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Distribution of Average Molar Weight in a Cascade for Separating Multicomponent Isotopic Mixtures

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ABSTRACT

The distribution of average molar weight of the process mixture in a cascade is an important characteristic in the separation of multicomponent isotopic mixtures. The average molar weight is shown to decrease monotonically along the enriching direction of the cascade. The composition of some intermediate weight isotopes may reach a maximum at some location in the cascade. The average molar weight can be used to judge the effectiveness of the separation cascade.

Key Words. Average molar weight; Separation cascade; Multicomponent isotopic mixtures

INTRODUCTION

The demand for stable isotopes is stimulating theoretical and experimental research on separation cascades for separating multicomponent isotopic mixtures. Cohen (1) and Benedict (2) nicely discussed the basic principles for binary multistage cascades since the process mixture for uranium separation is treated as a binary mixture. A. de la Garza et al. (3) discussed multicomponent isotope separation cascades with close separation in 1961. Since then, a number of authors have studied multicomponent isotope separation cascades with

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close separation (4–8). Not many papers have been published concerning gas centrifuge cascades for the separation of multicomponent isotopes. Gas centrifuge cascades have large separation factors. An extensive list of references to the relevent literature is provided by the authors in Ref. 9. Behavior that is special to multicomponent mixture separation cascades with large separation factors, such as the optimal feed position, has been discussed by Ying et al. (9). The average molar weight of the mixture is a characteristic parameter of multicomponent separation cascades. Understanding its distribution in a separation cascade is important for understanding and analyzing separation phenomena in the cascade.

In this paper the governing equations are used to derive the variation of the average molar weight and composition distribution of each component in the cascade. In the close-separation case, the average molar weight is shown to vary monotonically along the cascade, i.e., the average molar weight always decreases along the enriching direction of the lightest component. Five examples for separation cascades with large separation factors are given to show the same variation of the average molar weight distribution in the cascade. In some cases the intermediate weight component of the multicomponent mixture reaches its maximum composition in the middle of the cascade. An approximate relationship is developed. The average molar weight can easily be used to show the effectiveness of the cascade.

THEORETICAL ANALYSIS

Governing Equations for a Cascade

A general cascade schematic for the separation of multicomponent isotopic mixtures is shown in Fig. 1. The governing equations for the cascade are (3)

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

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

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

$$\frac{\partial(H_n C_{n,i})}{\partial t} = P_{n-1,i}^*(t) - P_{n,i}^*(t); \qquad i = 1, 2, \ldots, K; \quad n = 1, \ldots, N_F - 1, N_F + 1, \ldots, N \tag{4a}$$

$$\frac{\partial(H_n C_{n,i})}{\partial t} = P_{n-1,i}^*(t) - P_{n,i}^*(t) + FC_{Fi}; \qquad i = 1, 2, \ldots, K; \; n = N_F \tag{4b}$$

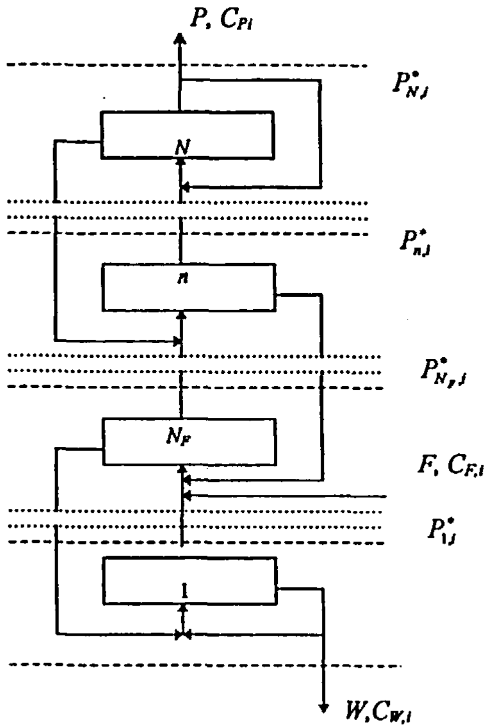

FIG. 1 A cascade scheme.

