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A Calculation Procedure for Designing Ideal Centrifugal Separation Cascades

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Abstract: A numerical method is presented for determining flow rates in centrifugal isotope separation cascades that satisfy the required "ideal cascade" conditions. This method is an iterative method, which in the iteration process uses the q-iteration method to give the concentration and distributions of components under a specified hydraulic status for a cascade; and then, based on the concentration distributions, solves the algebraic equations describing the hydraulic status of the cascade to improve the specified hydraulic status towards the required "ideal cascade" conditions. The material losses in the cascade pipes and centrifuges are taken into account. Numerical simulations are performed by taking an MARC cascade with variant stage separation factors as an example. The results from four cases with and without material losses are compared. The results demonstrate that the method works very well, and show that when the material losses are nonexistent or very small, the concentration distributions can exactly satisfy the "ideal cascade" condition; but when the material losses are large, the "ideal cascade" condition can not be satisfied, but the method can produce a solution that allows the condition to be approximately satisfied.

Keywords: Ideal cascade, isotope separation, numerical simulation, separation cascade

INTRODUCTION

The concept of ideal cascade is very important in isotope separation and was put forward initially in (1) for the binary separation of uranium

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isotopes. Here an ideal cascade is defined to satisfy two conditions (2): (a) the enriching separation factor and the stripping separation factor are equal and are constant at all stages; and (b) the concentrations of components have no mixing at a confluence point of an up-streaming and a down-streaming flow. Of all cascades that have the same external parameters (the feed, the withdrawals, and the component concentrations in the feed and the withdrawals), the ideal cascade has a total flow rate that is equal or close to the minimum, and the separative power of the cascade is the sum of the separative powers of all separation units. This is to say that to produce the same product, an ideal cascade needs the minimum separative power, and so, easily understandable, has the highest economic efficiency. Because of its importance, the concept of ideal cascade has been extended to the case of multicomponent isotope separation, such as the MARC cascade of De La Garza (3) (Matched Abundance Ratio Cascade), the Q-cascade (4,5) (for small separation factors), and the "Quasi-ideal Cascade" (for large separation factors) of Russian researchers (6) (Under certain circumstances, a MARC cascade can be derived from a quasi-ideal cascade (7,8)). These cascades have their individual ideal conditions; for example, the ideal condition for an MARC cascade is that at a confluence point in the cascade the ratio of the abundance of a component with respect to that of another component in a flow is equal to the ratio in another flow; whereas for a quasi-ideal cascade, the ideal condition is that the cuts of each component flow at all stages are identical, i.e., constant. For ease of reference, all "ideal" cascades that are different from the traditional ideal cascade in the sense of separating binary isotopes are all referred to as quasi-ideal cascades. Regardless of an ideal or a quasi-ideal cascade, they all satisfy an "ideal" state required. Therefore, if possible, it is always desirable to manage a cascade in reality to be as close to an ideal cascade as possible. This is an issue of cascade design.

The design of a cascade can be divided into two phases. One is to determine the concentration distribution of each component in the cascade under a given hydraulic status (i.e., the flow distribution is given, or in other words, the flows of all stages are known), and is important in analyzing the separation performance of a known cascade. Another phase, just on the contrary, is to determine the hydraulic status under a desired concentration condition, such as the no-mixing condition for an ideal cascade, or other requirements, such as the constant cuts of component flows for a quasi-ideal cascade. The latter phase is more difficult than the former one.

Numerical methods play an important role in designing cascades, as more practical factors are considered and new separation methods are explored. Currently all numerical methods (cf. (9,10,11,12)) perform

the work of determining concentration distributions in a cascade which requires the hydraulic status of the cascade to be specified, that is, solving the problems of the first phase. This is due to the fact that the algebraic equation system describing the concentration distributions is nonlinear and can only be solved by means of numerical approaches in general. As for solving problems arising from the second phase of cascade design, people are used to consider simple cases. In these simple cases, the concentration distributions can be given or obtained in advance, such as in an ideal cascade for binary separation, and thereby the hydraulic status can then be fairly easily determined and its analytical expressions can be derived. To deal with increasingly more and more complicated practical separation cases in multicomponent isotope separation, to just name a few, inclusion of material losses and impurity productions, consideration of variant stage separation factors, operation with intermediate feeds and withdrawals, and use of carrier gas, resorting to numerical approaches seems to be a good idea. This can be understood by considering material losses and impurity productions, which are functions of the concentrations distributions and hydraulic status, the concentration distributions are coupled with the hydraulic status. Therefore, neither the concentration distributions nor the hydraulic status can be known in advance or specified. Determining the concentration distributions must be done together with determining the hydraulic status, which means solving the problems of both the first phase and the second phase simultaneously in numerical ways. So the solution of problems of the second phase includes the solution of problems of the first phase. Here the so-called ''material losses'' and ''impurity production'' mean that corrosion is to some extent inevitable in gas centrifuges and in cascade pipes because the multicomponent mixture to be separated (i.e. the process gas) is often corrosive, which consumes the process gas and meanwhile accompanies the production of light impurities. An example often seen is that a process gas is of the fluoride kind and can usually react with the pipes and the separation units, as well as with the moisture leaking into the cascade, causing the loss of the process gas and production of the light impurity HF. We refer to both material losses and light impurity productions as material losses for simplicity. Without material losses taken into account, it is very easy to obtain an analytical expression of the flow distribution in the case of binary separation, but becomes much more difficult in the case of multicomponent isotope separation (13). With material losses taken into account, it is no longer an easy job to obtain analytical expressions for the flow distribution (1,8,14,15) even with some simplifications, such as using the assumption that the losses are proportional to the flow rates to decouple the concentrations with the hydraulic status. Such an assumption is invalid when the process gas contains a significant amount

of nonreactive components, for instance, in consideration of feeding a carrier gas. So clearly there is a demand for developing a numerical method to solve the problems of the second phase, without worrying about the existence of an analytical solution. However, to the authors' knowledge, apart from the above-mentioned simple cases in which analytical solutions can be obtained, numerical solution methods are hardly found in literature to handle the problems of the second phase (16).

The main subject of this paper is to present a numerical method that is able to determine the hydraulic status of a cascade and the concentration distributions satisfying the specified conditions. In Section 2 and Section 3, the algebraic equations are given describing the concentrations distributions and the hydraulic status. Section 4 briefly explains the q-iteration method of solving the algebraic equations for the concentration distributions under a specified hydraulic status, and Section 5 explains the method of determining the hydraulic status with the concentration distributions known. Using two quasi-ideal cascades as examples (one being an MARC cascade and one a cascade with constant cuts of component flows), in Section 6 how to obtain the hydraulic status is discussed that gives the concentration distributions satisfying the requirements. Numerical experiments are carried out in Section 7 for an MARC cascade to justify the numerical method, and the results are presented and the cases with and without material losses are compared.

