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A Second-Order Time-Accurate Method for Determination of Concentration Distribution of Multicomponent Mixtures in Separation Cascades

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ABSTRACT

An implicit method is presented to study the transient process of concentration distribution of multicomponent mixtures in separation cascades for gases. The problems that the first-order method (the so-called "transient approach") encounters have been overcome. The new method is second-order accurate in time and unconditionally stable. The nonlinear difference equations at each time step are solved by the q-iteration method. A simple cascade is studied as an example, and comparisons are made with the first-order "transient approach." The results show that the new method is advantageous over the "transient approach" in both accuracy and computational efficiency.

Key Words. Multicomponent isotope separation; Separation cascades; Transient process

INTRODUCTION

Analyzing and optimizing a separation process in a separation cascade for multicomponent mixtures requires knowledge of the concentration distribution of components in the cascade at steady state and at certain times during the transient process from one state to another. The concentration distribution

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in the cascade is described by a set of nonlinear difference equations for the steady state or by a set of nonlinear partial differential-difference equations for the transient process. The present study focuses on separation cascades for gases, such as cascades composed of gas centrifuges.

The solution of these equations has been studied for quite a long time, and the methods can be classified into two categories.

1. Iterative methods: The difference equations for the steady state are used in the solution. Iteration is necessary because of the nonlinearity in the equations and the impossibility of direct solution. A typical iterative method is outlined in Ref. 1.
2. Transient methods: The partial differential-difference equations are considered. By means of time marching, the concentration distribution at steady state is reached after a sufficient number of time steps. Transient methods are also useful in investigating transient processes of separation cascades if the time marching is accurate enough. The approach of Wu et al. (1) is a transient method. Since in this approach the time marching is explicit, we refer to it as the explicit transient approach (the ETA) here.

To the authors' knowledge, more attention has been paid to concentration distribution at steady state; iterative methods are better studied than transient methods. However, iterative methods usually suffer from some significant deficiencies, as mentioned in Ref. 2, which include consumption of considerable computer time, difficulties in choosing appropriate initial values, and low accuracy, so they may not work well or may even fail. All these may greatly restrict their applicability. The ETA proposed in Ref. 1 seems to be able to cope with the problems in choosing initial values and low accuracy. The reasons are that convergence to steady state is guaranteed as long as the time step is small enough, and the problem of low accuracy does not exist since the difference equations can be satisfied at each cascade stage to the required accuracy. The only problem with the ETA may be that for difficult cases, such as complicated cascades and a large number of isotope components, the time step has to be taken quite small, which results in a significant increase of computation time. As far as the concentration distribution at steady state is concerned, whether or not the transient process can be exactly resolved does not matter. So in order to reduce computation time, the time step can be chosen to be as large as possible, and some simplification (cf. Ref. 1) can also be used to speed up the computation. In cases where transient processes are to be studied, the ETA provides us a means for this purpose. But it is not accurate, especially at the initial stage of computation. This is due not only to its first-order accuracy but also to the way in which linearization is carried out. Furthermore, consumption of computation time may become more significant compared with the solution for steady state because large time steps and simplification cannot be employed for reasons of accuracy.

The work of the present study is to develop a method that is:

1. Second-order accurate in time. This means that with the same time step, an order of higher accuracy can be achieved than with ETA.
2. Unconditionally stable. This gives us flexibility in studying the transient process and steady state without worrying about the size of the time step.
3. Reasonably fast. This means that the computation time should not be large, or at least there should be no obvious increase compared with the ETA. This is important since the advantage of having an order of higher accuracy may be lost if the ETA can also obtain the same accuracy just by using a very small time step without spending more computation time. This method will be referred to as the implicit transient approach, the ITA for short.

THE PARTIAL DIFFERENTIAL-DIFFERENCE EQUATIONS

For simplicity, we consider a popular separation cascade as shown in Fig. 1. The number of stages is N. The cascade has one feed F at stage N_f and two withdrawals (the waste W and the product P at stage 1 and stage N, respectively). Assume that in the feed there are N_c components, the concentrations of which are $C_{i,F}$, $i = 1, 2, \ldots, N_c$. The partial differential-difference equations