where $P_n^*(t)$ is the net flow rate transported into the cascade above the nth stage, $P_{n,i}^*(t)$ is the net flow rate of the ith component transported into the cascade above the nth stage, K is the number of components in the mixture, 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, F is the feed flow rate at the N_Fth stage, and $C_{n,i}$, $C'_{n,i}$, and $C''_{n,i}$ are the feed, heads, and tails composition of the ith component in the nth stage. In Fig. 1, P is the product flow rate of the cascade, W is the tails flow rate of the cascade, and C_{Pi}, C_{Wi}, and C_{Fi} are the product, tails, and feed composition of the ith component. Equation (1) shows the total material balance in the cascade above the nth stage. Equation (2) reflects the material balance of the ith component in the cascade above the nth stage. Equation (3) is derived from the material balance of the nth stage. Equation (4a) or (4b) reflects the rate of change of the ith component in the nth stage.

Combining Eqs. (1) and (2) gives

$$\theta_n G_n (C'_{n,i} - C''_{n+1,i}) = P^*_{n,i} - P^*_n C''_{n+1,i};$$
$$i = 1, 2, \ldots, K; n = 1, \ldots, N - 1 \quad (5)$$

Equation (5) can be used to determine the change in tails composition between adjacent stages:

$$C''_{n+1,i} - C''_{n,i} = C'_{n,i} - C''_{n,i} - \frac{P^*_{n,i} - P^*_n C''_{n+1,i}}{\theta_n G_n},$$
$$i = 1, 2, \ldots, K; n = 1, \ldots, N - 1 \quad (6)$$

The separation factors in each separating unit are defined as (10)

$$\alpha_{ij} \equiv \frac{C'_i}{C'_j} \Big/ \frac{C_i}{C_j}; \qquad \beta_{ij} \equiv \frac{C_i}{C_j} \Big/ \frac{C''_i}{C''_j}; \qquad \gamma_{ij} = \alpha_{ij}\beta_{ij} \equiv \frac{C'_i}{C'_j} \Big/ \frac{C''_i}{C''_j} \quad (7)$$

The definitions in Eq. (7) can be used to obtain the following relationships:

$$C''_{n,j} = \frac{C'_{n,i}}{\sum_j \gamma_{ij} C'_{n,j}}, \qquad C'_{n,i} = \frac{C''_{n,i}}{\sum_j C''_{n,j}/\gamma_{ij}} \quad (8)$$

In this paper we discuss steady-state problems. Two approaches are used to obtain the steady-state solution. One approach is to solve the equations in steady state, i.e., assume $\partial/\partial t = 0$ in Eq. (4a) and (4b). Then use an iterative method to find the solution. Another approach is to solve the transient equation. Steady state is defined as the time when all the variables become constant. When the iterative approach is divergent, the transient approach is the only way to obtain the solution. The calculational methods are discussed in Wu et al. (12).

Variation of Average Molar Weight in the Cascade

The average molar weight of the mixture at the nth stage in the cascade, $\overline{M}_n$, is equal to $\sum_i M_i C_{n,i}$, where M_i is the molar weight of the ith component. Multiplying Eq. (6) by M_i and summing all the equations give the change of the average molar weight between adjacent stages:

$$\overline{M}''_{n+1} - \overline{M}''_n = \overline{M}'_n - \overline{M}''_n - \frac{\sum_i M_i P^*_{n,i} - P^*_n \overline{M}''_{n+1}}{\theta_n G_n}; \quad (9)$$
$$n = 1, \ldots, N - 1$$

where $\overline{M}'_n$ is the average molar weight of the heads flow at the nth stage, and $\overline{M}''_n$ is the average molar weight of the tails flow at the nth stage.