THE EQUATION SYSTEM FOR CONCENTRATION DISTRIBUTIONS

The centrifugal cascade considered here is as shown schematically in Fig. 1. The length of the cascade, i.e., the number of stages, is N, the feed F is fed into the cascade at stage N_F, and P, and W are respectively the product and the waste withdrawals.

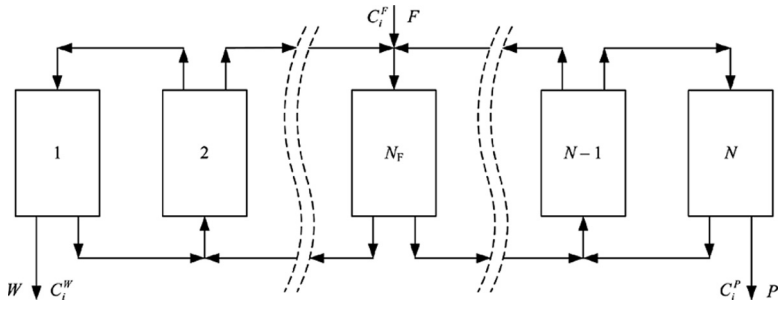

Figure 1. Schematic illustration of a centrifuge cascade.

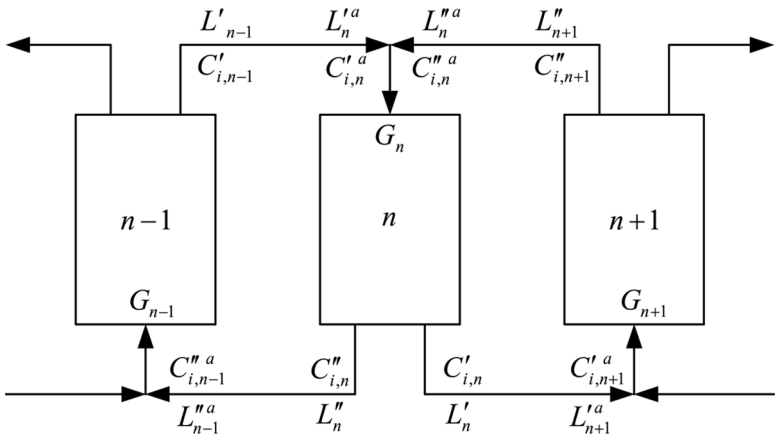

Figure 2. The flow and concentration quantities relevant to a stage and the neighboring two stages.

Assume that the process gas consists of N_c components, including the light impurities, which are numbered sequentially, according to their mole weights from 1 for the lightest component. At each stage, a mass conservation relation holds (cf. Fig. 2):

$$L'^{a}_{n}C'^{a}_{i,n} + L''^{a}_{n}C''^{a}_{i,n} - L'_{n}C'_{i,n} - L''_{n}C''_{i,n} - m_{i,n} = 0. \tag{1}$$

Here, L'_n and L''_n are the product flow (head flow) and the waste flow (tail flow), respectively, of the n-th stage, $C'_{i,n}$ and $C''_{i,n}$ are respectively the concentrations of the i-th component in the two flows, L'^{a}_{n} and L''^{a}_{n} are the two incoming flows at the influent point of the n-th stage, from respectively the product flow of the $(n-1)$-th stage and the waste flow of the $(n+1)$-th stage, $C'^{a}_{i,n}$ and $C''^{a}_{i,n}$ are the concentrations in the two incoming flows just before the influent point. G_n is the entering flow of the n-th stage:

$$G_n = \begin{cases} L'^{a}_{n} + L''^{a}_{n} + F & n = N_F \\ L'^{a}_{n} + L''^{a}_{n} & n \neq N_F \end{cases}, \tag{2}$$

with $C_{i,n}$ being the concentration of the i-th component:

$$C_{i,n} = \begin{cases} (C'^{a}_{i,n}L'^{a}_{n} + C''^{a}_{i,n}L''^{a}_{n} + C^{F}_{i}F)/G_n & n = N_F \\ (C'^{a}_{i,n}L'^{a}_{n} + C''^{a}_{i,n}L''^{a}_{n})/G_n & n \neq N_F \end{cases}.$$

For simplicity of expression, all quantities that are not defined or whose subscripts are out of range should be set to zero, for example for L'^{a}_{1}, L''^{a}_{n}.

Clearly, we have for the cascade in Fig. 1,

$$L''_1 = W, L'_N = P, C''_{i,1} = C_i^W, C'_{i,N} = C_i^P.$$

$m_{i,n}$ is the material loss of the i-th component at the n-th stage

$$m_{i,n} = a^l_{i,n} C_{i,n} G_n - \sum_{j=1}^{N_c} a^g_{ij,n} a^l_{j,n} C_{j,n} G_n.$$

The material losses (which, explained before, are the joint name of the corrosive consumptions and impurity productions of materials) are composed of two parts: the first term is the quantity of consumptions, with $a^l_{i,n}$ being the factor of losses for the i-th component; the second term is the quantity of productions, with $a^g_{ij,n}$ being the factor of productions, counting in the i-th component production from the loss of the j-th component. To make this clearer, take the separation of SiF_4 as an example. Without loss of generality, here the loss results only from the reaction of SiF_4 with the moisture leaking into the cascade to produce the light impurity HF: $SiF_4 + H_2O = SiOF_2 + 2HF$. There are three components in the natural SiF_4 ($^{28}SiF_4$, $^{29}SiF_4$, $^{30}SiF_4$), and two components in HF (^1HF, ^2HF) (Note that HF is considered to have two components here for the sake of explaining the meaning of the factors $a^l_{i,n}$ and $a^g_{ij,n}$; in practice it is not necessary to treat ^1HF and ^2HF separately as two components). Therefore, during the separation of SiF_4, the process gas has actually five components. If a loss of 1% SiF_4 and no loss of HF occur when flowing through the centrifuges of a stage, then $a^l_{i,n} = 0 (i = 1, 2)$, $a^l_{i,n} = 0.01 (i = 3, 4, 5)$. Because a loss of 1 mole of any component in SiF_4 would produce 2 moles of HF (whose 99.985% is ^1HF and only 0.015% is ^2HF), $a^g_{ij,n} = 0$ $(i = 1, 2, \ldots, 5;\ j = 1, 2)$, $a^g_{ij,n} = 0$ $(i = 3, 4, 5;\ j = 3, 4, 5)$, $a^g_{ij,n} = 2 \times 0.99985$ $(i = 1;\ j = 3, 4, 5)$, $a^g_{ij,n} = 2 \times 0.00015$ $(i = 2;\ j = 3, 4, 5)$. Note that here the quantities of loss and production are measured in terms of mole.

