FEATURES OF NONSTATIONARY MASS TRANSFER IN CASCADES FOR THE SEPARATION OF BINARY MIXTURES OF ISOTOPES

N. I. Laguntsov, E. V. Levin, and G. A. Sulaberidze

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We conducted a numerical investigation of the methods of organizing a process during the transitional period in the separation of binary mixtures of isotopes in cascades with a stripping section.

At the present time a number of problems relating to the choice of the most effective methods of organizing the separation of isotopes during the transition period remain unsolved. In what follows, we give the results of numerical investigations illustrating the features of nonstationary mass transfer in cascades for the separation of binary mixtures and also give some recommendations concerning methods for carrying out the transition processes.

The equation of nonstationary mass transfer in a multistage cascade when the independent variables used are $\tau = \varepsilon^2 t/h$ and $\ell = \varepsilon s$ will, irrespective of the specific separation method, have the form [1, 2]

$$\frac{\partial c}{\partial \tau} = -\frac{1}{\varepsilon L}\frac{\partial J}{\partial l}, \tag{1}$$

where $J = -\varepsilon L(\partial c/\partial \ell) + \varepsilon Lc(1 - c) + Tc$ is the transfer of the mixture being separated, in the direction from the waste toward the cascade product. In the enrichment sections the transfer T is equal to the extracted amount P, and in the stripping sections it is equal to the amount of waste W with a minus sign. The boundary conditions for the ends of the cascade, the flow input points, and the junction points of adjacent sections, obtained from the conditions of conservation of transfer of a component with due regard to the external flows of materials have the form given in [2], and as the initial condition we may use the condition that during the initial instants of time the cascade is filled with a uniform mixture of isotopes, for example, one which has the composition of the input flow.

The solution of the boundary-value problem was carried out by numerical methods described in [2, 3]. An analysis of the transition processes was carried out for cascades with different interstage flow profiles $L(\ell)$, depending on the mixture transfer value T in the cases of greatest practical interest, when the concentration of the input mixture is in the range from $c_F \ll 1$ to $c_F = 0.5$.

Nonstationary mass transfer takes the most complicated form in cascades in which there is not only enrichment but also stripping of the mixture to be separated. A significant influence is exerted on the nature and duration of the transition process at the ends of the cascade by the value of the variation of concentration at the input point, which in turn depends on the relation between the volumes and lengths of the stripping and enrichment parts of the cascade. In the stripping portion of the cascade the transfer to all stages (except the last one) is equal at the initial instant of time to

$$J_0 = \varepsilon L c_F (1 - c_F) - W c_F,$$

and at the waste point it takes on the value $J_W = -Wc_F$. Accordingly, there is a positive variation of the transfer, $\partial J/\partial \ell > 0$, leading to a decrease in the concentration as time increases. When the stripping portion of the cascade has a small volume in comparison with the enrichment portion, the disturbances in concentration which are caused by the variation of the transfer will quickly be transmitted to the input point, which leads to a decrease in the concentration c_f. As the volume of the stripping portion of the cascade increases (as the required degree of stripping increases), the decrease in the concentration c_f is compen-

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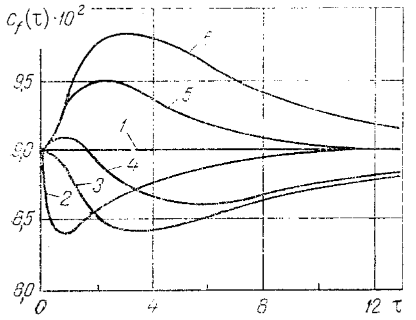

Fig. 1

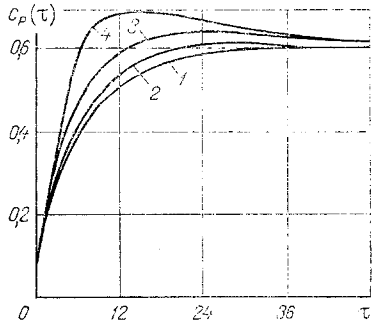

Fig. 2

Fig. 1. Concentration at the input point as a function of time for different values of the separation coefficient q_W ($c_F = 0.09$, $q_P = 15$): 1) $q_W = 1$; 2) 1.2; 3) 1.6; 4) 2.4; 5) 4.5; 6) 9.8.

Fig. 2. Concentration at the product end of the cascade as a function of time, using the example of the separation of neon isotopes ($q_P = 15$, $q_W = 33$), for different values of the parameter α: 1) $\alpha = 0.1$; 2) 0; 3) -0.1; 4) -0.17 (ideal cascade).

sated by the influence of the processes taking place in the enrichment portion, and its time-average value increases and may even become greater than the design value.

