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Calculational Methods for Determining the Distribution of Components in a Separation Cascade for Multicomponent Mixture

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ABSTRACT

The distribution of the mixture components in a cascade plays an important role in analyzing the performance of multicomponent isotope separation cascades. Two methods are proposed in this paper to calculate the distribution. The "iteration approach," in which a set of nonlinear steady-state equations are iteratively solved at each stage, has the advantage of high speed with high precision, but may not converge for complicated cascade structures. The "transient approach," which simulates the transient variations of the concentration distributions, is a generalized method which can be applied to any separation method, number of components, cascade structure, or number of feeds or products. The transient approach can not only determine the steady-state distribution, but can also examine the transient behavior. Some examples comparing the results of the two methods are given in the paper.

Key Words. Steady state; Transient; Centrifuge cascade; Multicomponent separation; Concentration distribution

INTRODUCTION

Determination of the component distribution in a multicomponent separation cascade is important for analyzing the separation process and optimiza-

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tion parameters. A common method used to determine the distribution is to solve a set of nonlinear equations for the cascade at steady-state by iteration. Although the calculational time used to obtain the solution is very short in some simple cascades, it is not easy to solve the equations if the cascade is complicated because it is difficult to determine the appropriate initial values that will lead to convergence. The "transient approach," which can avoid the shortcomings of the iteration approach, is a general method which has no restrictions on the number of components, the cascade structure, and the number of feeds or withdrawals. It simulates the transient process of concentration change. If the time step is small enough, stable solutions can always be found. The transient approach can be used in various separation processes such as chemical exchange, distillation, gaseous diffusion, thermal diffusion, and centrifuge, because these processes are governed by the same form of nonlinear partial differential-difference equations which have both differential and difference part in the equations. Another advantage of the transient approach is that it also predicts the transient behavior, which is important in the analysis of the properties of multicomponent separation.

Some authors (1, 2) have studied the distribution of components in a multicomponent cascade at steady-state, but no papers have been published using the iteration approach. The transient approach was used in some papers (3–5) to examine the transient behavior of minor isotopes when the separation factor is nearly unity. In this case the nonlinear partial differential-difference equations can be approximated as a set of partial differential equations. However, no articles were found analyzing the transient behavior in cases with large separation factors as in centrifuge cascades. The transient approach is used in this paper to analyze cascades with large separation factors. Some examples are given comparing the calculated results using the iteration approach and the transient approach. It is shown that the two methods give identical results at steady-state.

THE ITERATION APPROACH TO STEADY STATE

Steady-State Governing Equations for Gas Centrifuge Cascade

The analysis considers the separation of a multicomponent mixture with K components in a gas centrifuge cascade of N stages. The cascade is shown in Fig. 1.

The direction in which the lightest component is enriched is defined as the enriching direction; the opposite direction is the stripping direction. The governing equations at steady state in a cascade are (6)

$$\theta_n G_n - (1 - \theta_{n+1}) G_{n+1} = P_n^*; \qquad n = 1, 2, \ldots, N - 1 \qquad (1)$$

$$\theta_n G_n C'_{n,i} - (1 - \theta_{n+1}) G_{n+1} C''_{n+1,i} = P_{n,i}^*;$$

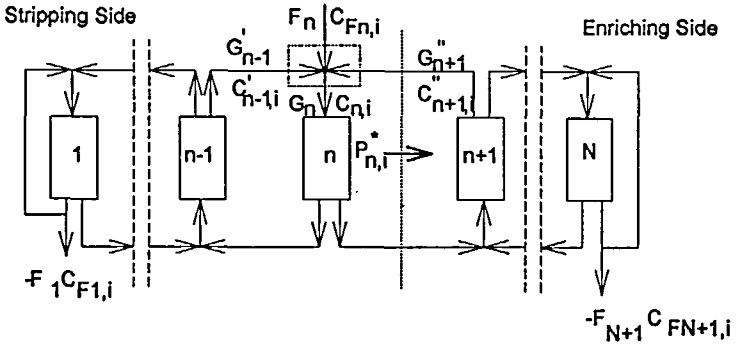

FIG. 1 Cascade scheme.