Having determined $C'_{i,n}$, $C''_{i,n}$, $C'^a_{i,n}$, and $C''^a_{i,n}$, the concentration distribution of each component is determined.

For the feed stage ($n = N_F$), Eq. (1) becomes:

$$L'^a_n C'^a_{i,n} + L''^a_n C''^a_{i,n} + F C_i^F - L'_n C'_{i,n} - L''_n C''_{i,n} - m_{i,n} = 0, \tag{3}$$

with C_i^F being the concentration of the i-th component in the feed. In the product and waste pipes, according to the mass conservation, we have

$$\begin{aligned} L'^a_n C'^a_{i,n} &= L'_{n-1} C'_{i,n-1} - m'_{i,n-1}, \quad (n = 2, 3, \ldots, N), \\ L''^a_n C''^a_{i,n} &= L''_{n+1} C'_{i,n+1} - m''_{i,n+1}, \quad (n = 1, 2, \ldots, N-1). \end{aligned} \tag{4}$$

The quantities $m'_{i,n}$ and $m''_{i,n}$ are, respectively, the mass losses at the n-th stage in the product and waste pipes. Similarly to the mass loss $m_{i,n}$ in the gas centrifuges, the mass losses in the pipes are made up of also two parts:

$$\begin{aligned} m'_{i,n} &= a'^{l}_{i,n} C'_{i,n} L'_n - \sum_{j=1}^{N_c} a'^{g}_{ij,n} a'^{l}_{j,n} C'_{j,n} L'_n, \\ m''_{i,n} &= a''^{l}_{i,n} C''_{i,n} L''_n - \sum_{j=1}^{N_c} a''^{g}_{ij,n} a''^{l}_{j,n} C''_{j,n} L''_n, \end{aligned} \tag{5}$$

with the first term being the quantity of losses and the second term the quantity of productions.

The separation characteristics of a stage are described by the following empirical formula:

$$\frac{C'_{i,n}/C''_{i,n}}{C'_{j,n}/C''_{j,n}} = \gamma_{0,n}^{M_j - M_i}. \tag{6}$$

Here $\gamma_{0,n}$ is the so-called overall separation factor of unit mass difference, which is variant from stage to stage in reality but assumed to be constant for simplicity in most studies; M_i and M_j are the molar weights of the i-th and the j-th components, respectively. Note that equation (6) holds for small difference between M_i and M_j (usually between two isotopic components of the same element). Because later in the paper the light impurity HF is involved, the mass difference between HF and another component for isotope separation can be very large. In this case, there is no evidence that equation (6) still holds, but experimental observations show that it can be used in numerical simulation as long as the mass difference between the light impurity and other components is taken to be large enough. However, too large a difference should not be used to avoid arithmetic problems. Here the difference is fixed at 20.

When the hydraulic status is known, that is, the hydraulic parameters (e.g., L'_n, L''_n, L'^{a}_n, and L''^{a}_n) of each stage and the feed are known, from Equations (1), (3), (4), and (6), plus the restriction condition of the concentrations:

$$\sum_{i=1}^{N_c} C_{i,n} = \sum_{i=1}^{N_c} C'_{i,n} = \sum_{i=1}^{N_c} C''_{i,n} = 1, \tag{7}$$

the concentration distributions in the cascade can be determined by, say, using the q-iteration method.

THE EQUATION SYSTEM DESCRIBING THE HYDRAULIC STATUS

In the above determining the concentration distributions with the hydraulic status known is explained. However, when material losses exist, the hydraulic parameters are coupled with concentrations, which implies that the hydraulic status cannot be specified in advance. This can be seen from the following. The four equations in (4) and (5) lead to

$$\begin{aligned} L_n'^{\,a} &= L'_{n-1}\left[1-\sum_{i=1}^{N_c}(a_{i,n-1}'^{\,l}C'_{i,n-1}-\sum_{j=1}^{N_c}a_{ij,n-1}'^{\,g}a_{j,n-1}'^{\,l}C'_{j,n-1})\right], \\ L_n''^{\,a} &= L''_{n+1}\left[1-\sum_{i=1}^{N_c}(a_{i,n+1}''^{\,l}C''_{i,n+1}-\sum_{j=1}^{N_c}a_{ij,n+1}''^{\,g}a_{j,n+1}''^{\,l}C''_{j,n+1})\right]. \end{aligned} \tag{8}$$

Obviously, the hydraulic quantity $L_n'^{\,a}$ is correlated to $C'_{i,n-1}$, and $L_n''^{\,a}$ to $C''_{i,n+1}$, $i=1,2,\ldots,N_c$. Even if the hydraulic parameters can be given prior to the concentration distributions, because of the nonlinearity of Eq. (6), the hydraulic parameters change as the iteration process proceeds for the determination of the concentrations. Using G_n, θ_n, L'_n, and L''_n $(n=1,2,\ldots,N)$ as the unknown hydraulic parameters, the conservation relations for these parameters include Eq. (2) and the following two:

$$\left(G_n-\sum_{i=1}^{N_c}m_{i,n}\right)\theta_n-L'_n=0, \tag{9}$$

$$\left(G_n-\sum_{i=1}^{N_c}m_{i,n}\right)-L'_n+L''_n=0, \tag{10}$$

where θ_n is the cut of the n-th stage, defined as

$$\theta_n=\frac{L'_n}{L'_n+L''_n}.$$

For a cascade of N stages, there are $4N$ unknowns, the solution of which needs one more equation, in addition to Eqs. (2), (9), and (10). This missing equation is provided by the condition that makes the cascade "ideal". In the case of binary separation, this condition

is just the ideal cascade condition:

$$\begin{aligned} C'_{1,n} &= \frac{1}{1+R_1} \quad n=1 \\ C''_{1,n} &= \frac{1}{1+R_N} \quad n=N \\ C''^{\,a}_{1,n+1} &= C'^{\,a}_{1,n-1} = C_1^F \quad n=N_F \\ C''^{\,a}_{1,n+1} &= C'^{\,a}_{1,n-1} \quad 2 \le n < N_F,\ N_F < n \le N-1, \end{aligned} \tag{11}$$