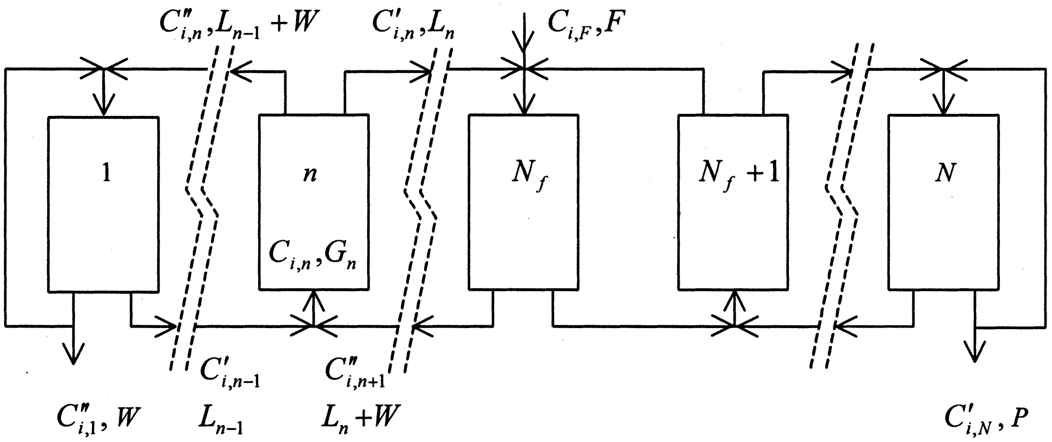

FIG. 1 A square separation cascade with one feed and two withdrawals.

are given by consideration of the mass conservation for the total mass and for each component:

$$F = P + W \tag{1}$$

$$\frac{\partial H_n C_{i,n}}{\partial t} = (L_n + a)C''_{i,n+1} + L_{n-1}C'_{i,n-1} - L_n C'_{i,n} - (L_{n-1} + b)C''_{i,n} + c \tag{2}$$

$$(L_n + L_{n-1} + b)C_{i,n} = L_n C'_{i,n} + (L_{n-1} + b)C''_{i,n} \tag{3}$$

with

$$a = \begin{cases} W & 1 \le n < N_f \\ -P & N_f \le n \le N \end{cases}, \quad b = \begin{cases} W & 1 \le n \le N_f \\ -P & N_f < n \le N \end{cases}, \quad c = \begin{cases} FC_{i,F} & n = N_f \\ 0 & \text{otherwise} \end{cases}$$

Note that $C'_{i,0} = C''_{i,1}$, and $C''_{i,N+1} = C'_{i,N}$. Here H_n is the holdup of the process gas in stage n; $C_{i,n}$, $C'_{i,n}$, and $C''_{i,n}$ are the inflow, head, and tail concentrations of the ith component; and L_n is the flow rate of the upflowing stream at the nth stage. L_n is taken to be constant here. Nonlinearity arises from the following well-known relationship (3):

$$\gamma_{ij} = \frac{C'_i}{C''_i}\bigg/\frac{C'_j}{C''_j} = \gamma_0^{Mj-Mi} \tag{4}$$

where γ_{ij} is called the separation factor, and γ_0 is called the unit separation factor and is constant for a given type of separation units and isotopes. M_i is the mole molecular weight of the ith component. By definition, the concentrations are subject to the following condition:

$$\sum_i C_{i,n} = \sum_i C'_{i,n} = \sum_i C''_{i,n} = 1 \tag{5}$$

SOLUTION METHODS

Outline of the Explicit Transient Approach

From Eqs. (4) and (5) we have

$$C''_i = C'_i \Big/ \sum_j \gamma_{ij} C'_j \tag{6}$$

Eliminating C and C'' from Eq. (2) by using Eqs. (3) and (6) yields

$$H_n \sum_j f_{ij} \frac{\partial C'_{j,n}}{\partial t} = g_i \tag{7}$$

where f_{ij} and g_i are given by

$$f_{ij} = \frac{\partial C_{i,n}}{\partial C'_{j,n}}$$

$$= \begin{cases} \dfrac{1}{L_n + L_{n-1} + b} \left[(L_{n-1} + b) \dfrac{\sum_j \gamma_{ij} C'_{j,n} - C'_{i,n}}{\left(\sum_j \gamma_{ij} C'_{j,n} \right)^2} + L_n \right] & i = j \\ -\dfrac{L_{n-1} + b}{L_n + L_{n-1} + b} \dfrac{\gamma_{ij} C'_{i,n}}{\left(\sum_j \gamma_{ij} C'_{j,n} \right)^2} & i \neq j \end{cases}$$

$$g_i = (L_n + a) \frac{C'_{i,n+1}}{\sum_j \gamma_{ij} C'_{j,n+1}} - L_n C'_{i,n}$$

$$- (L_{n-1} + b) \frac{C'_{i,n}}{\sum_j \gamma_{ij} C'_{j,n}} + L_{n-1} C'_{i,n-1} + c$$