$$i = 1, 2, \ldots, K;\ n = 1, 2, \ldots, N - 1 \tag{2}$$

$$C_{n,i} = \theta_n C'_{n,i} + (1 - \theta_n) C''_{n,i}; \qquad i = 1, 2, \ldots, K;\ n = 1, 2, \ldots, N \tag{3}$$

$$G_n C_{n,i} = G_{n-1} \theta_{n-1} C'_{n-1,i} + G_{n+1}(1 - \theta_{n+1}) C''_{n+1,i} + F_n C_{Fn,i};$$
$$i = 1, 2, \ldots, K;\ n = 2, \ldots, N - 1 \tag{4}$$

where P_n^* is the net flow rate transported toward the enriching direction at the nth stage in the cascade, $P_{n,i}^*$ is the net flow rate of the ith component toward the enriching direction at the nth stage in the cascade, θ_n is the cut of the nth stage, and G_n is the interstage flow rate of the nth stage. $C_{n,i}$, $C'_{n,i}$, and $C''_{n,i}$ are the feed, heads, and tails concentration of the ith components at the nth stage, respectively. $F_n (n = 1, \ldots, N)$ is the external feed or withdrawal rate at the feed of the nth stage, and F_{N+1} is the withdrawal at the head of the Nth stage. F_n is defined as:

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

where $n = 1, \ldots, N + 1$. F_n may be an independent feed or withdrawal or may come from or go into other cascades. $C_{Fn,i}$ is the concentration of the ith component corresponding to F_n at the nth stage.

Equation (1) reflects the total material balance in the cascade. Equation (2) reflects the material balance of the ith component. Equation (3) is derived from the material balance of the nth stage. Equation (4) reflects the material balance among stages: the $(n-1)$th, nth, $(n+1)$th and external feed or withdrawal rates F_n at the nth stage.

The boundary conditions are determined by the operating conditions. For the example shown in Fig. 1, the boundary conditions are

$$P^*_{N,i} = -F_{N+1}C_{FN+1,i}; \quad C_{FN+1,i} = C'_{N,i}; \quad P^*_N = -F_{N+1}; \quad i = 1, \ldots, K$$

$$P^*_{0,i} = F_1 C_{F1,i}; \quad C_{F1,i} = C''_{1,i}; \quad P^*_0 = F_1; \quad i = 1, \ldots, K \tag{5}$$

The separation factor, γ_{ij}, defined as 'heads to tails separation factor,' is (6)

$$\gamma_{ij} \equiv \frac{C'_i}{C'_j} \bigg/ \frac{C''_i}{C''_j} \tag{6}$$

γ_{ij} depends on the structure of the separation unit and the separation parameters which are assumed to be given in our calculation. The following relation can be derived from Eq. (6):

$$C''_{n,i} = \frac{C'_{n,i}}{\sum_j \gamma_{ij} C'_{n,j}}; \quad C'_{n,i} = \frac{C''_{n,i}}{\sum_j C''_{n,j} / \gamma_{ij}};$$

$$i = 1, \ldots, K; j = 1, \ldots, K; n = 1, \ldots, N \tag{7a,7b}$$

The separation factor, γ_{ij}, has the following relationship with the molar weight of the components (7):

$$\gamma_{ij} = \gamma_0^{M_j - M_i}; \quad i = 1, \ldots, K; j = 1, \ldots, K \tag{8}$$

where γ_0 is defined as the unit separation factor which depends on the separating condition. M_i and M_j are the molar weights of the ith and the jth components, respectively.

Numerical Solution of the Governing Equations

Equations (1), (2), (3), and (4) are a set of nonlinear equations. Three methods are usually used to obtain the solutions:

(a) Use Eq. (2) repeatedly, iterate from the enriching side (or the stripping side) to the stripping side (or the enriching side).

(b) Use Eq. (2) repeatedly, iterate from both the enriching side and the stripping side to the feed.

(c) Let Eq. (4) be the input values at every iteration. Then use Eq. (4) repeatedly until the material balance is satisfied.

All three methods have been used in our work. In this paper, Method (a) will be given as an example.

The Iteration Process

Our example has one product stream, one waste steam, and one feed stream. The known parameters are the number of components K, the total stages N, the interstage flow rate G_n ($n = 1, \ldots, N$), the feed F in the N_Fth stage, the concentration C_{Fi} ($i = 1, \ldots, K$) of the ith component in the feed, the product P in the heads of the Nth stage, and the waste W in the tails of the first stage. $P^*_{n,i}$ in Eq. (2) is equal to $-WC''_{1,i}$ when $n \leq N_F$, and equal to $PC'_{N,i}$ when $n > N_F$. The cut of the nth stage, θ_n, may be obtained from the following relations.

