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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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To cite this Article Ying, Chuntong , Wu, Hongjiang , Zhou, Mingsheng , Nie, Yuguang and Liu, Guangjun(1997) 'Analysis of Gas Centrifuge Cascade for Separation of Multicomponent Isotopes and Optimal Feed Position', Separation Science and Technology, 32: 15, 2467 — 2480

To link to this Article: DOI: 10.1080/01496399708000780

URL: http://dx.doi.org/10.1080/01496399708000780

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SEPARATION SCIENCE AND TECHNOLOGY, 32(15), pp. 2467–2480, 1997

Analysis of Gas Centrifuge Cascade for Separation of Multicomponent Isotopes and Optimal Feed Position

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ABSTRACT

Analysis of the concentration distribution in a gas centrifuge cascade for separation of multicomponent isotope mixtures is different from that in a cascade for separation of two-component mixtures. This paper presents the governing equations for a multicomponent isotope separation cascade. Numerically predicted separation factors for the gas centrifuge cascade agree well with the experimental data. A theoretical optimal feed position is derived for a short square cascade for a two-component mixture in a close-separation case. The optimal feed position for a gas centrifuge cascade for separation of multicomponent mixture is discussed.

Key Words. Gas centrifuge cascade; Multicomponent isotope mixture; Optimal feed position; Stable isotopes; Separation

1. INTRODUCTION

Stable isotopes are widely used in many fields, which thereby stimulates the research and development of gas centrifuge cascades for the separation of stable isotopes. There are 84 naturally occurring nonradioactive elements (including the very long decay period elements U and Th). Twenty-two of them have only one isotope. Forty-two of them have three or more isotopes. Gas centrifugation is the best method to produce the most stable isotopes. Many papers have discussed gas centrifuge cascades for two-

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component mixtures. However, only a few papers have been published concerning gas centrifuge cascades for the separation of multicomponent mixtures (1–4). Very few publications present experimental results for isotope separation cascades (5).

This paper presents the governing equations for multicomponent isotope separation cascades. A relationship between the separation factor of a gas centrifuge and mass difference, $\gamma_{ij} = \gamma_0^{M_j - M_i}$, is used. Two approaches are introduced to obtain steady-state solutions. Numerical results for the gas centrifuge cascade separation factors agree well with experimental data. A theoretical optimal feed position is derived for a short square cascade for a two-component mixture in a close-separation case. A relationship is obtained for the optimal feed position, N_{sopt}:N_{sopt}/N = P/F, where N is the total number of stages in the cascade, P is the product flow rate, and F is the feed flow rate. The optimal feed position in a gas centrifuge cascade is obtained numerically for separation of multicomponent mixtures. The numerical results show that the previous relationship does not apply in general for multicomponent separation. The optimal feed position is different for different components.

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2. GOVERNING EQUATIONS FOR GAS CENTRIFUGE CASCADE

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

The governing equations for the cascade are (6)

$$\theta_n G_n - (1 - \theta_{n+1})G_{n+1} = P_n^*(t), \qquad n = 1, \ldots, N-1 \tag{2.1}$$

$$\theta_n G_n C'_{n,i} - (1 - \theta_{n+1})G_{n+1}C''_{n+1,i} = P_{n,i}^*(t),\ i = 1, 2, \ldots, K; \quad n = 1, \ldots, N-1 \tag{2.2}$$

$$C_{n,i} = \theta_n C'_{n,i} + (1 - \theta_n)C''_{n,i}, \qquad i = 1, 2, \ldots, K;\ n = 1, \ldots, N \tag{2.3}$$

$$\frac{\partial(H_n C_{n,i})}{\partial t} = P_{n-1,i}^*(t) - P_{n,i}^*(t), \qquad i = 1, 2, \ldots, K; \quad n = 1, \ldots, N_F - 1, N_F + 1, \ldots, N \tag{2.4a}$$

$$\frac{\partial(H_n C_{n,i})}{\partial t} = P_{n-1,i}^*(t) - P_{n,i}^*(t) + FC_{Fi}, \quad i = 1, 2, \ldots, K;\ n = N_F \tag{2.4b}$$