Therefore, equation (7) is a partial differential-difference equation for C'. Note that f_{ij} is a function of C' and, therefore, Eq. (7) is nonlinear. The ETA approximates the left-hand side of Eq. (7) by finite difference, which gives

$$\frac{H_n}{\Delta t} \sum_j f_{ij}^{(m)} C'^{(m+1)}_{j,n} = g_i^{(m)} + \frac{H_n}{\Delta t} \sum_j f_{ij}^{(m)} C'^{(m)}_{j,n} \tag{8}$$

The superscripts in parentheses indicate the time level. Obviously, this approximation is at most first-order accurate in time. In order to avoid nonlinearity, linearization is carried out by evaluating its value at the previous time step, i.e., $f_{ij}^{(m)}$, which introduces an inaccuracy. Actually, it can be shown that this differencing approximates $\partial C_{i,n}/\partial t$ by

$$\frac{\partial C_{i,n}}{\partial t} \approx \frac{L_{n-1} + b}{L_n + L_{n-1} + b} \frac{\sum_j \gamma_{ij} C'^{(m+1)}_{j,n}}{\sum_j \gamma_{ij} C'^{(m)}_{j,n}}$$

$$\times \frac{C''^{(m+1)}_{i,n} - C''^{(m)}_{i,n}}{\Delta t} + \frac{L_n}{L_n + L_{n-1} + b} \frac{C'^{(m+1)}_{i,n} - C'^{(m)}_{i,n}}{\Delta t} \tag{9}$$

However, by comparing Eq. (3) with Eq. (9), we immediately notice that inaccuracy may come from the ratio $\sum_j \gamma_{ij} C_{i,n}^{\prime(m+1)} / \sum_j \gamma_{ij} C_{i,n}^{\prime(m)}$ unless it is equal to unity, which is, of course, not the case in general. This ratio is actually the ratio $q^{(m+1)}/q^{(m)}$, with $q = C'/C''$, which is defined later in the section about the q-iteration method. The two cases in which $q^{(m+1)}/q^{(m)} = 1$ are at steady state and $\gamma_0 = 1$. This is because $C_i^{\prime(m+1)} = C_i^{\prime(m)}$ at steady state, and $\gamma_{ij} = 1$ as well as $\sum_j C_{j,n}^{\prime(m+1)} / \sum_j C_{j,n}^{\prime(m)} = 1$ for $\gamma_0 = 1$. In all other cases we do not have $q^{(m+1)}/q^{(m)} = 1$. Furthermore, the larger γ_0 is, the larger the ratio, and therefore the less accurate the ETA. Obviously, the size of the time step Δt is a factor which affects accuracy since the ratio is larger with a larger Δt than with a smaller Δt. Another point we need to mention is that the concentrations of all components are coupled at the new time level and so an $N_c \times N_c$ algebraic system has to be solved for each stage, which may be time consuming if N_c is large.

If resolution of the transient process is not important, then $\partial C_{i,n}/\partial t$ in Eq. (2) can be simply replaced by $\partial C'_{i,n}/\partial t$. Actually, this is a good approximation if γ_0 is close to unity such as it is in gaseous diffusion separation processes. However, in separation process with large γ_0 (for instance, with gas centrifuges) this approximation is problematic since C is significantly different from C'. The benefit of replacing C by C' is that coupling among the concentrations is broken and, therefore, instead of solving an $N_c \times N_c$ algebraic system given by Eq. (8) for each stage to obtain the new values of C' at the new time level, the new values are directly given. This saves a lot of computation time. However, if we are only interested in the steady state, a much better way would be to use the q-iteration method. This will be briefly explained shortly when it is used to calculate the concentration distribution at steady state.

The Second-Order Implicit Transient Approach

The derivative in Eq. (2) is approximated by

$$\frac{\partial H_n C_{i,n}}{\partial t} = \frac{H_n}{\Delta t}\left(C_{i,n}^{(m+1)} - C_{i,n}^{(m)}\right) \tag{10}$$

