A METHOD FOR THE NUMERICAL INVESTIGATION OF NONSTATIONARY PROCESSES IN MULTISECTIONAL INSTALLATIONS FOR SEPARATING MULTICOMPONENT ISOTOPE MIXTURES

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UDC 621.039.31

On the basis of the Dufort–Frankel method for the integration of differential equations of parabolic type, we develop a method for the numerical calculation of nonstationary processes.

It is known that analytic solutions describing nonstationary processes in cascades for the separation of isotope mixtures have been constructed only for a limited number of problems. In the first place, these deal essentially with cascades for the separation of binary mixtures. In the second place, all known solutions relate either to cascade operation regimes without output or to a limited range of variation of the concentrations of the isotope components, and they have been obtained only for cascades consisting of one section which has a continuous profile.

The most promising method for the investigation of nonstationary processes appear to be methods for the numerical solution of the equations describing these processes [1-6]. However, even in this case the most successful calculation methods from the standpoint of universality and efficiency of use on modern computers have been developed, in the main, solely for the case of separation of binary isotope mixtures [4]. Attempts to carry these methods over to the calculation of nonstationary processes in cascades for the separation of multicomponent mixtures lead chiefly to results which are not universal and require large machine memories and long computation times.

We describe below a method for the calculation of nonstationary processes in cascade installations designed mainly for the separation of multicomponent isotope mixtures.

The fundamental system of equations describing the space–time process of accumulating an m-component mixture of isotopes in a cascade with low enrichment per separating stage can be represented in the form

$$\Gamma(s)\frac{\partial c_i(s,\ t)}{\partial t} = -\frac{\partial}{\partial s}J_i(s,\ t),\ i = 1,\ \ldots,\ m-1,\ \sum_{j=1}^{m} c_j(s,\ t)] = 1. \tag{1}$$

For most separation methods, it may be assumed with satisfactory accuracy that the residence time of the mixture in stage $\Gamma(s)$ is proportional to the interstage flow $L(s)$ entering this stage, i.e.,

$$\Gamma(s) = hL(s), \tag{2}$$

where h is proportionality constant which has the dimensions of time.

For simplicity, we shall confine our attention to an ordinary rectangularly sectioned cascade, i.e., a cascade which has one input flow F and two output flows P_1 and P_2 , which are removed at the ends of the cascade. The flow L(s) is regarded within the limits of each section as a constant which is independent of the number of stages in that section.

The transfer of the i-th component in any section of the cascade in the direction of the maximum enrichment in the lightest isotope can be written in the form

$$J_i(s,\ t) = -\frac{L}{2}\,\frac{\partial c_i(s,\ t)}{\partial s} + \frac{L}{2}\,c_i(s,\ t)\sum_{j=1}^{m}\varepsilon_{ij}c_j(s,\ t) + Tc_j(s,\ t), \tag{3}$$

where $\epsilon_{ij} = \epsilon_0(M_j - M_i)$ is the relative enrichment factor for the isotope pair i–j in the separation stage (the numbering of the isotopes is in increasing order of their mass numbers).

The transferred amount T of the mixture being separated is defined as P_2 for the stages of the cascade which are situated in the enriching portion (with respect to the lightest isotopes), and $P_2 - F = -P_1$ for the stripping portion.

Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 43, No. 3, pp. 456-462, September, 1982. Original article submitted June 17, 1981.

0022-0841/82/4303-1027$07.50

Equation (1), with the new variables $l=\varepsilon_0 s$, $\tau=t\varepsilon_0^2/2h$, and taking account of (2) and (3), gives us

$$\frac{\partial c_i}{\partial \tau}=\frac{\partial^2 c_i}{\partial l^2}-\frac{\partial c_i}{\partial l}\sum_{j=1}^{m}(M_j-M_i)c_j-c_i\sum_{j=1}^{m}(M_j-M_i)\frac{\partial c_j}{\partial l}-\frac{2T}{\varepsilon_0 L}\frac{\partial c_i}{\partial l}. \tag{4}$$