where $P_n^*(t)$ is the net flow rate transported into the cascade above the

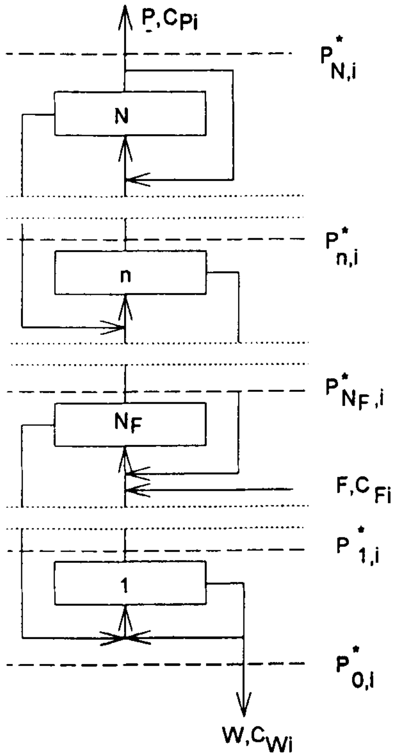

FIG. 1 A cascade scheme.

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nth stage, $P^*_{n,i}(t)$ is the net flow rate of the ith component transported into the cascade above the nth stage, N is the stage number in the cascade, θ_n is the cut of the nth stage, G_n is the interstage flow rate of the nth stage, H_n is the holdup of the nth stage, and $C_{n,i}$, $C'_{n,i}$, and $C''_{n,i}$ are the feed, heads, and tails concentration of the ith component in the nth stage. Equation (2.3) is derived from the material balance of the nth stage. Equation (2.4) reflects the change rate of the ith component in the nth stage.

The boundary conditions are determined by the operating conditions. In our example there is one product stream, one tails stream, and one feed stream. The boundary conditions are

$$P^*_{N,i} = PC_{Pi}, \qquad C_{Pi} = C'_{N,i}, \qquad P^*_N = P, \qquad i = 1, 2, \ldots, K \tag{2.5}$$

$$P^*_{0,i} = -WC_{Wi}, \qquad C_{Wi} = C''_{1,i}, \qquad P^*_0 = -W, \qquad i = 1, 2, \ldots, K \tag{2.6}$$

where P is the product rate, W is the tails rate, C_{Pi} is the concentration of the ith component in the product, and C_{Wi} is the concentration of the ith component in the tails.

In the example, the initial conditions are given as

$$C_{n,i}(t = 0) = C_{Fi}, \qquad i = 1, 2, \ldots, K; n = 1, \ldots, N \tag{2.7}$$

where C_{Fi} is the feed concentration of the ith component. Conditions (2.7) assume that the concentration of the ith component in the cascade in all stages has the same value as the concentration in the feed at the beginning of separation. For different cases the initial conditions may be different.

Equations (2.1)–(2.7) contain no simplifying assumptions. The governing equations, the boundary conditions, and the initial conditions are suitable for a close-separation cascade in which the separation factors are close to unity or for a cascade of stages with large separation factors.

The governing differential equations were solved explicitly. Equation (2.4) was written as the following difference equation:

$$\frac{H_n(C^{t+\Delta t}_{n,i} - C^t_{n,i})}{\Delta t} = P^*_{n-1,i}(t) - P^*_{n,i}(t), \tag{2.8a}$$

$$i = 1, 2, \ldots, K; n = 1, \ldots, N_F - 1, N_F + 1, \ldots, N$$

$$\frac{H_n(C^{t+\Delta t}_{n,i} - C^t_{n,i})}{\Delta t} = P^*_{n-1,i}(t) - P^*_{n,i}(t) + FC_{Fi}, \tag{2.8b}$$

$$i = 1, 2, \ldots, K; n = N_F$$

The separation factors in each separating unit are defined as

$$\alpha_{ij} \equiv \frac{C'_i}{C'_j}\bigg/\frac{C_i}{C_j}, \qquad \beta_{ij} \equiv \frac{C_i}{C_j}\bigg/\frac{C''_i}{C''_j}, \qquad \gamma_{ij} = \alpha_{ij}\beta_{ij} \equiv \frac{C'_i}{C'_j}\bigg/\frac{C''_i}{C''_j} \tag{2.9}$$

