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On a Criterion Efficiency for Multi-Isotope Mixtures Separation

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ABSTRACT

The criterion for efficiency for separation of binary isotopic mixtures is the well-known value function. However, in the case of multi-isotope separation, this value function does not exist. In this paper we develop a criterion to define the efficiency for separation of multi-isotope mixtures. It is based on the concept of the match-abundance ratio cascade (MARC) originally introduced by De La Garza for a ternary isotope mixture. The criterion has the property that, when applied to binary mixtures, it is the same as the value function. This approach has demonstrated that for obtaining the optimal parameters of a single stage in the cascade it is necessary to minimize the linear combination of the inverse values of "partial separative powers" for all mixture components. A numerical example using this efficiency criterion is presented using the separation of chromium isotopes by a single gas centrifuge.

INTRODUCTION

The primary interest in gas centrifuges was originally for the enrichment of the fissionable isotope U^{235} for use as fuel in nuclear power reactors. This is a binary separation between the two naturally existing isotopes of uranium, U^{235} and U^{238}. Today, much interest exists in multicomponent isotope separation for the cases of stable isotopes as well as reenrichment of the uranium that has been removed from power reactors and contains several new isotopes. Most of the literature has dealt with the binary separation case, and the metric for

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measuring the efficiency of the separation is called the "value function" developed by Cohen (1). Although numerous optimization strategies have been used, no corresponding metric has been developed for the case of multicomponent mixtures. In this paper we develop an efficiency criterion for separation of multicomponent mixtures that has the desirable property that it reduces to the value function in the case of binary mixtures. In this paper we illustrate the new criterion with calculations for the separation of chromium isotopes in a centrifuge.

The cost of work for isotope mixture separation composes the biggest part in the price of the enriched product. For series of isotope separation methods a good measure of the cost of isotope separation is the total number of separating units (elements) making up the separating cascade. If the total flow in a cascade is known and all its units are working under the same conditions, the whole number of separating elements n can be calculated as

$$n = \frac{\Sigma L}{G} \tag{1}$$

where ΣL is the total flow in the cascade and G is the flow entering each element of a cascade.

A cascade for isotope separation is usually constructed so that the head stream from a stage furnishes a part of a feed to the next stage up and a tail stream from the stage is a feed to the adjacent lower stage (so-called symmetric arrangement) (1). If one considers a generic cascade for binary mixture separation with a feed flow rate F and a concentration of a desired component C_{F} and releasing one product stream with flow rate P and concentration C_{P} and one waste stream with flow rate and concentration W and C_{W}, respectively, then the sum in Formula (1) can be derived by using the concept of "an ideal cascade." The idea of an ideal cascade was introduced by Karl Cohen (1) to get the most efficient cascade. In such a cascade the interstage flows that merge at each confluent point possess identical compositions:

$$C_{\mathrm{S}} = C'_{\mathrm{S}-1} = C''_{\mathrm{S}+1} \tag{2}$$

or

$$R_{\mathrm{S}} = R'_{\mathrm{S}-1} = R''_{\mathrm{S}+1} \tag{3}$$

where C_{S} is the concentration (mole fraction) of the desired component contained in the Sth stage feed flow. Primes (′ and ″) denote the enriched and depleted flows, respectively, and symbols without a prime pertain to the feed flow. R is the abundance ratio

$$R = C/(1 - C) \tag{4}$$

As has been shown in Refs. 1, 18, and 25, the nonmixing conditions for a desired isotope in the flow mixing points under certain conditions lead to the most efficient design of a cascade in which the total flow, and hence the total number of separating elements, is minimized. Calculating the total flow L for a symmetric ideal cascade, one can get the following expression for the total number of separating units in a cascade:

$$n = \Delta U/\delta U \tag{5}$$

where

$$\Delta U = PV(C_P) + WV(C_W) - FV(C_F) \tag{6}$$

$$V(C) = (2C - 1)\ln\{C/(1 - C)\} = \frac{R - 1}{R + 1}\ln(R) \tag{7}$$

where $V(C)$ is a separation potential.

