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Publisher *Taylor & Francis*

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Separation Science and Technology

Publication details, including instructions for authors and subscription information:

<http://www.informaworld.com/smpp/title~content=t713708471>

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Online publication date: 11 September 2000

To cite this Article Zeng, Shi and Ying, Chunlong(2000) 'A Method of Separating a Middle Component in Multicomponent Isotope Mixtures by Gas Centrifuge Cascades', *Separation Science and Technology*, 35: 14, 2173 — 2186

To link to this Article: DOI: 10.1081/SS-100102096

URL: <http://dx.doi.org/10.1081/SS-100102096>

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A Method of Separating a Middle Component in Multicomponent Isotope Mixtures by Gas Centrifuge Cascades

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ABSTRACT

Separating a certain middle component of isotopes from a multicomponent isotopic mixture is much more difficult than separating the two end isotope components. Numerical investigation revealed that by controlling the cut of a separation cascade, defined as the ratio of the product rate to the feed rate, it is always possible to separate a multicomponent mixture into two specified groups of components, a light group and a heavy group, in just one separation run. The cut is equal to the concentration sum in the feed of the components in the light group. The effects of the cascade length, shape, and feed location were studied. The results suggest that without using complicated cascades such as the M-cascade or the Q-cascade the separation of the middle component can be easily achieved.

Key Words. Multicomponent isotope separation; Gas centrifuge cascade; Middle component

INTRODUCTION

It is much more difficult to separate a middle component of isotopes from a multicomponent isotopic mixture than to separate an end component (i.e., the component with the largest molar weight and that with the smallest molar

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weight). For separating an end component, it is possible in principle to enrich the component to an arbitrarily high concentration within just one separation run, provided that the cascade is sufficiently long. But this does not apply to the separation of a middle component. Therefore, separating a middle component may require splitting the mixture into two parts such that in one part the middle component becomes an end component, then separating this part to enrich the middle component to the required concentration. In the best case, at least two runs are needed to enrich a middle component to a desired concentration.

Making a middle component an end component in only one separation run may be accomplished by Method A, using an M-cascade, or by Method B, using a Q-cascade. An M-cascade is the so-called Matched Abundance-Ratio cascade proposed by De La Garza (1). The abundance ratio is the ratio of the concentration of the component to that of a key component. At a mixing point in an M-cascade the abundance ratios of a component are equal for the joining streams. Let M^* represent the arithmetic average of the molar weights of the key component and the component whose abundance ratio is to be matched. By properly choosing the two components, those components whose molar weights are greater than M^* will appear only in the waste stream of the cascade, and those components whose molar weights are less than M^* will only appear in the product stream. Thus a middle component will be an end component if the molar weights of other components do not lie between the molar weight of the middle component and M^* . A Q-cascade is also a special cascade proposed in Refs. (2) and (3). For each component a parameter Q can be defined according to the separation factor relative to a specified component. Similar to an M-cascade, by properly choosing the parameter Q_s such that $Q > 0$ for the lighter components that are to appear in the product stream and $Q < 0$ for the heavier components that are to appear in the waste stream, it is possible to make a middle component become an end component after one separation run. Additional methods can be used to separate a middle component, e.g., Method C, using a cascade with an intermediate withdrawal at the position where the concentration of the middle component reaches maximum, or Method D using whatever cascade is available to get rid of the heavier (or lighter) components step by step.

Methods A and B have not been put into practice, perhaps because of their complexities, e.g., the stage flow rates are all different from stage to stage. Such a complex cascade is only suitable for separating one specific component in a multicomponent mixture; that is, a new cascade has to be designed if another component in the mixture is to be separated or a component in a new mixture is wanted. Moreover, the large-scale separation of stable isotopes is not yet carried out, as for the separation of uranium isotopes. Therefore, Methods C and especially D are more often used (4), because they are easily