Let the cut of the first stage be $\theta_1 = 0.5(1 - W/G_1)$. The others can be derived from

$$\theta_n = \begin{cases} 1 - \dfrac{G_{n-1}}{G_n}\theta_{n-1} - W/G_n; & n \leq N_F \\ 1 - \dfrac{G_{n-1}}{G_n}\theta_{n-1} + P/G_n; & n > N_F \end{cases} \qquad n = 1, \ldots, N \quad (9)$$

We start the iteration process from the stripping side.

(1) The initial concentrations $C''_{1,i}$ should be given, for example, let $C''_{1,i} = C_{Fi}$.

(2) From Eq. (2):

$$C''_{n+1,i} = \begin{cases} \left(G_n\theta_n \dfrac{C''_{n,i}}{\sum_j C''_{n,j}/\gamma_{ij}} + WC''_{1,i}\right)\Big/(1 - \theta_{n+1}); & n < N_F \\ \left(G_n\theta_n \dfrac{C''_{n,i}}{\sum_j C''_{n,i}/\gamma_{ij}} + WC''_{1,i} - FC_{Fi}\right)\Big/(1 - \theta_{n+1}); & n \geq N_F \end{cases}$$

$$i = 1, \ldots, K; j = 1, \ldots, K; n = 1, \ldots, N \quad (10)$$

Calculate the concentration stage by stage until the last stage $C''_{N,i}$. Then the head concentration of the Nth stage, C'_{Ni}, is obtained from Eq. (7a).

(3) Determine if the material balance relation is satisfied.

$$PC'_{N,i} + WC''_{1,i} - FC_{Fi} = 0; \qquad i = 1, \ldots, K \quad (11)$$

(4) If Eq. (11) is not satisfied, the initial values of step (m + 1) should be changed to

$$C''^{m+1}_{1,i} = \omega \frac{FC_{Fi}}{P\dfrac{C'^{m}_{N,i}}{C''^{m}_{1,i}} + W} + (1 - \omega)C''^{m}_{1,i}; \qquad i = 1, \ldots, K \quad (12)$$

where ω is a factor $0 < \omega < 1$.

(5) Repeat Steps (2), (3), and (4) until Eq. (11) is satisfied.

THE TRANSIENT APPROACH

Governing Transient Equations

The transient analysis considers the separation of a multicomponent mixture with K components in a gas centrifuge cascade of N stages. The definition of the enriching and stripping sides and the definition of symbols in the equations are the same as those for the steady-state case. The governing transient equations for a cascade are (6)

$$\theta_n G_n C'_{n,i} - (1 - \theta_{n+1})G_{n+1}C''_{n+1,i} = P^*_{n,i}; \quad i = 1, 2, \ldots, K; \; n = 1, 2, \ldots, N - 1 \qquad (13)$$

$$C_{n,i} = \theta_n C'_{n,i} + (1 - \theta_n)C''_{n,i}; \qquad i = 1, 2, \ldots, K; \; n = 1, 2, \ldots, N \qquad (14)$$

$$\frac{\partial(H_n C_{n,i})}{\partial t} = P^*_{n-1,i}(t) - P^*_{n,i}(t) + F_n C_{Fn,i}; \qquad i = 1, 2, \ldots, K; \quad n = 1, 2, \ldots, N \qquad (15)$$

H_n is the holdup of the nth stage. Equation (15) reflects the change rate of the ith component in the nth stage.

Because the flow change transient from the initial state to steady-state is very short, flow parameters such as θ_n and G_n are considered to be constant in time. Only the concentrations or parameters depending on the concentrations change with time.