Figure 1 shows the corresponding $c_f(\tau)$ curves for cascades with different degrees of separation q_W and identical values of q_P. It can be seen from the figure that as q_W increases the transition to increasing concentrations is complicated in form and initially takes place only in the first phase of the transition period. In tapered cascades the nonstationary variation of the concentration c_f has the same character as in cascades of constant width, but this character is more clearly marked for shorter duration.

The values of the increment in concentration at the input point are related to the relative stationary accumulation

$$\alpha = \frac{M_\infty - M_0}{M_0}, \tag{2}$$

where M_0 and M_∞ are the initial and final amounts of the enriched component in the cascade, which is dependent to a certain degree on the ratio of the volumes of the enrichment and stripping sections. The accumulation may be either negative or positive. In particular, for ideal cascades it is a simple matter to obtain an expression for the accumulation, which has the form

$$\alpha = \frac{(q_W - 1)(q_P + 1)\ln q_P - (q_P - 1)(q_W + 1)\ln q_W}{(q_W - 1)(R_F q_P - 1)\ln q_P - (q_P - 1)(R_F - q_W)\ln q_W}. \tag{3}$$

An analysis of formula (3) shows that the transition to $\alpha < 0$ takes place if the condition $q_W > q_P$ is satisfied. As a result of the investigations, it was found that a sufficient condition for an increase in the concentration c_f during the initial instants of time for cascades of any profile is the condition $\alpha \leqq 0$.

An increase in the concentration c_f is tantamount to an increase during the transition period in the effective length of the enrichment portion of the cascade. Simultaneously with this, in a number of cases the nonstationary value of the accumulation has a maximum, as does the quantity $c_f(\tau)$. As a result, the rate of increase of the concentration at the outlet end of the cascade increases. In Fig. 2 we show the characteristic graphs of $c_P(\tau)$ for cascades with different values of α, which were calculated for the same stationary values of the concentration c_P, c_W, and c_F. It can be seen from the figure that as the value of the relative accumulation decreases there is a reduction in the period of time it can take to reach the design concentration c_P, and for $\alpha < 0$, when at the initial instant of time there is a larger amount of the desired component in the cascade than in the stationary state, a maximum appears

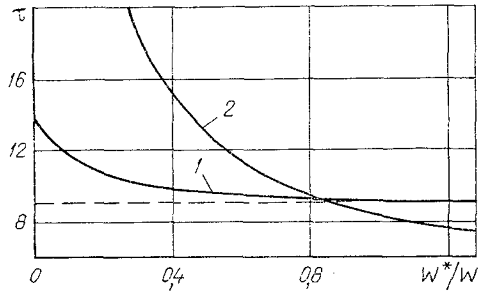

Fig. 3. Time required to reach the design concentration c_P (curve 1) and the design accumulation (curve 2) as functions of the ratio W*/W for a cascade operating with the product disconnected. The cascade parameters are: $\varepsilon L/P = 7.5$, $W/P = 6.379$, $\ell_0 = 10.66$, $\ell_P = 4.41$, $c_F = 0.09$.

in the function $c_P(\tau)$. For a sufficiently large value of the initial excess of the component (for example, when $\alpha < -0.15$ for an input concentration $c_F \ll 1$), the time-average value of the concentration $c_P(\tau)$ is found to be no lower than the design value. In these cases the extraction of the product may be continued during the entire time of operation of the cascade. It should be noted that when a constant concentration is maintained at one end of the cascade, the relative accumulation is always positive ($\alpha > 0$). Therefore, a cascade operating regime with continuous extraction of product does not permit overenrichment and is less efficient than a regime without extraction of product [1].

The most widely used regime for carrying out the transition period is the one in which the extraction of product is disconnected [1, 4, 5]. During the initial instants of time there is always a decrease in the concentration $c_W(\tau)$, as a result of which there is an increase in the rate of accumulation of the desired component, and the concentration c_P can be obtained in a much shorter time than when the cascade is operated with extraction of product.