performed. With Method C however, it is necessary to determine where the maximum is located not only theoretically but also experimentally. This introduces some complexities into the cascade operation. Furthermore, no matter how large the enrichment obtained for the middle component, it remains a middle component. This means that when a sufficiently high concentration is required, several runs have to be performed, using the intermediate withdrawal of the current run as the feed of the next run. Of course, if the concentration required is not very high or the concentration in the feed is high enough, it is possible to obtain the required concentration in just one run. Method D is perhaps the simplest method mentioned here. With this method, one or more heavier (or lighter) components are eliminated in several runs, and eventually the middle component becomes an end component or reaches the required concentration. Obviously, Methods C and D are, generally speaking, not able to separate a middle component in two runs; i.e., they do not work as efficiently as Methods A and B in terms of number of runs. Apart from this, Methods C and D do not require a special cascade to function; a simplest square cascade would work.

It is desirable to have a cascade, which can be as simple as a square cascade and as efficient as an M- or Q-cascade, that can turn any middle component into an end component in just one separation run. In this paper, by numerical experiments it is demonstrated that this can be realized simply by adjusting the cut of a cascade, defined to be the ratio of the product rate to the feed rate, regardless of the shape and feed location, as long as the cascade is sufficiently long.

THE DIFFERENCE EQUATIONS DESCRIBING THE CONCENTRATION DISTRIBUTION IN A CASCADE

The separation cascade considered here consists of gas centrifuges and is illustrated in Fig. 1. Here F , P , and W are the feed, the product, and the waste, respectively. Let there be N_c number of components in the multicomponent mixture, and N number of stages in the cascade. The concentration of the i th component in the feed is C_i^F . At stage n the concentrations of the i th component are $C_{i,n}$ in the entering flow, $C'_{i,n}$ in the head flow, and $C''_{i,n}$ in the tail flow. The interstage upflowing flow rate from stage n to $n + 1$ is denoted by L_n . Mass conservation for the i th component and the total mass at steady state yields

$$L_{n-1} C'_{i,n-1} + (L_n + a) C''_{i,n+1} + c = L_n C'_{i,n} + (L_{n-1} + b) C''_{i,n} \quad (1)$$

$$(L_n + L_{n-1} + b) C_{i,n} = L_n C'_{i,n} + (L_{n-1} + b) C''_{i,n} \quad (2)$$

$$F = P + W \quad (3)$$

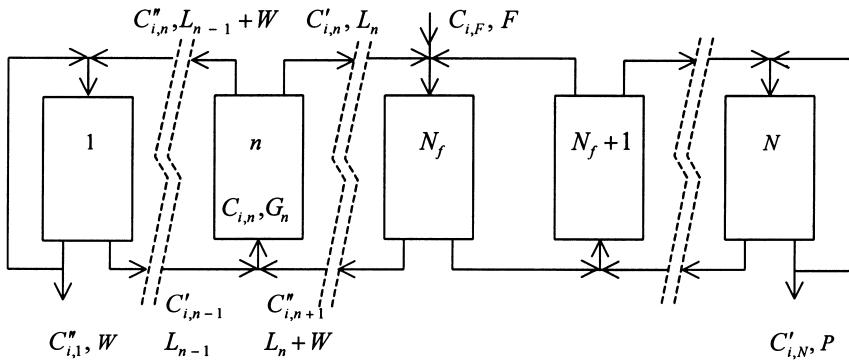


FIG. 1 A schematic illustration of a cascade.

with

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

Clearly $C'_{i,0} = C''_{i,1}$ is at the waste end of the cascade and $C''_{1,N+1} = C'_{i,N}$ at the product end. For gas centrifuges, the following relationship holds for the separation factor γ_{ij} (5):

$$\gamma_{ij} = \frac{C'_i}{C''_i} / \frac{C'_j}{C''_j} = \gamma_0^{M_j - M_i} \quad (4)$$

where M_i and M_j are the molar weights of the i th and the j th components, and γ_0 is the unit separation factor. Equations (1)–(4) are the set of difference equations describing the concentration distribution in a cascade at steady state. All quantities, except the concentrations C , C' , and C'' are specified or determined according to the cascade performance optimization or some practical considerations. The concentrations should satisfy the following constraint

