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CASCADES FOR SEPARATION OF MULTICOMPONENT ISOTOPE MIXTURES

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ABSTRACT

This article reviews and analyzes the state-of-the art on separation of multicomponent isotope mixtures in cascades. Different methods, which are applied to calculate the cascade configurations and designs as well as those used to describe some transient processes in cascades when separating multicomponent isotope mixtures are discussed and analyzed in detail. The main directions for the further development of isotopes fractionation using cascades are formulated.

INTRODUCTION

Production of many types of isotopes, which are widely used in different fields of science and technology, is tightly connected with separation of multicomponent isotope mixtures and in many instances with concentration and recovery of intermediate (by mass) components of desired purity. The theory of separation of multicomponent isotope mixtures is complicated by the necessity to account for the reciprocal influence of all components on the resultant transfer of desired isotopes. Despite numerous achievements in this field many problems still remain to be solved.

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The development of theory for separation of multicomponent isotope mixtures started in 1961 by A. de la Garza (1). Many published papers (1–10,13–18, 24–35) are devoted to the creation of theory for countercurrent cascade under the assumption that the single separation effect is small and does not depend upon composition of the mixture (component concentrations). This assumption is known to be valid for separation methods such as, gas diffusion, thermodiffusion, mass diffusion, and cryogenic distillation. The peculiarities of calculations for cascades with higher enrichment degrees at each stage that is observed, for example, in cascades composing ultracentrifuges or membrane separative elements have been reviewed (11,12,19–23).

The main goal of this study is to critically review and analyze the literature (published by both the authors of the present paper and other researchers) on separation of multicomponent isotope mixtures in cascades and to formulate the basic trends for further development of this particular field of Separation Science.

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GOVERNOR CASCADE EQUATIONS FOR STATIONARY OPERATING REGIME

Let us imagine a cascade, which is intended for separation of m isotope components ($m \geq 3$) or a m-component gas mixture, as a N-stages row consecutively connected by a countercurrent scheme where the flow recirculation can be realized. The flowsheet of the cascade under consideration is shown schematically in Fig. 1. The flux L_S with component concentrations $C_i\ (S)$ is entering into the S-th stage. The "upflowing" flux $L_S^+ = \Theta_S L_S$ with concentrations $C_i^+\ (S)$ from the S-th separation stage (where Θ_S is the cut of the stage) goes to the entrance of $(S + 1)$-th stage. The "downflowing" flux $L_S^- = (1 - \Theta_S)L_S$ having the composition of $C_i^-\ (S)$ is incoming to the entrance of $(S - 1)$-th stage. Let us consider a simple cascade where the feed F with composition of $C_{iF}(i = 1, 2, \ldots, m)$ is introduced into its entrance ($S = f$). The product flux P (product) with composition of $C_{iF}\ (i = 1, 2, \ldots, m)$ and the waste flux W (waste) with the component mole fractions of $C_{iW}(i = 1, 2, \ldots, m)$ are withdrawn from the cascade outlet in the points $S = N$ and $S = 1$, respectively.

It must be emphasized that the "product" and "waste" concepts are relative in the case of multicomponent mixture separation. We will consider that the product is collected from the outlet of cascade where the lightest isotope is concentrated while the waste is withdrawn from the opposite one. For the sake of definition, let us distribute the mixture components in the order of their growing molecular masses $M_i(\mathrm{i} = 1, 2, \ldots, m)$. The equations that allow calculation of cascade can be obtained by combining the mass-balance equation by the total flow with that by the flow of each component through the cross-section between S-th and $(S + 1)$-th stages:

$$\Theta_S L_S - (1 - \Theta_{S+1}) = P;$$

$$\Theta_S L_S C_i^+\ (S) - (1 - \Theta_{S+1})L_{S+1}C_i^-(S + 1) = PC_{iP}$$

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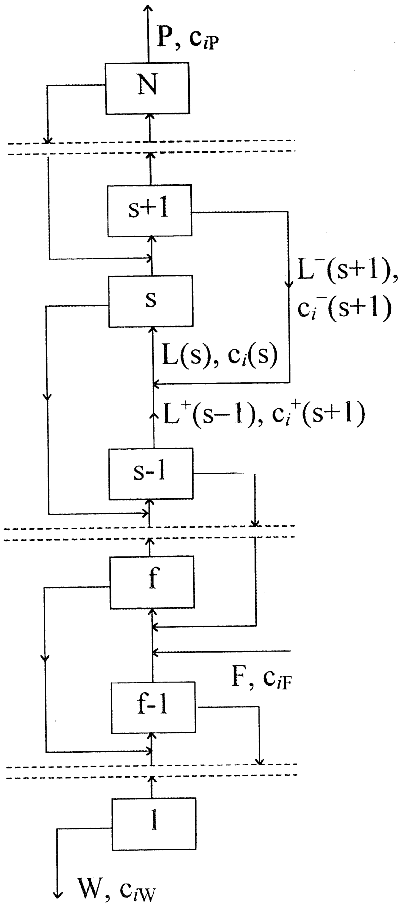

Figure 1. Schematic diagram of countercurrent cascade.

The main enrichment equation for the cascade can be written as follows:

$$C_i(S+1) - C_i(S) = \frac{\Theta_S L_S}{(1-\Theta_{S+1})L_{S+1}}\,\Delta_i^+(S) + \Delta_i^-(S+1) - \frac{P(C_{iP} - C_i(S))}{(1-\Theta_{S+1})L_{S+1}}, \qquad (1)$$

$$i = 1, 2, \ldots, m; \qquad S = f, \ldots, N;$$

where $\Delta_i^+ = C_i^+ - C_i = f_i(C_1, \ldots, C_m, \Theta)$ is the known function of concentrations and cut, which accounts for the changes of content of the i-th component in

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the mixture at a given stage and Δ_i^+ and $\Delta_i^- = C_i - C_i^-$ are connected by the following balance equation: $\Delta_i^- = \Theta/(1-\Theta)\Delta_i^+$.

For the stripping section of the cascade, the flux P and the concentration C_{iP} must be substituted with the flux W taken with an opposite sign and with the concentration C_{iW}, respectively. In this case, Eq. (1) takes the form:

$$\begin{aligned} C_i(S+1) - C_i(S) = {} & \frac{\Theta_S L_S}{(1-\Theta_{S+1})L_{S+1}}\Delta_i^+(S) \\ & + \Delta_i^-(S+1) + \frac{W(C_{iW} - C_i(S)}{(1-\Theta_{S+1})L_{S+1}} \end{aligned} \tag{2}$$

where $i = 1, 2, \ldots, m$, and $S = 1, 2, \ldots, f-1$.

For carrying out the cascade calculations, one needs to add to Eqs. (1) and (2) the following m linear-independent balance equations for the cascade as a whole:

$$FC_{iF} = PC_{iP} + WC_{iW}, \qquad i = 1, 2, \ldots, m-1; \tag{3}$$

$$F = P + W \tag{4}$$

In the case of small enrichment at a single separating stage, the value of Δ_i^+ for any separation method may be written as follows (2):

$$\Delta_i^+ = (1-\Theta)C_i \sum_{j=1}^{m} \varepsilon_{ij} C_j, \tag{5}$$

where ε_{ij} is the relative enrichment coefficient for the system of i and j components possessing the additive and anti-symmetric properties:

$$\begin{aligned} \varepsilon_{ij} &= -\varepsilon_{ji}, \\ \varepsilon_{ij} &= \varepsilon_{ik} + \varepsilon_{kj}. \end{aligned} \tag{6}$$

Small ε_{ij} value suggests that all cascade parameters are changing negligibly from stage to stage, and the flow of the isotope mixture passing through an arbitrary stage is much higher than the product flow. Under these conditions, $L_S \approx L_{S+1}$ and $P/L_S \approx 0$. Assuming this approximation to be valid for separation of any isotopes, it is easy to obtain the following correlation, $\Theta_s = 1/2$, from the balance equations for the whole mixture and for each component. The transfer from Eqs. (1) and (2) finite-difference equations to the differential ones, along with taking into account expression (5), gives after simple rearrangements the following equations:

$$\frac{dC_i}{dS} = C_i \sum_{j=1}^{m} \varepsilon_{ij} C_j - \frac{2P}{L(S)}(C_{iP} - C_i), \tag{7}$$

$$0 \le S \le S_P (S_P + 1 = N - f),$$

$$i = 1, 2, \ldots, m-1,$$

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$$\sum_{i=1}^{m} C_j = 1,$$

$$\frac{dC_i}{dS} = C_i \sum_{j=1}^{m} \varepsilon_{ij} C_j + \frac{2W}{L(S)} (C_{iW} - C_i), \tag{8}$$

$$0 \le S \le S_W (S_W = f),$$

$$i = 1, 2, \ldots, m,$$

$$\sum_{j=1}^{m} C_j = 1.$$

Here S_P and S_W are the numbers of stages in the enriching and the stripping sections of the cascade, respectively; S is the current number of a stage in both parts of the cascade. The stages in the stripping section are numbered from the bottom stage to the feed one, and in the enriching section, the numeration goes from the feed to the top stage.

The analysis of Eqs. (1) and (2) and (7) and (8) enables us to distinguish the following properties of cascade equations.

1. The above equations represent the system of both nonlinear finite difference and differential equations in respect to functions $C_i(S)$. Corresponding C_{iP} and C_{iW} values in these equations must be defined from the solution of these particular equations. The analytical solution of a similar system of equations is possible only in some specific cases. Under conditions of arbitrary distribution of the $L(S)$ value, the numerical solution of equations can be obtained in the following cases.
2. When the product withdrawal is equal to 0 ($P = 0$) the system of Eqs. (7) and (8) is easily integrated giving the following solution:

$$C_i(S) = \frac{C_i(0)}{\sum_{j=1}^{m} C_j(0) \exp(-\varepsilon_{ij} S)}, \tag{9}$$

$$i = 1, 2, \ldots, m,$$

where $C_i(0)$ is the concentration of the i-th component at the inlet of the zero stage (stage number $S = 0$).

As follows from relationship (9), the concentration distribution along the cascade stages does not depend on the flow distribution $L(S)$. The analysis of Eq. (9) shows that in a cascade of sufficient length, the concentrations of all intermediate components achieve the maximum value inside the cascade while the concentration of edge components are monotonously increasing to the end of cascade. The phenomenon of redistribution of components followed by their concentration in different cascade zones can be easily explained from the physical viewpoint. Indeed, the separation in a cascade can be considered to proceed under the

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action of the field of force with a constant stress. Interaction of components in this field results in ejecting of less "active" components by the most active ones out of the cascade ends. The less active components, in turn, affect the redistribution even more actively.

3. From the analysis of Eqs. (1) and (2) and (7) and (8), it directly follows that it is impossible to design a cascade for separation of a multicomponent mixture under no-mixing conditions at the entrances of all stages would be fulfilled for all component concentrations simultaneously. The last conclusion follows from the fact that the mass transfer of all components in the cascade is described by the system of m equations while only one parameter, $L(S)$, is responsible for providing of no-mixing conditions.
4. Since at arbitrary number of the mixture components enrichment of the lightest one, $\Delta_1 = C_1 \sum_{j=1}^{m} \varepsilon_{1j} C_j$, at each stage is always positive and enrichment of the heaviest component, $\Delta_m = C_m \sum_{j=1}^{m} \varepsilon_{mj} C_j$, is always negative, their separation and further recovery are not difficult. On the contrary, fractionation (and recovery) of the intermediate isotope components is complicated as their enrichment depends on the mixture composition. Therefore, at some distance from the cascade feed point, their concentration does not increase any more. This means that only a limited enrichment of the intermediate isotopes in the product flux can be obtained. The problem of separation and recovery of the intermediate isotope components can be solved through selection of the $L(S)$ profile providing the maximum content of the desirable component in the product (or waste) flux. It has been shown by Minenko (3) that in the cascade with the single product and waste fluxes the limited concentration of an arbitrary n-component at S_p, $S_w \gg 1$ is defined by

$$(C_{nP})_{\lim} = \frac{C_{nF}}{\sum_{j=1}^{n} C_{jF}} \tag{10}$$

In practice, it is often required to obtain the concentration of the intermediate component, which exceeds the value predicted by Eq. (10). In this case it is advisable either to introduce some additional fluxes in the zone of localization of desired component or to use more complicated cascade designs such as, double cascade and some others.

The problem of the cascade calculation using Eqs. (1) and (2) and (7) and (8) includes two approaches:

1. calculation of a cascade of specified configuration (check calculation), and
2. calculation of a cascade to be designed (design calculation).

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The first approach involves, as a rule, calculation of 1) simple or squared-off cascades with a specified number of stages; 2) the fluxes in stripping and enriching sections; 3) the feed flux F with component concentrations $C_{iF}(i = 1, 2, \ldots, m)$; and 4) one of the outgoing fluxes P or W. The relative enrichment coefficients ε_{ij} or enrichment functions $\Delta_i^+ \equiv C_i^+ - C_i = f(C_1, \ldots, C_m, \Theta)$ are considered to be known. The final aim of the calculation task is to define the component concentrations in the product flux, $C_{iP}(i = 1, 2, \ldots, m)$, in the waste flux $C_{iW}(i = 1, 2, \ldots, m)$, and the concentration distribution along the cascade, if necessary. If the enrichment at the cascade stages is not small, the cut distribution along the cascade Θ_S should also be specified. The calculations of this type are required to optimize the separation process through the change of the operating conditions and individual cascade parameters, and in the multipurpose use of the cascade, for example, for separation of a wide range of isotopes without significant modification of the cascade design. The main difficulty in carrying out check calculations is due to unknown values of concentrations C_{iP} and C_{iW} in Eqs. (1)–(2) and (7)–(8). Impossibility to solve analytically these equation systems calls for development of numerical simulation methods, which would be non-sensitive to the initial approximation of the component concentrations.

In the design calculation, one usually defines the parameters of the square or squared-off cascade with the specified concentration of one of the components, for example, the desirable component in the product or waste fluxes, and the value of the product (waste) flux. It is also implied that the cascade parameters under examination have to satisfy at best the conditions of its optimization. In the case of separation of binary mixtures, the design calculation of the squared-off cascades is based on the ideal cascade model. By the analogy, the design and optimization of cascades for separation of multicomponent mixtures is expediently carried out on the basis of the mathematical model, which is adequate for the separation process but allows simplification calculations. In this case, by means of an artificial choice of the flow profile, one can reach the most effective enrichment of the desirable component in the cascade.