For a separating cascade with small separation factors ($\alpha = R'/R''$ is close to 1), the value of δU is

$$\delta U = G(\alpha - 1)^2/8 \tag{8}$$

For the case when the degree of desired isotope enrichment per stage unit is not small, but α is independent of mixture concentrations and cut $\theta = G'/G$, the separative power for a single separating unit will be

$$\delta U = G\frac{\sqrt{\alpha} - 1}{\sqrt{\alpha} + 1}\ln\sqrt{\alpha} \tag{9}$$

The numerator in Formula (5) is defined by the external parameters of a cascade. The unit's separative characteristics do not enter into it. This value is called the "separation performance" of a cascade, and one may consider it as an external load. The denominator in the same formula, defined by Expression (8) or (9), is called the "separative power" of a unit and depends on separation properties of a single element. Thus, for a binary isotope mixture separation the process of optimization reduces to the definition of the working parameters providing a maximum value of the separative power δU.

In the case of a binary isotope mixture separation one may come to the Relations (5), (7)–(9) by another more formal way. Suppose that a stage is separating L_S moles with a concentration of desired isotope C_S into $\theta_S L_S$ moles with a concentration C'_S and $(1 - \theta_S)L_S$ moles with a concentration C''_S; θ_S is the cut of the stage. According to an idea of several of the authors (2), the separative power of a stage dU is a change in the value of mixture obtained by separation:

$$dU_S = L_S[\theta_S V(C'_S) + (1 - \theta_S)V(C''_S) - V(C_S)] \tag{10}$$

Assume that a symmetric ideal cascade has a feed flow F with the concentration of desired isotope C_F and one product flow P and one waste flow W with the concentrations of desired isotope C_P and C_W, respectively. Summing Eq. (10) over all stages of a cascade and taking into account that in an ideal cascade the separative powers of the stages are used in full, one can get ΣdU_S from the left side of the equation. Making the sum on the right side of Eq. (10), it is necessary to use the condition that the flows in an ideal cascade are connected without mixing of concentrations. This means that during summing, all members describing the interstage flows disappear except for three external ones with their own concentrations. As the result, one will get

$$\sum dU_S = PV(C_P) + WV(C_W) - FV(C_F) \tag{11}$$

So on the left side of Eq. (11) in the absence of mixing losses, one has the sum of stage separative powers and on the right side the separative power considering an ideal cascade as a single unit. This allows us to write

$$\Delta U = PV(C_P) + WV(C_W) - FV(C_F) \tag{12}$$

The separative power ΔU depends on neither the nature of the separation process being used nor on the scheme followed in the stage connections, but it merely depends on the external parameters of the cascade. That is, flow rates and compositions of the mixture coming into the cascade and out of it.

Now we shall consider the situation with the choice of a function $V(C)$. Formally, as it follows from Eqs. (11) and (12), the function $V(C)$ may be introduced in an arbitrary form, but from a practical point of view the choice of a function $V(C)$ should produce a form of the stage (element) separative power that is independent of the mixture concentrations. In this case if all separating units (elements) in the cascade have identical parameters, then

$$\sum dU_S = n\delta U \tag{13}$$

where δU is the separative power of the individual element and n is the total number of elements in the cascade. By combining the Relations (11), (12), and (13), we again obtain Expression (5).

The functional form of $V(C)$ is determined by requiring that the term in parentheses of Eq. (10) is a function of separation factors, and α does not depend on the cut θ or concentration C. The solution of this functional equation for the symmetric case may be obtained in Form (8) or (9), and the function $V(C)$ has Form (7) if the point of zero value is arbitrary placed at $C = 0.5$. Similar approaches exist for the definition of the separation potential and the separative power for the ideal cascade of asymmetric elements ($\theta \neq 0.5$) (2).