Using the Crank–Nicolson scheme, which is very popular in fluid dynamics and is second-order accurate in time and unconditionally stable, we have

$$\frac{H_n}{\Delta t}\left(C_{i,n}^{(m+1)} - C_{i,n}^{(m)}\right) = \frac{1}{2}\left(g_i^{(m+1)} + g_i^{(m)}\right) \tag{11}$$

which yields

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$$\begin{aligned}
&-\frac{L_{n-1}}{2}C_{i,n-1}^{\prime(m+1)}+\left(\frac{H_n}{\Delta t}\frac{L_n}{L_n+L_{n-1}+b}+\frac{L_n}{2}\right)C_{i,n}^{\prime(m+1)}\\
&+\left(\frac{H_n}{\Delta t}\frac{L_{n-1}+b}{L_n+L_{n-1}+b}+\frac{L_{n-1}+b}{2}\right)C_{i,n}^{\prime\prime(m+1)}-\frac{(L_n+a)}{2}C_{i,n+1}^{\prime\prime(m+1)}\\
&=\frac{L_{n-1}}{2}C_{i,n-1}^{\prime(m)}+\left(\frac{H_n}{\Delta t}\frac{L_n}{L_n+L_{n-1}+b}-\frac{L_n}{2}\right)C_{i,n}^{\prime(m)}\\
&+\left(\frac{H_n}{\Delta t}\frac{L_{n-1}+b}{L_n+L_{n-1}+b}-\frac{L_{n-1}+b}{2}\right)C_{i,n}^{\prime\prime(m)}+\frac{(L_n+a)}{2}C_{i,n+1}^{\prime\prime(m)}+1+c
\end{aligned}\tag{12}$$

For simplicity, we omit the superscripts and denote the above equation as

$$-\bar{\alpha}_{n-1}C'_{i,n-1}+\bar{\beta}_nC'_{i,n}+\delta_nC''_n-\gamma_{n+1}C''_{n+1}=r_n \tag{13}$$

The meanings of $\bar{\alpha}$, $\bar{\beta}$, δ, γ, and r are clear, but γ here is not to be confused with the separation factor γ_{ij} and γ_0. Equations (13), (4), and (5) constitute the set of equations to be solved for each component at every time step. Note that when Δt is taken to be infinity, this set of equations is just the set of difference equations for the steady state, and its solution is just as difficult as the solution of the steady-state equations. The implicit method makes sense only when the method of solving the set of nonlinear difference equations at each time step does not suffer from the problems mentioned in the Introduction. The q-iteration method proposed in Ref. 4 seems to be the method of choice.

Outline of the *q*-Iteration Method

Unlike the common iterative methods, iteration in the q-iteration method takes place over the ratios of the head and tail concentrations of an arbitrarily chosen component (for instance, the kth component), which are denoted as $q_{k,n}$, other than the concentrations themselves. So with specified values of $q_{k,n} = C'_{k,n}/C''_{k,n}$, we have

$$C'_{i,n}=q_{i,n}C''_{i,n} \tag{14}$$

where $q_{i,n}=q_{k,n}\gamma_0^{M_k-M_i}$. Substituting Eq. (14) into Eq. (13) gives

$$-\alpha_{n-1}C''_{i,n-1}+\beta_nC''_n-\gamma_{n+1}C''_{n+1}=r_n \tag{15}$$

with $\alpha_{n-1}=\bar{\alpha}_{n-1}q_{i,n-1}$, and $\beta_n=\bar{\beta}_nq_{i,n}+\delta_n$. The equation system given by Eq. (15) has a tridiagonal coefficient matrix and is easy to solve. Then $C'_{i,n}$ and $C_{i,n}$ are determined, respectively, by Eqs. (14) and (3). However, Condition (5) is not satisfied, so the values of $q_{k,n}$ need to be adjusted in the following way:

$$q_{k,n}=(1-\omega)q_{k,n}+\omega\frac{\sum\limits_i C_{i,n}}{\sum\limits_i \gamma_0^{M_k-M_i}C''_{i,n}} \tag{16}$$

Here ω is a weighting factor and $0 < \omega \leq 1$. The iteration is terminated when

$$\max_n \left(\left| \sum_i C_{i,n} - 1 \right|, \left| \sum_i C'_{i,n} - 1 \right|, \left| \sum_i C''_{i,n} - 1 \right| \right) \leq \varepsilon \qquad (17)$$

with ε a small given number. ε is fixed at 10^{-4} in our calculations.

CALCULATIONS AND COMPARISONS

The following cascade is used for our numerical experiments and comparisons. The number of stages is 100 with the feed at the 50th stage. The enterstage flow rate $G_n/F = 10$, and $P/F = 0.85$. This cascade is of course not practical, but it does not matter for the purpose of demonstrating the implicit method and making comparisons. The process gas is chosen to be Xe, which has nine stable isotopes and for which $\gamma_0 = 1.4$. We use a fairly long cascade and an element with quite a large number of stable isotopes to create sufficient difficulty for the methods. All computations were performed on a PC with a Pentium Pro-200 processor.