NUMERICAL METHODS FOR CALCULATION OF CASCADES WITH SPECIFIED CONFIGURATION

Calculation of a cascade of specified configuration (with a specified number of stages and flows in the enriching and stripping sections) is based on two approaches. The first one is advisable to use for calculation of so-called “long” cascades (for example, squared-off cascade of distillation columns). It is based on either analytical transitions enabling reduction of the problem to the algebraical transcendental equations (2,4–8) or on the direct integration of Eqs. (7)–(8) with subsequent iteration on the boundary conditions and balance Equations (9,25).

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This approach also involves the orthogonal-collocation technique consisting of the approximation of solution of differential equations by an interpolational Lagrangian polynomial (10).

Check calculations of the cascade with relatively high (non-small) enrichment of components at each stage are based either on the stage-by-stage calculation of the component concentrations from one end of the cascade to another by using Eqs. (1) and (2) with iterative refinement of the concentration values (11), or on solution of the matrix equations for the total and component balance over the cascade (12). Taking into account the features of computation process, the orthogonal-collocation technique (11) and the matrix technique (12) seem to be the most efficient for solution of the above problems. The description of these methods is given below.

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Cascade Calculation by Using Orthogonal Collocation Technique

The calculation methods reported previously (2,4–9,24) although possessing some advantages have significant deficiencies. First, they require considerable computer time. This is due to the need for cumbersome calculations at each iterative step in refining the concentration values. Second, these methods necessarily involve initial approximations of the component concentrations at the ends of the cascade that make them very sensible to the ratio of initial approximation to exact solution. Third, all these methods are characterized by low accuracy, (particularly for cascades with a high number of stages) due to accumulation of errors in calculating the component concentrations.

Let us now consider the method for calculation of cascades that is essentially free from the above deficiencies. For the sake of simplicity, let us consider an ordinary squared cascade with the feed, F, product, P, and waste, W, fluxes, respectively. The corresponding component concentrations in fluxes are C_{iF}, C_{iP} and C_{iW} ($i = 1, 2, \ldots, m$). The transfer of components along the cascade is described by the system of Eqs. (7)–(8) along with respective equations of the total and component balance (3)–(4) under assumption of the constant value of the flow L. Equations (7)–(8) and (3)–(4) should be complimented by the following boundary

$$C_i^{\mathrm{I}}(0) = C_{iW},\ C_i^{\mathrm{II}}(S_P) = C_{iP},\ i = 1, 2, \ldots, m \tag{11}$$

and continuity condition for component concentrations in the feed input cross-section

$$C_i^{\mathrm{I}}\,(S_W) = C_i^{\mathrm{II}}\,(\mathrm{O}) = C_{if},\ i = 1, 2, \ldots, m, \tag{12}$$

where C_{if} is the concentration of the i-th component in this cross-section, and superscripts I and II characterize the stripping and the enrichment sections of the cascade, respectively.

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As follows from the orthogonal-collocation method, solution of the differential equation is sought in the form of a polynomial or series. The interpolational Lagrangian polynomial used by Tarng and Anthony (35) as the approximating operator, can be written for the enrichment section as follows:

$$C_i(S) = \sum_{k=1}^{n} C^k_{Pi}\,(S)\,L_{Pk},\ i = 1, 2 \ldots, m, \tag{13}$$

where $L_{Pk} = T_n/[(S - S_k)T^1_n\,(S)]$ is the Lagrangian polynomial of the order $(n - 1)$; $T_n(S) = (S - S_1)\cdots(S - S_n)$ is the point polynomial of order n; S_k, $0 \le S \le S_p$, $k = 1, 2, \ldots, n$ is the collocation point, and $C^k_{Pi} = C_i(S_k)$, $i = 1, 2, \ldots, m$ is the corresponding ordinate of the collocation point.

Without loss of generality, it is assumed that the number of collocation points n chosen for the enriching and stripping sections is the same. Then, for the stripping section the Lagrangian polynomial determining $C_i(S)$ takes the form $C_i(S) = \sum_{k=1}^{n} C^k_{Wi}L_{Wk}\,(S)$, $i = 1,2, \ldots, m$. The real zeros of orthogonal polynomials are usually chosen as the collocation points to attain the highest calculation accuracy. In Fillipov et al. (10), n collocation points required are defined as follows: if $k = 1$, then $S_k = 0$; if $k = 2, \ldots, n = 1$, then $S_k = S^\nu\,(X_k + 1)/2$; if $k = n$, then $S_k = S$; here S^ν is the number of stages in the section; $X_k = \cos\{[2(n - k) - 1]\pi/2(n - 2)\}$ are the zeros of an orthogonal Chebyshev polynomial of order $(n = 2)$.

The boundary conditions (11) and the continuity conditions for the component concentrations in the cross-section of the feed flow are now written for the orthogonal collocation method as follows:

$$\begin{aligned} &S_1 = 0,\ C^1_{Pi}(S_1) = C_{if}; \quad S_n = S^P,\ C^n_{Pi}(S_n) = C_{iP};\\ &S_1 = 0,\ C^1_{Wi}(S_1) = C_{iW}; \quad S_n = S^W,\ C^n_{Wi}\,(S_n) = C_{if},\\ &i = 1,2, \ldots, m \end{aligned} \tag{14}$$

Taking into account that $L_k(S_j) = 0$ when $k \neq j$, $L_k(S_j) = 1$ when $k = j$, and after substitution of formula (13) into Eq. (7) for an arbitrary collocation point S_r, one obtains

$$\begin{aligned} &\sum_{k=1}^{n} C^k_{Pi}L^{(1)}_{Pk}\,(S_r) = C^r_{Pi}\left[\frac{2P}{L} + \sum_{j=1}^{m} \varepsilon_{ij}C^r_{Pj}\right] - \frac{2P}{L}\,C^n_{Pi},\\ &i = 1,2, \ldots, m; \quad r = 1,2, \ldots, n \end{aligned} \tag{15}$$

where $L^{(1)}_{Pk} = T^{(2)}_n(S_r)/[2T^{(1)}_n\,(S_r)]$ is the derivative of the Lagrangian polynomial at point S_r. Analogously, for the stripping section one can write

$$\begin{aligned} &\sum_{k=1}^{n} C^k_{Wi}L^{(1)}_{Wk}\,(S_r) = C^r_{Wi}\left[-\frac{2W}{L} + \sum_{j=1}^{m} \varepsilon_{ij}C^r_{Wj}\right] + \frac{2W}{L}\,C^{(1)}_{Wi},\\ &i = 1, 2, \ldots m;\ r = 1, 2, \ldots n \end{aligned} \tag{16}$$

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The material balance Eq. (3) and the conditions for selection of the collocation point (14) yield the following expression combining Eqs. (15) and (16):

$$FC_{iF} = PC^{n}_{Pi} + WC^{1}_{Wi},\ C^{n}_{Wi} = C^{1}_{Pi},\ i = 1, 2, \ldots, m \tag{17}$$

Thus, the set of differential Eqs. (7)–(8) reduces to a system of nonlinear algebraic equations with $(2n - 1)m$ unknowns for determining the component concentrations in the mixture at the collocation points and hence, in the cascade output fluxes. This system is solved by using the following method (36): instead of one nonlinear system with $(2n - 1)m$, m linear systems with $(2n - 1)$ unknowns are solved, constantly refining the concentration values in an iterative procedure. By calculating a squared-off cascade, one or more such nonlinear systems (according to the number of sections) and fitting conditions at the section boundaries analogous to the conditions at the feed flux cross-section must be added to Eqs. (15) and (16).

The method under consideration is realized as a software in FORTRAN for IBM PC and compatible computers. Let us illustrate the possibilities of the method in calculation of the squared-off cascade for separation of oxygen and nitrogen isotopes by cryogenic distillation of nitrogen oxide under assumption of absence of the isotope exchange (see Fig. 2). The cascade has two stripping and

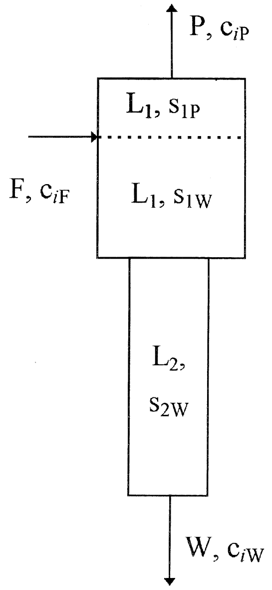

Figure 2. Schematic diagram of squared-off cascade for separation of nitrogen and oxygen isotopes by cryogenic distillation of NO.

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Table 1. Natural Abundance and Relative Enrichment Coefficients for Isotopic Mixture of Nitrogen Oxide (37)

Component, *i*	Molecule	C_{iF}, Mole Fraction	ε_{1i}
1	$^{14}N^{16}O$	$1-\sum_{j=2}^{6} C_{jF}$	0.000
2	$^{14}N^{17}O$	$0.36865\cdot10^{-3}$	0.017
3	$^{15}N^{16}O$	$0.364120\cdot10^{-2}$	0.025
4	$^{14}N^{18}O$	$0.203255\cdot10^{-2}$	0.034
5	$^{15}N^{17}O$	$0.135000\cdot10^{-5}$	0.042
6	$^{15}N^{18}O$	$0.745000\cdot10^{-5}$	0.059

one enrichment sections. In the stripping sections (numbered from the feed point), the section lengths and the circulation flows are as follows: $S_{1W} = 200$, $L_1/2W = 2\cdot10^3$ and $S_{2W} = 100$, $L_2/2W = 1\cdot10^3$. The length of the enrichment section is $S_p = 100$. The composition of the cascade feed and the values of relative enrichment coefficient for the components are presented in Table 1.

The calculated mole fractions for the mixture components at the ends of the cascade in the feed and product fluxes $F/W = 20$, $P/W = 19$, respectively, are presented in Table 2.

In the example considered, the component $i = 1$ concentrates at the "light" end of the cascade (in the product flux), and the rest components are extracted at the "heavy" end (in the waste flux). The main characteristics, which is reflecting the computational properties of the methods, are the number of manipulations of the function being calculated (the computational work) required to achieve the necessary accuracy in determining the desired unknowns (38). By calculating the cascades, the function being calculated is the relative or absolute residual, the sum of squares of the residuals, etc. For example, in Filippov et al. (10) this function is the sum of the relative residuals in the waste flux at the iterative steps μ and $\mu - 1$:

$$\Phi_1(\mu) = \sum_{i=1}^{m} [C_{iW}(\mu) - C_{iW}(\mu - 1)]/C_{iW}(\mu), \tag{18}$$

Table 2. Calculated Mole Fractions for Mixture Component at Cascade Ends

Component, *i*	Molecule	C_{iP}, Mole Fraction	C_{iW}, Mole Fraction
1	$^{14}N^{16}O$	$0.99920\cdot10^{0}$	$0.89400\cdot10^{0}$
2	$^{14}N^{17}O$	$0.11687\cdot10^{-8}$	$0.51523\cdot10^{-2}$
3	$^{15}N^{16}O$	$0.54968\cdot10^{-3}$	$0.62380\cdot10^{-1}$
4	$^{14}N^{18}O$	$0.12411\cdot10^{-3}$	$0.38293\cdot10^{-1}$
5	$^{15}N^{17}O$	$0.35480\cdot10^{-7}$	$0.26326\cdot10^{-4}$
6	$^{15}N^{18}O$	$0.31140\cdot10^{-7}$	$0.14841\cdot10^{-3}$

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where $C_{iW}(\mu)$ is the concentration of the i-th component in the waste flux at the μ-th iterative step. The function $\Phi_1(\mu)$ characterizes the convergence of the iterative procedure and serves as a criterion for ending this process. However, concluding the calculation on the basis of $\Phi_1(\mu)$ does not guarantee convergence of the problem to the exact solution. The exact solution is understood here to be the solution to which the iterative process tends with an increase of the number of collocation points in the cascade section. By denoting with C_{iW*} the concentration corresponding to exact solution, the degree of approximation to the exact solution can be estimated from the following formula:

$$\Phi_2(\nu) = \sum_{i=1}^{m} |[C_{iW*} - C_{iW}(\nu)]/C_{iW*}|, \tag{19}$$

where $C_{iW}(\nu)$ is the concentration of the i-th component in the waste flux determined with accuracy $\Phi_1(\mu)$ for ν collocation points in the section.

The results of numerical experiments show that only certain parameters such as, configuration of the cascade (number of sections, their lengths and flow values), the number of mixture components, mole fractions of components in the feed flux, and the relative enrichment coefficients, affect the convergence of the orthogonal-collocation method. The dependence of number of iterations on the relative accuracy of calculation of component concentrations in the waste flux obtained through the use of Eq. (18) with parameter values given above is shown in Fig. 3. The increase of number of iterations at higher accuracy on C_{iW} determina-

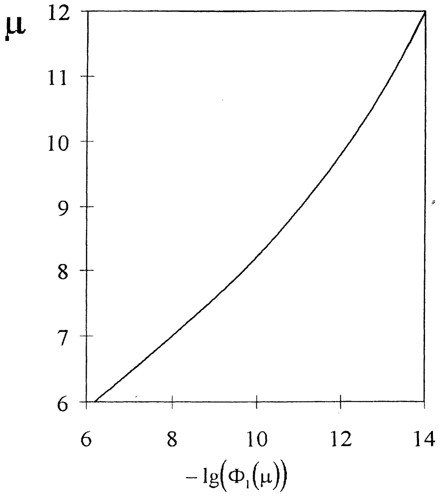

Figure 3. Dependence of number of iterations (μ) on specified accuracy of calculations of component concentrations in waste flow.

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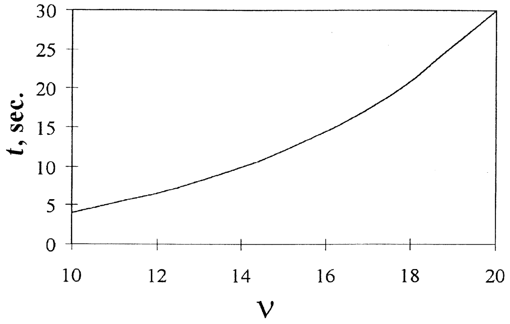

Figure 4. Dependence of calculation time (t) for single iterative step on number of collocation points per section of three-sectional cascade (ν).