Up to the present time the problem of defining a criterion efficiency which can be used for separation of multi-isotope mixtures has not been solved. In this case the difficulties of definition for the separative performance of the separative unit are connected with the problem that the no-mixing condition in a cascade is not applicable since it is not normally possible to match more than one concentration in a stream. The attempts to introduce the expressions for δU and $V(C_1, C_2, \ldots, C_\mathrm{m})$ by solving the functional equation

$$\delta U/L = \theta V(C_1', ..., C_\mathrm{m}') + (1 - \theta)V(C_1'', ..., C_\mathrm{m}'') - V(C_1, ..., C_\mathrm{m}) \quad (14)$$

or by extending the theory for binary mixture separation to the multi-isotope separation case (3–6) can be done formally. However, the results are inconvenient for practical use because of the nonvalidity of the expressions like Eqs. (12) and (13) obtained for a multi-isotope case. Note that in Eq. (14), $(C_1, C_2, \ldots, C_m)$ is the composition of the mixture and m is the number of components. In our opinion, a more basic approach to evaluation of the multicomponent separation performance is to use the theory for a multicomponent separating cascade which is an analog of the two-component “ideal cascade.” The theory of such cascades was established by De La Garza et al. (7, 8) and then developed in detail in a number of papers (9–17). The multicomponent analogy of an ideal cascade for binary isotope mixture separation is a cascade in which the abundance ratio between the nth and $(n + k)$th components $R_{n,n+k} = C_n/C_{n+k}$ [n and $(n + k)$ are designated as the “key” components] is matched whenever two streams come together; that is, at all interstage connections and feed points. Such a cascade is termed a “matched abundance ratio cascade” (MARC). Hence, for MARC one can write

$$R'_{n,n+k}(S - 1) = R_{n,n+k}(S) = R''_{n,n+k}(S + 1) \quad (15)$$

where S is the current number of a stage in the cascade.

The analysis of the papers cited above reveals the following important properties of MARC:

1. Its main characteristic is that it reduces to the no-mixing conditions used for the binary system.
2. When $k = 1$, MARC allows extraction in the product flow the key (nth) component together with the components with numbers $1, 2, \ldots, n - 1$, and “to depress” in this stream the components with the numbers $n + 1, n + 2, \ldots, m$. The possibility of changing the numbers of the components separated together in the product or waste streams, as the result of varying n, makes MARC promising for the solution of separation problems involving the production of mixtures enriched with components of intermediate weight.
3. As was demonstrated by Gadkary and Govind (18), the total interstage flow in MARC represents the minimum total interstage flow in a multicomponent cascade for a desired separation. It is assumed that in this case the values of the abundance ratio in the product and waste streams are defined.

4. It has been established in Refs. (10, 19, and 20) that MARC can be the base model for the squared-off approximation of the multicomponent cascade.

The properties of MARC described above allow it to be used for solving the problem of introduction of “a separative power” and a separation potential for multi-isotope mixtures. It should be noted that the first time this approach was used in the papers of De La Garza (7, 8); particular concern was with a multicomponent cost formula which could be used to price uranium containing isotope ^{236}U. As the theory was developed for gaseous diffusion plants, the mathematical analysis was performed for the differential equation governing the separation potential for an infinitesimal process of separation.

THEORY

The purpose of the present study is to obtain the multicomponent analog of separative power and a separation potential for the general case when the separation factors for the cascade stages are not necessarily small; for example, in a gas centrifuge cascade. We shall examine a counterflow symmetric MARC consisting of N steps and intended for separation of an m-component isotope mixture. Let the feed stream F with component concentrations C_j^{F} ($j = 1, 2, \ldots, m$) be fed into the input of the step with the number f. As a matter of convenience we shall consider a matched $R_{n,n+1}$ cascade in the case when the separation factors do not vary from stage to stage. For such a MARC, it has been shown (14, 16, 17) that the flow rates of streams entering each stage of the cascade can be expressed by the following formulas.
For the enriching section:

$$L_{\mathrm{S}} = P \sum_{\xi=1}^{\mu} C_j^{\mathrm{P}} \frac{\beta_j + 1}{\beta_j - 1} (1 - \beta_j^{\mathrm{S-N+1}}) \tag{16}$$