Figure 2 shows the error change during the transient process from an initial state, with concentrations of all components at each stage the same as those of

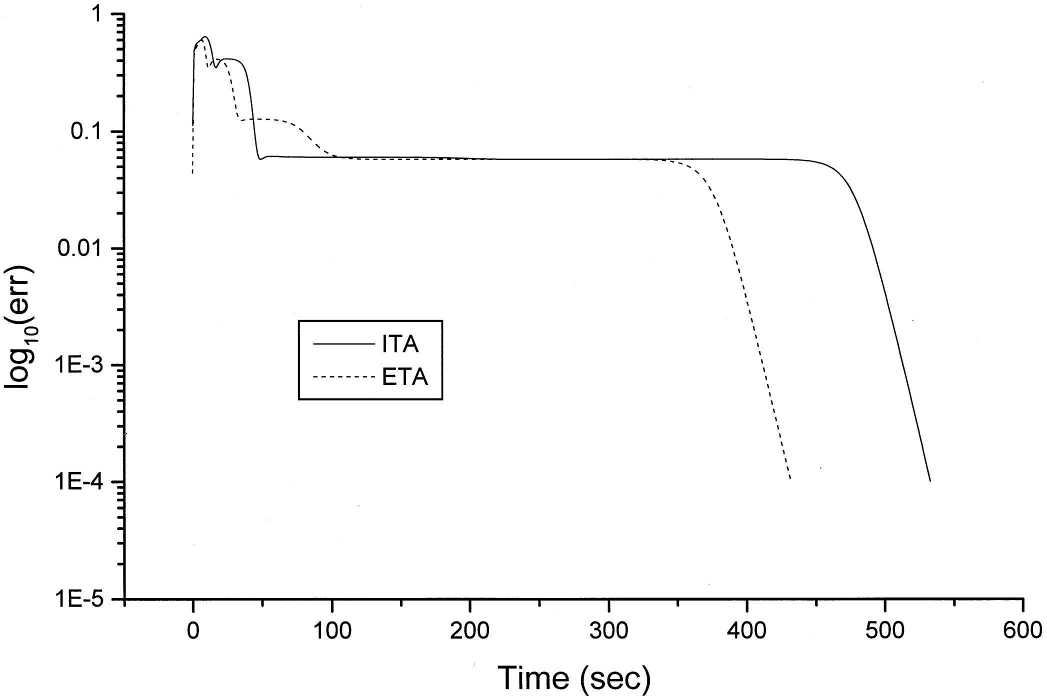

FIG. 2 The error change during the transient process.

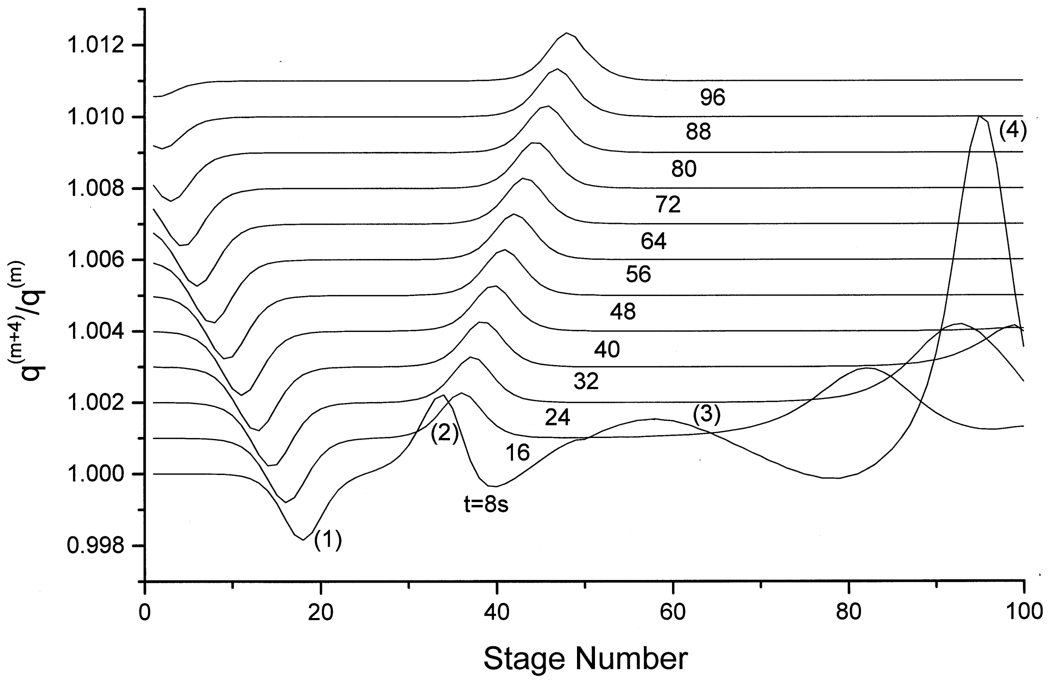

FIG. 3 The traveling of the $q^{(m+4)}/q^{(m)}$ peaks in the ETA case.