From the definition (2.9) it is not difficult to obtain the relations

$$C''_{n,i} = \frac{C'_{n,i}}{\sum_j \gamma_{ij} C'_{n,j}}, \qquad C'_{n,i} = \frac{C''_{n,i}}{\sum_j C''_{n,j}/\gamma_{ij}} \tag{2.10}$$

Two approaches were then used to obtain the steady-state solution. The first approach was to solve the steady-state equations, i.e., assume $\partial/\partial t = 0$ in Eq. (2.4). The second approach was to solve the transient

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equations. Steady-state was defined as the time when all the variables remained constant or the following condition was satisfied:

$$FC_{Fi} = PC_{Pi} + WC_{Wi}, \qquad i = 1, 2, \ldots, K \tag{2.11}$$

3. COMPARISON OF EXPERIMENTAL DATA WITH NUMERICAL RESULTS

A variety of isotopes were enriched using a gas centrifuge and a gas centrifuge cascade in our laboratory at Tsinghua University. Table 1 shows the numerical and experimental results for the separation of osmium isotopes.

In the experiments the osmium, in the form of OsO_4 gas, was separated in a gas centrifuge cascade. The numerical results are in good agreement with the experimental data. In the computations the gas centrifuge separation factor was related to the mass difference using $\gamma_{ij} = \gamma_0^{M_j - M_i}$, where M_j and M_i are the molecular weights of the jth and ith components, respectively. The relationship was verified by experiments and calculations (7).

TABLE 1

Numerical and Experimental Results for Gas Centrifuge Cascade

		^{184}Os	^{186}Os	^{187}Os	^{188}Os	^{189}Os	^{190}Os	^{192}Os
Experiment 1	C_{pi} (cal.)(%)	0.97	35.22	15.54	36.99	9.07	2.16	0.05
	C_{pi} (Exp.) (%)	0.91	35.63	15.77	36.47	8.57	2.13	0.50
	C_{Fi} (%)	0.22	12.67	8.91	40.76	22.31	12.99	2.14
	C_{Wi} (cal.) (%)	0.10	7.71	7.43	42.11	25.23	15.21	2.25
	C_{Wi} (Exp.) (%)	0.06	7.94	7.52	41.54	25.08	15.27	2.25
Experiment 2	C_{pi} (cal.) (%)	0.10	5.16	4.70	30.74	26.79	25.54	6.95
	C_{pi} (Exp.) (%)	0.13	5.28	4.74	30.57	26.17	25.23	7.88
	C_{Fi} (%)	0.05	2.85	2.81	20.80	22.13	28.84	22.51
	C_{Wi} (cal.) (%)	0.00	0.71	1.09	12.32	18.96	32.93	34.06
	C_{Wi} (Exp.) (%)	0.00	0.76	1.10	11.81	17.93	31.83	36.57

The separation factor per unit mass difference in the gas centrifuge, γ_0, depends on the operating conditions in the gas centrifuge. $\gamma_0 = 1.4$, used in the analysis, was obtained in a single gas centrifuge experiment. The numerical results using the steady-state equations and the transient equations were identical. In some cases the iterative process necessary to solve the steady-state equations did not converge well for long cascades of gas centrifuges with large separation factors or large numbers of components. However, solving the transient equations required a large amount of CPU time to obtain accurate steady-state solutions. Convergence was not a problem when the time step was sufficiently small.

4. OPTIMAL GAS CENTRIFUGE CASCADE FEED POSITION

Feed position is an important parameter for cascade operation. A square gas centrifuge cascade was chosen as a simple example. Figure 2 shows the dependence of the product concentration of ^{182}W on the feed position for the square gas centrifuge cascade.

In Figure 2, N_s is the feed position, i.e., the number of stages below the feed position, and N is the total number of stages in the cascade, $N = 15$. The difference between the highest and the lowest product concentrations is about 10%.