where

$$R_1 = \frac{C_1^F}{C_2^F}\prod_{n=1}^{N_F}\frac{1}{\beta_n},\ R_N = \frac{C_1^F}{C_2^F}\alpha_N \prod_{n=N_F+1}^{N}\beta_n,$$

and α_n and β_n are respectively the enriching and stripping factors of the n-th stage. In the case of multicomponent separation, it is the quasi-ideal cascade condition. To be specific, for an MARC cascade, the condition is:

$$\begin{aligned} &R''_{j,n} && n=1 \\ &R'_{j,n} && n=N \\ &R_{j,n} = R'_{j,n-1} = R''_{j,n+1} = R_j^F && n=N_F \\ &R_{j,n} = R'_{j,n-1} = R''_{j,n+1} && 2 \le n < N_F, N_F < n \le N-1, \end{aligned} \tag{12}$$

where $R_{j,n}$ is the ratio of the concentrations of a chosen pair of components, the j-th and the the k-th component, at the confluent point of the n-th stage: $R'_{j,n} = C'^{\,a}_{j,n}/C'^{\,a}_{k,n}$, $R''_{j,n} = C''^{\,a}_{j,n}/C''^{\,a}_{k,n}$, and $R_j^F = C_j^F/C_k^F$. But for the quasi-ideal cascade requiring constant cuts of component flows in (6), the condition is:

$$\frac{L'_n}{G_n}\frac{C'_{j,n}}{C_{j,n}} = \text{const} \quad (1 \le n \le N), \tag{13}$$

namely, the cuts of the component flow of the j-th component are constant at all stages. It is worth pointing out that, when the separation factor is constant, the above equation implies the cuts of the component flows of all other components are also constant:

$$\frac{L'_n}{G_n}\frac{C'_{i,n}}{C_{i,n}} = \text{const} \quad (1 \le n \le N), \quad i \ne j.$$

When material losses take place, the above equation becomes

$$\frac{L'^{\,a}_{n+1}}{G_n}\frac{C'^{\,a}_{j,n+1}}{C_{j,n}} = \varphi_j = \text{const}, \quad n = 1, 2, \ldots, N. \tag{14}$$

Of course, other types of quasi-ideal cascades can be constructed by posing other conditions, based on requirements.

SOLUTION OF THE CONCENTRATION DISTRIBUTIONS

With a known hydraulic status, the concentration distributions in a cascade can be readily determined by using the q-iteration (12,17), which is described briefly as follows. For the i-th component, the four algebraic equations which its concentration satisfies are (1), (3), (4), and (6), and can be written as the following algebraic system:

$$\mathbf{Ax} = \mathbf{r}. \tag{15}$$

Here

$$\begin{aligned}\mathbf{x} &= \{\mathbf{x}_1, \ldots, \mathbf{x}_n, \ldots, \mathbf{x}_N\} \\ &= \{x_{1,1}, x_{2,1}, x_{3,1}, x_{4,1}, \ldots, x_{1,n}, x_{2,n}, x_{3,n}, x_{4,n}, \\ &\quad \ldots, x_{1,N}, x_{2,N}, x_{3,N}, x_{4,N}\} \\ &= \{C'_{i,1}, C''_{i,1}, C'^{\,a}_{i,1}, C''^{\,a}_{i,1}, \ldots, C'_{i,n}, C''_{i,n}, C'^{\,a}_{i,n}, C''^{\,a}_{i,n}, \\ &\quad \ldots, C'_{i,N}, C''_{i,N}, C'^{\,a}_{i,N}, C''^{\,a}_{i,N}\},\end{aligned}$$

$$\begin{aligned}\mathbf{r} &= \{\mathbf{r}_1, \ldots, \mathbf{r}_n, \ldots, \mathbf{r}_N\} \\ &= \{r_{1,1}, r_{2,1}, r_{3,1}, r_{4,1}, \ldots, r_{1,n}, r_{2,n}, r_{3,n}, r_{4,n}, \ldots, r_{1,N}, r_{2,N}, r_{3,N}, r_{4,N}\},\end{aligned}$$

and $\mathbf{A}$ is a block tri-diagonal matrix:

$$\mathbf{A} = \begin{bmatrix} \mathbf{B}_1 & \mathbf{C}_1 & & & & & & \\ \mathbf{A}_2 & \mathbf{B}_2 & \mathbf{C}_2 & & & & & \\ & \ddots & \ddots & \ddots & & & & \\ & & \mathbf{A}_n & \mathbf{B}_n & \mathbf{C}_n & & & \\ & & & \ddots & \ddots & \ddots & & \\ & & & & \mathbf{A}_{N-1} & \mathbf{B}_{N-1} & \mathbf{C}_{N-1} \\ & & & & & \mathbf{A}_N & \mathbf{B}_N \end{bmatrix},$$

with $\mathbf{A}_n, \mathbf{B}_n, \mathbf{C}_n$ are all 4×4 block matrices. From Eqs. (1), (3), (4) and (6) the non-zero elements of $\mathbf{A}_n, \mathbf{B}_n, \mathbf{C}_n$ can be obtained. Eqs. (1) and (3) are

$$B_{11,n}x_{1,n} + B_{12,n}x_{2,n} + B_{13,n}x_{3,n} + B_{14,n}x_{4,n} = r_{1,n},$$

where

$$B_{11,n} = L'_n,\ B_{12,n} = L''_n,\ B_{13,n} = -L'^{\,a}_n,\ B_{14,n} = -L''^{\,a}_n,$$

$$r_{1,n} = \begin{cases} FC_i^F - m_{i,n} & n = N_F \\ -m_{i,n} & n \neq N_F. \end{cases}$$

Eq. (6) is

$$B_{21,n}x_{1,n} + B_{22,n}x_{2,n} = r_{2,n},$$

with

$$B_{21,n} = 1,\ B_{22,n} = -q_{i,n},\ r_{2,n} = 0,$$

where

$$q_{i,n} = \frac{C'_{m,n}}{C''_{m,n}}\gamma_{0,n}^{M_m - M_i}.$$

According to (4), we have

$$A_{31,n}x_{1,n-1} + B_{33,n}x_{3,n} = r_{3,n},$$
$$C_{42,n}x_{2,n+1} + B_{44,n}x_{4,n} = r_{4,n},$$

where

$$A_{31,n} = -L'_{n-1}, \quad B_{33,n} = L'^{\,a}_n,$$
$$r_3 = -m'_{i,n-1},$$
$$C_{42,n} = -L''_{n+1}, \quad A_{44,n} = L''^{\,a}_n,$$
$$r_{4,n} = -m''_{i,n+1}.$$