For the stripping section:

$$L_{\mathrm{S}} = W \sum_{j=1}^{m} C_j^{\mathrm{W}} \frac{\beta_j + 1}{\beta_j - 1} (1 - \beta_j^{\mathrm{S}}) \tag{17}$$

where P is the cascade product stream; W is the cascade waste stream; C_j^{P} and C_j^{W} are the concentrations of jth component in the product and waste streams, respectively; $\beta_j = \alpha_{j,n+1}/\sqrt{\alpha_{n,n+1}}$, $j = 1, 2, \ldots, m$, and $\alpha_{j,n+1}$ is the overall stage separation factor for a pair of components with the numbers j and $n + 1$.

The values of $R^{\mathrm{P}}_{n,n+1}$, $R^{\mathrm{W}}_{n,n+1}$, and $R^{\mathrm{F}}_{n,n+1}$, are connected with the values N and f (defined in the previous paragraph) by the expressions

$$N - f + 1 = \frac{1}{\ln \beta_n} \ln \frac{R^{\mathrm{P}}_{n,n+1}}{R^{\mathrm{F}}_{n,n+1}} \tag{18}$$

$$f = \frac{1}{\ln \beta_n} \ln \frac{R^{\mathrm{F}}_{n,n+1}}{R^{\mathrm{W}}_{n,n\ +1}} \tag{19}$$

The ratios of the cascade product rate to the cascade feed and waste rates to the cascade feed rate are given by

$$\frac{P}{F} = \sum_{j=1}^{m} C^{\mathrm{F}}_j \frac{1 - \beta_j^{-\mathrm{f}}}{1 - \beta_j^{-\mathrm{N}-1}} \tag{20}$$

$$\frac{W}{F} = \sum_{j=1}^{m} C^{\mathrm{F}}_j \frac{\beta_j^{\mathrm{N}-\mathrm{f}+1} - 1}{\beta_j^{\mathrm{N}+1} - 1} \tag{21}$$

and the concentrations of the individual components in the cascade product and waste streams are written as

$$C^{\mathrm{P}}_i = C^{\mathrm{F}}_i \frac{1 - \beta_i^{-\mathrm{f}}}{1 - \beta_i^{-\mathrm{N}-1}} \Bigg/ \sum_{j=1}^{m} C^{\mathrm{F}}_j \frac{1 - \beta_j^{-\mathrm{f}}}{1 - \beta_j^{-\mathrm{N}-1}} \tag{22}$$

$$C^{\mathrm{W}}_i = C^{\mathrm{W}}_i \frac{\beta_i^{\mathrm{N}-\mathrm{f}+1} - 1}{\beta_i^{\mathrm{N}+1} - 1} \Bigg/ \sum_{j=1}^{m} C^{\mathrm{F}}_j \frac{\beta_j^{\mathrm{N}-\mathrm{f}+1} - 1}{\beta_j^{\mathrm{N}+1} - 1} \tag{23}$$

It should be noted that for many isotope separation processes the stage separation factor for the pair of components can be expressed as a function of the difference between their molecular masses. For example, for the case of gas centrifugation it is appropriate to use the following approximation (16):

$$\alpha_{ik} = \alpha_0^{\mathrm{M}_k - \mathrm{M}_i} \tag{24}$$

where α_0 is the overall stage separation factor per unit mass difference; M_k and M_i are the molecular masses of the component with the numbers k and i. Thus, in the case of the isotope separation by gas centrifuges, β_j will be transformed into the expression

$$\beta_j = \alpha_0^{\mathrm{M}^* - \mathrm{M}_j}, \qquad (j \neq \mathrm{n}) \tag{25}$$

where $M^* = (M_n + M_{n+1})/2$.