the feed, to the steady state. The error here is defined as

$$err = \left(\sum_i (FC_{i,F} - WC''_{i,1} - PC'_{i,N})^2 \right)^{1/2}$$

The steady state is thought to be reached when $err \leq 10^{-4}$. The ratio $H_n/(F\Delta t)$ is taken to be 15, which is nearly the smallest value that the ETA can use for stability consideration, or in another words, the largest value of Δt for a given value of H_n. Hereafter we assume $H_n = 0.3$ g without loss of generality, and so $\Delta t = 0.02$ second. The difference is clear from Fig. 2. The ETA predicts that the transient process will take 432 seconds whereas the ITA predicts 532 seconds. Another big difference is that the curve for the ETA has two flat areas while that for the ITA has only one.

To understand the above phenomena, the ratio $q^{(m+4)}/q^{(m)}$ is plotted in Fig. 3 for the ETA and in Fig. 4 for the ITA against the stage number at different times. In the two figures we have shifted all curves except the two for $t = 8$ seconds by a distance of 0.001 second between the two curves of successive time steps in the direction of the ordinates to avoid overlapping. The ratio of the quantity q for two successive time steps is not used in the figures because we want to keep the value of the ratio large enough to show the difference more clearly due to plotting accuracy. We see that there are peaks (and valleys) traveling along the cascade in both directions. The transient process

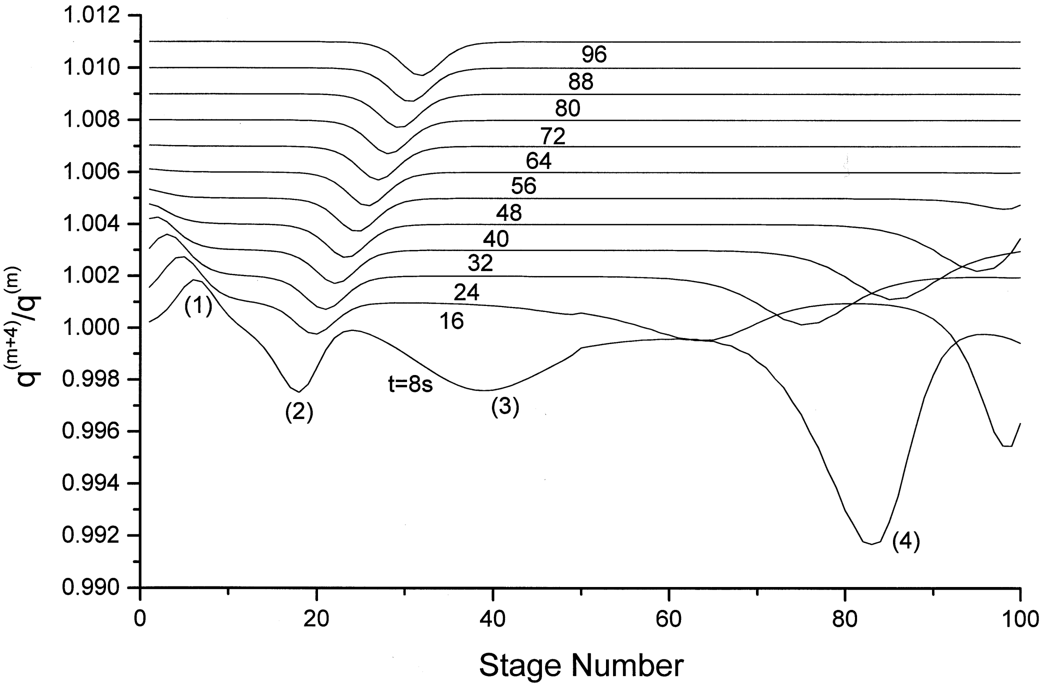

FIG. 4 The traveling of the $q^{(m+4)}/q^{(m)}$ peaks in the ITA case.