Close-Separation Cascade

A close-separation cascade is one in which the separation factors are very close to unity. In a close-separation cascade the difference Eq. (6) may be approximated by the following differential equation:

$$\begin{aligned}\frac{dC_{n,i}}{dn} &= C'_{n,i} - C''_{n,i} - \frac{P^*_{n,i} - P^*_{n,i}C_{n,i}}{\theta_n G_n} \\ &= C'_{n,i} - \frac{C'_{n,i}}{\sum_j \gamma_{ij} C'_j} - \frac{P^*_{n,i} - P^*_n C_{n,i}}{\theta_n G_n} \\ &= C_{n,i} \ln \gamma_0 \sum (M_j - M_i) C_j - \frac{P^*_{n,i} - P^*_n C_{n,i}}{\theta_n G_n} \qquad (10) \\ &= C_{n,i} \ln \gamma_0 (\overline{M}_n - M_i) - \frac{P^*_{n,i} - P^*_n C_{n,i}}{\theta_n G_n}; \\ i &= 1, 2, \ldots, K;\ n = 1, \ldots, N-1\end{aligned}$$

The close-separation condition $\gamma_{ij} - 1 \ll 1$ or $\gamma_{ij} - 1 = \ln \gamma_{ij}$ and the approximation $C_{n,i} \approx C''_{n+1,i}$ were used to derive Eq. (10). In addition, γ_{ij} was related to the differences of the molar weights using (10)

$$\gamma_{ij} = \gamma_0^{M_j - M_i} \qquad (11)$$

where γ_0 is the heads-to-tails separation factor per unit molar weight difference. According to the definition of γ_{ij}, γ_0 must be greater than 1; therefore, $\ln \gamma_0 \geq 0$. We should emphasize that Expression (11) is only correct for the separation of multicomponent isotopic mixtures.

Multiplying Eq. (10) by M_i and summing over all the components gives

$$\frac{d\overline{M}_n}{dn} = \ln \gamma_0 \left(\overline{M}_n^2 - \sum_i M_i^2 C_{n,i} \right) - \frac{\sum_i M_i (P^*_{n,i} - P^*_n C_{n,i})}{\theta_n G_n}; \quad n = 1, 2 \ldots, N \qquad (12)$$

According to the Cauchy inequality:

$$\left(\sum_i a_i b_i \right)^2 \leq \left(\sum_i a_i^2 \right) \left(\sum_i b_i^2 \right) \qquad (13)$$

Letting $a_i = M_i C_{n,i}^{1/2}$ and $b_i = C_{n,i}^{1/2}$ gives

$$\left(\sum_i M_i C_{n,i} \right)^2 \leq \left(\sum_i M_i^2 C_{n,i} \right) \left(\sum_i C_{n,i} \right) = \sum_i M_i^2 C_{n,i} \qquad (14)$$

that is,

$$\left(\overline{M}_n^2 - \sum_i M_i^2 C_{n,i}\right) \leq 0 \tag{15}$$

The condition $\sum_i C_{n,i} = 1$ was used for the derivation of Eq. (14).

If there are no external withdrawals from the cascade, the second term on the right side of Eq. (12) equals zero. In that case

$$\frac{d\overline{M}_n}{dn} = \ln\gamma_0\left(\overline{M}_n^2 - \sum_i M_i^2 C_{n,i}\right) = -\ln\gamma_0 \sum_j \sum_{1\leq i<j} C_{n,i}C_{n,j}(M_i - M_j)^2 \leq 0 \tag{16}$$

Equation (16) shows that the average molar weight of the process mixture in the cascade monotonically decreases with the stage number, i.e., toward the enriching direction, when there is no withdrawal from the separation cascade.