The total flow in MARC, $\Sigma_{\mathrm{S}=1}^{N} L_{\mathrm{S}}$, will be obtained from Expressions (16) and (17) by combining them with the balance equations:

$$F = P + W \tag{26}$$

$$FC^{\mathrm{F}}_i = PC^{\mathrm{P}}_i + WC^{\mathrm{W}}_i \tag{27}$$

Simple transformations lead to the following result:

$$\sum_{S=1}^{N} L_S = \sum_{j}^{m} \frac{PC_i^{P} \ln R^{P}_{n,n+1} + WC_i^{W} \ln R^{W}_{n,n+1} - FC_i^{F} \ln R^{F}_{n,n+1}}{\dfrac{\beta_j - 1}{\beta_j + 1} \ln \beta_n} \tag{28}$$

If all elements of the cascade have the identical productivity G, the expression for the total number of the separating units in the cascade per unit of the product flow may be written as

$$\frac{n}{P} = \sum_{j=1}^{m} \frac{V_j(C_j^{P}, R^{P}_{n,n+1}) + \dfrac{W}{P} V_j\,(C_j^{W}, R^{W}_{n,n+1}) + \dfrac{F}{P} V_j\,(C_j^{F}, R^{F}_{n,n+1})}{\delta U_j} \tag{29}$$

where

$$V_j(C_j, R_{n,n+1}) = C_j \ln R_{n,n+1} \tag{30}$$

$$\delta U_j = G \frac{\beta_j - 1}{\beta_j + 1} \ln \beta_n \tag{31}$$

When the number of components is two, Expressions (29)–(31) become identical to ones obtained for the two-component ideal cascade. For the case of “a difficult separation” ($\alpha_0 = 1 + \epsilon_0 \approx 1$), these expressions are transformed into Formula (8).

It is obvious that δU_j can be considered as a stage separative power relevant to the jth component, and V_j may be interpreted as a partial separation potential for the jth component.

It is easy to find that for the case of a gas centrifuge, δU_j will be given as

$$\delta U_j = G \frac{(\alpha_0)^{M^* - M_j} - 1}{(\alpha_0)^{M^* - M_j} + 1} \ln(\alpha_0)^{M_{n+1} - M^*} \tag{32}$$

Analyzing Relations (20)–(23) for this case and taking into account Expressions (18) and (19), one may come to the conclusion that the numerator in Eq. (29) does not depend on the characteristics of a single separation element and only depends on the values of $R^{P}_{n,n+1}$, $R^{W}_{n,n+1}$, $R^{F}_{n,n+1}$ and the mass differences of the components $M^* - M_i$ and $M_{n+1} - M_n$.

Thus, in the case of multi-isotope separation for obtaining the optimal parameters of a single stage in the cascade, it is necessary to minimize the linear combination of the value $1/\delta U_j$ for all components.

RESULTS AND DISCUSSION

In this section we present an application of the above-defined “separative power” for the multi-isotope case. As an example, we consider the separation

TABLE 1
Separation of Chromium Isotopes. Centrifuge and Process Gas Data

Length of the centrifuge	15 m
Diameter of the centrifuge	0.5 m
Peripheral speed	800 m/s
Process gas	CrO_2F_2
Average gas temperature	315 K
Gas pressure at the wall	50 torr
Molecular weight	122 g/mol
Viscosity	1.40×10^{-5} kg/m·s
Heat capacity	636 J/kg·k
Thermal conductivity	0.00675 w/m·k
Ratio of specific heats	1.12
Schmidt number	0.75
Isotopic composition of the feed:	
Cr-50 4.35%	
Cr-52 83.79%	
Cr-53 9.50%	
Cr-54 2.36%	

of natural chromium isotopes in the form of CrO_2F_2 by a gas centrifuge described in Ref. 21 with the parameters shown here in Table 1.