$$\sum_i C_{i,n} = \sum_i C'_{i,n} = \sum_i C''_{i,n} = 1 \quad (5)$$

THE SOLUTION METHOD

The solution of the difference equations may use classical iterative methods, which are outlined in Ref. (6). Because these methods encounter problems in choosing appropriate initial values or consuming large amount of computation time, here the q -iteration method proposed in Ref. (7) is em-

ployed for better computation performance. The transient method (cf. Ref. 8) based on the q -iteration method can also be used. It is only used here to cope with very the few situations in which the q -iteration method does not work well. Because the q -iteration method is new an explanation follows.

Let $q_{k,n} = C_{k,n}'/C_{k,n}''$ for the k th component, which can be chosen arbitrarily. Because the difference equations contain nonlinearity, which arises from Eq. (4), iteration is necessary for the solution. Starting the solution from a given initial value of $q_{k,n}$ for each stage, we have from Eq. (4)

$$C_{i,n}' = q_{i,n} C_{i,n}'' \quad (6)$$

with $q_{i,n} = q_{k,n} \gamma_0^{M_k - M_i}$. Substituting Eq. (6) into Eq. (1) gives

$$\begin{aligned} -L_{n-1} q_{i,n-1} C_{i,n-1}'' + [L_n q_{i,n} + (L_{n-1} + b)] C_{i,n}'' \\ - (L_n + a) C_{i,n+1}'' = -c \end{aligned} \quad (7)$$

which is just a linear equation system with a tridiagonal coefficient matrix and is easily solved. After C'' is obtained, C and C' are readily calculated from Eqs. (2) and (6). However Constraint (5) is unlikely to be satisfied, because the value of $q_{k,n}$ for each stage is assumed. To improve the value of $q_{k,n}$

$$\sum_i q_{k,n} \gamma_0^{M_k - M_i} C_{i,n}'' = \sum_i C_{i,n} \quad (8)$$

is required; that is, $\sum_i C_{i,n}' = \sum_i C_{i,n}$, which implies $\sum_i C_{i,n}'' = \sum_i C_{i,n}$ because of Eq. (2). A new value of $q_{k,n}$ can be derived from Eq. (8):

$$q_{k,n} = \frac{\sum_i C_{i,n}}{\sum_i \gamma_0^{M_k - M_i} C_{i,n}''} \quad (9)$$

It is unlikely that this value of $q_{k,n}$, denoted by $q_{k,n}^{\text{new}}$, is equal to the previous value of $q_{k,n}$, denoted by $q_{k,n}^{\text{old}}$. Therefore, the next step of iteration uses a new value of $q_{k,n}$, which is given by

$$q_{k,n} = (1 - \omega) q_{k,n}^{\text{old}} + \omega q_{k,n}^{\text{new}} \quad (10)$$

Here ω is the weighting factor, and $0 < \omega \leq 1$. The iteration carries on until a termination criterion is satisfied, which here is defined by

$$\max_n (|\sum_i C_{i,n} - 1|, |\sum_i C_{i,n}' - 1|, |\sum_i C_{i,n}'' - 1|) \leq \varepsilon \quad (11)$$

where ε is a given small number. In our numerical investigations, $\omega = 0.8$ and $\varepsilon = 10^{-8}$. Equation (12) shows that the constraint is also satisfied. On convergence, we obtain

$$\sum_i C_{i,n} = \sum_i C_{i,n}' = \sum_i C_{i,n}'' \quad (12)$$

Summing up Eq. (1) over i and taking into account Eq. (12) give

$$-L_{n-1} \sum_i C_{i,n-1} + (L_n + L_{n-1} + b) \sum_i C_{i,n} - (L_n + a) \sum_i C_{i,n+1} = d \quad (13)$$

where $d = F$ for $n = N_F$ because $\sum_i C_i^F = 1$, but $d = 0$ for all other values of n . Equation (13) can be viewed as a linear equation system with respect to variables $\sum_i C_{i,n}$, $n = 1, 2, \dots, N$. Because $F \neq 0$, and the diagonal element $L_n + L_{n-1} + b$ is no smaller than the sum of the absolute values of the two off-diagonal elements L_{n-1} and $L_n + a$, the coefficient matrix is not singular and the equation system has a unique solution. It is readily verified that the solution is 1.