In the case without withdrawals, the composition differential Eq. (10) becomes

$$\frac{dC_{n,i}}{dn} = C_{n,i} \ln \gamma_0(\overline{M}_n - M_i) \tag{17}$$

Equation (17) shows that:

(a) The composition of the lightest component of the mixture in the cascade increases monotonically along the enriching direction since the average molar weight $\overline{M}_n$ is always $\geq M_1$ in the cascade. From Eq. (17), $dC_{n,1}/dn \geq 0$.

(b) The composition of the heaviest component of the mixture in the cascade decreases monotonically along the enriching direction since the average molar weight $\overline{M}_n$ is always $\leq M_K$ in the cascade. From Eq. (17), $dC_{n,K}/dn \leq 0$.

(c) When the average molar weight of the mixture at stage n, $\overline{M}_n$, is equal to the molar weight of the ith component, M_i, the composition of the ith component reaches its maximum because in this case $dC_{n,i}/dn = 0$. In this case the average molar weight of the mixture at the first stage, $\overline{M}_1$, is greater than M_i. The average molar weight of the mixture in the cascade decreases with stage number n. When the average molar weight of the mixture at the nth stage, $\overline{M}_n$, reaches $\overline{M}_i$, then $dC_{n,i}/dn = 0$ and the composition of the ith component reaches its maximum.

(d) The composition distribution has no minimum value since the average molar weight of the mixture, $\overline{M}_n$, in the cascade is always decreasing. Therefore, since the composition decreases from the first stage, then for any component $\overline{M}_1 < M_j$, so $\overline{M}_n$ will never be equal to M_j because $\overline{M}_n$ is decreasing with stage number n.

In summary, the compositions of the lightest and heaviest components vary monotonically along the cascade, the compositions of some intermediate components may reach a maximum value in the cascade but not at the end stages, and the compositions of all component have no minimum value in the cascade.

The separation of binary mixtures for separation cascades with external withdrawals will be considered first. The composition differential equation for close-separation cascades is (11)

$$\frac{dC_n}{dn} = (\gamma - 1)C_n(1 - C_n) - \frac{P_{Ln}^* - P_n^* C_n}{\theta_n G_n} \tag{18}$$

where C_n is the composition of the light component in the nth stage, P_{Ln}^* is the net upflow rate of the light component in the cascade at the nth stage, P_n^* is the net upflow rate of the process gas mixture in the cascade at the nth stage, and

$$\gamma = \frac{C_n'}{1 - C_n'} \Bigg/ \frac{C_n''}{1 - C_n''}$$

Let M_1 be the molar weight of the light component and M_2 be the molar weight of the heavy component. Then the average molar weight of the gas mixture in stage n, $\overline{M}_n$, is equal to

$$\overline{M}_n = M_1 C_n + M_2(1 - C_n) \tag{19}$$

Differentiating Eq. (19) with respect to n gives

$$\frac{d\overline{M}_n}{dn} = (M_1 - M_2)\frac{dC_n}{dn} \tag{20}$$

For a given process gas mixture, $M_1 - M_2$ is constant and always less than zero. Because the stages in the cascade are connected with each other as shown in Fig. 1, the composition of the light component is always increasing along the enriching direction, therefore

$$dC_n/dn \geq 0 \tag{21}$$

Then,

$$d\overline{M}_n/dn \leq 0 \tag{22}$$

Inequality (22) shows that the average molar weight of the process gas mixture in the cascade decreases monotonically along the enriching direction of the cascade regardless of any external withdrawals.

For the separation cascade of multicomponent gas mixtures, Eq. (12) must be used. It has been proven that the first term on the right side of Eq. (12) is less than zero. Because $P_n^*/(\theta_n G_n)$ is usually much less than $\ln \gamma_0$ in actual square cascades, the absolute value of the second term is usually less than the absolute value of the first term, so $d\overline{M}_n/dn \leq 0$ for the close-separation cascade in most cases of square cascades. However, Inequality (22) is still not proven for the general case.