Figure 1 demonstrates the results of calculations for α_0 versus feed rate in a gas centrifuge. One can see the smooth dependence obtained for the feed flows beginning with 50 mg/s. Figure 2 shows results of calculations for α_0 versus θ with the feed flow to the gas centrifuge $G = 50$ mg/s, the end-to-end

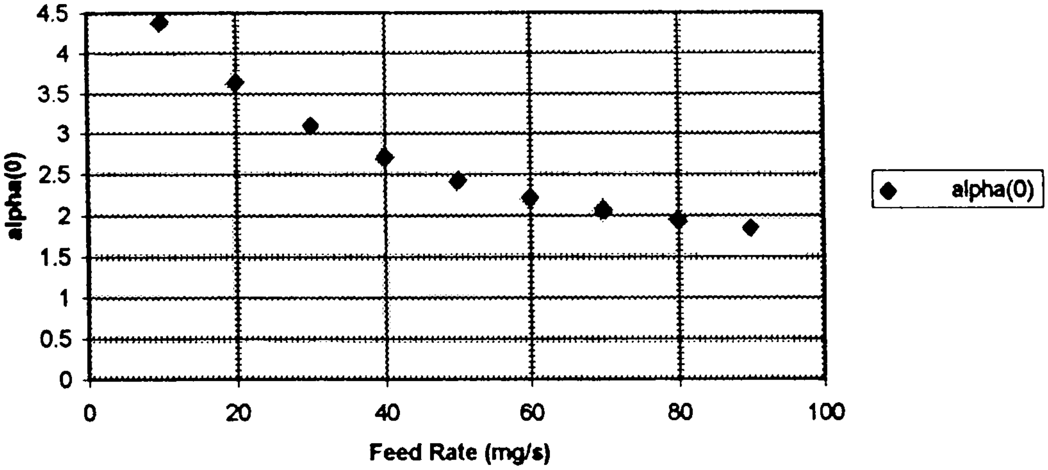

FIG. 1 α_0 vs feed rate for chromium.

TABLE 2
Concentrations for Stages 2 and 3

	Stage 2	Stage 3
Cr-50	16.2%	37.5%
Cr-52	79.4%	60.7%
Cr-53	4.0%	1.7%
Cr-54	0.4%	0.1%

temperature difference 5.66 K, and the scoop drag 5512 dynes. These are the optimal values of these parameters as shown in Table 4 of Ref. 21. The results show that over the range of cut from 0.25 to 0.75, α_0 remains essentially constant. For a very low or a very high cut, some variation exists.

The dependence of α_0 on feed concentration was next investigated. To do this, the optimal scoop drag and temperature difference were maintained, and three feed rates, 40, 60, and 80 mg/s, were used for three different sets of feed concentrations. The normal feed concentration in Table 1 was used and the concentrations to the next two stages in the cascade shown by Szady (22) were used. These concentrations are given in Table 2. The results of these calculations are shown in Table 3.

The information shown in Fig. 2 and Table 3 justifies the claim made in the Introduction that the "separative power" for the multi-isotope case defined in this work is independent of the characteristics of the separating unit.

Now we consider enrichment of chromium 50 from a naturally occurring four isotope mixture in MARC of the centrifuges described above with the following data in the definitions of the previous sections:

TABLE 3
α_0 vs Feed Concentrations

Feed rate (mg/s)	Concentrations	α_0
40	Normal	2.09604
	Stage 2	2.11951
	Stage 3	2.16474
60	Normal	1.80097
	Stage 2	1.80607
	Stage 3	1.81905
80	Normal	1.60757
	Stage 2	1.60827
	Stage 3	1.61355