THE IDEA FOR SEPARATING A MIDDLE COMPONENT

For ease of reference, the components in a multicomponent isotopic mixture are numbered according to their molar weights in an ascending order. The components are divided into two groups, a light group and a heavy group. The molar weight of any component in the light group is smaller than that of any component in the heavy group; i.e., the sequence number of a component in the light group is always smaller than that of a component in the heavy group. Depending on some practical considerations, the middle component can be put in either group: if it goes into the light group it is the heaviest component; if it belongs to the heavy group it is the lightest component. Therefore, if the two groups can be separated, that is, if the light group appears only in the product and the heavy group only in the waste, the middle component must be an end component in the product or the waste. To evaluate to what extent the two groups are separated, we define a function D

$$D = \frac{P}{F} \sum_{i=1}^{N'_c} C'_{i,N} + \frac{W}{F} \sum_{i=N'_c+1}^{N_c} C''_{i,1} \quad (14)$$

where N'_c is the number of components in the light group. It is clear that

$$D \leq \frac{P}{F} \sum_{i=1}^{N_c} C'_{i,N} + \frac{W}{F} \sum_{i=1}^{N_c} C''_{i,1} = \frac{P+W}{F} = 1 \quad (15)$$

The equality holds only when the two groups are completely separated, i.e.,

$$\frac{P}{F} = \sum_{i=1}^{N'_c} C_i^F, \quad \frac{W}{F} = \sum_{i=N'_c+1}^{N_c} C_i^F \quad (16)$$

and also

$$\sum_{i=1}^{N_c} C'_{i,N} = 1, \quad \sum_{i=N_c+1}^{N_c} C''_{i,1} = 1 \quad (17)$$

This means that the function D reaches its maximum when the two groups are completely separated. The first equation in Eq. (16) suggests that if the cascade cut, defined to be P/F , is taken to be $\sum_{i=1}^{N_c} C_i^F$, i.e., the concentration sum in the feed of the components in the light group, then the cascade may be able to separate the components into the two intended groups, which will be shown in the numerical experiments in the following section.

Note that Eq. (16) implies that the concentration of the j th component in the light group (i.e., $1 \leq j \leq N_c$) is

$$C'_{j,N} = C_j^F / \sum_{i=1}^{N_c} C_i^F \quad (18)$$

and that of the j th component in the heavy group (i.e. $N_c < j \leq N_c$) is

$$C''_{j,1} = C_j^F / \sum_{i=N_c+1}^{N_c} C_i^F \quad (19)$$

Equations (18) and (19) are obtained by Minenko (9) as a limiting property of infinitely long cascades, which means that the maximum concentration for the j th component achieved at the two ends of a cascade cannot exceed the values given by Eqs. (18) and (19). According to Eq. (16), the two equations can be interpreted as the following more explicit forms

$$C'_{j,N} = C_j^F / (P/F), \quad C''_{j,1} = C_j^F / (1 - P/F) \quad (20)$$

Note that the j th component is in either the light group or the heavy group. If a fraction ϕ of it is in the light group, with $0 < \phi \leq 1$, then Eq. (20) should be modified as

$$C'_{j,N} = \phi C_j^F / (P/F), \quad C''_{j,1} = (1 - \phi) C_j^F / (1 - P/F) \quad (21)$$

NUMERICAL EXPERIMENTS

Without loss of generality, the process gas is taken to be Xe, which has nine stable isotopes. The natural concentrations of the nine components are presented in Table 1.

To make an end component in one separation run, the component ^{130}Xe is chosen because its concentration is small and it is definitely hard to separate.