The boundary conditions are determined by the operating conditions. The boundary conditions for the example shown in Fig. 1 are

$$P^*_{N,i} = -F_{N+1}C_{FN+1,i}; \qquad C_{FN+1,i} = C'_{N,i}; \qquad P^*_N = -F_{N+1}; \qquad i = 1, \ldots, K$$

$$P^*_{0,i} = F_1 C_{F1,i}; \qquad C_{F1,i} = C''_{1,i}; \qquad P^*_0 = F_1; \qquad i = 1, \ldots, K$$

Substituting Eq. (13), Eq. (14), and the boundary conditions into Eq. (15) gives

$$H_n \frac{\partial C_{n,i}}{\partial t} = [1 - \delta(n, 1)]\{\theta_{n-1} G_{n-1} C'_{n-1,i} - (1 - \theta_n) G_n C''_{n,i}\}$$
$$- [1 - \delta(n, N)]\{\theta_n G_n C'_{n,i} - (1 - \theta_{n+1}) G_{n+1} C''_{n+1,i}\}$$
$$+ F_n C_{Fn,i} + \delta(n, N) F_{N+1} C_{FN+1,i};$$
$$i = 1, 2, \ldots, K;\ n = 1, 2, \ldots, N \qquad (16)$$

where $\delta(n, nx)$ is defined as

$$\delta(n, nx) = \begin{cases} 0, & n \neq nx \\ 1, & n = nx \end{cases}$$

Substituting Eq. (7a) into Eq. (15) gives

$$C_{n,i} = \theta_n C'_{n,i} + (1 - \theta_n) \frac{C'_{n,i}}{\sum_j \gamma_{ij} C'_{n,j}};$$
$$i = 1, \ldots, K;\ j = 1, \ldots, K;\ n = 1, \ldots, N \quad (17)$$

From Eq. (17):

$$\frac{\partial C_{n,i}}{\partial t} = \sum_j \frac{\partial C_{n,i}}{\partial C'_{n,j}} \cdot \frac{\partial C'_{n,j}}{\partial t}; \qquad i = 1, \ldots, K;\ j = 1, \ldots, K;$$
$$n = 1, \ldots, N \quad (18)$$

where

$$\frac{\partial C_{n,i}}{\partial C'_{n,j}} = a(i, j) = \begin{cases} \theta_n + (1 - \theta_n) \dfrac{\sum_l \gamma_{il} C'_{n,l} - C'_{n,i}}{\left(\sum_l \gamma_{il} C'_{n,l}\right)^2} & (i = j) \\ -(1 - \theta_n) \dfrac{\gamma_{ij} C'_{n,i}}{\left(\sum_l \gamma_{il} C'_{n,l}\right)^2} & (i \neq j) \end{cases}$$
$$i = 1, \ldots, K;\ j = 1, \ldots, K;\ l = 1, \ldots, K;$$
$$n = 1, \ldots, N \quad (19)$$

Substituting Eq. (7a) and Eq. (19) into Eq. (16) gives

$$H_n \sum_j a(i, j) \frac{\partial C'_{n,j}}{\partial t}$$
$$= [1 - \delta(n, 1)]\left\{\theta_{n-1} G_{n-1} C'_{n-1,i} - (1 - \theta_n) G_n \frac{C'_{n,i}}{\sum_l \gamma_{il} C'_{n,l}}\right\}$$

$$-[1-\delta(n,N)]\left\{\theta_n G_n C'_{n,i} - (1-\theta_{n+1})G_{n+1}\frac{C'_{n+1,i}}{\sum_l \gamma_{il} C'_{n+1,l}}\right\}$$

$$+F_n C_{Fn,i} + \delta(n,N)F_{N+1}C_{FN+1,i};$$

$$i = 1,\ldots,K;\ j = 1,\ldots,K;\ l = 1,\ldots,K;\ n = 1,\ldots,N$$

Equation (20) is a set of partial differential and difference equations governing the transient concentration variation of a multicomponent cascade. The finite difference method can be used to solve Eq. (20).

Finite Difference Approach

a) The finite difference representation of the time derivative in Eq. (20) is

$$\frac{\partial C'_{n,i}}{\partial t} = \frac{C'^{m+1}_{n,i} - C'^{m}_{n,i}}{\Delta t} + O(\Delta t); \qquad i = 1,\ldots,K;\ n = 1,\ldots,N \tag{21}$$