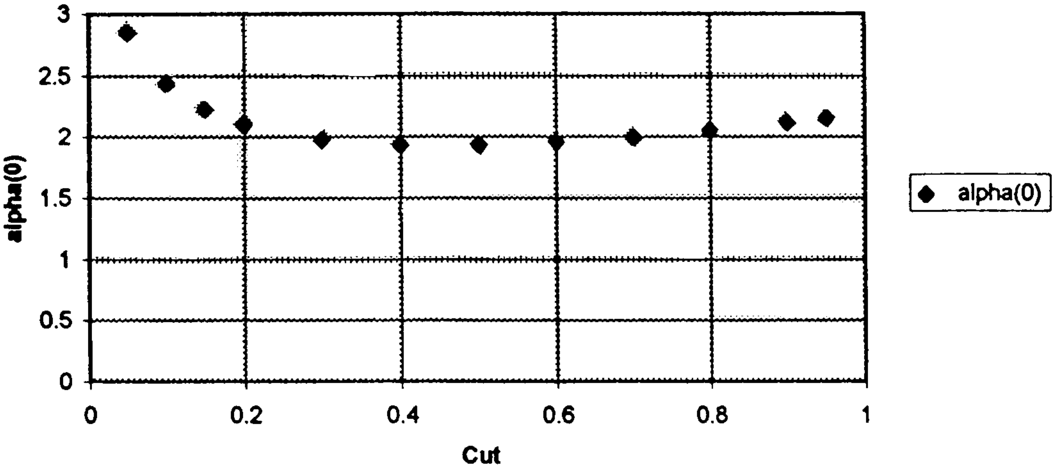

FIG. 2 α_0 vs cut for chromium.

- Desired isotope Cr-50 ($n = 1$)
- $M^* = 51$
- Values of abundance ratio in the product, waste and feed flows are

$$R_{12}^{\mathrm{P}} = 51.4565425$$

$$R_{12}^{\mathrm{W}} = 0.0005145$$

$$R_{12}^{\mathrm{F}} = 0.0514565$$

For these values of R_{12}, the concentrations of the desired isotope in the product and waste flows are

$$C_1^{\mathrm{P}} = 0.980934$$

$$C_1^{\mathrm{W}} = 0.000450$$

The results of calculations of the partial separative powers and n/P versus feed flow with a cut of 0.5 are presented in Figs. 3 and 4. In Fig. 3, Series j corresponds to δU_j. It is of interest to note that the positions of the maxima for the partial separative powers are close to each other. Also, the positions of optimal values for n/P and δU_j are approximately coincident. So if one wants to find the optimal conditions for multi-isotope separation in a single gas centrifuge, it is enough to optimize the partial separative power for the desired component:

$$\delta U_n = \frac{G}{2}(M_{n+1} - M_n)\frac{\alpha_0^{\frac{\mathrm{M}_{n+1}-\mathrm{M}_n}{2}} - 1}{\alpha_0^{\frac{\mathrm{M}_{n+1}-\mathrm{M}_n}{2}} + 1}\ln \alpha_0 \tag{33}$$