TABLE 1
Natural Concentrations of Stable Isotopes of Xe

^{124}Xe	^{126}Xe	^{128}Xe	^{129}Xe	^{130}Xe	^{131}Xe	^{132}Xe	^{134}Xe	^{136}Xe
0.00093	0.0009	0.01917	0.2644	0.0408	0.2118	0.2689	0.1044	0.0887

The following experiments show that it is possible by simply adjusting the cut of the cascade to make ^{130}Xe an end component in only one separation run for: (a) different lengths of cascades if the cascades are long enough; (b) different shapes of cascades; and (c) different feed locations. For other cases similar experiments can be conducted.

Experiment 1: Different Lengths of Cascades

Here we consider a square cascade with constant entering flow rate $G_n = L_{n-1} + L_n + b$ at each stage and $G_n/F = 10$. The unit separation factor $\gamma_0 = 1.4$. Cascades with five different lengths ($N = 5, 11, 21, 41$, and 61) are investigated. The feed locations are at the centers of the cascades. The goal is for ^{130}Xe to be contained in the waste, and thus $N'_c = 4$. The components whose molar weights are smaller than that of ^{130}Xe are ^{124}Xe , ^{126}Xe , ^{128}Xe , and ^{129}Xe and should appear in the products. Figure 2 plots the values of the function D versus the cascade cut P/F for the above different values of N . It is clearly seen that for all lengths there exist maximums, which all occur at the same location $F/P = 0.2854$. This value is exactly the sum of the concentrations of the components in the light group. Is this a coincidence? To be more convincing, another component, ^{132}Xe , is chosen and it, as well as all other components with lighter molar weights, should be in the product; that is, the light group consists of components ^{124}Xe , ^{126}Xe , ^{128}Xe , ^{129}Xe , ^{130}Xe , ^{131}Xe , and ^{132}Xe and thus $N'_c = 7$. The sum of concentrations of these components in the feed is 0.8069. A similar plot to Fig. 2 is given in Fig. 3. The curve for $N = 41$ is not distinguishable from that for $N = 61$ in the figure. Examining the curves we find that the maximums occur at the cascade cuts being equal to 0.8069 for all lengths except for $N = 5$. When a cascade is too short, like $N = 5$ here, the maximum may not take place at the exact expected location. The reason is that the stripping section of the cascade, which is to the left of the feed location, may not be able to completely separate the components belonging to the light group, and the enriching section, which is to the right of the feed location, may not be able to completely separate the components belonging to the heavy group. Therefore, both sections should not be too short. Table 2 presents the maximum values of the function D corresponding to the curves in Figs. 2 and 3.

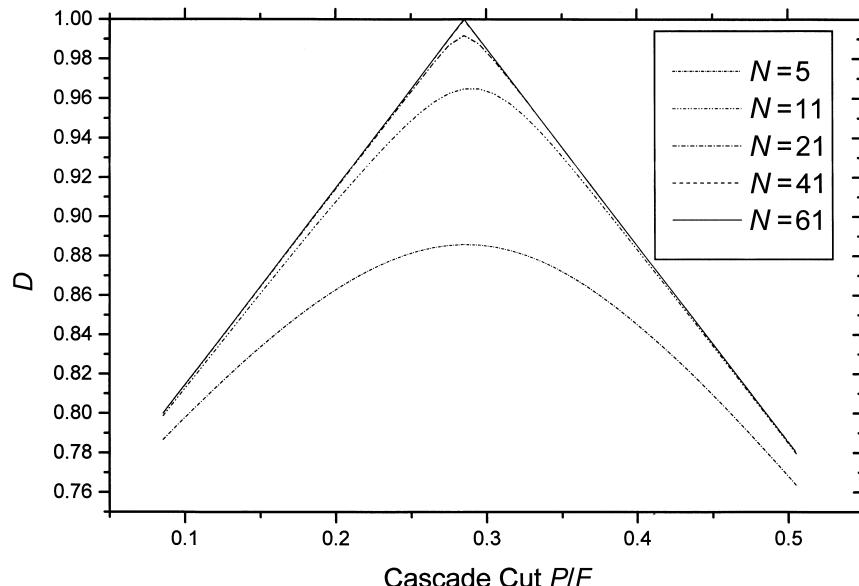


FIG. 2 The value of D as a function of cascade cut P/F for different cascade lengths. The light group contains the components ^{124}Xe , ^{126}Xe , ^{128}Xe , and ^{129}Xe .