The condition for the joining of two adjacent sections with numbers ν and $\nu+1$ can be obtained from the law of conservation of matter, which in our case, taking account of the equality of the concentrations at the boundaries of these sections, can be written as conservation of the transferred mass of each component:

$$-\frac{L^{\nu}}{2}\frac{\partial c_i^-}{\partial l}+\frac{L^{\nu}}{2}c_i\sum_{j=1}^{m}(M_j-M_i)c_j+\frac{T^{\nu}}{\varepsilon_0}c_i=-\frac{L^{\nu+1}}{2}\frac{\partial c_i^+}{\partial l}+\frac{L^{\nu+1}}{2}c_i\sum_{j=1}^{m}(M_j-M_i)c_j+\frac{T^{\nu+1}}{\varepsilon_0}c_i,$$

where L^{ν} and $L^{\nu+1}$ are values of the flow L in sections ν and $\nu+1$. Dividing this equation by c_i and subtracting from it the analogous condition for the k-th component, we find, after replacing the subscript k with the subscript j, that

$$L^{\nu}\left(\frac{\partial}{\partial l}\ln\frac{c_i}{c_j}\right)^{-}-L^{\nu+1}\left(\frac{\partial}{\partial l}\ln\frac{c_i}{c_j}\right)^{+}=(M_j-M_i)(L^{\nu}-L^{\nu+1}), \tag{5}$$

where the + and − signs denote the right and left derivatives at the points under consideration. At the input point this condition has the form

$$L^{\nu}\frac{\partial c_i^-}{\partial l}-L^{\nu+1}\frac{\partial c_i^+}{\partial l}-(L^{\nu}-L^{\nu+1})c_i\sum_{j=1}^{m}(M_j-M_i)c_j=\frac{2F}{\varepsilon_0}(c_i^F-c_i). \tag{6}$$

At the ends of the cascade we have the equations

$$J_i(0,\tau)=-P_1 c_i(0,\tau), \tag{7}$$

$$J_i(l_p,\tau)=P_2 c_i(l_p,\tau), \tag{8}$$

where $l_p=\epsilon_0 s_p$; s_p is the total number of stages in the cascade.

Taking account of (3), after some transformations analogous to those used in deriving (5), we can represent these conditions in the form

$$\left.\frac{\partial}{\partial l}\ln\frac{c_i}{c_j}\right|_{l=0}=(M_j-M_i), \tag{9}$$

$$\left.\frac{\partial}{\partial l}\ln\frac{c_i}{c_j}\right|_{l=l_p}=(M_j-M_i). \tag{10}$$

In order to solve the resulting boundary-value problem, we must specify the initial distribution of the concentrations of all the components along the cascade; in the simplest case, where the cascade is initially filled with a homogeneous input mixture which has concentration c_i^F, this can be written as

$$c_i(l,0)=c_i^F. \tag{11}$$

Thus, the (m − 1) equations of the form (4), with the boundary conditions (5), (6), (9), and (10), the initial condition (11), and the identity $\sum_{j=1}^{m}c_j(l,\tau)\equiv 1$, completely defines the system for finding any concentration of an m-component mixture at each point of the cascade at any instant of time.

The expressions obtained here can easily be generalized to the cases of cascades of any profile (with any type of distribution of the flow L(s)) and cascades which have several input flows and output flows. In the case of the calculation of cascades whose operation is based on the use of invertible separation methods, in the boundary conditions we must take account of the accumulation of isotopes in the phase-reversal vessels.

To solve such problems, we can use various network-type finite-difference methods. However, all of these have been investigated fairly thoroughly only in the case of linear equations [7]. At the same time, the questions of the stability and convergence of these methods in the case of nonlinear boundary-value problems are, in practice, open problems.

Among all the methods for the integration of these equations, explicit numerical schemes are of the greatest interest, since they enable us to carry out the calculations efficiently with a computer which has a small operating memory. The transition to implicit schemes requires at each calculated time layer the solution of large systems of nonlinear algebraic equations. If we wish to obtain sufficient accuracy in the results, this may require cumbersome computing procedures. At the same time, most of the explicit schemes have the serious defect of becoming unstable outside of a limited range of variation of the spatial and time steps of the integration.

However, as is known [8], for the solution of linear boundary-value problems of parabolic type there exists an explicit method which is absolutely stable, i.e., is stable regardless of how the magnitude of the integration steps approaches zero. This method is known in the literature as the Dufort–Frankel method.

The idea of the method is that in symmetric finite-difference relations approximating a partial differential equation, the central term in the second space derivative is averaged over the two adjacent time layers. The only defect of this method is that its error of approximation Ψ with respect to the desired value of x is of the order of $\Psi = -\frac{\Delta\tau^2}{\Delta^2}\ddot{x} + O\cdot(\Delta\tau^2+\Delta^2)$, where $\Delta\tau$ is the integration step with respect to time and Δ is step with respect to space, as a result of which, when $\Delta\tau$ is large, there will be undesirable oscillations in the solution.