Separation Cascade with Large Separation Factors

For a separation cascade with large separation factors, Eq. (9) gives the change of the average molar weight between adjacent stages. When there are no withdrawals from the cascade, the change of the average molar weight between adjacent stages becomes

$$\overline{M}''_{n+1} - \overline{M}''_n = \overline{M}'_n - \overline{M}''_n; \qquad n = 1, \ldots, N-1 \tag{23}$$

It is reasonable to say that $\overline{M}'_n \leq \overline{M}''_n$ because the lightest component of the mixture is enriched in the heads flow of the nth stage and the other components satisfy the relationship

$$\gamma_{ij} = \gamma_0^{M_j - M_i} \tag{11}$$

where γ_0 is the heads-to-tails separation factor per unit molar weight difference and $\gamma_0 \gtrsim 1$. Therefore,

TABLE 1
Process Gas and Separation Cascade Operating Data

	Example				
	1	2	3	4	5
Process gas	UF_6	CrO_2F_2	WF_6	OsO_4	Xe
Number of stages [N]	20	20	20	20	20
γ_0	1.15	1.30	1.40	1.40	1.40
F/G	0.10	0.10	0.10	0.10	0.10
P/F	0.3	0.5	0.35	0.3	0.2
N_F	4	16	4	6	2
Isotopic composition		(^{16}O, 100.%)		(^{16}O, 100.%)	
of the feed:	^{234}U, 0.02%	^{50}Cr, 4.31%	^{180}W, 0.135%	^{184}Os, 0.018%	^{124}Xe, 0.096%
	^{235}U, 0.9%	^{52}Cr, 83.76%	^{182}W, 26.41%	^{186}Os, 1.59%	^{126}Xe, 0.09%
	^{236}U, 0.4%	^{53}Cr, 9.55%	^{183}W, 14.40%	^{187}Os, 1.64%	^{128}Xe, 1.919%
	^{238}U, 98.68%	^{54}Cr, 2.38%	^{184}W, 30.64%	^{188}Os, 13.3%	^{129}Xe, 26.44%
			^{186}W, 28.41%	^{189}Os, 16.1%	^{130}Xe, 4.08%
				^{190}Os, 26.4%	^{131}Xe, 21.18%
				^{192}Os, 40.95%	^{132}Xe, 26.89%
					^{134}Xe, 10.44%
					^{136}Xe, 8.87%

TABLE 2
Distribution of Average Molar Weight in Separation Cascade

	Example				
	1	2	3	4	5
Process gas	UF_6	CrO_2F_2	WF_6	OsO_4	Xe
$\underline{M}_1$	237.99	52.24	184.57	190.94	131.54
$\underline{M}_2$	237.99	52.18	184.21	190.59	130.74
$\underline{M}_3$	237.99	52.15	183.94	190.32	130.16
$\underline{M}_4$	237.99	52.12	183.72	190.11	129.75
$\underline{M}_5$	237.99	52.10	183.54	189.94	129.46
$\underline{M}_6$	237.99	52.09	183.41	189.76	129.28
$\underline{M}_7$	237.99	52.08	183.30	189.59	129.17
$\underline{M}_8$	237.98	52.08	183.21	189.46	129.10
$\underline{M}_9$	237.98	52.07	183.12	189.35	129.06
$\underline{M}_{10}$	237.98	52.07	183.04	189.27	129.03
$\underline{M}_{11}$	237.98	52.07	182.97	189.19	129.02
$\underline{M}_{12}$	237.98	52.06	182.90	189.11	129.01
$\underline{M}_{13}$	237.98	52.06	182.83	189.04	129.00
$\underline{M}_{14}$	237.98	52.05	182.76	188.98	128.99
$\underline{M}_{15}$	237.98	52.04	182.69	188.91	128.99
$\underline{M}_{16}$	237.97	52.02	182.61	188.84	128.98
$\underline{M}_{17}$	237.97	52.00	182.53	188.77	128.97
$\underline{M}_{18}$	237.95	51.97	182.45	188.67	128.96
$\underline{M}_{19}$	237.94	51.94	182.38	188.55	128.94
M_{20}	237.91	51.88	182.30	188.39	128.90

$$\overline{M}''_{n+1} - \overline{M}''_n \leq 0; \qquad n = 1, \ldots, N - 1 \qquad (24)$$

Inequality (24) shows that the average molar weight of the process gas mixture decreases monotonically with stage number when there are no withdrawals from the cascade. In other words, the average molar weight decreases monotonically along the enriching direction.