The optimal feed position was investigated by first considering a square cascade for separating a two-component mixture and then considering a square cascade having stages with large separation factors.

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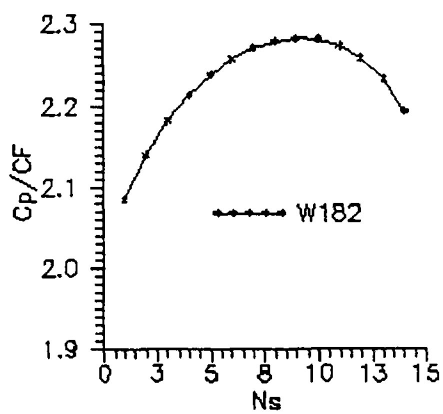

FIG. 2 Product concentration of ^{182}W in a square gas centrifuge cascade.

4.1. Optimal Feed Position of Square Cascade for Separation of Two-Component Mixture

A. Optimal Feed Position in a Close-Separation Cascade

A close-separation cascade is one in which the separation factors are very close to unity. A concentration gradient equation in the cascade in this case is given as (8)

$$\frac{dC}{dn} = 2\epsilon C(1 - C) - \frac{2(P_1^* - P^*C)}{G} \tag{4.1}$$

where ϵ is the enrichment factor, $\epsilon = (1 - \theta)(\gamma - 1)$ and the other variables are defined in Section 2. When θ is approximately equal to 0.5, $\epsilon = (\gamma - 1)/2$. Analysis of the short square cascade with consideration that C is close to 0.5 assumes that $C(1 - C) \approx$ constant. Figure 3 shows a schematic of the cascade.

In Fig. 3, N_E is the number of stages above feed position; F, P, and W are the feed, product, and tails flow rates, respectively; C_F, C_P, and C_W are the feed, product, and tails concentrations, respectively; and C_f is the concentration at the entrance of the stage into which the feed is supplied.

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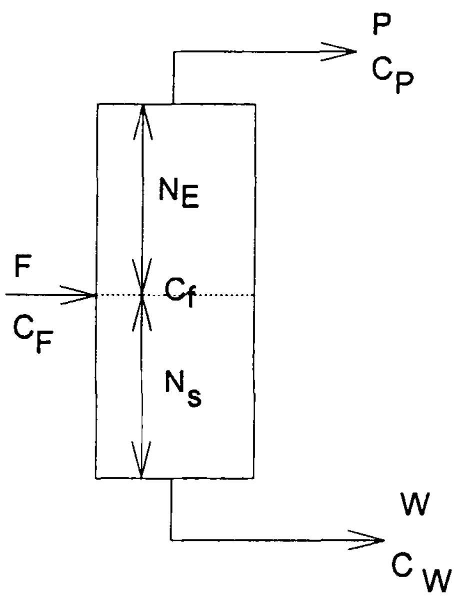

FIG. 3 A cascade schematic.

Integrating Eq. (4.1) over n gives two equations (8):

$$C_P - C_f = \frac{G\epsilon\overline{C_F(1 - C_F)}}{P}(1 - e^{-2P/GN_E}) \tag{4.2}$$

$$C_f - C_W = \frac{G\epsilon\overline{C_F(1 - C_F)}}{W}(1 - e^{-2W/GN_s}) \tag{4.3}$$

In the cascade, feed, product, and tails flow rates and concentrations must satisfy the material-balance relations

$$F = P + W \tag{4.4}$$

$$FC_F = PC_P + WC_W \tag{4.5}$$

Combining Eqs. (4.2)–(4.4) to eliminate C_P, C_f, and W gives

$$C_P = C_F + \frac{G\epsilon\overline{C_F(1 - C_F)}}{F} \times \left(\frac{1}{\Theta} - e^{-2(1-\Theta)F/GN_s} - \frac{1-\Theta}{\Theta}e^{-2\Theta F/G(N-N_s)}\right) \tag{4.6}$$

where $N = N_E + N_s$ and $\Theta = P/F$. The optimal feed position is obtained when the product concentration C_P reaches a maximum, i.e., $dC_P/dn = 0$

$$N_{sopt} = \frac{P}{F}N \quad \text{or} \quad \frac{N_{sopt}}{N} = \frac{P}{F} \tag{4.7}$$

Using previous equations gives

$$C_f = C_F \tag{4.8}$$

Equation (4.8) shows that the optimal product concentration occurs when there is no mixing loss at the cascade feed position.