In the present study we use a finite-difference network method constructed by analogy with the Dufort–Frankel method. For the transition to finite differences, we cover the entire region of integration with a uniform orthogonal network: $l = k\Delta$ $(k = 0, 1, \ldots, N)$, $\tau = n\Delta\tau$ $(n = 1, 2, \ldots)$. The symmetric finite-difference relations approximating the derivatives at the nodes of the network have the form [8]:

$$\frac{\partial c}{\partial l} = \frac{c^n_{k+1} - c^n_{k-1}}{2\Delta}, \tag{12}$$

$$\frac{\partial^2 c}{\partial l^2} = \frac{c^n_{k+1} - (c^{n+1}_k + c^{n-1}_k) + c^n_{k-1}}{\Delta^2}, \tag{13}$$

$$\frac{\partial c}{\partial \tau} = \frac{c^{n+1}_k - c^{n-1}_k}{2\Delta\tau}, \tag{14}$$

where n is the number of the time layer and k is the number of the space node in the calculation network.

At the junction points of the sections and at the ends of the cascade, we cannot use relations of the form (12). Here we must use one-sided expansions with a sufficiently high order of accuracy. Practical calculations have shown that satisfactory results are obtained by using the following relations for one-sided derivatives:

$$\frac{\partial c^-}{\partial l} = \frac{-2c^n_{k-3} + 9c^n_{k-2} - 18c^n_{k-1} + 11c^n_k}{6\Delta}, \tag{15}$$

$$\frac{\partial c^+}{\partial l} = \frac{2c^n_{k+3} - 9c^n_{k+2} + 18c^n_{k+1} - 11c^n_k}{6\Delta}, \tag{16}$$

which are obtained by superposition of the expansions of the function c in Taylor's series at the points with subscripts k − 1, k − 2, k − 3 and k + 1, k + 2, k + 3, respectively.

It follows from the expressions (13), (14) that the difference scheme requires us to calculate, in addition, the values of the function at the second time layer. These calculations must be carried out with greater accuracy than is required for the solution in the remaining region. However, if for our initial conditions we take conditions of the form (11), then the second time layer consists of initial data, except for the points of introduction of the external flows, the points of junction between the sections, and the ends of the cascade, which are calculated by using the expansions (15) and (16).

The above-described method was used for investigating transition processes in rectangularly sectioned cascades. The numerical investigations show that the process of calculation remains stable over a fairly wide range of variation of the spatial and time steps of the integration. We give below some results of the numerical investigation of a cascade with a rectangular profile, using as an example the separation of a mixture of tungsten isotopes with mass numbers M_i = 180, 182, 183, 184, and 186. The concentrations of the components in the input flow were assumed to be equal to the concentrations of the isotopes in the natural mixture: c_i^F = 0.0014, 0.2646, 0.1440, 0.3060, and 0.2840. The parameters of the cascade were taken to be the following: $2P_1/\varepsilon_0 L = 1.538$; $2P_2/\varepsilon_0 L = 1.004$; $l_F = 7.5$; $l_p = 8.4$. The calculations were carried out with steps Δ = 0.075 and $\Delta\tau$ = 0.0147. The stages were counted in the direction of enrichment of the light end of the isotope mass spectrum. The investigations were carried out for the following operating regimes: an open cascade, i.e., with the output flows connected at all times; and a closed cascade, in which the output flows were shut off (total recycle) at some nonzero instants of time.

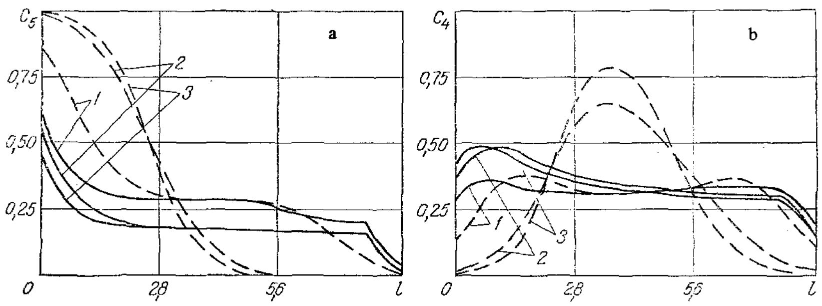

Fig. 1. Space–time distribution of the concentrations of components with mass numbers 186 (a) and 184 (b): solid curves represent the case of an open cascade; dashed curves represent the case of a closed cascade; dimensionless time τ: 1) 1; 2) 3; 3) ∞.