shown in Fig. 2 has a close relationship with the traveling of the peaks. First, let us look at these peaks for the ETA. Four peaks can be clearly identified, as indicated in Fig. 3. Peaks 1, 3, and 4 move out of the cascade at about 96, 40, and 16 seconds, respectively, which correspond to the first, the second, and the third decline of the error. One can estimate that Peak 2 goes out of the cascade at 346 seconds, which is just about the time that the last decline of the error takes place. A similar phenomenon happens in the ITA. Where there are also four peaks, and they look like the four peaks of the ETA case but are upside down. Peak 4 moves out of the cascade at about 24 seconds, and Peaks 1 and 3 nearly vanish at 48 seconds. Peak 2 is expected to go outside of the cascade at about 450 seconds. Again, the disappearances of peaks correspond to the decline of error. Since Peaks 1 and 3 travel out of the cascade at almost the same time, the third decline of the error as for the ETA (cf. Fig. 2) does not show up. These differences result from the initial condition which satisfies Eqs. (1), (2), (3), and (5) but not (4). The ETA gives large changes in C and C'' relative to the initial values at the first time step, no matter how small Δt is, but the change in C' are small. This does not seem to be reasonable, since it is C' and C'' that should have big changes due to separation when the concentration of the entering flow of a stage is C. At this point, the results from the ITA appear to be reasonable, i.e., C has small changes but C' and C'' have large changes that satisfy Eq. (4). The concentration distributions of a component at the first time step for the ETA and the ITA are plotted in Fig. 5. By using the results at the first time step from the ETA as the initial condition, both methods have the same error change histories. Of course, the initial condition used before may not exist in practice, but a very appropriate initial condition is not easily imposed. This implies that the ETA may not be very reliable in resolving the transient process if the initial condition is not carefully specified, unless γ_0 is close to unity.

Now we show that only when $q^{(m+1)}/q^{(m)} = 1$ will the ETA give the same result as the ITA. As mentioned before, in two special cases $q^{(m+1)}/q^{(m)} = 1$: steady state and $\gamma_0 = 1$. We see from Fig. 2 that as steady state is approached in the former case, the curve from the ETA resembles that from the ITA. In the latter case the error changes are presented in Fig. 6 for different values of γ_0. Again, the curves for different γ_0 are shifted to give more clarity. Clearly, as $\gamma_0 \to 1$, the results of the ETA gradually approach those of the ITA. So when γ_0 is close to unity, the influence of the initial condition may be negligible.

From the point of view of computational cost, the ITA is not more expensive than the ETA, although it is implicit. To demonstrate this briefly, we make the following comparisons of timing which were carried out by using the computer's internal clock to measure the elapsed time used for the calculations to satisfy the required accuracy from the same initial condition. There is

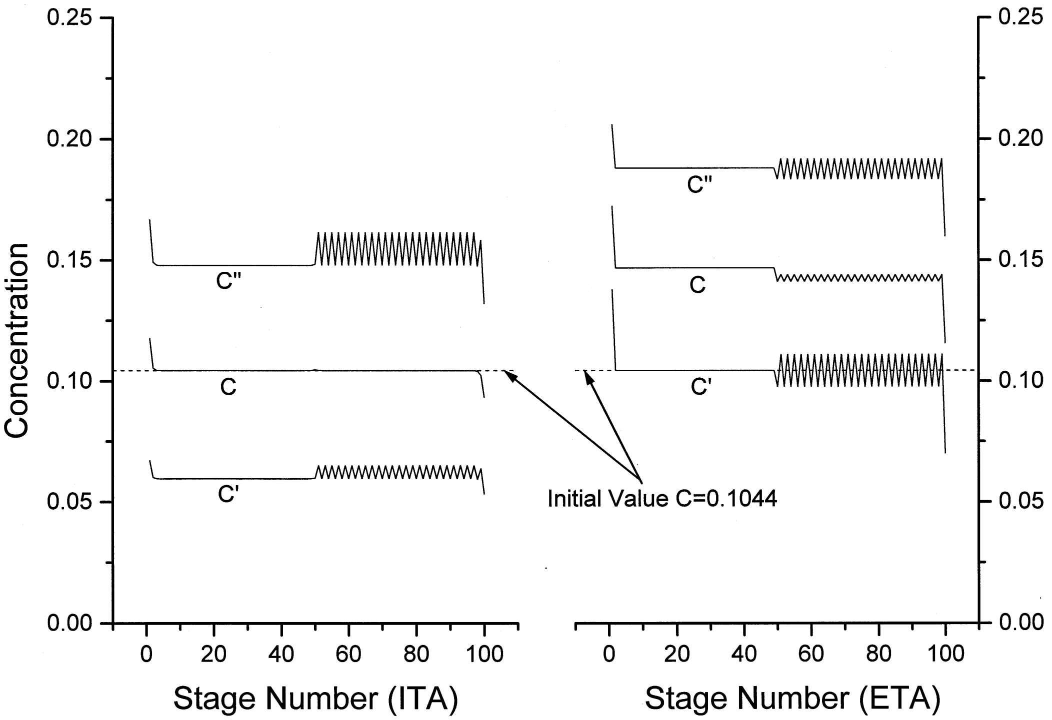

FIG. 5 C, C', and C'' distribution at the first time step for the Component ^{134}Xe.