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tion is associated with the need to refine the concentration value at intermediate collocation points. This refinement is required to provide the continuity of concentration at the junction of enrichment and stripping sections and to satisfy the external component-by-component balance equation. Note that the number of iterations required to achieve the specified accuracy $\Phi_1(\mu)$ does not depend on the number of collocation points in the sections of the squared-off cascade.

The number of collocation points in a cascade section, the number of cascade sections, and the number of components in the mixture influence the dimensionality of equation system (15)–(17) and hence, the computation time (see Fig. 4). An increase of computation time by one iterative step is due to the increase of calculations volume of intermediate concentration values with number of collocation points in the section.

Figure 5 shows $\Phi_2(\nu)$ versus ν dependence calculated by using Eq. (19). The calculations were made for the accuracy of $\Phi_1(\mu) = 10^{-10}$, and the values ob-

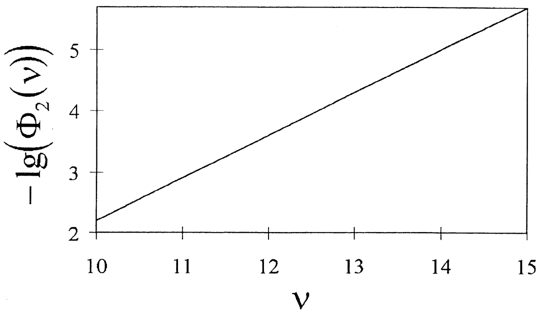

Figure 5. Dependence of degree of closeness to accurate solution on number of collocation points per cascade section (ν).

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tained with 20 collocation points per section are taken as C_{iW*}. For 16–20 collocation points per section, $\Phi(\mu) < 10^{-10}$ and its relative change appears to be ~1%, which indicates that approximation to the solution with $\nu = 20$ is sufficiently good.

From the known value of required accuracy, one can determine the optimal number of collocation points and iterations and the total time for computer calculation of the problem through analysis of dependencies obtained (see Fig. 5).

The use of cascade calculations, which require the specification of initial approximations for the desired quantities, the number of iterations will also depend on the deviation of the initial approximations from the values corresponding to exact solution. This property of the computational process can be called the "sensitivity" of computation technique to the choice of the initial approximations. The orthogonal-collocation method developed for cascade calculation is not sensitive to the choice of the initial approximations (see Fig. 6). This is confirmed by the result of calculations carried out by using initial concentration of all components, which is equal to 0, 1, and to the concentration in the natural isotope mixture. The number of iterations appears to be essentially the same in each case. In the method described above, the concentrations of components in the feed flux are the most convenient to choose as an initial approximation. For comparison, the analogous dependence for the calculation method based on integration of Eqs. (7)–(8) with subsequent iterative refinement of the concentrations C_{iP} and C_{iW} is shown in Fig. 6 (25). As seen, when the relative deviation of the initial approximations from the solution approaches one, the number of iterations tends to infinity, and it is impossible to obtain the solution. In such methods, the choice of initial approxima-

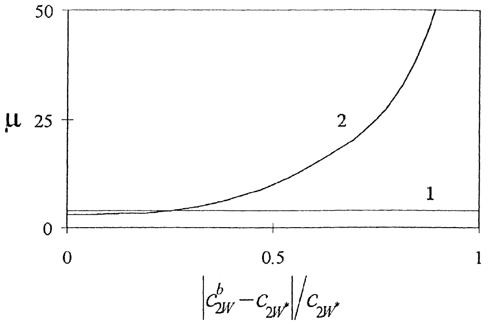

Figure 6. Influence of deviation of initial approximation of target-component ($^{14}N^{17}O$) concentration C^b_{2W} from accurate solution C_{2W*} on number of iterations for various methods: 1) orthogonal collocation, $\Phi_1(\mu)$, $\Phi_2(\nu) \leq 10^{-8}$; 2) numerical integration of set of equations reported (25).

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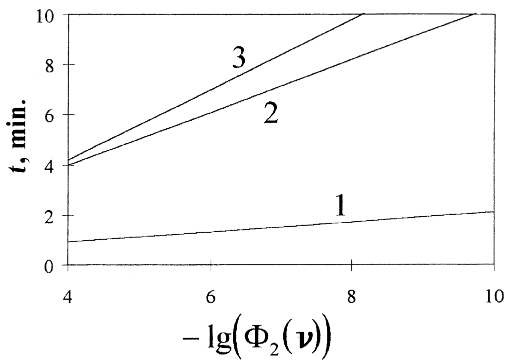

Figure 7. Dependence of calculation time for squared-off cascade (t) on accuracy: 1) orthogonal collocation method; 2) solution of overall and component-by-component balance at each stage; 3) numerical integration of set of transfer equations.

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tions is especially important for components with a small concentration in the initial mixture, the deviation for which may be considerable, and hence, the convergence of the iterative process may be difficult.

By solving physical problems such as cascade calculation, the same result may be obtained using different calculation methods based on the use of a function different from that described by Eq. (18). The number of iterations required for cascade calculation with a specified accuracy characterizes the efficiency of the method (in particular, the convergence of the iterative process). Nevertheless, this parameter is unsuitable for the comparison of methods differing by the function under calculation, even with the same structure of equations describing the component transfer. Therefore, it seems more reasonable to compare different calculation methods in terms of the time required for cascade calculations with a specified accuracy (see Fig. 7). The orthogonal-collocation method permits approximately 5-fold reduction in the calculation time of the squared-off cascade compared with other existing methods.

Calculation of Cascade of Specified Configuration by Matrix Method

It is advisable to use the system of Eqs. (1)–(4) for the stage-by-stage calculation of a cascade. Application of the matrix method suggests to be more convenient to use the following balance equations for the mixture component fluxes,

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LC_i, at the entrance of each stage (see Fig. 1) instead of Eqs. (1)–(4):

$$\begin{aligned}
&L_1C_i(1) - (1 - \Theta_2)L_2C_i^-(2) - [W - (1 - \Theta_1)L_1]C_{iW} = 0,\\
&i = 1, 2, \dots, m\\
&L_SC_i(S) - \Theta_{S-1}L_{S-1}C_i^+(S-1)\\
&\qquad\qquad - (1 - \Theta_{S+1})L_{S+1}C_i(S+1) - \delta_{Sf}FC_{iF} = O, \qquad (20)\\
&S = 2, 3, \dots, N-1;\ i = 1, 2, \dots, m\\
&L_NC_i(N) - \Theta_{N-1}C_i^+(N-1) - [P - \Theta_NL_N]C_{iP} = 0,\\
&i = 1, 2, \dots, m
\end{aligned}$$

where

$$\delta_{Sf} = \begin{cases} 0, S \neq f,\\ 1, S = f, i = 1, 2, \dots, m; S = 1, 2, \dots, N \end{cases}$$

To make the solution of the above system more stable, it seems reasonable to substitute the equation for the k-th component, having the maximum concentration in the initial mixture, by the following connecting equation:

$$1 - \sum_{i=1}^{m} C_i(S) = 0, S = 1, 2, \dots, N. \qquad (21)$$

Concentrations of the i-th components $C_i^+(S)$, $C_i^-(S)$, and $C_i(S)$ in the fluxes Θ_SL_S, $(1-\Theta_S)L_S$, L_S, respectively, are bound by the following relationships:

$$\begin{aligned}
&\Delta_{i,S}^+ = C_i^+(S) - C_i(S) = f_i(C_1(S), \dots, C_m(S), \Theta_S),\\
&\Delta_{i,S}^- = C_i(S) - C_i^-(S) = \frac{\Theta_S}{1-\Theta_S}\Delta_{i,S}^+, \qquad (22)\\
&i = 1, 2, \dots, m; S = 1, 2, \dots, N
\end{aligned}$$

where $f_i(C_1, \dots, C_m, \Theta)$ is the enrichment function of the i-th component. A more detailed form of this function is determined by the separation method and the mode of cascade operation.

To close system (20) comprising $m \times N$ equations, it is necessary to eliminate coefficients $\Theta_S (S = 1, 2, \dots, N)$ from unknowns. For this one needs to specify the value of one of these coefficients (for example, Θ_1) and determine the other Θ_S (under conditions of known distribution of L_S) by using the following standard relations:

$$\Theta_{S+1} = \begin{cases} 1 - \dfrac{W}{L_{S+1}} - \dfrac{\Theta_SL_S}{L_{S+1}}, S = 1, 2, \dots, f\\[2ex] 1 + \dfrac{P}{L_{S+1}} - \dfrac{\Theta_SL_S}{L_{S+1}}, S = f, f+1, \dots, N \end{cases} \qquad (23)$$

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Note that system (20) may be generalized to the case of arbitrary number of feed, product, and waste fluxes. Solution of nonlinear algebraic equations with accounting for the concrete form of the enrichment function (21) can be realized by all numerical methods applicable for solution of systems of nonlinear equations. Taking into account the convergence rate of the iterative process and the solution stability, a satisfactory result can be provided by a damped form of the Newton's method (47).

After signifying the unknowns $C_i(S)$ in the order of increasing subscript numbers i and the stage number S as a vector $\vec{x}$ with dimension of $m \times N$, the iterative process for this method will be written as follows:

$$\begin{aligned} \vec{x}^{\nu+1} &= \vec{x}^{\nu} + \Delta\vec{x}^{\nu}, \\ F'(\vec{x}^{\nu})\Delta\vec{x}^{\nu} &= -\omega F(\vec{x}^{\nu}), \end{aligned} \tag{24}$$

where $F(\vec{x}^{\nu})$ are the values of the left-hand sides in (20)–(21) at a point with known coordinates $\vec{x}^{\nu}$; $F'(\vec{x}^{\nu})$ is the Jacoby matrix for the system at that point; $\Delta\vec{x}^{k} = \vec{x}^{k+1} - \vec{x}^{k}$ and ω is the damping coefficient. Because each S-th equation of system (20)–(21) contains unknown values of concentrations in the fluxes entering only $(S - 1)$-th, S-th and $(S + 1)$-th stages, the Jacoby matrix belongs to the type of strip matrices with strip width of $(3 \times m)$. Hence, during iterations (24) the strip structure of the matrix persists (48). As it follows from the practical calculations, this matrix is suitable to represent in the form of the square submatrices of the dimension $m \times m$. All elements of the Jacoby matrix, which do not belong to these submatrices, equal zero. The schematic of this matrix is shown in Fig. 8.

Determination of the $(\nu + 1)$-th approximation to $\vec{x}^{\nu+1}$ in the iterative process (24) is reduced to the solution of nonlinear algebraic equations of $A\Delta\vec{x}^{\nu} = \vec{b}$ type with unknowns $\Delta\vec{x}^{\nu}$. Determination of $\Delta\vec{x}^{\nu}$ can be preferably carried out through the use of the Gauss elimination algorithm formulated in such a way that the elements of the Jacobian, which do not appear in the strip of this width, are disregarded in the calculations. The last circumstance reduces the run time substan-

s = 1	A_{11}	A_{12}	0	0	0
s = 2	A_{21}	A_{22}	A_{23}	0	0
s = 3	0	A_{32}	A_{33}	A_{34}	0
…	…	…	…	…	…
…	…	…	…	…	…
…	…	…	…	…	…
s = N	0	0	0	0	$A_{N\,N-1}$

Figure 8. Jacoby matrix for Eqs. (20) and (21) (A_{ij} are submatrices).

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tially. The calculation scheme for the squared-off cascade by the method proposed includes:

1. specification of the initial data: m, N, f, F, P, C_i^F, $\Delta_{i,S}^+$, L_S, Θ_1;
2. definition of the cut at stages: $\Theta_S(S = 2, \ldots, N)$;
3. solution of the system Eqs. (20)–(21).

This method provides a better convergence than the stepwise method. It does not require the initial data close to unknowns, and often reduces the run time. At the same time, it requires a certain increase in the computer memory. For example, calculation of the cascade of 50 stages with two feed fluxes of different composition and three waste fluxes requires solution of the system (20)–(21) with 250 equations that needs several iterations. The concentrations in one of the feed fluxes have been chosen as an initial approximation for the unknown concentrations at each stage. The calculations of the squared-off cascades for separation of a wide-range of multicomponent mixtures have shown that the use of the above initial approximations permits determination of unknown concentrations with accuracy of $<0.1\%$ in 10 or less iterations. The method considered is particularly effective for the calculation of cascades with a relatively high enrichment at stages.

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DESIGN CALCULATIONS OF CASCADES FOR SEPARATION OF MULTICOMPONENT MIXTURES

Q-Cascade and Its Properties

Calculation of a cascade with defined product flux and given limitations on concentrations in the product and waste fluxes for the target isotope implies determination of the following parameters: 1) number and length of stages; 2) flows entering each stage of section; 3) waste flux W; and 4) feed flux F. The values of these parameters have to be, as a rule, optimized by certain criteria. At the same time, the effective numerical methods permitting calculation of the flux profile for a given cascade and concentrations of the multicomponent isotope mixture components at cascade ends do not exist. For this reason, it seems reasonable to use the uninterrupted-profile cascade as the preliminary model. The theory allows for determination of lengths of the stripping and enrichment sections of this cascade, the flow distribution, $L(S)$, and component concentrations along the cascade length, if the product flux and concentration of the desirable isotope in the product and waste fluxes are known. The following procedure reduces the cascade calculation to approximation of the model cascade by the squared-off cascade followed by its optimization.

The most common method of calculation of model cascades is based on the application of relationships proposed by Kucherov and Minenko (2) by means of

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substitution of variables in Eqs. (7) and (8). The essence of this conversion becomes evident from the following example, which describes the enrichment section of the simple cascade. Concentrations $C_i(S)$ are substituted by functions $\varphi_i(S)$ and system of Eq. (7) is reduced to the single Volterra equation, which can be written for one of these functions as follows:

$$\varphi_{\mathrm{i}}(S) + \sum_{j=1}^{m} \frac{2PC_{jP}}{L(S)} \int_0^S \varphi_i(t) \exp[\varepsilon_{ij}(S - t)]dt = \sum_{j=1}^{m} \frac{C_{iF}L_F}{L(S)} \exp(\varepsilon_{ij}S), \tag{25}$$

and must be complemented with $(m - 1)$ relations having the following simple form:

$$\varphi_j(S) = \varphi_i(S)\exp(\varepsilon_{ij}S), \tag{26}$$

where L_F and C_{iF} are the flux and the concentrations in the cascade feedflow at $S = 0$. After substitution $S = S_P - 1$, Eqs. (25) and (26) will be written as follows:

$$\varphi_i(l) - \sum_{j=1}^{m} \frac{PC_{iP}}{L(l)} \int_0^l \varphi_i(t)\exp[\varepsilon_{ij}(t - l)]dt = \sum_{j=1}^{m} \frac{C_{jP}L(0)}{L(l)} \exp(-\varepsilon_{ji}l), \tag{27}$$

$$\varphi_j(l) = \varphi_i(l)\exp(\varepsilon_{ji}l), \tag{28}$$

where $L(0)$ is the flow corresponding to the conditions $P = 0$, i.e., at the product outlet of the cascade.