The composition distribution for each component and the average molar weight in square separation cascades was calculated for some examples (UF_6, CrO_2F_2, WF_6, OsO_4, and Xe). The separation and process gas data are given in Table 1.

The distributions of the average molar weights in the separation cascade are listed in Table 2 and shown in Fig. 2 (note that the average molar weight in the figure represents the average molar weight of the elements which have several isotopes). Although the distribution patterns differ, the average molar weights all decrease monotonically with stage number for all the examples.

The composition distribution of some of the components in the cascade is shown in Figs. 3–5 for Examples 3, 4, and 5, respectively. Two maximums are

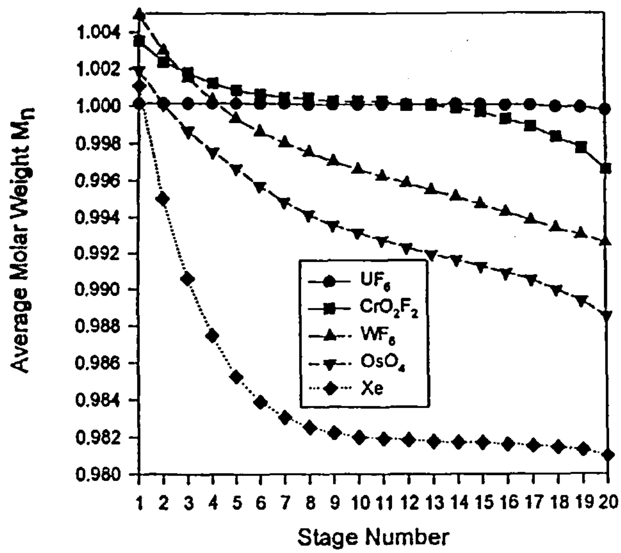

FIG. 2 Average molar weight distribution in different cascades normalized by average molar weight in feed.

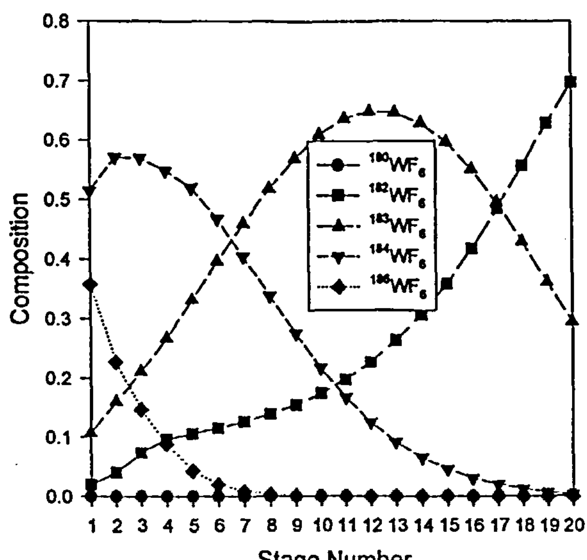

FIG. 3 Composition distribution of W in a cascade.