As an example, a cascade is analyzed with the following variables: $\epsilon = 0.002$, $F/G = 0.1$, and $N = 10$. The mixture is natural UF_6 where the concentration of ^{235}U is 0.00711, $C_F = 0.00711$. The optimal feed position was calculated for nine different values of P/F. The numerical results are listed in Tables 2a and 2b.

Table 2a shows that N_{sopt}/N is equal to P/F. In Table 2b, C_f is the concentration inside the cascade at the feed stage, C_{fn} is the concentration inside the cascade which is closest to the feed concentration, and N_n is the number of stages between C_{fn} and the tails. When C_{fn} equals C_F or N_{sopt} equals N_n, as in Table 2a, then there is no mixing loss at the feed point.

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TABLE 2a
Optimal Feed Position for a Short Square Cascade
$\varepsilon = (\gamma - 1)/2 = 0.002$, $F/G = 10$, $N = 10$, $C_F = 0.00711$

P/F	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9
N_{sopt}	1	2	3	4	5	6	7	8	9
N_{sopt}/N	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9

TABLE 2b
Concentration Inside the Short Square Cascade

P/F	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9
N_{sopt}	1	2	3	4	5	6	7	8	9
C_{fs}	.00711	.00711	.00711	.00711	.00711	.00711	.00711	.00711	.00711
Nn	1	2	3	4	5	6	7	8	9
C_{fn}	.00711	.00711	.00711	.00711	.00711	.00711	.00711	.00711	.00711

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B. Cascade of Stages with Large Separation Factors

A cascade was analyzed with following parameters: $\gamma \equiv C'/(1 - C')/C''/(1 - C'') = 1.41$, $F/G = 0.1$, and $N = 10$. Natural UF_6 is fed into the cascade. Numerical results were obtained using the transient approach. They are listed in Tables 3a and 3b.

Table 3a shows that N_{sopt}/N is not equal to P/F and that the value of N_{sopt}/N is increasing with P/F. Table 3b shows that the concentration at the entrance of the optimal feed stage is approximately equal to C_F.

4.2. Optimal Feed Position of Square Cascade for Multicomponent Mixtures

A. Close-Separation Cascade

A square cascade for separation of WF_6 was analyzed with the following parameters: $\gamma_0 = 1.02$, $F/G = 0.1$, and $N = 10$. Numerical results are listed in Tables 4a and 4b.

TABLE 3a
Optimal Feed Position

$\gamma = 1.41$, $F/G = 0.1$, $N = 10$, $C_F = 0.00711$

P/F	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9
N_{sopt}	3	5	6	7	7	8	9	9	9
N_{sopt}/N	0.3	0.5	0.7	0.7	0.8	0.8	0.9	0.9	0.9

TABLE 3b
Concentrations Inside the Cascade

P/F	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9
N_{sopt}	3	5	6	7	7	8	8	9	9
C_{fs}	.00707	.00770	.00751	.007778	.00658	.00730	.00640	.00751	.00673
Nn	3	5	6	7	7	8	8	9	9
C_{fn}	.00707	.00770	.00751	.007778	.00658	.00730	.00640	.00751	.00673

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TABLE 4a
Optimal Feed Positions of a Cascade for Separation of Multicomponent Mixtures