Functions $\varphi_i(l)$ are connected with concentrations and the flow within the cascade by the following relation:

$$C_i(l)L(l) = \frac{C_{iP}L(0) + 2PC_{iP} \int_0^l \varphi_i(t)dt}{}. \tag{29}$$

After substitutions

$$C_i(l)L(l) = G_i(l), \tag{30}$$

$$\sum_{j=1}^{m} G_j(l) = L(l), \tag{31}$$

one obtains

$$C_i(l) = \frac{G_i(l)}{\sum_{j=1}^{m} G_j(l)}. \tag{32}$$

Note that relation (27) can be easily generalized for the squared-off cascade as well as for the case of several feed and waste fluxes.

For a given $L(S)$, Eq. (27) has a single solution that can be obtained, for example, by using consecutive approximations permitting concentrations to be found of the mixture components by formulae (29) and (32).

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In practice it seems more attractive to use Eqs. (29)–(31) for determination of distribution $L(l)$ in the cascade that allows one to solve the problem of searching the desired enrichment for the target component. One of the characteristic functions $\varphi_n(l)$ defines unconditionally distributions $L(l)$ and $C(l)$ along the length of cascade and the rest $(m-1)$ of the functions are defined by relation (28). If the characteristic function enables us to calculate the integral in formula (29), one can obtain the analytical ratio for the flow distribution $L(l)$ and concentrations $C_i(l)$.

Presentation of the characteristic function in the exponential form (13) permits a simple and suitable mathematical model to be obtained, which can be used to solve many separation problems:

$$\varphi_i(l) = \exp(Q_i l), \tag{33}$$

where Q_i are the constants, which according to Eq. (28) have to be connected by the following relation:

$$Q_i - Q_j = \varepsilon_{ij}. \tag{34}$$

Therefore, only a single Q_i value can be selected as an arbitrary one. The model cascades satisfying relation (33) are called Q cascades.

To minimize the total flow in the cascade, it is natural to introduce the following condition:

$$L(0) = 0. \tag{35}$$

Substituting Eq. (33) for Eqs. (29), (30), and (31) and accounting for Eq. (35), one obtains:

$$G_i(l) = \frac{2PC_{iP}}{Q_i}[1 - \exp(-Q_j l)]. \tag{36}$$

$$L(l) = 2P\sum_{j=1}^{m}\frac{C_{jP}}{Q_j}[1 - \exp(-Q_j l)]. \tag{37}$$

Equations (36) and (37) represent the distributions of component flow G_i and the mixture flow L in the enrichment section of the Q cascade.

The analogous equations can be written for the stripping section of the Q cascade:

$$G_i(l) = \frac{2WC_{iW}}{Q_i}[\exp(Q_j l) - 1], \tag{38}$$

$$L(l) = 2W\sum_{j=1}^{m}\frac{C_{jW}}{Q_j}[\exp(Q_j l) - 1], \tag{39}$$

where $l = S_w - S$.

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Through the use of relations (36)–(39) and (32) one can obtain concentrations in the product C_{iP} and in the waste C_{iW} fluxes from concentrations in the feed point C_{if}:

$$C_{iP} = \frac{Q_i C_{if}}{1 - \exp(-Q_i S_P)} \Big/ \sum_{j=1}^{m} \frac{Q_j C_{if}}{1 - \exp(-Q_j S_P)}, \tag{40}$$

$$C_{iW} = \frac{Q_i C_{if}}{\exp(Q_i S_W) - 1} \Big/ \sum_{j=1}^{m} \frac{Q_j C_{if}}{\exp(Q_j S_W) - 1}. \tag{41}$$

It is necessary to emphasize that in general case concentrations C_{if} in the feed point of the cascade does not equal the concentrations in the feed stream C_{iF}. Using the material balance Eqs. (3) and (4), one can expel C_{if} from Eqs. (40) and (41) and express C_{iP} and C_{iW} through C_{iF} values as follows:

$$C_{iP} = \frac{1 - \exp(Q_i S_W)}{\exp(-Q_i S_P) - \exp(Q_i S_W)} C_{iF} \Big/ \sum_{j=1}^{m} \frac{1 - \exp(-Q_j S_W)}{\exp(-Q_j S_P) - \exp(Q_j S_W)} C_{jF} \tag{42}$$

$$C_{iW} = \frac{\exp(-Q_i S_P) - 1}{\exp(-Q_i S_P) - \exp(Q_i S_W)} C_{iF} \Big/ \sum_{j=1}^{m} \frac{\exp(-Q_j S_P) - 1}{\exp(-Q_j S_P) - \exp(Q_j S_W)} C_{jF} \tag{43}$$

By using formulae (37) and (39) and the evident relation $\sum_{j=1}^{m} C_j = 1$, it is easy to obtain the following ratios:

$$\frac{W}{P} = \sum_{j=1}^{m} \frac{1 - \exp(-Q_j S_P)}{\exp(Q_j S_W) - \exp(-Q_j S_P)} C_{jF}, \tag{44}$$

$$\frac{F}{P} = \sum_{j=1}^{m} \frac{\exp(Q_j S_W) - 1}{\exp(Q_j S_W) - \exp(-Q_j S_P)} C_{jF}. \tag{45}$$

The above formulae demonstrate that the Q cascades allow concentrating isotopes of definite masses under conditions of an appropriate choice of Q_i value. Indeed, we assume, for the sake of definition that for the i-th isotope parameter $Q_i > 0$, and the length of cascade is sufficiently high, i.e., both S_P and S_W are $\gg 1$. Estimation of the separation effect shows that the behavior of isotopes in the cascade is different depending on the Q_n sign. For example, if $Q_i > 0$ one can easily obtain from Eq. (42) the following estimation relations taking into account that the denominators standing in expressions for concentrations C_{iF} and C_{jF} are identical:

$$\frac{C_{iP}}{C_{jP}} \approx \frac{C_{iE}}{C_{jF}}. \tag{46}$$

If $Q_i < 0$, from expression (31) it follows that:

$$\frac{C_{iP}}{C_{jP}} \gg \frac{C_{iF}}{C_{jF}}. \tag{47}$$

For the stripping section of cascade the same estimations are made when $Q_i < 0$.

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For the majority of separation methods, the coefficients of reciprocal enrichment are proportional to the mass difference of isotopes to be separated. Hence, if one needs some intermediate isotope with number "n" to be obtained in the product flow, Q_n value should be expressed as follows:

$$Q_n = k\varepsilon_{n,n+1} = k\varepsilon_0\,(M_{n+1} - M_n), \tag{48}$$

where k is the arbitrary constant lying in the range $0 < k < 1$; ε_0 is the enrichment coefficient for the pair of isotopes with mass difference of one; M_n is the molecular mass of the n-th component of the mixture.

According to expressions (46) and (47) concentrations of isotopes with numbers $j < n$ will be increasing in the course of separation in the same manner as concentration of the n-th isotope whereas concentrations of all isotopes with numbers $j > n$ will be sharply decreasing. Hence, Q cascades permit separation of all isotopes of a given element into two groups.

As directly follows from the results obtained, concentration of an intermediate isotope in the product flow cannot be unlimitedly increased in a single Q cascade. This conclusion evidently follows from the condition that the sum of all component concentrations must not exceed one.

The length of the stripping section of Q cascade depends upon the maximum concentration of an intermediate isotope in the product flow (16). The formula describing the concentration ratio of arbitrary i-th and n-th components in the product flow of Q cascade can be written as follows:

$$\frac{C_{iP}}{C_{nW}} = \frac{\exp(Q_i S_W) - 1}{\exp(Q_n S_W) - 1} \cdot \frac{\exp(Q_n S_W) - \exp(-Q_n S_P)}{\exp(Q_i S_W) - \exp(-Q_i S_P)} \cdot \frac{C_{iF}}{C_{nF}} \tag{49}$$

If n is the number of the target component and the mixture components are enumerated in the order of growth of their masses, then the Q_i values are chosen so that $Q_i > 0$ for $n \geq i$ and $Q_i < 0$ for all $i > n$. In this case for cascades with an enrichment section of sufficient length (i.e., for any i in compliance with formula (49) $\exp(|\,Q_i\,|\,S_P) \gg 1$), one can approach a limited value of the target isotope concentration Cmax\\$nP$ for a given number of stages in the stripping section, S_W:

$$C_{nP}^{\max} \approx \frac{C_{nF}}{\sum\limits_{j=1}^{m} \gamma_i C_{iF}}, \tag{50}$$

where

$$\gamma_i = \frac{1 - \exp(-Q_i S_W)}{1 - \exp(-Q_n S_P)}. \tag{51}$$

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In this case, the number of stages S_W and the concentration of desirable isotope in the waste flow C_{nW} are connected by the following relation:

$$C_{nW} \approx \frac{C_{nF}\exp(-Q_n S_W)}{\sum\limits_{i=1}^{n} C_{iF}\exp(-Q_n S_W) + \sum\limits_{i=n+1}^{m} C_{iF}}. \tag{52}$$

In the limited case when $S_W \rightarrow \infty$ formula (50) transforms to (10) and at $S_W \rightarrow 0$ we obtained a simpler expression for $C_{nP}^{\max}$:

$$C_{nP}^{\max} \approx \frac{C_{nF}}{\sum\limits_{i=1}^{n} \dfrac{Q_i}{Q_n} C_{iF}} \tag{53}$$

under conditions that $Q_i/Q_n \geq 1$ ($i = 1, 2, \ldots, n$). Hence, the maximum possible $C_{nP}^{\max}$ value in a cascade with stripping section is defined by the following relation:

$$\frac{C_{nF}}{\sum\limits_{i=1}^{n} \dfrac{Q_i}{Q_n} C_{iF}} < C_{nP}^{\max} < \frac{C_{nF}}{\sum\limits_{i=1}^{n} C_{iF}}. \tag{54}$$

In the case when the desirable component is enriched in the opposite part of the cascade (in the waste flow W) together with other components having numbers $i = n + 1, \ldots, m$, relation (54) must be rewritten as follows:

$$\frac{C_{nF}}{\sum\limits_{i=n}^{m} \dfrac{Q_i}{Q_n} C_{iF}} < C_{nW}^{\max} < \frac{C_{nF}}{\sum\limits_{i=n}^{m} C_{iF}}. \tag{55}$$

If concentration of the desirable component C_{nP} is determined from Eqs. (50), (51), and (52), one can obtain $C_{nW}^{\max}$ value corresponding to the minimum length of the stripping section, $(S_W)_{\min}$. Because we have assumed that $\exp(| Q_n | S_P) \gg 1$, hence, when $C_{nW} \rightarrow C_{nW}^{\max}$, the total flow in the cascade must unlimitedly increase. The desired C_{nP} value can be obtained for arbitrary $S_W > (S_W)_{\min}$ values. Besides, it is clear that along with the increase of S_W the S_P value will be decreasing. When $S_W \rightarrow \infty$ (i.e., when $C_{nW} \rightarrow 0$) the total flow will also increase unlimitedly. Consequently, the optimal S_W (or C_{nW}) value corresponding to the minimum of $\sum L$ will be in the interval between $(S_W)_{\min}$ and $S_W \rightarrow \infty$. Existence of the optimal length for the stripping section in cascades separating multicomponent mixtures differ them from those used to separate binary isotope mixtures. In the last case, the minimum of $\sum L$ always corresponds to $S_W \rightarrow \infty$ without any limitation for the choice of utmost concentration for desired isotope. The utmost concentrations for the intermediate mixture components at the ends of Q cascade (defined by formu-

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Table 3. Natural Concentration of Tungsten Isotopes

Component Number, *i*	1	2	3	4	5
Isotope	W-180	W-182	W-183	W-185	W-186
C_{iF}, %	0.13	26.30	14.30	30.67	28.60

lae (50)–(52)) essentially depend on their initial abundance in the isotope mixture. Thus, for the tungsten isotope mixture of the natural abundance, shown in Table 3, the utmost concentrations for W-183 ($i = 3$) are $C_{3P}^{\max} \approx 35.11\%$ and $C_{3W}^{\max} = 19.43\%$, and those for W-185 ($i = 4$): $C_{4P}^{\max} \approx 42.95\%$ and $C_{4W}^{\max} \approx 51.75\%$, respectively.

The following routs can be used to obtain the target components at higher than the utmost concentrations at the ends of the simple cascade:

1. to switch an additional product flow inside the cascade in the point of localization (concentration) of the target component. The analysis shows that in this case the concentration of purpose component does not exceed too much that predicted by relation (10), and
2. to use two consecutively connected Q cascades as shown in Fig. 9.

The total flow is an important integral characteristics of the separating cascade. By multiplying Eqs. (37) and (38) with dl and then integrating both expressions along the stripping and enriching parts of the cascade one can obtain the fol-

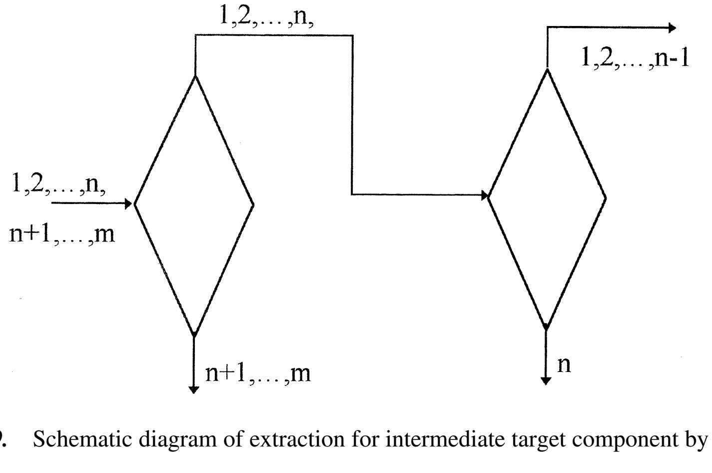

Figure 9. Schematic diagram of extraction for intermediate target component by double cascade.