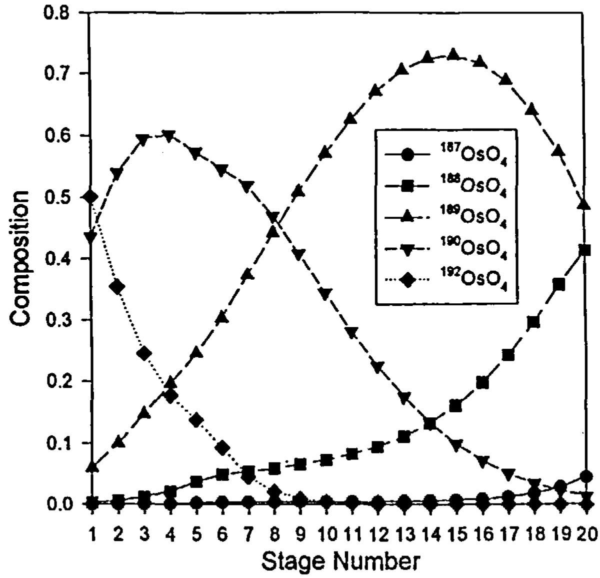

FIG. 4 Composition distribution of some Os isotopes in a cascade.

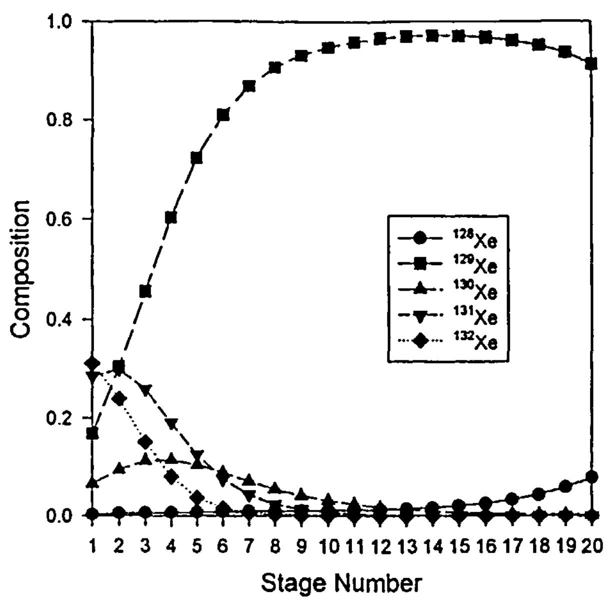

FIG. 5 Composition distribution of some Xe isotopes in a cascade.

TABLE 3
Separation of Chromium Using CrO_2F_2 for Different Total Stage Numbers N

N	20	16	12	8
$\underline{N_F}$	16	12	8	6
$\underline{M_1}$	52.24	52.24	52.23	52.21
$\underline{M_2}$	52.18	52.18	52.18	52.16
$\underline{M_3}$	52.15	52.15	52.14	52.12
$\underline{M_4}$	52.12	52.12	52.11	52.08
$\underline{M_5}$	52.10	52.10	52.09	52.05
$\underline{M_6}$	52.09	52.09	52.07	52.02
$\underline{M_7}$	52.08	52.08	52.05	51.97
$\underline{M_8}$	52.08	52.07	52.03	51.91
$\underline{M_9}$	52.07	52.06	52.01	
$\underline{M_{10}}$	52.07	52.05	51.98	
$\underline{M_{11}}$	52.07	52.04	51.94	
$\underline{M_{12}}$	52.06	52.02	51.89	
$\underline{M_{13}}$	52.06	52.00		
$\underline{M_{14}}$	52.05	51.98		
$\underline{M_{15}}$	52.04	51.94		
$\underline{M_{16}}$	52.02	51.88		
$\underline{M_{17}}$	52.00			
$\underline{M_{18}}$	51.97			
$\underline{M_{19}}$	51.94			
M_{20}	51.88			
$C_{P,i}$:				
^{50}Cr	8.61%	8.61%	8.56%	8.40%
^{52}Cr	89.66%	89.59%	89.33%	88.03%
^{53}Cr	1.64%	1.72%	2.02%	3.33%
^{54}Cr	0.075%	0.076%	0.082%	0.226%
$C_{W,i}$:				
^{50}Cr	0.002%	0.010%	0.059%	0.213%
^{52}Cr	77.86%	77.93%	78.19%	79.49%
^{53}Cr	17.46%	17.37%	17.07%	15.77%
^{54}Cr	4.68%	4.68%	4.68%	4.53%