Because Eq. (6) is nonlinear, the algebraic system (15) is nonlinear, and has to be solved by an iterative method. Different from ordinary iterative methods using the concentration C as the variable of iteration, the q-iteration method uses the ratio $q_{m,n} \equiv C'_{m,n}/C''_{m,n}$ as the iteration variable, that is, the ratio of the concentrations of a specified component (here the m-th component) in the product and waste flows. The ratio of the i-th component, according to (6), is

$$q_{i,n} \equiv C'_{i,n}/C''_{i,n} = q_{m,n}\gamma_{0,n}^{M_m - M_i}. \tag{16}$$

Clearly, under the assumption that $q_{m,n}$ $(n = 1, \ldots, N)$ are known, one can see that Eq. (6) is a linear equation with respect to the concentration. So solving Eq. (15) is easy. Then the concentration distributions of all components are obtained with assumed values of $q_{m,n}$. Definitely the assumed values are unlikely the correct values, so the obtained C', C'', $C'^{\,a}$, and $C''^{\,a}$ do not satisfy Condition (7), and the assumed values of $q_{m,n}$ need to be improved. With the improved values of $q_{m,n}$, Eq. (15) is solved again to give improved C', C'', $C'^{\,a}$, and $C''^{\,a}$. This

process is just the q-iteration, which is outlined below, and goes on until Condition (7) is satisfied.

1. Specify the initial value of $\mathbf{q} \equiv \{q_{m,1}, q_{m,2}, \ldots, q_{m,n}, \ldots, q_{m,N}\}$, and set the number of iterations $n_{\text{iter}} = 0$.
2. Solve equation system (15) for each component to obtain the concentration distributions $\{C'_{i,1}, C''_{i,1}, C'^{\,a}_{i,1}, C''^{\,a}_{i,1}, \ldots, C'_{i,n}, C''_{i,n}, C'^{\,a}_{i,n}, C''^{\,a}_{i,n}, \ldots, C'_{i,N}, C''_{i,N}, C'^{\,a}_{i,N}, C''^{\,a}_{i,N}\}$, $(i = 1, \ldots, N_c)$.
3. Improve $\mathbf{q}$:

$$\mathbf{q}_i = (1-\omega)\mathbf{q} + \omega \frac{\sum_{i=1}^{N_c} [\theta \mathbf{C}'_i + (1-\theta)\mathbf{C}''_i]}{\sum_{i=1}^{N_c} \gamma_0^{M_m - M_i} \mathbf{C}''_i},$$

where θ, $\mathbf{C}'_i$, $\mathbf{C}''_i$ and γ_0 are respectively $\theta \equiv \{\theta_1, \ldots, \theta_n, \ldots, \theta_N\}$, $\mathbf{C}'_i \equiv \{C'_{i,1}, \ldots, C'_{i,n}, \ldots, C'_{i,N}\}$, $\mathbf{C}''_i \equiv \{C''_{i,1}, \ldots, C''_{i,n}, \ldots, C''_{i,N}\}$, and $\gamma_0 \equiv \{\gamma_{0,1}, \ldots, \gamma_{0,n}, \ldots, \gamma_{0,N}\}$, ω is the under-relaxation factor, $0 < \omega \leq 1$. Set the number of iterations $n_{\text{iter}} := n_{\text{iter}} + 1$. In above formula the addition, multiplication, division and power are performed element-wise, for instance,

$$\theta \mathbf{C}'_i = \{\theta_1 C'_{i,1}, \ldots, \theta_n C'_{i,n}, \ldots, \theta_N C'_{i,N}\}.$$

4. Check whether either one of the following conditions is satisfied:

$$\max_n \left(|\sum_{i=1}^{N_c} C_{i,n} - 1|, |\sum_{i=1}^{N_c} C'_{i,n} - 1|, |\sum_{i=1}^{N_c} C''_{i,n} - 1| \right) \leq \varepsilon, \tag{17}$$

$$n_{\text{iter}} \leq N_{\text{itermax}}. \tag{18}$$

Here ε is a given small number and is set to 10^{-6} in later numerical experiments. N_{itermax} is the maximum number of iteration. If one of the above two conditions is satisfied, the iteration is terminated; if not, go back to (step 2).

SOLUTION OF THE HYDRAULIC EQUATIONS

Before the q-iteration has converged, the concentration distributions change in every iteration. Without material losses, the hydraulic parameters involved in the coefficient matrix and the right hand side of Eq. (15) are not functions of concentrations, in which case the concentration distributions can be obtained through enough number of q-iteration steps, independently of the hydraulic parameters. With

material losses, however, the hydraulic parameters are functions of concentrations, in which case the hydraulic status needs to be determined at every iteration step, that is, it is necessary to solve the hydraulic equations (2), (9), (10), and (12) as well if we would like to design an MARC cascade.

Eqs.(2), (9) and (10) can be written as

$$\begin{aligned} & G_n - \left(1 - \sum_{i=1}^{N_c} a'^{l}_{i,n-1} C'_{i,n-1} - \sum_{i=1}^{N_c}\sum_{j=1}^{N_c} a'^{g}_{ij,n-1} a'^{l}_{j,n-1} C'_{j,n-1}\right) L'_{n-1} \\ & - \left(1 - \sum_{i=1}^{N_c} a''^{l}_{i,n+1} C''_{i,n+1} - \sum_{i=1}^{N_c}\sum_{j=1}^{N_c} a''^{g}_{ij,n+1,} a''^{l}_{j,n+1} C''_{j,n+1}\right) L''_{n+1} \\ & = \begin{cases} F & n = N_F \\ 0 & n \neq N_F \end{cases}, \end{aligned} \tag{19}$$

$$\left(1 - \sum_{i=1}^{N_c} a^{l}_{i,n} C_{i,n} - \sum_{i=1}^{N_c}\sum_{j=1}^{N_c} a^{g}_{ij,n} a^{l}_{j,n} C_{j,n}\right) G_n \theta_n - L'_n = 0, \tag{20}$$

$$\left(1 - \sum_{i=1}^{N_c} a^{l}_{i,n} C_{i,n} - \sum_{i=1}^{N_c}\sum_{j=1}^{N_c} a^{g}_{ij,n} a^{l}_{j,n} C_{j,n}\right) G_n - L'_n - L''_n = 0. \tag{21}$$