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lowing formula for the total flow:

$$\begin{aligned}\Sigma L = 2\sum_{i=1}^{m}\Bigg\{ & \frac{PC_{iP}[\exp(-Q_iS_P - 1] + WC_{iW}[\exp(Q_iS_W - 1]}{Q_i^2} \\ & + \frac{PC_{iP}S_P - WC_{iW}S_W}{Q_i}\Bigg\}\end{aligned} \tag{53}$$

The following sequence of operations can be recommended to calculate a Q cascade for the definite values of C_{nP}, C_{nW}, and P if n is the target component number; and all ε_{ij} and C_{iF} values are known:

1) to set up Q_n values (or k value, to be exact, as follows from Eq. (48)) for the target component;
2) to define the rest of Q_i in accordance with relation (34);
3) to solve system Eqs. (42) and (43) for $i = n$ to find S_P and S_W;
4) to find the rest of $C_{iP}(i \neq n)$ and $C_{iW}(i \neq n)$ in line with Eqs. (42) and (43), and the values of product and feed flows by using Eqs. (44) and (45), and
5) to find the distribution of flows in the stripping and enriching sections of the cascade in conformity with Eqs. (37) and (39) and to calculate the total (sum) flow in the cascade in accordance with relation (53).

Matched Abundance Ratio Cascade (MARC)

The need to set up parameter Q_n requires determination of k value in expression (48). The results of calculations of cascades under consideration show that the sum ΣL should be taken with coefficient 0.5 to minimize the total flow. The analysis of relations (27)–(29) demonstrates that the case $Q_n = (1/2)$ $\varepsilon_{n,n+l(l=1,\ldots,m-n)}$ corresponds to the cascade, in which the abundance ratio between n- and $(n + 1)$-th components in two flows, which are mixed at the inlet of each stage, is the same. The theory of such cascade was developed in detail for $m = 3$ in the classic article by de la Garza et al. (1) where it was named the matched abundance ratio cascade (MARC) or briefly as matched R cascade. At $l = 1$, we have the most efficient MARC, which allows separation in the product flow of the target (n-th) component along with components number $1, 2, \ldots, n - 1$, and suppresses in this flow the components with numbers $n + 1, n + 2, \ldots, m$. If

$$Q_n = \frac{1}{2}\,\varepsilon_0\,(M_{n+1} - M_n), \tag{54}$$

it follows from relation (34) that for arbitrary i

$$Q_i = \varepsilon_0\left(\frac{M_n + M_{n+1}}{2} - M_i\right), \tag{55}$$

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and, in particular,

$$Q_{n+1} = -Q_n. \tag{56}$$

Rearranging Q_i in the form of (55) and taking into account the balance Eqs. (3) and (4) one can obtain from Eqs. (42) and (43) the following relations:

$$R^P_{n,n+1} \equiv \frac{C_{nP}}{C_{n+1,P}} / \frac{C_{nF}}{C_{n+1,F}} = \exp(Q_n S_P), \tag{57}$$

$$R^W_{n,n+1} \equiv \frac{C_{nW}}{C_{n+1,W}} / \frac{C_{nF}}{C_{n+1,F}} = \exp(-Q_n S_W). \tag{58}$$

Equations (57) and (58) clearly show that in the cascade under consideration the abundance ratios $R_{n,n+1} = C_n/C_{n+1}$ in the inlets of separating stage are identical, i.e., the conditions of nonmixing of relative concentrations $R_{n,n+1}$ are fulfilled.

As follows from relations (57) and (58), setting up the relative concentrations $R_{n,n+1}$ in the product and waste flows allows one to determine invariantly the numbers of stages S_P and S_W and hence, by using formulae (42)–(45), (37), and (39) one can entirely calculate the cascade.

Simultaneously with definition of Q_i in the form of Eq. (54), expression (53) for the total flow is essentially simplified. In fact, from Eqs. (40) and (41) in line with conditions (56) we have

$$PC_{iP}\,[\exp(-Q_i S_P) - 1] = -\,PC_{n+1,P}\,\frac{C_{if}}{C_{n+1,f}} \cdot \frac{Q_i}{Q_n}\,[\exp(Q_n S_P) - 1], \tag{59}$$

$$WC_{iW}\,[\exp(Q_i S_W)_P - 1] = -WC_{n+1,W}\,\frac{C_{if}}{C_{n+1,f}} \cdot \frac{Q_i}{Q_n}\,[\exp(-Q_n S_W) - 1] \tag{60}$$

By summarizing Eqs. (59) and (60) and accounting for Eqs. (4), (5), (57), and (58), one obtains the following equality, which is valid for arbitrary *i*:

$$PC_{iP}\,[\exp(-Q_i S_P) - 1] + WC_{iW}\,[\exp(Q_i S_W) - 1] = 0 \tag{61}$$

Besides, it directly follows from relations (57) and (58) that

$$\frac{PC_{iP} - WC_{iW}\,S_W}{Q_i} = \frac{PC_{iP} \ln R^P_{n,n+1} + WC_{iW} \ln R^W_{n,n+1} - FC_{iF} \ln R^F_{n,n+1}}{Q_i Q_n} \tag{62}$$

Inserting relations (61) and (62) in (53) and substituting Q_n and Q_i in accordance with (54) and (55) one obtains

$$\sum L = \frac{2}{\varepsilon_0^2} \sum_j^m \frac{PC_{jP} \ln R^P_{n,n+1} + WC_{jP} \ln R^W_{n,n+1} - FC_{iF} \ln R^F_{n,n+1}}{(M_{n+1} - M_n)\left(\dfrac{M_{n+1} - M_n}{2} - M_j\right)} \tag{63}$$

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Equation (61) along with Eqs. (57), (58), and (3), (4) can be easily transformed into the following form

$$PC_{iP}(R^{P}_{n,n+1})^{-\beta_i} + WC_{iW}(R^{W}_{n,n+1})^{-\beta_i} - FC_{iF}\,(R^{F}_{n,n+1})^{-\beta_i} = 0, \qquad (64)$$

where $\beta_i = Q_i/Q_n$.

Relation (64) connecting the flows and concentrations in MARC is called the H balance (1). One needs to take into account that it includes $(m - 2)$ independent relations therefore, when $i = n$ and $i = n + 1$, it transforms into the balance equation for n and $n + 1$ components. Equation (64) together with m balance Eq. (4) and evident relations $\Sigma_{j=1}^{m} C_{jP} = 1$ and $\Sigma_{j=1}^{m} C_{jW} = 1$ compose the system of $m - 2 + m + 2 = 2m$ independent algebraic equations possessing $(2m + 3)$ unknown parameters ($2m$ outgoing concentrations and P, W, and F flows). Hence, the design calculations of MARC suggest the definition of at least $(2m + 3) - 2m = 3$ parameters (for example, C_{nP}, C_{nW}, and P). The rest of the parameters will be defined from the solution of the above mentioned system of algebraic equations.

In the separation practice, one can meet the variants of cascade calculations of specified number of separating stages, i.e., the cascades with known total flow. In this case the system of algebraic equations must be complemented by Eq. (63), which suffices to determine only two parameters (for example, C_{nP} and P) for its solution.

Note that the considered Q cascade for separation of binary mixture is reduced to the ideal cascade without mixing of concentrations at entrances of the stages. This directly follows from formulae (57) and (58). It is easy to show that in this case, expression (63) for the total flow also converts into the well-known expression for the binary cascade. It is important to emphasize that as it has been proposed by Levin (18), extraction of the intermediate components can be carried out by means of so-called M^* cascades using somewhat other considerations, which lead to the same results.

Calculation of MARC with Additional Product Flows

The problem of calculation of multicomponent cascades (used for separation of multicomponent mixtures) with additional product flows is connected with the situation when the concentration of intermediate components achieves maximum inside the cascade. Hence, their concentrations in an additional product flow can be higher than in the flow at the end of cascade. The approach described at the end of the previous paragraph can be used to calculate MARC with additional product flows (see Fig. 10). Taking into account the condition used above on nonmixing of relative concentrations $R_{n,n+1} = C_n/C_{n+1}$ we can write equations for the

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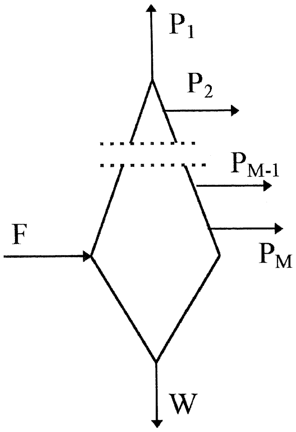

Figure 10. Schematic diagram of *m*-component MARC having one feed, one waste, and *M* withdrawal points.

total and single component balances connected by external flows and component concentrations in these flows (so-called productivity equations (1,17)):

$$\sum_{l=1}^{M} P_l + W - F = 0, \tag{65}$$

$$\sum_{l=1}^{M} P_l C_{jP_l} + W C_{jW} - F C_{jF} = 0. \tag{66}$$

where $j = 1, 2, \ldots, m$

The H balance equations will be the same as in the previous paragraph

$$\sum_{l=1}^{M} P_l C_{jP_l} (R_{n,n}^{P_l} + 1)^{-\beta_i} + W C_{iW} (R_{n,n+1}^{W})^{-\beta_i} - F C_{lF} (R_{n,n+1}^{F})^{-\beta\mathrm{i}} = 0, \tag{67}$$

where $j = 1, 2, \ldots, m; i \neq n, n + 1$.

Considering flow L as uninterrupted along the cascade length and following Borisevich et al. (17), one can write the following relation for the section in MARC starting from the tail product flow until the entering point of additional product flow P_k:

$$C_{iPk} = C_{nPk} \frac{\sum_{l=1}^{M-1} P_l C_{iP_l} [1 - (R_{n,n+1}^{P_l} / \mathrm{R}_{n,n+1}^{P_k})^{-\beta\mathrm{i}}]}{\sum_{l=1}^{M} P_l C_{lP_l} [1 - (R_{n,n+1}^{P_l} / R_{n,n+1}^{P_k})^{-1}]}, \tag{68}$$

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where $i = 1, 2, \ldots, m$; $i \neq n, n+1$; $k = 2, 3, \ldots, M$, and $R^{P_l}_{n,n+1} = C_{nP_l}/C_{(n+1)P_l}$ and C_{iP_l} are the i-th component concentration in the product flow P_l.

If necessary, one can use expression for the total flow in the cascade, which is written as follows:

$$\sum L = 2 \sum_{j=1}^{m} \frac{\sum_{l=1}^{M} P_l C_{jP_l} \ln R^{P_l}_{n,n+1} + WC_{jW} \ln R^{W}_{n,n+1} - FC_{jF} \ln R^{F}_{n,n+1}}{Q_i Q_n} \tag{69}$$

Equations (64)–(68) compose the system for calculation of MARC with several product flows. In the simplest case when $M = 2$, these equations take the following form:

$$P_1 + P_2 + W - F = 0, \tag{70}$$

$$P_1 C_{jP_1} + P_2 C_{jP_2} + WC_{jW} - FC_{jF} = 0, \tag{71}$$

$$\begin{aligned} P_1 C_{iP_1} (R^{P1}_{n,n+1})^{-\beta i} + P_2 C_{iP_2} (R^{P_2}_{n,n+1})^{-\beta i} \\ + WC_{iW}(R^{W}_{n,n+1})^{-\beta i} - FC_{iF}(R^{F}_{n,n+1})^{-\beta i} = 0 \end{aligned} \tag{72}$$

$$\frac{C_{iP2}}{C_{iP1}} = \frac{C_{nP2}}{C_{nP1}} \frac{1}{\beta_i} \frac{(R^{P_1}_{n,n+1})^{\beta i} - (R^{P_2}_{n,n+1})^{\beta i}}{R^{P_1}_{n,n+1} - R^{P_2}_{n,n+1}} (R^{P_1}_{n,n+1})^{1-\beta i}, \tag{73}$$

where $i = 1, 2, \ldots, m$; $i \neq n, n+1$.

In this case, if the total flow is not defined we have $(3m - 1)$ independent equations comprising $(3m + 3)$ unknown parameters: C_{iP1}, C_{iP2}, C_{iW}, P_2/P_1, W/P_1, F/P_1 (concentrations C_{iF} are supposed to be defined). Hence, to solve this system we have to select $(3m + 3) - (3m - 1) = 4$ parameters (for example, P_2/P_1, C_{nP1}, C_{nP2}, C_{nW}). If in addition, the total flow is defined we have $3m$ equations including only three parameters. The system of equations can be solved by one of the well-known calculation methods such as, e.g., the Newton method.

The results of calculations for MARC with an additional product flow exemplify separation of five-components isotope mixture of tungsten of natural abundance. The intermediate isotope, W-183 ($n = 3$, $C_{3F} = 0.143$), was selected as the target component. The flow ratio P_2/P_1 (which is varied from 0 to 1) was chosen as the desired parameter to calculate MARC with an additional product flow to achieve the concentrations: $C_{3P1} = 32.0\%$; $C_{2P2} = 43.0\%$, and $C_{3W} = 1.2\%$. The results of these calculations are presented in Table 4.

The results of calculations for MARC with a given total flow and the same values of concentrations are presented in Table 5.

The results of calculations demonstrate the possibility of obtaining in the additional product flow concentration of the target component higher than in the flow at the end of the cascade. The value of additional product flow depends on the concentration of the target component in it as well as on the composition of initial mixture to be separated and on concentration of the target component in the

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Table 4. Results of Calculation for MARC with Additional Product Flow and Given Concentrations $C_{3P1} = 32.0\%$, $C_{2P2} = 43.0\%$, and $C_{3W} = 1.2\%$

P_2/P_1	0.043	0.060	0.080	0.100	0.120	0.160	0.200	0.204	0.208
W/P_1	5.764	6.008	6.368	6.766	7.251	8.590	12.33	13.65	22.88
$\varepsilon_0 S_{P_1}$	2.331	1.732	1.551	1.439	1.358	1.246	1.168	1.162	1.155
$\varepsilon_0(S_{P_1} - S_{P_2})$	5.912	5.917	5.925	5.933	5.941	5.955	5.967	5.971	5.973
$\varepsilon_0 S_W$	1.499	1.437	1.531	1.575	1.619	1.707	1.759	1.805	1.814
$(\varepsilon_0^2 \Sigma L)/P_1$	17.17	17.34	18.35	19.11	20.22	23.12	29.70	31.76	45.25

Here S_{P_1} is the total number of stages in the enriching section of MARC; S_{P_2} is the number of stages between the feed point and the point of the additional product flow P_2; and S_w is the number of stages in the stripping section.