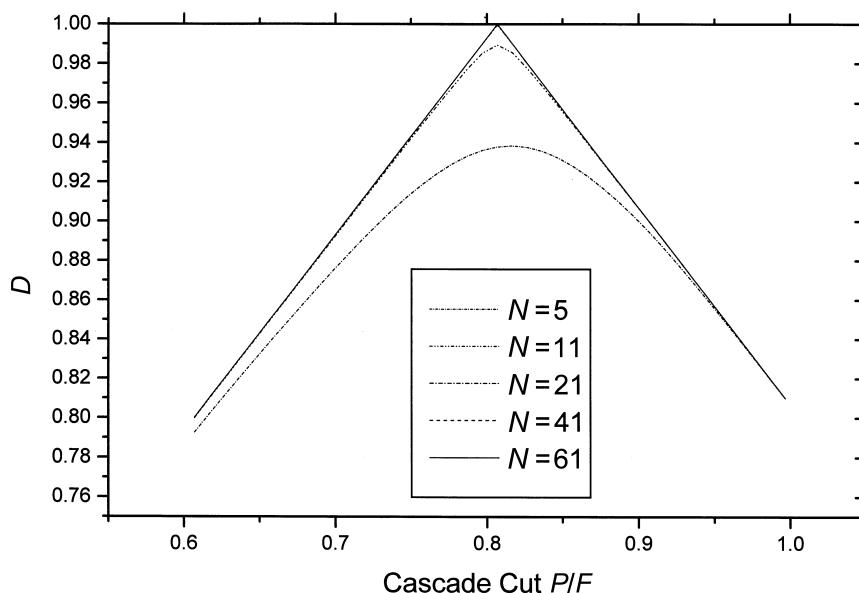


FIG. 3 The value of D as a function of cascade cut P/F for different cascade lengths. The light group contains the components ^{124}Xe , ^{126}Xe , ^{128}Xe , ^{129}Xe , ^{130}Xe , ^{131}Xe , and ^{132}Xe .

TABLE 2

The Maximum Values of the Function D Obtained with Different Cascade Lengths for Two Divisions for the Light Group $N'_c = 4$ and $N'_c = 7$

N	5	11	21	41	61
$N'_c = 4$	0.885856	0.964870	0.991783	0.999580	0.999980
$N'_c = 7$	0.938425	0.989388	0.999530	0.999997	0.999998

As long as a cascade is sufficiently long, the following observation is true: *Divide a multicomponent mixture into two groups of components, a light group with N'_c components and a heavy group. If the cascade cut P/F is taken to be the concentration sum $\sum_{i=1}^{N'_c} C_i^F$ in the feed of the components in the light group, then the product of the cascade will consist of only the components in the light group and the waste only of the components in the heavy group, or in other words, the concentration sum in the feed for the components appearing in the product is equal to the cascade cut P/F.* Thus if a middle component is an end component in either group, it will be an end component after separation. Comparing Fig. 2 and Fig. 3, we find that when the cascade is sufficiently long (for example, the two cases for $N = 61$ here), the shapes of the two corresponding curves of the function D are identical. Actually, the shapes of such curves for all components should be identical. This is easily explained according to the previously described observation: For a given component, the curve of the function D reaches maximum at the cascade cut P/F being equal to $\sum_{i=1}^{N'_c} C_i^F$. When the cascade cut is smaller than this value, the product consists of components 1 to j , and the waste consists of components j to N_c , with $1 \leq j \leq N'_c$. Thus Equations (20) and (21) can be written together as