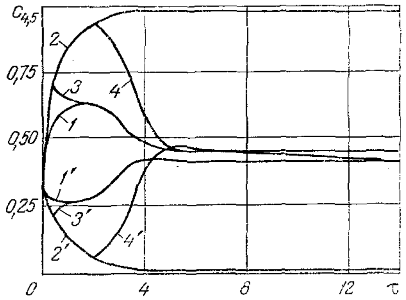

Fig. 2. Variation of the concentrations of components with mass numbers 186 and 184 (curves with primed numbers) as functions of dimensionless time τ at the point where the output P_1 is removed: 1, 1') open cascade; 2, 2') closed cascade and cascade with permanently connected output P_2; 3, 3' and 4, 4') output P_1 connected at $\tau = 0.5$ and $\tau = 2.0$, respectively.

Figure 1 shows the space–time distributions of the concentrations of isotopes with mass numbers 186 and 184 for an open cascade (solid curves) and a closed cascade (dashed curves). This gives a good illustration of the fact [9] that in the stationary state in total-recycle operation, the components with highest and lowest mass numbers attain maximum enrichment at the ends of the cascade, and those with intermediate mass numbers at intermediate points of the cascade.

The numerical calculations show that during the initial period the cascade has a definite inertia to external perturbations. Thus, for example, the space–time distribution of the concentrations for open and closed cascades in the initial period are similar (curves 1 in Fig. 1), although in the stationary state the profiles of the concentrations for these two regimes are radically different. In Fig. 2, which shows how the concentrations of the isotopes with mass numbers 186 and 184 (the curves with primed numbers) at the point of the output P_1 vary as functions of time, curves 1, 2, and 1', 2' are practically identical up to $\tau = 0.125$, although they represent different regimes of cascade operation.

It is also interesting to note that for the initial stage of the transition period, at the ends of the cascade we observe maximum rates of accumulation and maximum gradients of the concentrations of the components with highest and lowest mass numbers, whereas when the process is carried out with internal output, we find removal of a certain amount of isotopes. The result of combining these two processes, taking account of the competing influences of the other components of the mixture (the mixture is characterized by the fact that most of the components in it are present in approximately the same quantities) is clearly illustrated in Fig. 2. Here we observe clearly expressed maxima and minima in the curves, and asymptotic behavior is observed only after some time has elapsed.

We should also point out that the numerical calculations confirmed the natural fact that the stationary distribution of concentrations in cascades does not depend on their initial profile. The shape of this profile is essential only for determining the duration of the transition period.

In the separation of binary mixtures in cascades with a rectangular profile, it was noted [4] that for maximum reduction of the transition period it is most advisable to use the method of conducting the process with constantly connected waste and the product must be connected after the calculated value of the concentration is reached at the product point.

It is found that in the separation of multicomponent mixtures a conclusion analogous to this is not always valid. Calculations show that when there is no output P_1, the variation of the concentrations at the point of application of P_1 depends only slightly on whether or not there is an output P_2. However, turning P_1 on at different nonzero instants of time does not reduce the transition period and may even, on the contrary, increase it (see curves 4 and 4′ in Fig. 2). This may be explained by the fact that the distributions of the concentrations of the intermediate components along the cascade are of radically different form for the cases of closed and open cascades (see Fig. 1b).

The time required for calculating these examples by the above-described method on an ES-1033 computer is 10-15 min. The error in the solutions in relation to the known stationary distributions was no more than 1.5%.

All of the above-described results were obtained for a constant integration step with respect to time. However, here there is an additional possibility for reducing the amount of calculation work and the time needed for it by gradually increasing $\Delta\tau$, which is permissible, since the solution is asymptotic.

In conclusion, it should be noted that the method proposed in this study makes it possible to analyze fairly thoroughly the transition processes taking place in multisectional installations for the separation of multicomponent isotope mixtures and provides an opportunity for conducting investigations on the choice of the optimal strategy for carrying out the process in the cascade.

NOTATION

$c_i(s, t)$, concentration of the i-th component at the s-th stage at time t; c_i^F, concentration of the i-th component in the input flow; $j_i(s, t)$, amount of the i-th component transferred in the s-th stage at time t; T, amount of mixture transported in the cascade; F, input flow; P_1, P_2, output flows; ϵ_0, enrichment factor for components whose mass numbers differ by one; M_i, mass number of the i-th component; l_F, coordinate of the point of application of the input; l_p, total length of the cascade.

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