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main product flow P_1. In the above example, P_1 has reached the value of ~20% of the main product flow at a relative increase of the target component concentration in this flow (compared with C_{3P_1}) >30%. The analysis of the results obtained has shown that introduction of additional product flow leads to an increase of both the total flow and the number of stages. It is important to note that the value of P_2/P_1 is limited from above. The existence of the utmost P_2/P_1 value can be explained as follows: this ratio increases until the moment when the given concentration of the target component cannot be reached in any section of the cascade.

Design Calculations for Squared-Off Cascades Based on Continuous Profile Cascades

Calculations of the squared-off cascade (SOC) for the fixed extraction degree (product flow) and given restrictions on the range of concentrations of the target isotope in the product and waste flows consist of determination of the following parameters: the number, the length, and the values of intermediate flows in sections, and the ratio of waste to product flows W/P. In this case, the values of the parameters must, as a rule, correspond to the optimal ones meeting some definite criteria.

The profile of cascade under design and the concentrations of components

Table 5. Calculated Results for MARC with $\varepsilon_0^2 L/P_1 = 20$, $C_{3P_1} = 32.0\%$, $C_{2P_2} = 43.0\%$, and $C_{3W} = 1.2\%$

$\varepsilon_0 S_{P_1}$	$\varepsilon_0(S_{P_1} - S_{P_2})$	$\varepsilon_0 S_W$	P_2/P_1
7.188	1.371	5.939	0.117

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of a multicomponent isotope mixture at its ends cannot be determined analytically. Hence, the methods used to directly integrate the mass-transfer equations for calculation of cascades (to obtain a fixed enrichment of intermediate component) are not always effective and some of them are not applicable at all.

It is convenient to use the above-described Q cascade as a model cascade of continuous profile to solve the calculation problem for intermediate components. The principle possibility of approximation of a Q cascade by a squared-off cascade has been demonstrated (13). One of possible criteria for optimal approximation along with technique for calculation of SOC has been proposed (24). The problem of approximating a part of Q cascade operating in the range of concentrations from C_{ib} to C_{il} ($i = 1, 2, \ldots, m$, where m is the number of mixture components) by a section of constant width was also formulated by Kolokol'tsov et al. (24). According to Kolokol'tsov et al. (24), one needs first to find the values of flow L and number of stages S in the section providing the minimum deviations of concentrations at the end of the square section to be calculated. In this case, it is assumed that the concentrations at the beginning of section and in the product flow of cascade C_{iP} coincide with the relative concentrations of a Q cascade. The sum of relative deviations of component concentrations in the joint points of sections can be easily used as an approximation criterion, Φ:

$$\Phi = \sum_{i=m}^{m} \left| \frac{C_{il} - C_i}{C_{il}} \right|. \tag{74}$$

The calculation of single sections can be done by direct integration of Eqs. (7) and (8). For concentrations at the beginning of each section one can write:

$$C_i(0) = C_{ib}; \quad i = 1, 2, \ldots, m - 1, \tag{75}$$

Hence, the calculation of section is reduced to solution of Eqs. (7) and (8) with respective initial conditions, i.e., it is reduced to the ordinary Cauchi problem. Since the concentration values in each point of section with defined concentrations in the product and the waste flows, C_{iP} and C_{iW}, respectively, depend on coordinate S (starting from the beginning of section) and on the flow L the problem is reduced to the search of S and L values that will minimize function (74). Figure 11 shows distributions of relative deviations for concentrations $(C_{il} - C_i)/C_{il} = (\Delta C_i/C_{il})$ and functional (74) with respect to the relative flow $\varepsilon_0 L/2P$ in a constant width cascade approximating a Q cascade for the six-component isotope mixture of krypton: $M_i = 78, 80, 82, 83, 84, 86$; $C_{iF} = 0.00354, 0.0227, 0.1156, 0.1155, 0.5690, 0.1737$. As seen from Fig. 9, the relative deviations of concentrations are not high and do not exceed 3% in the point corresponding to the minimum of functional (74).

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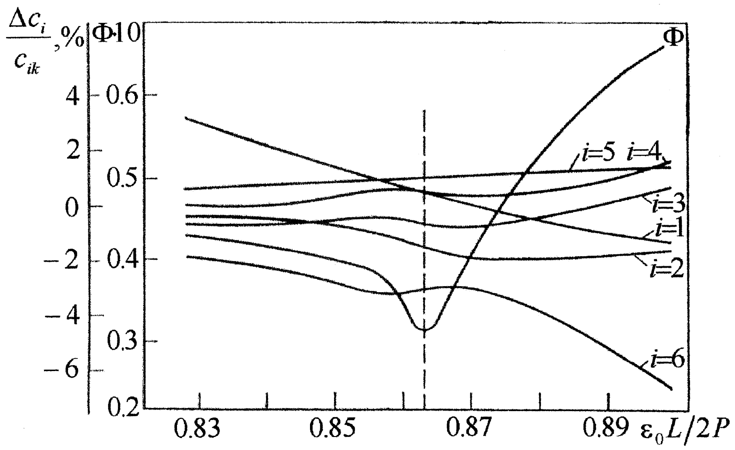

Figure 11. Dependence of relative deviation of concentrations and function of residuals vs. relative flow $\varepsilon_0 L/2P$.

A method for approximate calculation and optimization of SOC by using some criterion was proposed (25). The method is based on the use of a target optimization function, which accounts for both the optimality criterion and the sum of deviations of component concentrations at the joints of the product and waste sections. For the total flow chosen as a criterion, the target function has the following form:

$$\Phi = K_1 \sum |C_{if}(P) - C_{if}(W)| + K_2 \frac{\sum L_{\mathrm{SOC}}}{L_Q}, \tag{76}$$

where $C_{if}(P)$, $C_{if}(W)$ $(i = 1, 2, \ldots, m)$ are the concentrations in the feed point determined as the result of integration of system (7)–(8) along the lengths of the product and waste parts in the direction from the ends of the cascade to its feed point, and ΣL_{SOC}, ΣL_Q are the total flow of the SOC and the Q cascade, respectively.

The target approximation of the cascade is obtained in the course of optimization of cascade parameters, for which the value of Φ function is minimal. At each optimization stage, system (7)–(8) is integrated with only the following boundary conditions:

$$C_{iP} = C_{iP}^{Q}, \quad C_{iW} = C_{iW}^{Q} \quad i = 1, 2, \ldots, m, \tag{77}$$

where C_{iP}^{Q}, C_{iW}^{Q} are the concentrations at the ends of Q cascade to be approximated.

The first term of function (77) determines the accuracy of calculations of SOC corresponding to the optimal Q cascade and having the same component

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concentrations at the ends. The second term characterizes the degree of SOC approximation to the optimal one and is inverse to the efficiency of the cascade profile form.

The accuracy of calculation increases if the terms of function (76) are of the same order of magnitude. The last condition is achieved through the use of normalizing coefficients K_1 and K_2. Their ratio depends on 1) the number of components; 2) the composition of initial mixture; 3) the concentration of target component in the initial mixture; and 4) the degree of its enrichment. The dependencies of mixture concentrations in the product and waste flows and the value of $1/\eta_\Phi$ for isotopic mixtures of different composition were studied by Frolov et al. (25) to determine the range of variation of K_1/K_2 ratio. It was found that for mixtures of different composition containing from three to six components the ratio K_1/K_2 varies from 10 to 30.

Figure 12 shows the dependencies of concentrations of the target component C_2 in the three-component mixture determined in the product and waste flows and the values of inverse efficiency of the cascade form on the K_1/K_2 ratios. The calculations were performed for separation of the three-component model mixture of isotopes with component concentrations in the feed stream: $C_{1F} = 0.2$, $C_{2F} = 0.3$, and $C_{3F} = 0.5$. The isotopes are enumerated in the order of increasing masses. The second isotope is the target one. For the mixture under study the values of the form efficiency and the concentration of the target isotope in the product and waste flows are evidently close to the optimal ones (in the given range of K_1/K_2 variations) for the SOC obtained as the result of approximation. For high K_1/K_2 values the second term in Eq. (76) can be neglected, and the SOC resulting due to approximation is close to that computed

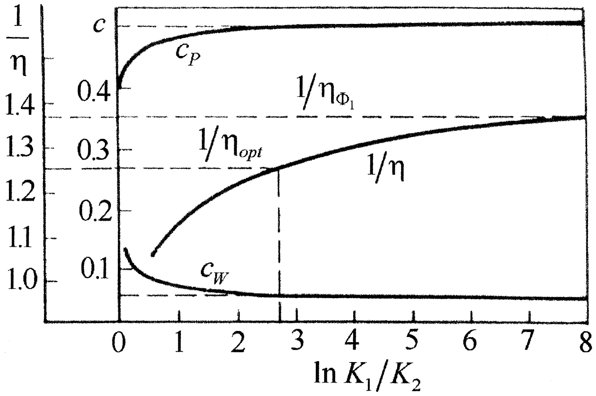

Figure 12. $1/\eta$ and concentration of target component intermediate of model three-component mixture in product flow (C_P) and waste flow (C_W) of square cascade vs. K_1/K_2.

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according to criterion (74) with efficiency equal to $\eta_{\Phi 1}$. For low K_1/K_2, the condition for continuity of component concentrations at the feed point can be disturbed. In this case to make the calculation more accurate one needs to solve the following system of equations:

$$\begin{aligned} &C_{if}(P) = C_{if}(W), \\ &PC_{iP} + WC_{iW} = FC_{iF}, \quad i = 1, 2, \ldots, m. \end{aligned} \tag{78}$$

The concentrations of the target isotope in the product and waste flows obtained from this solution can substantially differ from the above given values (see Fig. 10).

Thus the scheme for calculation of the optimal SOC based on the method proposed is as follows: calculation and optimization of the Q cascade; simultaneous calculation and optimization of the SOC approximating this Q cascade; the refinement of concentrations of the target isotope in the product and waste of the cascade obtained.

It is desirable to optimize the Q cascade and the SOC by a unified criterion. The approach described can also be used to calculate optimal cascades, which have several feed and product flows.

MARC with Arbitrary Enrichment at Each Stage

It is reasonable to assume that MARC (R cascade) can also be effectively used for separation of isotopic and non-isotopic (gaseous) mixtures when the enrichment effect at a separating stage is not small.

Let us examine a counter-flow symmetric MARC consisting of N stages, which is intended for separation of an m-component mixture. The flux of the initial mixture, F, with component concentrations C_{jF}($j = 1, 2, \ldots, m$) is fed into the input of stage number f. To calculate separation characteristics of R cascade one needs to determine 1) the product (P) and the waste (W) flows exiting the N-th and the first stages, respectively; 2) the component concentrations in these streams (C_{jP} and C_{jW}); 3) the distributions of the mixture flow (L_S) along the cascade length; 4) the concentrations of components $C_j(S)$; and 5) the cut Θ_S.

In the case under consideration, the concentration distribution along the length of the cascade can be obtained by solving the system of Eq. (20). Here the cut coefficients $\Theta(S)$, $S = 2, 3, \ldots, N - 1$ must be chosen to fulfil the non-mixing conditions:

$$R^{+}_{nk}(S - 1) = R_{nk}(S) = R^{-}_{nk}(S + 1), \tag{79}$$

where

$$R_{nk} = \frac{C_n(S)}{C_k(S)}, R^{+}_{nk}(S - 1) = \frac{C^{+}_n(S - 1)}{C^{+}_k(S - 1)}, R^{-}_{nk}(S + 1) = \frac{C^{-}_n(S + 1)}{C^{-}_k(S + 1)},$$

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$C_n(S)$, $C_n^+(S)$, $C_n^-(S)$ are the concentrations of the n-th component in the fluxes entering and leaving the S-th stage, respectively, n and k are the arbitrary numbers of components of the mixture under separation (for the target components $n \neq k$). In addition, for each separation method the concentrations C_i^+, C_i^- depend on the composition of the mixture at the inlet into stage and on Θ_i value at this stage. The cut of the flux in the first stage is fixed arbitrary (20). The non-mixing conditions for the relative concentrations of the n-th and k-th components in the feed flow and in the flow at the inlet into the f-th stage written as follows:

$$R_{nk}^F = R_{nk}(f) \tag{80}$$

are determined by a proper selection of the corresponding waste flow W.

The flows at the inlet of each cascade stage, L_S, are calculated with the help of the following material balance equations in an arbitrary cross-section of the cascade (12,20):

$$\mathrm{L_S} = \begin{cases} W/(1 - \Theta_S),\ S = 1 \\ \Theta_{S-1}L_{S-1}/(1 - \Theta_S) + W/(1 - \Theta_S),\ S = 2, 3, \ldots, f \\ \Theta_{S-1}L_{S-1}/(1 - \Theta_S) - P/(1 - \Theta_S),\ S = f + 1, f + 2, \ldots, N - 1 \\ \Theta_{S-1}L_{S-1},\ S = N \end{cases} \tag{81}$$

Equations (20), (79), and (80) along with (81) and the known functions C_i^+ $(C_1, \ldots, C_m, \Theta)$, C_i^- $(C_1, \ldots, C_m, \Theta)$ can be solved for unknowns $C_i(S)$, Θ_s, W, $S = 2, 3, \ldots, N - 1$; $i = 1, 2, \ldots, m$ by using the modified damped Newton method (12). As it is known, the convergence of numerical methods in solution of nonlinear algebraic equations mainly depends on either the structure of equations or the form in which they are written. The numerical experiments have shown (20) that the Newton method is the most stable when Eqs. (79) and (80) are written in the following form:

$$1 - \frac{R_{nk}^+(S - 1)}{R_{nk}^-\,(S + 1)} = 0, \qquad 1 - \frac{R_{nk}^F}{R_{nk}(f)} = 0,$$

and the ratio W/F is the unknown.