The fourth equation, namely Eq. (12), cannot be directly used in the solution, because it is an equation regarding the concentrations rather than the hydraulic parameters. It is a little tricky to get round of this problem in a simple way. Here the following equation, which is independent of (19)–(21), is used to replace Eq. (12)

$$C_{i,n}\left(G_n - \sum_{l=1}^{N_c} m_{l,n}\right) - C'^{a}_{i,n-1} L'^{a}_{n-1} - C''^{a}_{i,n+1} L''^{a}_{n+1} = 0. \tag{22}$$

The above equation is a relation of mass conservation at the n-th stage and is the same as

$$\widetilde{C}_{i,n}\left(G_n - \sum_{l=1}^{N_c} m_{l,n}\right) - \widetilde{C}'^{a}_{i,n-1} L'^{a}_{n-1} - \widetilde{C}''^{a}_{i,n+1} L''^{a}_{n+1} = 0, \tag{23}$$

where

$$\widetilde{C}_n = \sum_{i=1}^{j} C_{i,n} \Big/ \sum_{i=1}^{N_c} C_{i,n},$$
$$\widetilde{C}'_n = \sum_{i=1}^{j} C'^{a}_{i,n+1} \Big/ \sum_{i=1}^{N_c} C'^{a}_{i,n+1}, \tag{24}$$
$$\widetilde{C}''_n = \sum_{i=1}^{j} C''^{a}_{i,n-1} \Big/ \sum_{i=1}^{N_c} C''^{a}_{i,n-1}.$$

Therefore, when concentrations are known, the solution of Eqs. (19)–(21) and (23) gives the hydraulic parameters. Of course, because Eq. (20) is nonlinear, iteration is necessary in the solution.

SATISFACTION OF THE QUASI-IDEAL CONDITION

There is an obvious problem in the solution of the hydraulic equations: since Eq. (23) and (12) are not equivalent, the hydraulic parameters obtained from the solution of equations (19)–(23) do not lead Eq. (12) to be satisfied automatically. Then the question is: how to take Eq. (12) into account in the solution of (19)–(23).

To allow the concentration distributions in the cascade to satisfy the required quasi-ideal condition, the key is to make the hydraulic status meet the corresponding requirement. Clearly, it is impossible to specify such a status in advance, but fairly easy to acquire this status through the following procedure. Suppose that a hydraulic status is specified rather arbitrarily. Because the hydraulic status is known, the concentration distributions can be derived. At this stage of calculation it is not necessary to obtain accurately the concentration distributions, so performing one or two steps of the q-iteration is sufficient. Now with the derived concentration distributions, the corresponding hydraulic status can be found by solving the hydraulic equations. One can imagine that if the derived concentration distributions are modified according to the quasi-ideal condition before being used in the solution of the hydraulic equations, the hydraulic status obtained would be closer to the wanted status than that initially specified. Then the specified hydraulic status can be replaced by the current status and the q-iteration can be invoked to generate new concentration distributions. This procedure is repeated until convergence, i.e., Condition (17) holds. We implement this procedure in the following manner.

In usual separation, it is desired that the components whose molar weight $M_i \geq M^*$ are enriched at the waste end of the cascade, whereas

those whose molar weight $M_i < M^*$ are enriched at the product end. So here for the MARC cascade considered we take $k = j + 1$, which gives $M^* = (M_j + M_k)/2$. After having obtained the concentration distributions by using one or two steps of q-iteration, the separation factor of a stage for the chosen pair of components can be estimated by:

$$\tilde{\gamma}_{jk,n} = \frac{C'^a_{j,n+1}/C''^a_{j,n-1}}{C'^a_{k,n+1}/C''^a_{k,n-1}}. \tag{25}$$

Note that in the calculation the concentrations use the values at the confluent point $C'^a_{\cdot,n+1}$ and $C''^a_{\cdot,n-1}$ other than $C'_{\cdot,n}$ and $C''_{\cdot,n}$, since the matching location of the abundance ratio is at the confluent point. The difference of this separation factor from $\gamma_{jk,n} = \gamma_{0,n}^{M_k - M_j}$ lies in that Eq. (25) reckons in the influence of the pipes. When material losses are naught,

$$\tilde{\gamma}_{jk,n} = \gamma_{jk,n} = \frac{C'_{j,n}/C''_{j,n}}{C'_{k,n}/C''_{k,n}}.$$

It is usually required that the separation between the chosen pair of components be symmetrical, so we impose at a stage (here the feed stage)

$$\tilde{\alpha}_{jk,n} = \tilde{\beta}_{jk,n} = \sqrt{\tilde{\gamma}_{jk,n}}\ (n = N_F),$$

and

$$\begin{aligned} &\tilde{\alpha}_{jk,n} = \tilde{\beta}_{jk,n+1}, \tilde{\beta}_{jk,n} = \tilde{\gamma}_{jk,n}/\tilde{\alpha}_{jk,n}\ (1 \le n < N_F), \\ &\tilde{\beta}_{jk,n} = \tilde{\alpha}_{jk,n-1}, \tilde{\alpha}_{jk,n} = \tilde{\gamma}_{jk,n}/\tilde{\beta}_{jk,n}\ (N_F < n \le N) \end{aligned} \tag{26}$$

because of the matching requirement. Of course, the requirement for symmetrical separation is not necessary. If needed, other types of separation can be requested, for instance, $\tilde{\alpha}_{jk,n} = \tilde{\gamma}_{jk,n}^{3/8}$ and $\tilde{\beta}_{jk,n} = \tilde{\gamma}_{jk,n}^{5/8}$ at $n = N_F$. Then the abundance ratios at all confluent points and at the two cascade ends can be readily calculated

$$\begin{aligned} &R_{j,N_F} = R_j^F; \\ &R_{j,n} = R_{j,n+1}/\tilde{\beta}_{jk,n+1}\ (1 \le n < N_F);\ R''_{j,1} = R_{j,1}/\tilde{\beta}_{jk,1}; \\ &R_{j,n} = R_{j,n-1}\tilde{\beta}_{jk,n}\ (N_F < n \le N);\ R'_{j,N} = R_{j,N-1}\tilde{\alpha}_{jk,N}. \end{aligned} \tag{27}$$