$C'^{m}_{n,i}$ is the concentration of the ith component at the head of the nth stage at the mth time step. Neglecting $O(\Delta t)$ in Eq. (21), Eq. (20) becomes

$$H_n \sum_j a(i,j)^m \frac{C'^{m+1}_{n,l} - C'^{m}_{n,l}}{\Delta t}$$

$$= [1-\delta(n,1)]\left\{\theta_{n-1}G_{n-1}C'^{m}_{n-1,i} - (1-\theta_n)G_n\frac{C'^{m}_{n,i}}{\sum_l \gamma_{il}C'^{m}_{n,l}}\right\}$$

$$-[1-\delta(n,N)]\left\{\theta_n G_n C'^{m}_{n,i} - (1-\theta_{n+1})G_{n+1}\frac{C'^{m}_{n+1,i}}{\sum_l \gamma_{il}C'^{m}_{n+1,l}}\right\}$$

$$+F_n C^{m}_{Fn,i} + \delta(n,N)F_{N+1}C^{m}_{FN+1,i} \tag{22}$$

Rearranging gives

$$\frac{H_n}{\Delta t}\sum_j a(i,j)^m C'^{m+1}_{n,j}$$

$$= [1-\delta(n,1)]\left\{\theta_{n-1}G_{n-1}C'^{m}_{n-1,i} - (1-\theta_n)G_n\frac{C'^{m}_{n,i}}{\sum_l \gamma_{il}C'^{m}_{n,l}}\right\}$$

$$-[1-\delta(n,N)]\left\{\theta_n G_n C'^{m}_{n,i} - (1-\theta_{n+1})G_{n+1}\frac{C'^{m}_{n+1,i}}{\sum_l \gamma_{il}C'^{m}_{n+1,l}}\right\}$$

$$+ F_nC^m_{Fn,i} + \delta(n, N)F_{N+1}C^m_{FN+1,i} + \frac{H_n}{\Delta t}\sum_j a(i, j)^m C'^m_{n,j};$$

$$i = 1, \ldots, K; j = 1, \ldots, K; n = 1, \ldots, N \tag{23}$$

The initial values at time step 0 are determined by practical conditions. For example, if the feed concentration is C_{Fi}, the initial values can be chosen as $C'^0_{n,i} = C_{Fi}$. Equation (23) is then used repeatedly until steady-state is reached. Two kinds of steady-state criterion may be used: (1) Each component in the cascade satisfies the material balance, that is:

$$\sum_{n=1}^{N+1} F_nC_{Fn,i} = 0; \qquad i = 1, \ldots, K \tag{24}$$

(2) The concentration difference during the mth and the $(m + 1)$th time step is small enough so that $\delta = (C^{m+1}_{n,i} - C^m_{n,i})/C^{m+1}_{n,i}$ has achieved the required accuracy.

b) Accuracy and stability of the explicit approach. The explicit approach to the numeral solution of Eq. (20) involves estimation of new concentrations at time step $m + 1$ based on values at time step m. The method given by Eq. (21) is a first-order method. Decreasing Δt can reduce the error. With modern computer technology, the computational time is acceptable for quite small Δt.

In addition to the accuracy, the stability of the algorithm also limits Δt. For small separation factors Eq. (20) can be replaced by a partial differential equation, in which case Δt is limited by (6):

$$\Delta t < \min_{1\leq n\leq N}\left(\frac{H_n}{G_n}\right) \tag{25}$$

Equation (25) was also used in this work to estimate the maximum time step for large separation factors.

c) Choice of parameters. Parameters such as G_n, H_n, and F_n in Eq. (23) are given for practical cascades. The cut of the nth stage, θ_n, may be obtained by two methods: (1) Let the cut of the first stage be $\theta_1 = 0.5(1 + F_1/G_1)$; the others can then be derived from

$$\theta_n = 1 - \frac{G_{n-1}}{G_n}\theta_{n-1} - \sum_{i=n}^{N+1} F_i/G_n; \qquad n = 1, \ldots, N \tag{26}$$

(2) Let the cut of the Nth stage be $\theta_N = 0.5(1 - F_{N+1}/G_N)$; the others can then be derived from

$$\theta_{n-1} = \frac{G_n}{G_{n-1}} - \frac{G_n}{G_{n-1}}\theta_n - \sum_{i=n}^{N+1} F_i/G_{n-1}; \quad n = N-1, \ldots, 1 \tag{27}$$

EXAMPLES

Both the iteration approach and the transient approach can determine the distribution of components in a multicomponent cascade at steady-state. Consider the separation of Cr isotopes which contains four components, Cr-50, Cr-52, Cr-53, and Cr-54, as an example. The natural concentrations of each component are 0.0431, 0.8376, 0.0955, and 0.0238, respectively.