The solution of system of Eqs. (20), (79), and (80) has been found in the analytical form (21–23) for the case when the relative separation factors

$$\alpha_{ik} = \frac{R_{ik}^+}{R_{ik}^-} = \frac{C_i^+}{C_k^+}\cdot\frac{C_k^-}{C_i^-}; \quad \alpha_{ik}^+ = \frac{R_{ik}^+}{R_{ik}} = \frac{C_i^+}{C_k^+}\cdot\frac{C_k}{C_i}; \quad \alpha_{ik}^- = \frac{R_{ik}}{R_{ik}''} = \frac{C_i}{C_k}\cdot\frac{C_k^-}{C_i^-}$$

(where $i = 1, \ldots, m$, and k denotes an arbitrary target component) do not depend on either composition of the mixture or the cut, i.e., their values remain constant along the cascade length.

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Let us introduce the following notations:

$$G_i = C_i L, \tag{82}$$

$$G_i^{+} = C_i^{+}\Theta L, \tag{83}$$

$$G_l^{-} = C_i^{-}(l - \Theta)L, \tag{84}$$

$$\beta_i = G_i^{+}/G_i^{-}, \tag{85}$$

and the following evident relationships:

$$\alpha_{ik}^{+} = \frac{R_{ik}^{+}}{R_{ik}} = \frac{G_i^{+}\cdot G_k}{G_k^{+}\cdot G_i}, \tag{86}$$

$$\alpha_{ik}^{-} = \frac{R_{ik}}{R_{ik}^{-}} = \frac{G_i\cdot G_k^{-}}{G_k\cdot G_i^{-}}, \tag{87}$$

$$\alpha_{ik} = \alpha_{ik}^{+}\cdot\alpha_{ik}^{-} = \frac{R_{ik}^{+}}{R_{ik}^{-}} = \frac{G_i^{+}\cdot G_k^{-}}{G_k^{+}\cdot G_i^{-}}, \tag{88}$$

$$L = \sum_{j=1}^{m} G_j, \quad \Theta L = \sum_{j=1}^{m} G_j^{+}, \quad (1 - \Theta)L = \sum_{j=1}^{m} G_j^{-}, \tag{89}$$

It is easy to obtain the following expressions connecting β_i and β_k values with relative separation factors:

$$\beta_i = \frac{\alpha_{ik}^{+}(\alpha_{ik}^{-} - 1)}{(\alpha_{ik}^{+} - 1)}, \quad i \neq k \tag{90}$$

$$\beta_k = \frac{(\alpha_{ik}^{-} - 1)}{\alpha_{ik}^{-}(\alpha_{ik}^{+} - 1)}. \tag{91}$$

Thus, the separation factors are defined with respect to the k-th component, and β_k has the same value for an arbitrary $i \neq k$. As it follows from Eqs. (90) and (91)

$$\beta_i = \alpha_{ik}\beta_k. \tag{92}$$

Taking into account the above-introduced notations, the balance equation for the component fluxes at the inlet of an arbitrary S-th stage has the following form:

$$G_i^{+}(S - 1) + G_i^{-}(S + 1) = G_i^{+}(S) + G_i^{-}(S), \quad i = 1, 2, \ldots, m. \tag{93}$$

If the relative separation factors do not change from stage to stage, the β_i and β_k values will also be constant along the length of the cascade. Equations (93) and (85) can then be written as follows:

$$G_i^{+}(S + 1) - (\beta_i + 1)G_i^{+}(S) + \beta_i G_i^{+}(S - 1) = 0. \tag{94}$$

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Equation (94) intends a nonlinear second order finite-difference equation regarding unknown functions $G+/i\ (S)$. The boundary conditions for them are written as follows:

$$
\begin{aligned}
&G_i^+(0) = G_i^+(N+1) = 0,\\
&G_i^+(f-1) + G_i^+(f+1)\cdot\frac{1}{\beta_i} = \left(1+\frac{1}{\beta_i}\right)G_i^+(f) + FC_{iF},\\
&G_i^+(N) = PC_{iP},\\
&G_i^+(1) = WC_{iW}\beta_i, \quad i = 1, 2, \ldots, m.
\end{aligned}
\tag{95}
$$

Besides, the solutions of Eq. (94) must also coincide at $S = f$. The stages with numbers $S = 1$ and $S = N$ are the end stages of the cascade. Hence, this makes it possible to formally write: $G_i^+\ (0) = G_i^+(N+1) = 0$.

The fundamental solution of Eq. (94) for the constant values of coefficients β_i and β_k can be presented in the following form:

$$G_i^+(S) = A_i\omega_1^S + B_i\omega_2^S, \tag{96}$$

where A_i and B_i are the integration constants, and $\omega_{1,2}$ are the roots of characteristic equations

$$\omega^2 - (\beta_i + 1)\omega + \beta_i = 0 \tag{97}$$

having the following values

$$\omega_1(\beta_i) = \beta_i, \tag{98}$$

$$\omega_2(\beta_i) = 1. \tag{99}$$

Using the boundary conditions (95) to determine constants A_i and B_i, one can obtain:

for enriching section of cascade

$$G_i^+(S) = PC_{iP}\,\frac{\beta_i}{\beta_i - 1}\,(1 - \beta_i^{S-N-1}), \tag{100}$$

$$PC_{iP} = FC_{iF}\,\frac{1 - \beta_i{}^{-f}}{1 - \beta_i{}^{-N-1}}, \quad S = f, \ldots, N, \tag{101}$$

for stripping section

$$G_i^+(S) = WC_{iW}\,\frac{\beta_i}{\beta_i - 1}\,(\beta_i^S - 1), \tag{102}$$

$$WC_{iW} = FC_{iF}\,\frac{\beta_i^{N+1-f}}{\beta_i^{N+i} - 1}, \quad S = 1, \ldots, f-1. \tag{103}$$

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From Eqs. (100)–(103), one can easily obtain the following relations:

$$\frac{P}{F} = \sum_{j=1}^{m} C_{jF}\,\frac{1-\beta_j^{-f}}{1-\beta_j^{-N-1}}, \tag{104}$$

$$\frac{W}{F} = \sum_{j=1}^{m} C_{jF}\,\frac{\beta_i^{N+1-f}-1}{\beta_j^{N+1}-1}, \tag{105}$$

$$C_{iP} = C_{iF}\,\frac{1-\beta_i^{-f}}{1-\beta_i^{-N-1}} / \sum_{j=1}^{m} C_{iF}\,\frac{1-\beta_j^{-f}}{1-\beta_j^{-N-1}}, \tag{106}$$

$$C_{iW} = C_{iF}\,\frac{\beta_i^{N+1-f}-1}{\beta_i^{N+1}-1} / \sum_{j=1}^{m} C_{iF},\,\frac{\beta_j^{N+1-f}-1}{\beta_j^{N+1}-1}. \tag{107}$$

If C_{iF} and β_i values and the numbers of stages in the enriching and stripping sections are defined, one can calculate in conformity with formulae (104)–(107) the values of the following parameters: P/F, W/F, C_{iP}, and C_{iW}. Then by using Eqs. (100)–(103), the following expressions describing $L(S)$, $C_i(S)$, and $\Theta(S)$ distributions can be written:

$$L = \sum_{j=1}^{m} G_j = \sum_{j=1}^{m} G_j^{+}\,\frac{1+\beta_j}{\beta_j}, \tag{108}$$

$$C_i = \frac{1+\beta_i}{\beta_i}\,\frac{G_i}{L}, \tag{109}$$

$$\Theta = \sum_{j=1}^{m} \frac{G_i^{+}}{L}. \tag{110}$$

Let us now consider MARC with non-mixing condition (79). It follows directly from Eq. (79) (21,23) that

$$\alpha_{nk}^{+} = \alpha_{nk}^{-} = \sqrt{\alpha_{nk}}, \tag{111}$$

where $\alpha_{ik}^{+} \neq \alpha_{ik}^{-} (i \neq n)$. Because relationship (91) is valid for arbitrary i including $i = n$, by taking into account (101) and (92) one obtains:

$$\beta_k = \frac{1}{\sqrt{\alpha_{nk}}} \tag{112}$$

$$\beta_i = \frac{\alpha_{ik}}{\sqrt{\alpha_{nk}}}. \tag{113}$$

The separation factor for each pair of mixture components can be expressed for many isotope separation methods as a function of the difference between their

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molecular masses. For example, for a gas centrifuge (GC) technique the following approximation can be used:

$$\alpha_{ik} = \alpha_0^{(M_k - M_i)}, \tag{114}$$

where α_0 is the separation factor per unit of mass difference, M_k and M_i are the masses of the target and the i-th components, respectively.

The coefficients β_i and β_k for GC-MARC can be expressed as follows:

$$\beta_k = \alpha_0^{(M_k - M_i)/2}, \tag{115}$$

$$\beta_i = \alpha_0^{(M^* - M_i)}, \ (i \neq k), \tag{116}$$

where

$$M^* = \frac{M_n + M_k}{2}, \tag{117}$$

and n and k are the numbers of the target components. Equations (100)–(107) together with (115)–(117) permit calculation of the variables of interest for the matched abundance ratio cascade.

As seen from Eqs. (106) and (107) when the number of stages in both enriching and stripping sections become unlimitedly large, the components with molecular mass less than M^* accumulate only in the cascade product flux. The components with masses higher than M^* concentrate only in the cascade waste flux. This circumstance defines the choice of components with numbers n and k. When $k = n + 1$, the cascade under consideration (similar to the case of small enrichment at the stage) permits both to extract in the product flux the target (n-th) component together with components with numbers $1, 2, \ldots, n - 1$ and "to depress" in this flow component with numbers $n + 1, \ldots, m$.

In conclusion one needs to emphasize that the main advantage of using MARC calculations to obtain an approximate cascade design is that the solution is explicit, regardless of the number of components under consideration. These calculations can be easily carried out either on a PC or even on a programmable calculator.

TRANSIENT PROCESSES IN CASCADES SEPARATING MULTICOMPONENT ISOTOPE MIXTURES

Mathematical Modeling of Mass Transfer

One of the important problems in the theory of multicomponent isotope separation is the study of distinctive features of transient processes existing in starting operating period of cascades preceding establishment of a steady state. The

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importance of this problem deals with sufficiently high duration of transient processes in the majority of separation methods. Hence, they may compose a big part of exploitation time of operating installations.

The mathematical model for separation process in isotope approximation (big number of separating stages, small value of enrichment coefficients, and their independence on current concentrations of the mixture) may be presented in the form of a set of differential equations in partial derivatives (26–32):

$$H(S)\frac{\partial C_i(S,t)}{\partial tl} = -\frac{\partial}{\partial S} J_i(S,t),\ i = 1, \ldots, m-1,$$
$$\sum_{j=1}^{m} C_j = 1. \tag{117}$$

Here $C_i(S,t)$ is the mole fraction of i-th component of the mixture at the cascade stage number S; t is the time; $H(S)$ is the retardation (delay) of the substance flow at the stage; and m is the number of the mixture components. For the majority of separation methods one can presume with good accuracy that the delay $H(S)$ is proportional to the flow $L(S)$ incoming S-th stage, i.e., $H(S) = hL(S)$, where h is the coefficient of proportionality having the dimension of time. The value of transfer $J_i(S,t)$ for the mixture components in the direction of the cascade stages with growing numbers can be written as follows:

$$J_i(S,t) = \frac{L(S)}{2}\left[C_i(S,t)\sum_{j=1}^{m} \varepsilon_{ij} C_j(S,t) - \frac{\partial C_i(S,t)}{\partial S}\right] + TC_i(S,t) \tag{118}$$

where ε_{ij} is the enrichment coefficient for the component pairs with numbers i and j (the components are enumerated in the order of their growing masses).

For definiteness, let us consider a separation in cascade of a constant width, i.e., a cascade with one feed flow and two withdrawals $P_1 \equiv W$ and $P_2 \equiv P$ at the cascade ends. In this case, the transfer value for the mixture under separation is defined as P_2 for the cascade stages in the enriching section (related to the lightest isotope) and that of the cascade stripping section is defined as $(P_2 - F)$.

The boundary conditions for the points where the flow $L(S)$ and the transfer T are interrupted are written in the following form at the ends of the cascade:

$$\frac{\partial C_i}{\partial S} = C_i \sum_{j=1}^{m} \varepsilon_{ij} C_j, \tag{119}$$

In the internal points of the cascade where the flow $L(S)$ also has the break off

$$J_i^+ = J_i^-, \tag{120}$$

and for the feed-point

$$J^- + FC_{iF} = J^+. \tag{121}$$

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The signs "+" and "−" denote the transfer J_i at the right- and left-hand sides of the points being considered.

In the two-phase separation methods such as distillation or chemical isotope exchange, these equations must be complemented by additional expressions characterizing the loading of the volumes of the phase reversal with a given component, $E(dC_i/dt)$, where E is the delay of substance in the respective volume. The initial condition must be defined in the form of distribution of component concentrations along the cascade length at zero time ($t = 0$) to solve the boundary problem

$$C_i(S,0) = \Psi_i(S). \tag{122}$$

In the simplest case when the cascade is filled with a homogeneous mixture of composition C_{iF}, the initial condition (122) will be written as follows:

$$C_i(S,0) = C_{iF} \tag{123}$$

Therefore, Eq. (117) along with boundary conditions (119–121) and initial condition (122) define the full set of equations desired to search all concentrations of the m-th component mixture in every point of the cascade at arbitrary moment of time.

The modeling of transient processes in a cascade is connected with well-known difficulties even for the case of separation of binary isotopic mixtures when the set of Eq. (117) reduces to a single equation in partial derivatives. If the number of components is more then two, probably the only opportunity to solve the problem consists of numerical integration of equations that, in turn, suggests the development of effective algorithms and computing codes.