Connecting the product flow when the design concentration is approached may have different effects on the laws governing the further variation of the concentration at the product end, depending on a number of parameters. In ideal and near-ideal tapered rectangular-sectioned cascades, owing to the relatively high variations in the transfer J, there is a temporary decrease in the concentration $c_P(\tau)$, which was pointed out in [2]. The insufficiency of accumulation in comparison with the design value after the extraction of product is connected will also lead to a decrease in the concentration, and this takes place independently of the profile of the cascade and lasts a longer time. As the investigations showed, if there is a 10% or greater shortfall in the concentration of the desired component, the length of the transition period will be no less than in the case of operation with the product connected at all times. In those cases in which the accumulation is close to the design value, the concentration changes only slightly after the product is connected (this was pointed out in [2, 4, 6]) and, lastly, when there is an excess of the desired component, just as in the case of operation with constant product, the function $c_P(\tau)$ will have a maximum, and in tapered cascades the maximum in the concentration may be preceded by the formation of a minimum.

An important role in the operation of a cascade with disconnected product is played by the value of the transfer of material in the stripping sections, due to the waste flow W*. It follows from Fig. 3 that as W* increases the rate of accumulation will increase faster than the rate of variation in the concentration, and for certain values of W* the design accumulation will be reached sooner than the design concentration. In those cases in which the value of α approaches zero or is negative, this feature is realized over the entire range of variation of W*.

As the waste value increases, the time required for reaching the design concentration decreases, approaching a limit (the dashed curve in Fig. 3). The existence of this limit is due to the fact that the input flow in the cascade increases, and this increase causes a decrease in the deviation of the "zero" point (at which the concentration value coincides with the concentration of the input mixture) with respect to the input point. In these cases the processes of transfer of the components in the enrichment portion approach in character the processes taking place in cascades fed from an infinite reservoir. In order to determine the transition characteristics at the end of the cascade in this case we may use solutions known from the literature. In particular, for sufficiently long cascades of constant width, good results are obtained with the limiting solution obtained for a cascade of infinite length [1]:

$$c_P(\tau) = \frac{1}{2}\left[1 + \operatorname{erf}\left(\frac{\sqrt{\tau}}{2}\right)\right] - \left(\frac{1}{2} - c_F\right)\exp\left[-c_F(1 - c_F)\tau\right]\left\{1 + \operatorname{erf}\left[\left(\frac{1}{2} - c_F\right)\sqrt{\tau}\right]\right\}. \quad (4)$$

The nature of the variation of the time required for reaching the concentration c^P as a function of the waste value W^* in the transition region, where the main variation in this quantity takes place, is determined by the mass-transfer processes that take place in a cascade operating in a closed regime (with zero waste and zero product). In such a regime the stationary concentration at the end of the cascade depends on the position of the "zero" point with respect to the coordinate $\ell = \ell_0 - \ln q^P$. For positive displacements of the "zero" point toward the product end of the cascade ($\Delta\ell > 0$) the design value of the concentration at that end cannot be attained. If we introduce a quantity characterizing this deviation as

$$\gamma = \exp(\Delta l), \tag{5}$$

then the condition for attaining the design concentration is the condition $\gamma \leqq 1$. In cascades of constant width, by virtue of the fact that in a closed regime the accumulation α is equal to zero, it is a simple matter to obtain an expression for γ which has the form

$$\gamma = R_P \frac{1 - \exp[l_0(c_F - 1)]}{\exp[c_F l_0] - 1}. \tag{6}$$

In most cases, when $\gamma \leqq 0.5$-0.6, the transition region of curve 1 in Fig. 3 is not large, and the limiting minimum time required for reaching the concentration c^P is realized for near-zero values of W^*. If at the same time the parameters of the cascade are such that the design relative accumulation α also does not exceed zero, then there is a possibility of using a closed regime in which there is no need of any expenditure for initial raw material to feed the cascade. Such a regime can be used for secondary startups of cascades, when the cascade at the initial instant of time is already filled with an enriched mixture of isotopes. In these cases, as can be deduced from formula (6), an increase in the concentration c^F leads to a substantial decrease in the parameter γ.

In conclusion we can formulate some recommendations concerning the methods of organizing the process of separation during transitional periods. If the initial filling of the cascade ensures a sufficient excess amount of the component being enriched ($\alpha < 0$), then as a result of the temporary superenrichment of the product flow it can be recommended that waste and product flows should last equal lengths of time, equal to the stationary design values, and the product can be accumulated for some time in order to obtain the required average concentration. In those cases in which the excess of this component is small and $\gamma \leqq 0.5$-0.6, it is effective to operate the cascade in the closed regime, making it possible to economize on the flow of the original raw material. In all other cases the process of separation during the transition period must be carried out without any product flow, and the waste flow should be made sufficiently large to make sure that the design accumulation is obtained not later than the design concentration. The minimum value of W^* should be selected either from the condition of economy of flow rate of the input mixture (the point of intersection of curves 1 and 2 in Fig. 3) or from the condition of minimum duration of the transition period (the transition of curve 1 to the limiting solution). In the example shown in Fig. 3, these conditions are satisfied almost simultaneously. The design product and waste values in both cases should be given after the concentration $c^P(\tau)$ reaches the value c^P.