$$C'_{i,N} = \phi C_i^F / (P/F), \quad C''_{i,1} = (1 - \phi) C_i^F / (1 - P/F) \quad (22)$$

with $\phi = 1$ for $i \neq j$, and $0 < \phi \leq 1$ for $i = j$, and

$$\sum_{i=1}^j C'_{i,N} = 1, \quad \sum_{i=j}^{N_c} C''_{i,1} = 1 \quad (23)$$

Therefore,

$$\begin{aligned} D &= \frac{P}{F} \sum_{i=1}^{N'_c} C'_{i,N} + \frac{F-P}{F} \sum_{i=N'_c+1}^{N_c} C''_{i,1} \\ &= \frac{P}{F} \sum_{i=1}^j C'_{i,N} + \frac{P}{F} \sum_{i=j+1}^{N'_c} C'_{i,N} + \frac{F-P}{F} \sum_{i=N'_c+1}^{N_c} C''_{i,1} \end{aligned}$$

On the right-hand side of the above equation, the first term is equal to 1 because of Eq. (23), and consequently the second term is zero because the product at the current cascade cut does not contain components $j + 1$ to N'_c , and the third term gives

$$\frac{F - P}{F} \sum_{i=N'_c + 1}^{N_c} C''_{i,1} = \sum_{i=N'_c + 1}^{N_c} C_i^F$$

because of Eq. (22). The function D behaves like

$$D = \frac{P}{F} + \sum_{i=N'_c + 1}^{N_c} C_i^F$$

Similarly, if the cascade cut is greater than $\sum_{i=1}^{N'_c} C_i^F$, the function D behaves like

$$D = -\frac{P}{F} + 1 + \sum_{i=1}^{N'_c} C_i^F$$

The slopes of the curves for the function D versus the cascade cut P/F are 1 for P/F smaller than $\sum_{i=1}^{N'_c} C_i^F$ and -1 for a value larger than $\sum_{i=1}^{N'_c} C_i^F$. Clearly, this behavior of the function D is independent of any particular component.

In reality, cascades cannot be infinitely long. So a question arises: how can a cascade be considered to be sufficiently long? The answer may vary and should be given according to practical considerations. For the purpose of demonstration, a cascade that gives at least three 9s for the value of the function D is sufficiently long. Hence, based on the results in Table 2, a cascade with 41 stages is sufficiently long, but not too long to be practical. Experiments 2 and 3 are carried out for cascades of this length, which are meant to briefly show the effects of cascade shapes and feed locations. Detailed investigations (e.g., optimization) for a real cascade in use should be made to gain the best separation.

Experiment 2: Different Shapes of Cascades

The square cascade is examined in Experiment 1. Now consider two strange cascades whose entering flow rates G_n are given, respectively, by

$$\frac{G_n}{F} = \begin{cases} 9 + n & 1 \leq n \leq N_f \\ 51 - n & N_f < n \leq N \end{cases} \quad \text{and} \quad \frac{G_n}{F} = \begin{cases} 31 - n & 1 \leq n \leq N_f \\ -11 + n & N_f < n \leq N \end{cases}$$

Here the feed location $N_f = 21$. Figure 4 plots three cases of cascade shapes, two defined by the above two distributions of G_n and one corresponding to the square cascade $G_n/F = 10$ used in Experiment 1, which are respectively re-