Known from the literature, approaches to solving this problem are based on the use of the finite-difference (grid) models as well as the differential models for equations describing the non-steady processes in the cascades. The former has an essential disadvantage consisting of the fact that implicit schemes used are rather stable but require the solution of large sets of nonlinear algebraic equations at every time-step to be computed. The use of traditional explicit schemes (free of this disadvantage) is limited by the inadmissible low value of the integration step by time (27). The second approach is usually settled by the reason that modern methods for numerical integration of the ordinary differential equations are better developed than those used to solve large systems of nonlinear algebraic equations. The difference models (methods of lines) may be considered the limiting case of the grid models when one of the grid sizes (the integration step by time) tends to zero. For example, the boundary problem for cascades with the phase reversal volumes has been reduced (28) by means of the asymptotic transformations to the Cauchy problem for the system of ordinary differential equations. This permitted the set of equations to be obtained with well-conditioned matrices of coefficients admitting for use their solution simple integration methods such as, for example,

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Euler's method. However, approbation of this technique for the non-reversal separation methods did not give good results.

Transition to the set of ordinary differential equations has been accomplished (29) by changing the spatial derivatives into symmetric finite-difference relations. The solution of the set obtained is carried out by the Runge-Kutta-Gill method or similar techniques requiring relatively small integration steps for their stability. The numerical technique for integration of the set of Eqs. (117)–(122) free from the above disadvantages was developed (30,31). The gist of the method is briefly as follows. An absolutely stable method for solution of boundary problems of the parabolic type equations is described (39). This method is stable for all kinds of laws at integration steps tending to zero and is known as the DuFort-Frankel method. The idea is to use symmetric finite-difference relations to approximate differential equations in partial derivatives. The finite-difference grid method constructed as analog of the DuFort-Frankel method has been used (30,31). To make the transition to the finite differences with new variables $l = \varepsilon_0 S$, $\tau = \varepsilon_0 t/2h$ (ε_0 is the scaling enrichment coefficient defined for the majority of separation methods as the enrichment coefficient per unit mass difference) the whole region of integrating is covered by the regular orthogonal grid: $1 = k\Delta(k = 0, 1, \ldots, K)$, $\tau = n\Delta\tau(n = 1, 2, \ldots)$, where Δ is the integration step by the spatial variables, $\Delta\tau$ is the integration step by time, and k is the number of spatial points in the calculation grid.

The symmetric finite-difference relations approximating derivatives in the points of the grid are written as follows:

$$\frac{\partial C_i}{\partial l} = \frac{C^n_{i,k+1} - C^n_{i,k-1}}{2\Delta}, \tag{124}$$

$$\frac{\partial^2 C_i}{\partial l^2} = \frac{C^n_{i,k+1} - (C^{n+1}_{i,k} - C^{n-1}_{i,k}) + C^n_{i,k-1}}{\Delta^2}, \tag{125}$$

$$\frac{\partial C}{\partial \tau} = \frac{C^{n+1}_{i,k+1} - C^{n-1}_{i,k}}{2\Delta\tau} \tag{126}$$

In the points where the feed flow and the flow $L(S)$ are interrupted as well as at the cascade ends relations (124) are not valid. Here one needs to use the one-side decompositions with sufficient level of accuracy. The calculations in practice have shown that the following relations (obtained from superposition of functions C_i decomposed in the Taylor series in the points with indices $k-1$, $k-2$, $k-3$, and $k+1$, $k+2$, $k+3$, respectively) for one-side derivatives give satisfactory results:

$$\frac{\partial C_i^+}{\partial l} = \frac{2C^n_{i,k+3} - 9C^n_{i,k+2} + 18C^n_{i,k+1} - 11C^n_{i,k}}{6\Delta}, \tag{127}$$

$$\frac{\partial C_i^-}{\partial l} = \frac{-2C^n_{i,k-3} + 2C^n_{i,k-2} - 18C^n_{i,k-1} + 11C^n_{i,k}}{6\Delta}. \tag{128}$$

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As it follows from expressions (125) and (126), the difference scheme requires additional calculations to determine the value of functions by the second time layer. These calculations have to be carried out with a higher accuracy than that required for the rest region. However, when using expressions (123) as initial conditions, the second time layer will consist of the initial data except 1) the feed-point, 2) the interruption-points of flow $L(S)$, and 3) the cascade ends. The computation experiments were carried out for the squared-off cascade after determination of the stability region of this method. The results obtained demonstrated that the main operating parameters of the cascade weakly influence the stability, i.e., the technique is reliable and effective within a wide range of problems under study. The integration step $\Delta\tau$ is big enough, and its value is of the same order as Δ. A comparison of this method with the explicit one-step method having integration step $\Delta\tau < (\Delta)^2$ makes clear the advantages of the method for $\Delta \ll 1$.

The numerical calculations have shown that both methods are stable within a wide range of the spatial and time steps of integration. Some results of numerical experiments for a simple cascade separating tungsten isotopes ($M_i = 180, 182, 183, 184, 186$) are presented below. Concentrations in the feed flow equaled to isotope concentrations in the natural mixture: ($C_{iF} = 0.0015, 0.2646, 0.1440, 0.3060, 0.2840$) in the order of growing mass numbers. The cascade parameters were chosen as follows:

$$2W/\varepsilon_0 L = 1.538; \quad 2P/\varepsilon_0 L = 1.004; \quad l_F = 7.5; \quad l_P = 8.4.$$

The calculations were carried out with steps values $\Delta = 0.075$ and $\delta\tau = 0.0147$. The stages were numbered in the direction of enrichment of the light species of isotope mass spectrum. The study was performed for the following cases: 1) open cascade (with product and waste flows being continuously switched on, $W \neq 0$, $P \neq 0$); 2) closed cascade ($W = 0$, $P = 0$); and 3) cascade with periodically switched internal flows (at time moments $\neq 0$).

Figure 13 shows the spatial-time distributions of isotope concentrations with mass numbers 184 and 186 for the open (solid lines) and closed (dashed lines) cascades. As seen, at the steady state for regime corresponding to $P = 0$, components with extreme mass numbers reach the maximum enrichment, whereas the intermediate components achieve the maximum enrichment in the intermediate points of the cascade. The numerical calculations have shown that the cascade has some inertia to the external perturbations in the initial period of time. For example, the spatial-time distributions of concentrations for open and closed cascades in the initial period have similar characteristics (see curves 1 in Fig. 11). After achieving the steady state, the concentration profiles for these two regimes substantially differ from each other.

Figure 14 demonstrates the concentration-time histories for isotopes with mass numbers 184 and 186 (dashed curves) in the point of withdrawal of waste flow. As seen, although curves 1, 2 and 1′, 2′ corresponding to different modes of

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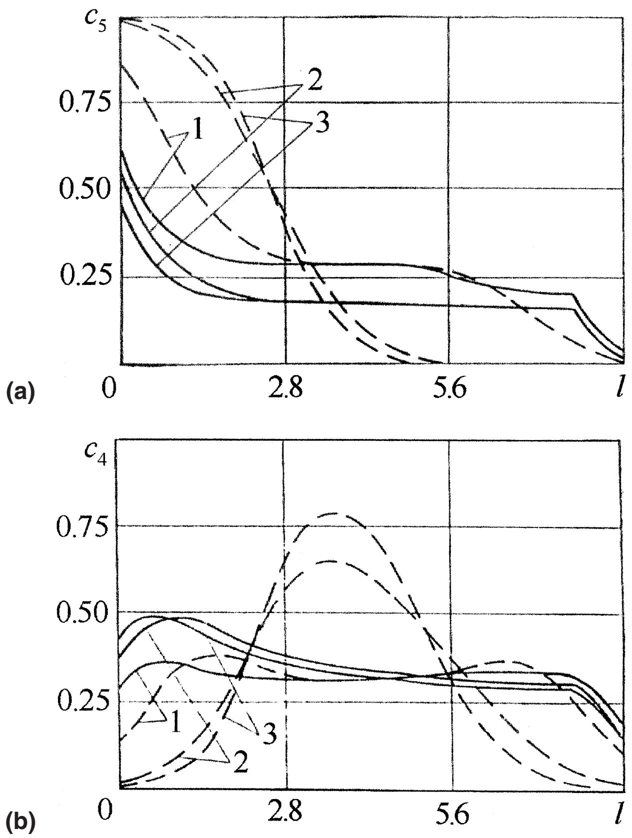

Figure 13. Spatial-time distribution of components with mass numbers 186 (a) and 184 (b) for open cascade (solid curves); closed cascade (dashed curves) at dimensionless time $\tau = 1$ (1); 3 (2), and ∞ (3).

cascade operation do not essentially differ from each other until $\tau = 0.125$. It is also interesting to note that in the initial part of the transient period, the maximum rate of substance accumulation and the maximum gradients of concentrations for components with extreme mass numbers are observed. Besides, a definite quantity of isotopes leaves cascade in the product and the waste flows if the process is carried out with external flows. The result of combination of these two processes by taking into account the competitive influence of intermediate mixture components (approximately equal concentrations of components is the main feature of this mixture) is also illustrated by Fig. 14. Here, clearly pronounced maxima and minima are seen on the curves having an asymptotic feature, which develops only

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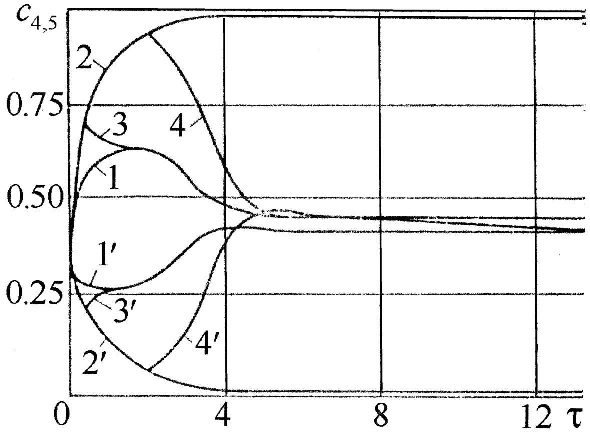

Figure 14. Component concentrations with masses 186 and 184 (dashed numbers) plotted vs. dimensionless time τ at points of carrying out P_1: 1, 1′ for open cascade; 2, 2′ for closed cascade, and closed cascade with constantly switched on P_2; 3, 3′ and 4, 4′ for switching on P_1 at $\tau = 0.5$ and 2.0.

after a certain period of time. Note also that the steady distribution of concentrations in cascades does not depend on its initial profile. The features of this profile define only the duration of the transient period.

The results of calculations on separation of binary mixture in the cascades of constant width (40) have shown that the most expedient way to maximally shorten the transient period is to operate with constantly switched on waste flow. The product flow should be switched on only after reaching the calculated value of concentration. This conclusion appeared to be non-valid for separation of multicomponent isotope mixtures. The calculations have shown that the deviation of concentrations in the point of withdrawal of waste flow weakly depends on the fact of whether the product flow exists or not. However, switching on the product flow in the time moments $t > 0$ may increase the duration of transient process (see curves 4 and 4′ in Fig. 14). This can be explained by distribution of concentrations of intermediate components along the length of the cascade, which have in fact different profiles for the open and closed cascades (see Fig. 13b).

The methods described can be effectively used in the study of non-steady mass transfer processes in cascades to minimize the duration of the starting period. In addition, several original methods for operation during the starting period including those based on switching off the product and the waste flows at defined moments of cascade operation and "operational procedure" (operation with accumulation of the target isotope within the cascade volume) are described by Laguntsov et al. (32).

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FUTURE WORK

In conclusion, we would like to distinguish the following problems related to separation of multicomponent mixtures, which in our opinion are of great theoretical and practical importance:

development of theory for separation of multicomponent molecular mixtures accompanied by interphase isotope exchange (33,34);
development of theory for calculation of multicomponent non-symmetrical cascades (14) and "cascades with losses";
investigation of cascades with a given number of stages to find a non-steady regime of separation that can provide a higher concentration of the target isotope in the product flow due to variation of either internal or external parameters of the cascade such as the product, the waste, and the feed flows;
development of new calculation methods for optimization of cascades by different criteria such as the total flow, the cascade length, and some others.

Finally, we would like to touch upon the problem of determination of the value function and separative power of cascades for the multicomponent mixtures. It is known that the notions of the value function and the separative power in the case of binary system is based on the non-mixing concept and on the theory of separation in ideal cascade (41). For practice, these notions permit the minimum number of separating elements required for carrying out a desired separation to be estimated. The main difficulty in introducing the value function and the separative power for multicomponent system is connected with inapplicability in this case of non-mixing condition (see above).

By using the MARC theory in the case of infinitesimal separation effect de la Garza et al. (1) derived the multicomponent analogs for "value function" and "separative power". Note that these analogs do not allow in general case the minimum number of separating elements to be calculated because unlike a bi-component ideal cascade, MARC does not minimize the total cascade flow. An attempt to obtain a common expression for the value function based on different approaches has been made (42–46). The main result obtained was as follows: the value function can be generated as the set of terms $(C_j - C_i)\ln C_i/C_j$. In the case of binary mixture, it reduces to the classic expression $V(C) = (2C - 1)\ln C/(1 - C)$. In the context of the above reasoning, this function is inconvenient for practical usage, because it is evident now that this problem also requires further study.

It is necessary to note that several recent studies devoted to the further development of various techniques for calculation of squared-off cascades have been published (47–50). Also, in practice of stable isotope production by a gas centrifuge cascade the problem arising in calculation of a cascade with losses has been solved in our recent publication (51). The efficiency criterion for separation of multicomponent isotope mixtures on the basis of the so-called *R* cascade has been introduced by Wood et al. (52).

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NOMENCLATURE

n	is number of mixture components
C_i	is mole fraction of component i in mixture
L	is stream of isotope mixture entering cascade stage [mol/s]
S	is index number of stage in cascade
N	is total number of cascade stages
f	is number of stage which feed stream enters in
S_P	is total number of stages in enriching part of cascade
S_W	is total number of stages in stripping part of cascade
F	is rate of feed flow entering cascade [mol/s]
P	is product flow rate being withdrawn from cascade [mol/s]
W	is waste flow rate leaving cascade [mol/s]
M_i	is molecular mass of component i
R_{ij}	is abundance ratio of pair of components i and j
G_i	is flux of component i entering stage [mol/s]
G_i^+, G_i^-	are fluxes of component i leaving stage [mol/s]
$H(S)$	is retardation (delay) of flow substance at stage S
J_i	is transfer for component i [mol/s]
T	is transfer for isotope mixture [mol/s]
t	is time [s]

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Greek

α_{ij}	is stage separation factor for pair of components i and j
α_0	is overall separation factor per unit mass difference
ε_{ij}	is stage enrichment coefficient for pair of components i and j
δ	is relative error
ν	is number of iterations
θ	is cut

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