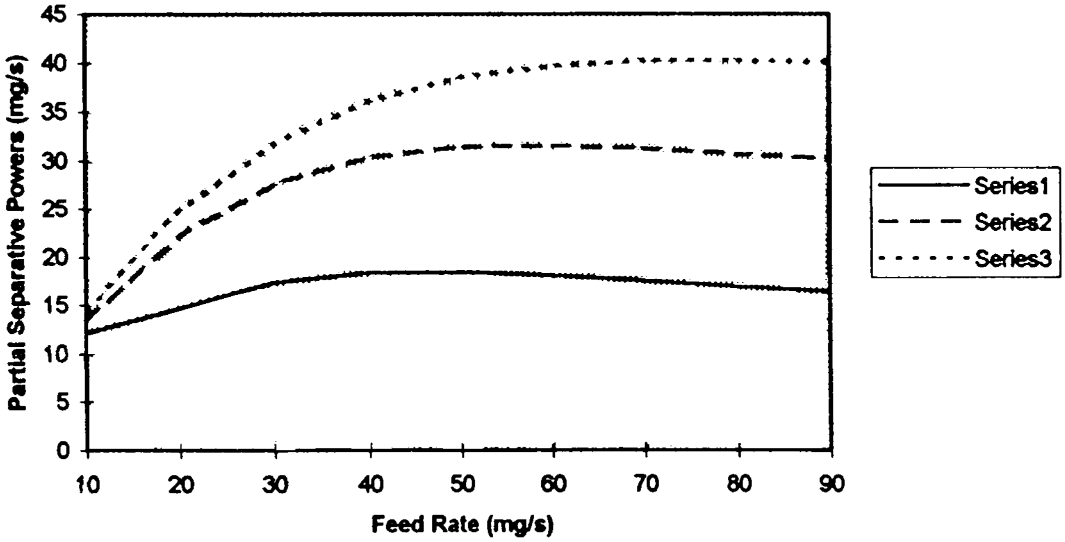

FIG. 3 Partial separative power vs feed rate.

For the case of the difficult separation ($\alpha_0 = 1 + \epsilon_0 \approx 1$), Expression (33) will be simplified to

$$\delta U_n = G\varepsilon_0^2(M_{n+1} - M_n)^2/8 \tag{34}$$

In Refs. 23 and 24 the separation of multicomponent isotope mixtures by a

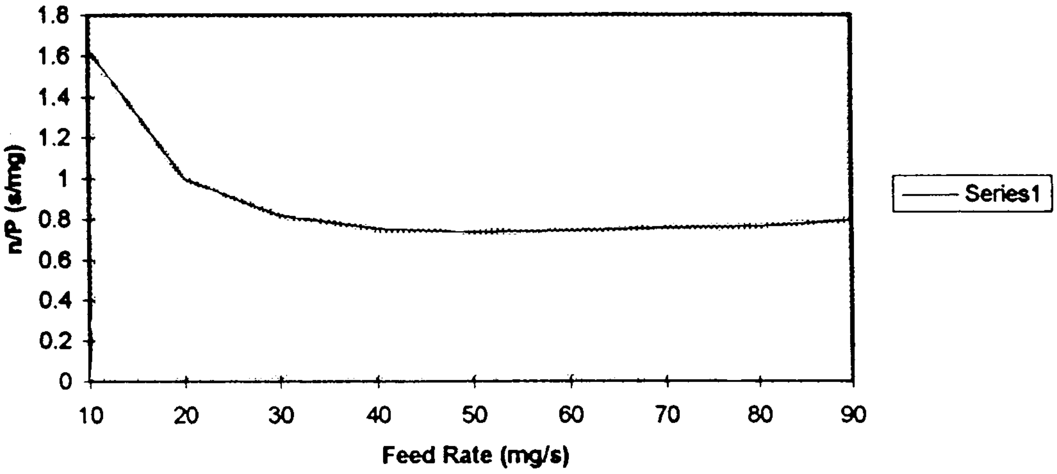

FIG. 4 *n*/*P* vs feed rate.

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gas centrifuge was considered, and the above result confirms the validity of the formula

$$e = G\varepsilon_0^2/8 \tag{35}$$

which was used as a criterion efficiency in both of those papers without a theoretical background.

CONCLUSIONS

A formula to calculate a “separative power” for the case of a multi-isotope separation in a cascade with arbitary separation factors of its units is proposed in this article. In contrast to the earlier proposed formulas for calculation of separation characteristics of multicomponent mixtures, the ones obtained here are simpler and more intuitively obvious. The criterion developed here has the desirable property that when the number of components is two, the expressions obtained become identical to ones for binary separation.

The approach developed dictates an easy strategy for optimizing the process of multicomponent isotope mixture separation. The optimal parameters of a separating unit of a cascade may be found by the optimization of the “partial separative power” of the desired isotope. The calculations based on this approach and made for real gas centrifuges for chromium-50 enrichment have demonstrated the validity of the formulas and strategy proposed for the case of multi-isotope separation.

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