Consider a square cascade as shown in Fig. 2. There is one product stream ($F_{N+1} = -P$), one waste stream ($F_1 = -W$), and one feed stream ($F_{N_F} = F$). Assume a total of 15 stages which have same interstage flow rate G and same stage holdup H. The ratio of P to F is 0.2, the ratio of F to G is 0.2, $\gamma_0 = 1.12$, the feed is at the 12th stage, that is, $N_F = 12$, and the ratio of H to G is $0.03h$. The value of H actually only influences time step Δt and has no effect on the calculation result at steady-state. Equations (9) and (26) are used to chose θ_n. Table 1 shows the results of the two methods where C_{Pi} is the component concentrations in the product stream at steady-state and C_{Wi} is the component concentrations in the waste stream at steady-state.

The calculation reaches steady-state when the material balance relation $\sum_{n=1}^{N+1} F_n C_{Fn,i} < 10^{-6}$ for $i = 1, \ldots, K$. With a 486-66 MHz PC, the iteration approach required a total of 12 steps and 0.2 second. With the same machine and the same accuracy, the transient approach using $\Delta t = 0.02h$, needed about 1000 steps and a total of 20 seconds to reach steady-state. If $\partial C'/\partial t$ was used instead of $\partial C/\partial t$ in Eq. (16), the calculation time was reduced to

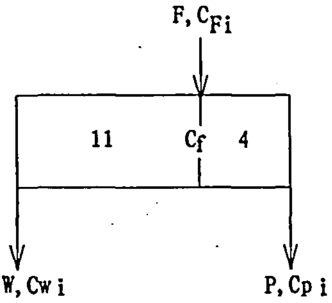

FIG. 2 Cascade schematic.

TABLE 1
Calculational Result

	Method	Cr-50	Cr-52	Cr-53	Cr-54
C_{Pi}	Iteration approach	0.075793	0.84224	0.069679	0.012285
	Transient approach	0.075795	0.84224	0.069679	0.012285
C_{Wi}	Iteration approach	0.034926	0.83644	0.10196	0.026679
	Transient approach	0.034926	0.83644	0.10196	0.026679

12 seconds without changing the calculation result. Although the iteration approach has some advantages in short, square cascades, the iteration may not converge for inappropriate initial values for a long cascade or a large separation factor. The transient approach cannot only predict the component distribution at steady-state, it can also predict the transient behavior. Figure 3 shows the variation of the concentration of Cr-50 in different stages (*n*) of a cascade with time. The result shows that the concentration changes rapidly

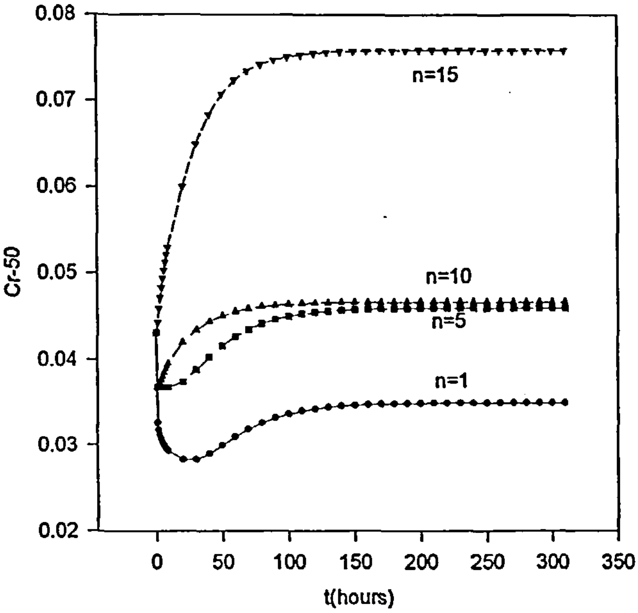

FIG. 3 Variation of Cr-50 concentration calculated using the transient approach.

at the beginning of separation. On the stripping side, a large decrease occurs in the Cr-50 concentration at the beginning of separation, $n = 1$.

CONCLUSION

Both the iteration approach and the transient approach can give component distribution along a multicomponent cascade. The iteration approach has the advantages of short calculational time and high precision, but the iteration approach may diverge for an improper choice of initial value if the cascade is complicated. The transient approach can be more easily applied to a wide range of cascade designs, especially for cascades with complicated structures, such as interconnected cascades. The results in Ref. 6 were all obtained using the transient approach. In addition, only the transient approach can predict the transient behavior of a separation process, which is useful when analyzing the properties of multicomponent separation.

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