$$
\theta_n C_{n,i} - (1 - \theta_n) C_{n-1,i} = P^*_n; \quad n = 1, 2, \ldots, N - 1; i = 1, 2, \ldots, K
$$

where $P^*_n$ is the flow transferred in the enrichment direction at the $n$th stage in the cascade, $P^*_n,i$ is the flow of the $i$th component in the enrichment direction at the $n$th stage in the cascade, $\theta_n$ is the cut of the
nth stage flow, and $L_n$ is the interstage flow rate of the $n$th stage; $C_{n,i}$, $C'_{n,i}$ and $C''_{n,i}$ are the concentrations in the feed flow, product flow and waste flow of the $i$th component at the $n$th stage, respectively. $F_n$ is the feed or product flow at the $n$th stage, and $F_{N+1}$ is the product flow at the $n$th stage. $F_n$ is defined as

$$F_n = \begin{cases} 0 & \text{No feed and no withdrawal} \\ > 0 & \text{Feed} \\ < 0 & \text{Withdrawal} \end{cases}$$

where $n = 1, 2, ..., N + 1$. $C_{Fn, i}$ is the concentration of the $i$th component corresponding to $F_n$ at the $n$th stage. Equation (1) reflects the overall material balance at the cascade. Equation (2) shows the material balance of the $i$th component. Equation (3) is obtained from the material balance at the $n$th stage. Equation (4) shows the material balance between the stages with the numbers $(n - 1)$th, $n$th, $(n + 1)$th, and the external feed or product flows $F_n$ at the $n$th stage.

The boundary conditions are determined by the operating conditions of the cascade. For the example shown in figure 1, the boundary conditions look as follows:

$$P_{N+1} = -F_{N+1}C_{F_{N+1}}; C_{F_{N+1}} = C_{N+1}; P_N = -F_{N+1}$$

$$P_i = -F_iC_{F_i}; C_{F_i} = C_{i}; P_e = F_i$$

(5)

In this research model, the process of isotope separation is carried out by the molecular kinetic methods namely gas centrifuge [15]-[17]. In addition, the separation factor does not depend on a cut ratio $\theta_n$ of the flow and the feed flow of a separation element. Therefore, a separation factor $q_{i,j}$ can be approximated by the following relation:

$$q_{i,j} = \frac{M_j - M_i}{M_i}$$

(6)

where $q_0$ is the separation factor per unit mass difference, $M_i$ and $M_j$ are the molar masses of the $i$th and $j$th components, respectively [18].

Let us set the cut of the flow at a first stage of the cascade is equal to $\theta_1 = 0.5(1 - W/L_1)$. In this case, other cuts will be calculated by the formula (7):

$$\theta_n = \begin{cases} 1 - \frac{L_n - \theta_n - 1}{L_n} - \frac{W}{L_n}; n \leq N_F \\ 1 - \frac{L_n - \theta_n - 1}{L_n} - \frac{P}{L_n}; N_F < n \leq e n = 2, ..., N \\ 1 - \frac{L_n - \theta_n - 1}{L_n} - \frac{W}{L_e}; e < n \leq N_F \end{cases}$$

(7)

where $e$ is the position of the point of an additional flow.

The specific amount of a substance per each component of the mixture at the $n$th stage is determined by the relation (8). It characterizes the rate of change in the amount of the $i$th component at the $n$th stage of a cascade:

$$H_n \frac{\delta(C_{n,i})}{\delta t} = P_{n-1,i}(t) - P_{n,i}(t) + F_n C_{F,i}$$

(8)

In this model, we assume that $\theta_n$, $L_n$ and $H_n$ are time constant. Only concentrations or concentration-dependent parameters are changing over time.

Substituting the boundary conditions (5) and equations (1-4) in the equation (9) we obtain:

$$H_n \frac{\delta(C_{n,i})}{\delta t} = [1 - \delta(n, 1)] [\theta_n - L_n C_{n-1,i} + (1 - \theta_n) L_n C_{n,i}]$$

$$- [1 - \delta(n, N)] [\theta_n L_n C_{n,i} + (1 - \theta_n) L_n C_{n+1,i}] + F_n C_{F,i} + \delta(n, N) F_{N+1} C_{F_{N+1}}$$

(9)

where $\delta(n, x)$ is the Kronecker symbol.
The solution of this equation reduces to replacing $C_{n,i}$ and $C'_{n,i}$ by $C'_{n,i}$. Further, the resulting system of differential equations describing the change in concentration during the transient in the cascade can be solved by the finite difference method [10].