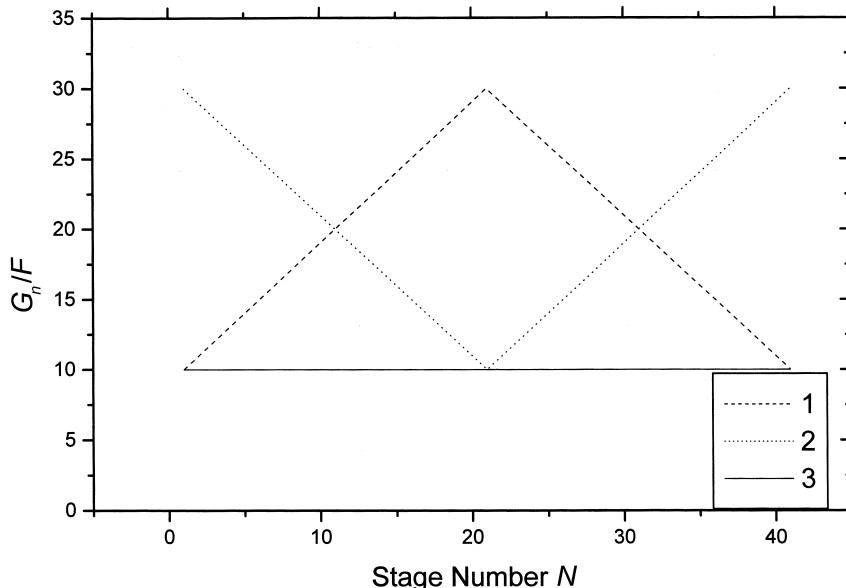


FIG. 4 The different shapes of cascades used.

ferred to as Shape 1, Shape 2, and Shape 3. A figure like Fig. 2 can be plotted about the values of D against the cascade cut for $N'_c = 4$. Because the three curves for the three cascade shapes are indistinguishable, which is expected and explained in Experiment 1, the figure is not presented here and only the maximum values of D are given for a comparison in Table 3.

The results indicate that the shape of a cascade only slightly affects the separation of a multicomponent mixture into a light and a heavy group in a desired way. It is natural to conclude that, as long as the length of the cascade is adequate, its shape has little effect. However, because the cascade is finitely long, the three values do have some slight differences; the larger the value is, the better the separation of the two groups is. This suggests that, for a finitely

TABLE 3
The Maximum Values of the Function D Obtained with Different Cascade Shapes
for $N'_c = 4$

Shape	1	2	3
Maximum D	0.999705	0.999687	0.999580

TABLE 4

The Maximum Values of the Function D Obtained with Different Feed Locations
for $N'_c = 4$

N_f	11	21	31
Maximum D	0.999087	0.999580	0.999137

long cascade, properly designing the cascade shape may be necessary to achieve better separation performance.

Experiment 3: Different Feed Locations

Here we still use the square cascade in Experiment 1 with $N = 41$ and take $N'_c = 4$. In the previous experiments, $N_f = 21$ was studied. Here the feed locations $N_f = 11$ and $N_f = 31$ are considered. These locations are chosen to prevent the stripping or the enriching sections from becoming too short. Again, because the curves for the values of the function D versus the cascade cut are hard to distinguish from each other, the maximum values of the function D are given in Table 4.

The results show that the feed location has little influence. For a finitely long cascade, choosing a proper feed location may help in achieving a better separation performance.

CONCLUSIONS

Numerical experiments were carried out by using the q -iteration method to solve the finite difference equations describing the concentration distribution in a cascade to investigate the separation of a middle component in a multi-component mixture. It was found that without using complicated cascades, such as an M- or a Q-cascade, the mixture can be separated into two groups of components, a light group and a heavy group, in a desired way. The key point is only to control the cascade cut P/F such that it is equal to the concentration sum in the feed of the components in the light group. The effects of the cascade length, shape, and feed location were studied. The results show that the longer the cascade, the more complete the separation of the two groups. For a sufficiently long cascade, the influences of cascade shape and feed location are negligible. However, in some cases it may be necessary to take into account the effects of the cascade shape and the feed location for performance optimization. Because controlling the cascade cut is a common task for any cascade operation and is much easier than designing and constructing an M- or a Q-cascade just for a specific middle component and a specific multicom-

ponent mixture, this method can be easily applied to the separation of any middle component in any multicomponent mixtures.

ACKNOWLEDGMENT

This research is partially supported by the National Natural Science Foundation of the People's Republic of China under contract No. 59676020.

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Received by editor January 14, 2000