present in Fig. 3: one is for ^{183}W and the other is for ^{184}W. The maximum of ^{183}W appears between Stages 2 and 3. In Table 2 the average molar weight of W at Stage 2 is 184.21 and the average molar weight of W at Stage 3 is 183.94. Therefore, the maximum composition appears at the nth stage, where $\overline{M}_n \approx M_i$. All the maximums of the composition curves in Figs. 3–5 correspond to

$$\overline{M}_n \approx M_i \tag{25}$$

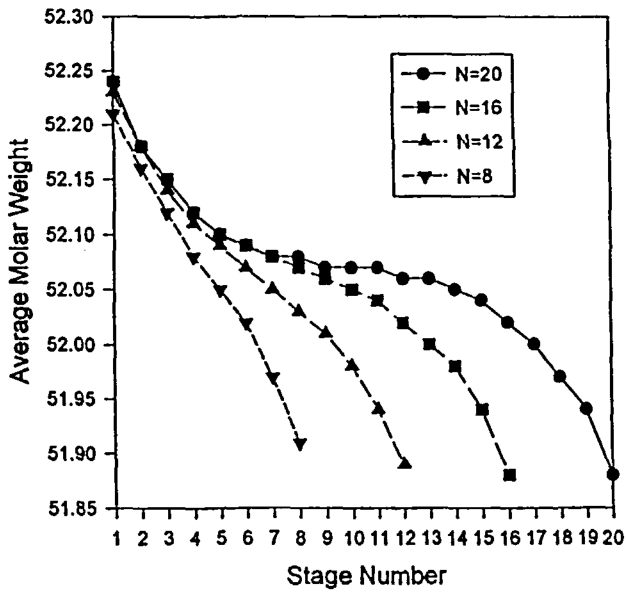

FIG. 6 Average molar weight distribution for different total stage numbers for the separation of chromium.

Application of the Concept of Average Molar Weight to Designing Separation Cascades

The concept of average molar weight can be easily used to check the design of a separation cascade. The results for the separation of chromium will be used as an example. The average molar weight curve for CrO_2F_2 in Fig. 2 shows that it varies very little in the middle of the separation cascade. Therefore, some portion of the cascade is not necessary for the separation of chromium under the given operating conditions. Table 3 lists calculated results for different total numbers of stages under identical operating conditions, i.e., under the same F/G, P/F, displayed in Table 1. The results show that when the total number of stages is reduced from 20 to 12, C_{Pi} and C_{Wi} for all components are nearly identical. Therefore, for the given operating conditions, only 12 stages are needed for the separation. In Fig. 6 the average molar weight curves are plotted for different total numbers of stages. The curves show how the distribution of the average molar weight changes with the total stage number, N, of the separation cascade. The average molar weight curve for $N = 12$ is best because the average molar weight changes uniformly from one stage to another. When $N = 8$ C_{Pi} and C_{Wi} change too much to obtain the desired values.

CONCLUSION

The average molar weight is a characteristic parameter for separation cascades used to separate multicomponent isotopic mixtures. The average molar weight at the nth stage in the cascade, $\overline{M}_n$, decreases monotonically with stage number. In other words, $\overline{M}_n$ decreases along the enriching direction. The composition of the lightest and the heaviest components varies monotonically along the separation cascade. The composition of the intermediate components may sometimes reach its maximum value in the cascade, but not at the end stages. There is no minimum composition of any component in the cascade. The average molar weight is a good parameter for judging cascade design.

The conclusion is proven in some special cases, such as when there are no withdrawals from the separation cascade or $P_n^*/(\theta_n G_n) \ll \ln \gamma_0$. It is correct for many examples including the example given in this paper. However, the conclusion is still not proven for the general case.

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