At the moment we know that the rough values of $C_{i,n}$, $C'_{i,n}$, $C''_{i,n}$, $C'^a_{i,n}$, and $C''^a_{i,n}$ from the q-iteration, which are now modified to be used in the solution of the hydraulic equations. Using the concentrations of the k-th component at the confluent point we calculate the concentrations of the j-th component, indicated with a hat: $\widehat{C}_{j,n} = C_{k,n}R_{jk,n}$,

$\widehat{C}'^{a}_{j,n} = C'^{a}_{k,n} R_{jk,n}$, $\widehat{C}''^{a}_{j,n} = C''^{a}_{k,n} R_{jk,n}$. Replace $C_{j,n}$, $C'^{a}_{j,n}$, and $C''^{a}_{j,n}$ in Eq. (24) by $\widehat{C}$, namely

$$\begin{aligned}
\widetilde{C}_n &= \frac{\sum_{i=1}^{j-1} C_{i,n} + \widehat{C}_{j,n}}{\sum_{i=1}^{j-1} C_{i,n} + \widehat{C}_{j,n} + \sum_{i=j+1}^{N_c} C_{i,n}}; \\
\widetilde{C}'_n &= \frac{\sum_{i=1}^{j-1} C'^{a}_{i,n+1} + \widehat{C}'^{a}_{j,n+1}}{\sum_{i=1}^{j-1} C'^{a}_{i,n+1} + \widehat{C}'^{a}_{j,n+1} + \sum_{i=j+1}^{N_c} C'^{a}_{i,n+1}}; \\
\tilde{C}''_n &= \frac{\sum_{i=1}^{j-1} C''^{a}_{i,n-1} + \hat{C}''^{a}_{j,n+1}}{\sum_{i=1}^{j-1} C''^{a}_{i,n-1} + \hat{C}''^{a}_{j,n+1} + \sum_{i=j+1}^{N_c} C''^{a}_{i,n-1}}.
\end{aligned} \tag{28}$$

Then after the solution of the hydraulic equations, go back to the q-iteration to find the concentration distributions again. This goes on until convergence:

1. Specify the initial hydraulic status of the cascade: $G_n, \theta_n, L'_n, L''_n$, and concentration distributions: C', C'', C'^{a}, C''^{a};
2. Perform one step of q-iteration and update C', C'', C'^{a}, C''^{a};
3. If Condition (17) or (18) is satisfied, terminate the computation; or continue;
4. Calculate $\tilde{\gamma}_{jk,n}$, $\tilde{\alpha}_{jk,n}$, $\tilde{\beta}_{jk,n}$ from Eqs. (25) and (26);
5. Calculate the ratios $R_{j,n}$ at the confluent points from Eq. (12);
6. Calculate $\widetilde{C}$ from (28);
7. Solve the hydraulic equations (19)–(21) and (23) to obtain $G_n, \theta_n, L'_n, L''_n$, and return to step 2.

Table 1. The overall separation factors $\gamma_{0,n}$ of unit difference at all stages

n	1	2	3	4	5	6	7	8	9	10	11
$\gamma_{0,n}$	1.359	1.410	1.439	1.447	1.369	1.402	1.390	1.377	1.425	1.359	1.406
n	12	13	14	15	16	17	18	19	20	21	
$\gamma_{0,n}$	1.408	1.431	1.409	1.401	1.438	1.450	1.423	1.447	1.380	1.393	

Table 2. The molar weights M_i and the concentrations C_i^F in the feed

i	1	2	3	4	5
M_i	20	134	135	136	138
C_i^F	0.0	0.1	0.3	0.2	0.4

A little explanation is in place. In the solution of the hydraulic equations, not all cascades need consideration of how to satisfy the quasi-ideal condition by modifying the concentrations distributions, depending on the form of the condition. Before the end of this section, using the quasi-ideal cascade in (6), we discuss a little how to handle this quasi-ideal condition. This cascade requires Condition (14) be satisfied, i.e.,

$$\varphi_j C_{j,n} G_n - C'^a_{j,n+1} L'^a_{n+1} = 0.$$

Different from an MARC cascade, not only included are the quantities of concentrations, but also the hydraulic parameters. For a condition posed this way, we simply use it to replace Eq. (23) as one of the hydraulic equation. In this manner there is no need for a tricky treatment of the condition as for an MARC cascade, and so the solution process is simpler.

NUMERICAL EXPERIMENTS

Here the numerical experiments are meant for demonstrating the feasibility of the implementation in a numerical method of the above ideas. Consider a cascade of length $N = 21$, with the feed stage at $N_F = 7$. The overall separation factors of unit mass difference $\gamma_{0,n}$ for the stages are given in Table 1.

In practice the separation factors are hardly identical for different stages, so in the table the separation factors are given at random,

Table 3. The material loss factors $a^l_{i,n}$, $a'^l_{i,n}$, $a''^l_{i,n}$ and production factors $a^g_{ij,n}$, $a'^g_{ij,n}$, $a''^g_{ij,n}$ for the four cases considered

	$a^l_{i,n}$		$a'^l_{i,n}$		$a'^l_{i,n}$		$a^g_{ij,n}$, $a'^g_{ij,n}$, $a''^g_{ij,n}$	
$i(j)$	1	2,3,4,5	1	2,3,4,5	1	2,3,4,5	1(2,3,4,5)	other
Case 1	0	0	0	0	0	0	0	0
Case 2	0	10^{-7}	0	1.1×10^{-7}	0	1.2×10^{-7}	1.0,1.3,0.4,0.7	0
Case 3	0	10^{-5}	0	1.1×10^{-5}	0	1.2×10^{-5}	1.0,1.3,0.4,0.7	0
Case 4	0	10^{-3}	0	1.1×10^{-3}	0	1.2×10^{-3}	1.0,1.3,0.4,0.7	0

Table 4. The values of $R_{j,n}$ at the confluent points and the concentrations in the product and waste withdrawals for case 1

n	1	2	3	4	5	6	7	8	9	10	11
$R'^{a}_{j,n}$	–	.09269	.1215	.1919	.2544	.3597	.5000	.6950	.9481	1.411	1.751
$R_{j,n}$	.06111	.09269	.1215	.1919	.2544	.3597	.5000	.6950	.9481	1.411	1.751
$R''^{a}_{j,n}$	.06111	.09269	.1215	.1919	.2544	.3597	.5000	.6950	.9481	1.411	1.751

n	12	13	14	15	16	17	18	19	20	21
$R'^{a}_{j,n}$	2.780	3.471	5.713	6.891	11.21	14.25	23.58	28.86	49.36	54.95
$R_{j,n}$	2.780	3.471	5.713	6.891	11.21	14.25	23.58	28.86	49.36	54.95
$R''^{a}_{j,n}$	2.780	3.471	5.713	6.891	11.21	14.25	23.58	28.86	49.36	–

		i	1	2	3	4	5
P	.578776	$C'_{i,N}$	.000000	.172604	.513114	.311035	.003247
W	.421224	$C''_{i,N}$	.000000	.000239	.007174	.047434	.945153