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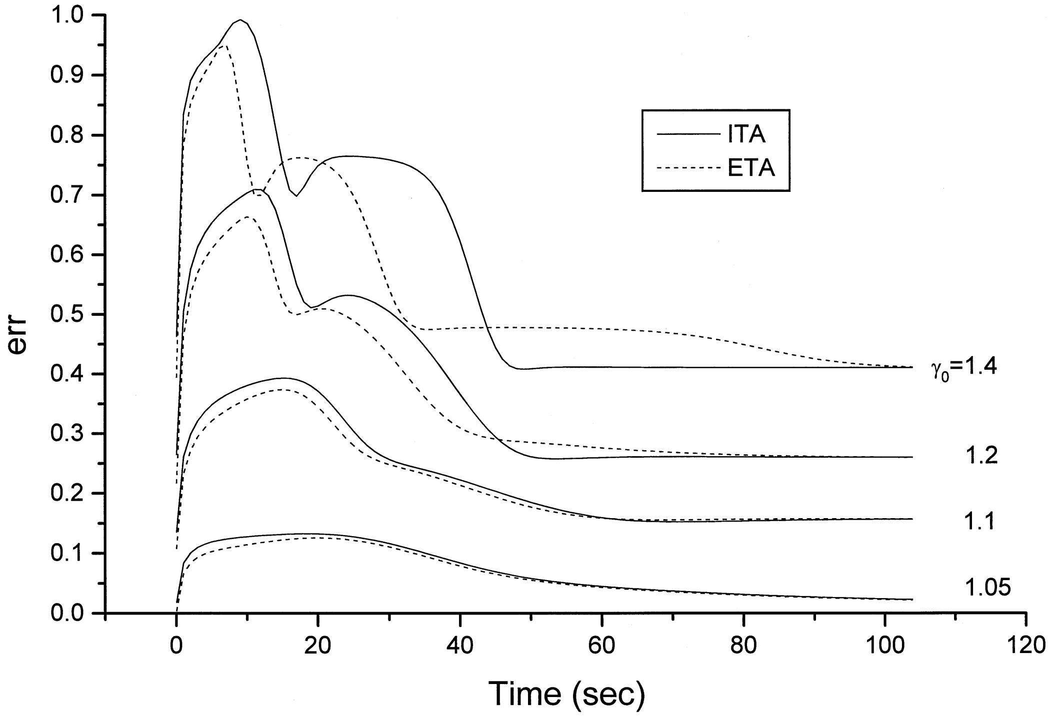

FIG. 6 The error changes for different values of γ_0.

only one process running on the computer during timing, and no input/output takes place. With $\Delta t = 0.02$ second the ETA uses 935 seconds and the ITA spends 674 seconds, which are roughly comparable. Reducing Δt by a factor of 10, the time used by the ETA is increased to 9332 seconds, a 10 times increase. But the ITA uses only 2517 seconds. This is not surprising, since no matter how small the time step is, the ETA has to do the same amount of work at every time step to solve N_c equations for each stage, while the ITA needs to do a smaller amount of work because the solutions at two successive time steps are closer with smaller Δt and consequently the number of iterations over nonlinearity is reduced. Using the ITA, one does not have to use small time steps because of its higher accuracy and unconditional stability. For this test problem, the transient process can still be quite correctly resolved even when $\Delta t = 3$ seconds although some little wiggles start to appear in the error–time plot. In this case the computation costs 36 seconds. If only the steady state is of interest, the time step Δt can be taken very large or the ratio $H_n/\Delta t$ can be set to zero. So the time marching process is skipped and only iterations over nonlinearity are performed. Now the ITA is just the q-iteration method and it consumes only 1.65 seconds to obtain the solution for the steady state.

To sum up, we conclude that the ITA is advantageous over the ETA for both accuracy and computational cost. The ITA can study transient processes with high accuracy, and it also serves as an efficient method for the investigation of steady state.

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