NOTATION

c, nonstationary value of the concentration at an arbitrary point of the cascade, mole fraction; $c^f(\tau)$, $c^P(\tau)$, $c^W(\tau)$, nonstationary values of the concentration at the entry point of the input flow, at the product end of the cascade, and at the waste end of the cascade; $h = H(s)/L(s)$, residence time of the substance being separated in a stage of the cascade, sec; $H(s)$, inventory of the substance in a stage of the cascade, moles; $L(s)$, interstage flow of the cascade, moles/sec; ℓ_0, total length of the cascade; ℓ_P, length of the enrichment portion of the cascade; $R = c/(1 - c)$, relative concentrations; $q^P = R^P/R^F$, $q^W = R^F/R^W$, stationary degrees of separation in the enrichment and stripping portions of the cascade; s, number of the separating stage of the cascade; t, time, sec; ε, enrichment factor.

LITERATURE CITED

1. G. D. Rabinovich, R. Ya. Gurevich, and G. I. Bobrova, Thermodiffusive Separation of Liquid Mixtures [in Russian], Nauka i Tekhnika, Minsk (1971).

2. N. I. Laguntsov, E. V. Levin, and G. A. Sulaberidze, "Computer calculation of nonstationary processes in multistage cascades for the separation of two-component mixtures of isotopes," Inzh.-Fiz. Zh., 31, No. 3, 506-513 (1976).
3. E. V. Levin, N. I. Laguntsov, and G. A. Sulaberidze, "A method for the numerical investigation of nonstationary processes in multisectional installations for the separation of multicomponent mixtures of isotopes," Inzh.-Fiz. Zh., 43, No. 3, 456-463 (1982).
4. A. M. Rozen, Theory of Separation of Isotopes in Columns [in Russian], Atomizdat, Moscow (1960).
5. S. Villani (ed.), Enrichment of Uranium [Russian translation], Énergoatomizdat, Moscow (1983).
6. V. M. Vetsko, O. A. Devdariani, E. V. Levin, et al., "Simulation of transitional processes in cascade installations for the separation of multicomponent mixtures of stable isotopes," Isotopenpraxis, 18, No. 8, 288-293 (1982).

GENERALIZATION OF THE KINETICS OF CHEMICAL DISSOLUTION UNDER CONDITIONS OF GAS FORMATION

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A generalization has been made of the experimental data on the chemical dissolution of several nonferrous metals and carbonates in solutions of hydrochloric acid in the presence of forced motion of the liquid.

In a number of papers [1-5] it has been shown on the basis of experimental studies that a gaseous phase has a considerable effect on the kinetic relationships for mass transfer, intensifying or retarding the rate of dissolution depending on the particular conditions. These processes have been studied most fully under conditions corresponding to an external problem, i.e., when a solid object dissolves in a stagnant or moving liquid. The same cannot be said for the case of the internal problem, i.e., during the motion of the solvent inside hollow objects of tubular or rectangular cross section. At the same time, chemical dissolution during the motion of a reagent along channels and tubes is quite frequently encountered in processes of chemical milling, in the production of parts for waveguide channels, in the removal of salt incrustations in heat-exchange tubes, etc. This has prompted the authors to consider certain aspects of this problem in more detail on the basis of investigations carried out earlier [6-8].

During the flow of a reagent in a channel or tube a stream is formed whose hydrodynamics are greatly complicated (especially in the layers near the wall) as a result of the gaseous products which are liberated at the reaction surface. The gas bubbles, which are generated on the surface at which the phases interact, grow until some diameter is reached, and then under the influence of the hydrodynamic head of the liquid and the buoyancy forces they break away and are carried off in the core of the flow. The periodic processes of growth and separation leads to a partial disturbance of the diffusion layer and to the turbulization of this layer, a reduction in the resistance to mass transfer, and an intensification of the dissolution process in the case of reactions which are controlled by diffusion. The liquid velocity also shows the same qualitative effects on mass transfer. However, in quantitative terms, the effects of the action of the gas phase and of a forced flow are not the same and differ depending on the concentration. At small concentrations of the reagent the hydrodynamic effects caused by the reduction of the thickness of the diffusion boundary layer predominate. At relatively large concentrations, on the other hand, the effect of turbulization of the layer by the gas bubbles predominates. It is obvious that at some critical concentration the mass flow rate of the gas becomes so large that the rate of the heterogeneous phase interaction

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