Table 5. The values of $R_{j,n}$ at the confluent points and the concentrations in the product and waste withdrawals for Case 2

n	1	2	3	4	5	6	7	8	9	10	11
$R'^{a}_{j,n}$	–	.09269	.1215	.1919	.2544	.3597	.5000	.6950	.9481	1.411	1.751
$R_{j,n}$	.06111	.09269	.1215	.1919	.2544	.3597	.5000	.6950	.9481	1.411	1.751
$R''^{a}_{j,n}$	.06111	.09269	.1215	.1919	.2544	.3597	.5000	.6950	.9481	1.411	1.751

n	12	13	14	15	16	17	18	19	20	21	
$R'^{a}_{j,n}$	2.790	3.471	5.713	6.891	11.21	14.25	23.58	28.86	49.36	54.95	
$R_{j,n}$	2.790	3.471	5.713	6.891	11.21	14.25	23.58	28.86	49.36	54.95	
$R''^{a}_{j,n}$	2.790	3.471	5.713	6.891	11.21	14.25	23.58	28.86	49.36	–	–

		i	1	2	3	4	5
P	.578777	$C'_{i,N}$	.000016	.172602	.513107	.311027	.003247
W	.421221	$C''_{i,N}$	$.18 \times 10^{-8}$	.000239	.007174	.047435	.945151

Table 6. The values of $R_{j,n}$ at the confluent points and the concentrations in the product and waste withdrawals for Case 3

n	1	2	3	4	5	6	7	8	9	10	11
$R'^{a}_{j,n}$	–	.09296	.1218	.1925	.2550	.3605	.5009	.6961	.9497	1.414	1.754
$R_{j,n}$	.06129	.09295	.1218	.1924	.2549	.3604	.5007	.6962	.9498	1.414	1.754
$R''^{a}_{j,n}$	.06129	.09294	.1218	.1924	.2549	.3603	.5009	.6963	.9499	1.414	1.754

n	12	13	14	15	16	17	18	19	20	21
$R'^{a}_{j,n}$	2.795	3.478	5.723	6.902	11.23	14.27	23.61	28.89	49.42	55.01
$R_{j,n}$	2.795	3.477	5.722	6.902	11.23	14.27	23.60	28.89	49.42	55.01
$R''^{a}_{j,n}$	2.795	3.477	5.722	6.901	11.23	14.27	23.60	28.89	49.42	-

		i	1	2	3	4	5
P	.578838	$C'_{i,N}$	.001600	.172449	.512453	.310263	.003236
W	.420926	$C''_{i,N}$	$.18 \times 10^{-6}$	.000241	.007202	.047569	.944988

Table 7. The values of $R_{j,n}$ at the confluent points and the concentrations in the product and waste withdrawals for Case 4

n	1	2	3	4	5	6	7	8	9	10	11
$R'^{a}_{j,n}$	–	.1242	.1604	.2492	.3191	.4400	.5901	.7921	1.097	1.644	2.044
$R_{j,n}$	.08066	.1208	.1557	.2380	.3082	.4208	.5661	.7995	1.100	1.642	2.036
$R''^{a}_{j,n}$	.08066	.1203	.1524	.2348	.3002	.4100	.5784	.8096	1.107	1.641	2.013

n	12	13	14	15	16	17	18	19	20	21	
$R'^{a}_{j,n}$	3.244	3.991	6.516	7.740	12.50	15.65	25.69	31.15	53.07	58.99	
$R_{j,n}$	3.207	3.971	6.422	7.705	12.34	15.59	25.49	31.13	52.96	58.99	
$R''^{a}_{j,n}$	3.182	3.899	6.369	7.568	12.22	15.38	25.35	30.97	52.91	-	

		i	1	2	3	4	5
P	.582453	$C'_{i,N}$	.141785	.158495	.453072	.244268	.002379
W	.398581	$C''_{i,N}$	.000019	.000375	.010693	.062313	.926600

simulating the fluctuation in reality. The fluctuation results from small manufacture discrepancy, differences in flow rates, and cuts at different stages, etc. Of course the range of fluctuation in the table is exaggerated somewhat. Without loss of generality, it is assumed that the process gas consists of five components, with the first component being the impurity, produced from the corrosive consumption of other components. The molar weights and the concentrations in the feed of the five components are given in Table 2.

For MARC cascade, we consider four cases: one without material losses and three with material losses. In Table 3, the material loss factors and production factors are given. Note that these factors are meant only for theoretical research, not aiming at practical situations.

In Tables 4–6, the values of $R_{j,n}$ (taking $j = 4, k = 5$) at the confluent points are presented for the four cases, as well as the concentrations in the product and waste withdrawals. Tables are used here instead of more intuitionistic curve plots because from the latter it is hard to see clearly the differences of values at the confluent points.

It can be seen from Tables 4–7 that, as the material losses increase, matching the abundance ratios exactly is no longer possible at the confluent points. Therefore, for MARC cascades the abundance ratios cannot always be matched in any case.

CONCLUSIONS

A numerical method is presented for the calculation and design of quasi-ideal cascades. This method implements a process of solving hydraulic status in the q-iteration method of determining the concentration distributions with a given hydraulic status. Material losses can be handled in the method.

If hydraulic parameters are included in the condition that defines the quasi-ideal cascade, for instance, the condition that requires constant cuts of component flows, then the condition can be used as an equation in the system of hydraulic equations describing the cascade hydraulic status. But if the condition is one posed on concentrations, for instance, the condition in an MARC cascade that requires the match of abundance ratios, it cannot be directly used as one of the hydraulic equations, and needs some special treatments: using a relation of mass conservation instead of the condition as one of the hydraulic equations, then using the condition to adjust the concentration distributions obtained by the q-iteration, and using the adjusted distributions in the calculation of the coefficient matrix of the hydraulic equation system.

Numerical experiments are carried out to verify the method for an MARC cascade in four different cases of material losses. The results show that this method is feasible and effective. It is found that the match of the abundance ratios is possible when the material losses are naught or small; the match is impossible when the material losses become large, but in this case an approximate match is given by the method.

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