$\gamma_o = 1.02$, $F/G = 0.1$, $N = 10$

	W-180		W-182		W-183		W-184		W-186	
C_{Fi}	0.00135		0.264		0.144		0.305		0.284	
P/F	Nsopt	Nsopt/N	Nsopt	Nsopt/N	Nsopt	Nsopt/N	Nsopt	Nsopt/N	Nsopt	Nsopt/N
0.1	1	0.1	1	0.1	1	0.1	1	0.1	1	0.1
0.2	2	0.2	2	0.2	1	0.1	2	0.2	1	0.1
0.3	3	0.3	3	0.3	2	0.2	4	0.4	2	0.2
0.4	4	0.4	4	0.4	3	0.3	6	0.6	3	0.3
0.5	5	0.5	5	0.5	4	0.4	9	0.9	4	0.4
0.6	6	0.6	6	0.6	5	0.5	9	0.9	5	0.5
0.7	7	0.7	7	0.7	6	0.6	9	0.9	6	0.6
0.8	8	0.8	8	0.8	7	0.7	9	0.9	7	0.7
0.9	9	0.9	9	0.9	8	0.8	9	0.9	9	0.9

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TABLE 4b
Concentrations Inside the Cascade

	W-180				W-182				W-183				W-184				W-186			
C_{Fi}	0.00135				0.264				0.144				0.305				0.284			
P/F	Nsopt	C_{fsi}	Nn	C_{fni}	Nsopt	C_{fsi}	Nn	C_{fni}	Nsopt	C_{fsi}	Nn	C_{fni}	Nsopt	C_{fsi}	Nn	C_{fni}	Nsopt	C_{fsi}	Nn	C_{fni}
0.1	1	.00137	1	.00137	1	.2679	1	.2679	1	.1455	1	.1455	1	.3060	2	.3053	1	.2789	1	.2789
0.2	2	.00135	2	.00135	2	.2672	2	.2672	1	.1437	1	.1437	2	.3065	4	.3054	1	.2877	1	.2877
0.3	3	.00134	3	.00134	3	.2668	3	.2668	2	.1440	2	.1440	4	.3062	6	.3047	2	.2871	2	.2871
0.4	4	.00134	4	.00134	4	.2667	4	.2667	3	.1440	3	.1440	6	.3056	7	.3046	3	.2867	3	.2867
0.5	5	.00134	5	.00134	5	.2665	5	.2665	4	.1440	4	.1440	9	.3028	7	.3050	4	.2866	4	.2866
0.6	6	.00134	6	.00134	6	.2666	6	.2666	5	.1440	5	.1440	9	.3038	8	.3052	5	.2867	5	.2867
0.7	7	.00135	7	.00135	7	.2668	7	.2668	6	.1439	6	.1439	9	.3048	9	.3048	6	.2869	6	.2869
0.8	8	.00136	8	.00136	8	.2672	8	.2672	7	.1437	7	.1437	9	.3056	9	.3056	7	.2876	7	.2876
0.9	9	.00137	9	.00137	9	.2677	9	.2677	9	.1435	9	.1435	9	.3052	9	.3052	9	.2787	8	.2887

Table 4a shows that for a short square close-separation cascade, the values of N_{sopt}/N are approximately equal to P/F except for ^{184}W. Table 4b shows that the concentration at the entrance of the optimal feed stage, C_{fsi}, is approximately equal to C_{Fi}.

B. Cascade of Gas Centrifuges with Large Separation Factors

The parameters of a square cascade of gas centrifuges are $\gamma_0 = 1.12$, $F/G = 0.1$, and $N = 15$. Numerical results are listed in Tables 5a and 5b.

Table 5a shows that $N_{sopt}/N \neq P/F$ and that N_{sopt}/N increases with P/F. Table 5b shows that when the product concentration of the ith component reaches its extreme value, i.e., the optimal feed position is found, the concentration of the ith component at the feed point inside the cascade, C_{fsi}, is sometimes not equal to the feed concentration of the ith component, C_{Fi}. For example, to obtain the maximum concentration of ^{183}W at the product discharge for $P/F = 0.5$, the optimal feed position is $N_{sopt} = 6$. However, the concentration of ^{183}W at the entrance of stage 6 inside the cascade, C_{fsi}, is equal to 0.1591 while the feed concentration of ^{183}W is 0.1440. The stage at which the entrance concentration is closest to the feed concentration is $N_n = 4$. A second result shown in Table 5b is that the optimal feed position for different isotopes may be different. There is no common optimal feed position suitable for all components.