The criterion to find the stationary component concentrations at the cascade is the fulfillment of the condition (10) simultaneously for all the outgoing flows from the cascade, where $t$ is the time step.

$$\sum_{i} \frac{C_{i,j}(t^{+\Delta t})-C_{i,j}(t)}{C_{i,j}(t^{+\Delta t})} \leq 10^{-2}$$

(10)

Based on this algorithm, a computer code was created using the open-source NumPy library for the Python programming language. This library is supported the multidimensional arrays and high-level mathematical functions designed to work with the multidimensional arrays. The developed code was tested in a series of calculation for various cascade schemes and separated mixtures. The results of the computational experiments demonstrated that the developed code is effective in a wide range of problems under investigation since it is based on a general mathematical model of a symmetric countercurrent cascade with an arbitrary number of outgoing flows and components of the separated mixtures.

3. Formatting the text Results and discussion

A square cascade with an additional flow included at one of the stages between the point of the feed flow stage and the last stage from which the light fraction is extracted as the object of study. The mixture to be separated is tungsten (in the form of WF$_6$) with the natural concentration of components. The scheme of the cascade under consideration is presented in Figure 2. The main assumptions used in the work are as follows:

- At the initial time moment, the concentrations at all stages of the cascade are the same and equal to the natural concentration of the mixture being separated.
- The time to reach the stationary regime in concentrations is much longer than that of to establish stationary flows.
- The cut $\theta_n$ at each stage does not change in time.
- The interstage flow rate $C'_{n,i}$ for each stage is constant in time and is the same for each stage.
- The holdup at the stage $H_n$ does not depend on time and is the same for each stage.

![Figure 2. Scheme of a squared cascade with an additional product flow](image)

The cascade parameters are: the number of stage $N_f = 40$, the feed point $f = 16$, the additional flow point $e = 33$, the ratio of the feed flow to the product flow for the cascade as a whole $F/P = 2.47$, the product flow is normalized to the inter-stage flow rate $P/L_n = 0.0493$, the separation factor per unit mass $q_0 = 1.16306$, the holdup on the stage related to the value of the inter-stage flow rate $H_n/L_n = 0.03$.

For the conditions described above, calculation were carried out to determine the pattern for the
dependence on the time to achieve the stationary mode since the start-up of the additional withdrawal flow counted from the start of the cascade work as a whole.

**Figure 3.** Dependence of the stationary time in a cascade on the time to switch-on an additional product flow

The obtained dependence is presented in Figure 3. As follows from its analysis, it is possible to find the optimal time moment providing. According to the dependence presented in Figure 3, this time is about 13 hours. Moreover, the time to reach the stationary mode is 28.6 hours. The total time to go to the stationary is 42.38 hours, which is 32.5% more than the optimal value.

One of the possible reasons that can explain the revealed patterns is that at the moment of switch-on of the additional flow, a redistribution of concentrations in the cascade occurs since the three-stream cascade that has not yet reached the stationary becomes four-stream with other stationary concentrations. Thus, including switch-on an additional flow at a certain time point, it can be achieved that this “perturbation” can favorably influence the distribution of concentrations, leading to a reduction in the time for the cascade to reach stationary concentrations. However, this assumption requires further verification which is planned in an additional research.

**4. Conclusion**

In the paper it is analyzed the patterns of mass transfer in a square separation cascade for multicomponent isotope separation with an additional product flow. The relationship between the cascade steady state time and the time of a switch-on moment for an additional flow is discovered. It was shown that the steady state cascade time has a non-linear dependence on the time to switch-on the additional product flow. The analysis of this dependence allows us to determine that there is the moment to switch-on of an additional flow corresponding to the minimum time for the cascade with its stationary concentrations. The optimal moment to start-up the additional product flow gives the possibility to achieve a significant reduction of the steady state cascade time in the case when the additional product flow occurs. In the considered in the paper example for separation of the tungsten isotope mixture, the decrease in the cascade steady state time to the stationary was 32.5%.

The revealed patterns can be used in the search for the optimal conditions for a separation campaigns. This can be especially attractive for cases when the time of a separation campaign is too long or limited for some manufacturing reason. Under these conditions, reduction in the time to its stationary concentrations can significantly affect the efficiency of the production of intermediate isotopes. In addition, the revealed regularities are also valid for other diffusion separation methods, in which the reduction of the transition process time is one of the key problems due to their relatively long duration.

**Acknowledgments**

This research was supported by the grant of the Russian Foundation for Basic Research no. 19-58-53002 GFEN_a and NSFC Grant No. 11911530087.
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