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TABLE 5a

Optimal Feed Positions of a Cascade with Large Separation Factors

γ_0=1.12, F/G=0.1, N=15

	W-180		W-182		W-183		W-184		W-186	
C_{Fi}	0.00135		0.264		0.144		0.305		0.284	
P/F	N_{sopt}	N_{sopt}/N	N_{sopt}	N_{sopt}/N	N_{sopt}	N_{sopt}/N	N_{sopt}	N_{sopt}/N	N_{sopt}	N_{sopt}/N
0.1	7	.467	1	.067			1	.067	1	.067
0.2	9	.600	5	.333	1	.067	3	.200	1	.067
0.3	10	.667	7	.467	1	.067	6	.400	2	.133
0.4	11	.733	8	.533	2	.133	9	.600	3	.200
0.5	12	.800	9	.600	6	.400	1*	.067	4	.267
0.6	12	.800	10	.667	8	.533	1*	.067	6	.400
0.7	13	.867	11	.733	10	.667	5*	.333	8	.533
0.8	13	.867	12	.800	12	.800	10*	.667	10	.667
0.9	14	.933	14	.933	14	.933	13*	.867	13	.867

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TABLE 5b
Concentrations Inside the Cascade

	W-180				W-182				W-183				W-184				W-186			
C_{Fi}	.00135				.264				.144				.305				.284			
P/F	Nsopt	C_{fsi}	Nn	C_{fni}	Nsopt	C_{fsi}	Nn	C_{fni}	Nsopt	C_{fsi}	Nn	C_{fni}	Nsopt	C_{fsi}	Nn	C_{fni}	Nsopt	C_{fsi}	Nn	C_{fni}
0.1	7	.00167	6	.00138	1	.27332	1	.27332					1	.32617	3	.30433	1	.23382	1	.23382
0.2	9	.00449	8	.00121	4	.30892	3	.27894	1	.15859	1	.15859	3	.34330	6	.30307	1	.26810	1	.26810
0.3	10	.00130	10	.00131	6	.30034	5	.27428	1	.13997	1	.13997	6	.34259	9	.29718	2	.26513	2	.26513
0.4	11	.00124	11	.00124	8	.29416	7	.26886	2	.13452	3	.14740	9	.33277	11	.30232	3	.27354	3	.27354
0.5	12	.00126	12	.00126	9	.26877	9	.26877	6	.15914	4	.14053	1*	.33221	1	.33221	4	.29326	4	.29326
0.6	12	.00104	13	.00142	10	.24845	10	.24845	8	.15634	7	.14839	1*	.28867	2	.30655	6	.29260	6	.29260
0.7	13	.00116	13	.00117	11	.23349	12	.26751	10	.15276	9	.14349	5*	.33728	3	.30651	8	.30693	9	.26900
0.8	13	.00100	14	.00146	12	.22381	13	.26256	12	.14956	11	.13900	10*	.36293	6	.31439	10	.33145	10	.28854
0.9	1 4	.00124	14	.00124	14	.26606	14	.26606	13	.14005	13	.14082	13*	.33830	9	.29686	13	.29657	13	.29657

5. CONCLUSIONS

A. The relationship between the gas centrifuge separation factor and the mass difference, $\gamma_{ij} = \gamma_0^{M_j - M_i}$, can be used to analyze cascades for separating multicomponent isotopes.
B. Two methods were used to calculate the steady-state concentration distribution in the cascade for separation of multicomponent isotope mixtures. The numerical results agree well with experimental data. Solving the transient equations was an effective method for analyzing a multistage cascade with large separation factors.
C. The optimal feed position was investigated for various cases. The relationships $N_{sopt}/N = P/F$ and $C_{fi} = C_{Fi}$ can be used to determine the optimal feed position only for some special cases. For many situations, the optimal feed position must be determined by solving all of the equations.

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ACKNOWLEDGMENTS

Financial support for this work was provided by the National Natural Science Foundation of China (No. 59676020) and the Science Foundation of China Nuclear Energy Industry Co.

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Received by editor October 30, 1996
Revision received January 1997