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# Comparative Study of the Model and Optimum Cascades for Multicomponent Isotope Separation

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The possibility of estimating the minimum total flow in a cascade with concentrations of a target component given in the product and waste flows by means of a model match abundance ratio cascade (MARC) is studied. The parameters required to describe MARC characteristics are the total number of separation stages, the feed flow location, and the  $M^*$  parameter, which is equal to a half-sum of mass numbers of the target and the supporting components. Specific research carried out independently in two scientific labs in China and Russia has demonstrated that the integral parameters of the MARC, optimized by the  $M^*$  parameter, are very close to that of the optimum by the minimum total flow cascade found by means of numerical optimization. The calculation is performed for separation of krypton isotopes when the end component  $^{78}\text{Kr}$  and the intermediate component  $^{83}\text{Kr}$  are considered to be the targets. It paves the way to use the optimized MARC parameters for two purposes: first, for fast and easy evaluation of the real cascade parameters and second, as an initial guess in its further direct numerical optimization, thereby allowing significant savings in computation time.

**Keywords** isotope; model cascade; optimum cascade; separation

## INTRODUCTION

There is considerable interest in the theoretical and practical aspects of finding the optimum cascades parameters for the separation of multicomponent isotope mixtures with the given concentrations of a target component in its external flows. As the optimum cascade, we will consider in this paper a cascade, whose parameters provide the minimum total flow without any other additional conditions imposed on its internal or external characteristics. The common principles for optimization of such cascades have been developed in (1,2). However, there are problems implementing these in practice due to the necessity of initiating the process with a reasonable initial guess and to

carry out the time-consuming calculation of cascade parameters on each optimization step. It is noteworthy that the most difficult problems occur when one needs to calculate the optimum parameters of the cascade with the fixed values and/or limitations on the withdrawal concentrations of a target component. With such cases, it is reasonable to question the extent of the set of optimum parameters for the quasi-ideal cascade. This is the analog of the ideal case for the multicomponent isotope mixture separation and for which it is possible to find its optimum parameters (3,7,8,10–12), coincides with that for the optimum cascade. If there is agreement between the estimates for these cases, as it is in the case of the binary isotope mixture, then the method can provide a better tool for the estimation of optimum cascade parameters rather than a more difficult process of optimization. The study of this feasibility is just the subject of the present paper.

## THEORY

According to the definition, in the quasi-ideal cascade, the component flow cuts  $\phi_i$  are constant over all cascade stages (3), i.e.,

$$\begin{aligned}\phi_i &= G'_i/G_i = \text{const}, \\ (1 - \phi_i) &= G''_i/G_i = \text{const}, \\ i &= 1, 2, \dots, N_c,\end{aligned}\quad (1)$$

where  $G'_i$ ,  $G''_i$  are the  $i$ th component flows in the enriched and depleted outlet flows from a stage, respectively;  $G_i$  is the  $i$ th component flow entering a stage;  $i$  is the sequence number of a component,  $i = 1, 2, \dots, N_c$ ; and  $N_c$  is the number of components in the separating mixture.

The condition equivalent to (1) for existence of a quasi-ideal cascade is the constancy of head and tail stage separation factors:  $\gamma'_{ik} = \frac{C'_k C_k}{C'_k C_i} = \text{const}$ ,  $\gamma''_{ik} = \frac{C_i C''_k}{C_k C''_i} = \text{const}$ , where  $k$  is a number designated as the supporting component;  $C_i$ ,  $G'_i$ ,  $G''_i$  are concentrations of the  $i$ th component in the feed, product, and waste flows respectively.

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As already known, the notions of the product and waste flows in cascades for multicomponent isotope separation are relative. It is therefore possible to identify “the light and heavy ends” of a cascade, where the lightest and heaviest together with other accompanying components are enriching. The light end withdrawal  $P$  and the heavy end withdrawal  $W$  of a quasi-ideal cascade are determined by the equations as follows (3,4):

$$\frac{P}{F} = \sum_{i=1}^{N_c} C_{Fi} \frac{1 - \beta_i^{-N_F}}{1 - \beta_i^{-N_S-1}}, \quad \frac{W}{F} = \sum_{i=1}^{N_c} C_{Fi} \frac{1 - \beta_i^{N_S-N_F+1}}{1 - \beta_i^{N_S+1}}$$

The concentrations of the  $i$ th component  $C_{Pi}$  and  $C_{Wi}$  in these flows are found by the following formulas:

$$\begin{aligned} C_{Pi} &= C_{Fi} \frac{1 - \beta_i^{-N_F}}{1 - \beta_i^{-N_S-1}} \left/ \sum_{j=1}^{N_c} C_{Fj} \frac{1 - \beta_j^{-N_F}}{1 - \beta_j^{-N_S-1}} \right., \\ C_{Wi} &= C_{Fi} \frac{1 - \beta_i^{N_S-N_F+1}}{1 - \beta_i^{N_S+1}} \left/ \sum_{j=1}^{N_c} C_{Fj} \frac{1 - \beta_j^{N_S-N_F+1}}{1 - \beta_j^{N_S+1}} \right., \\ i &= 1, 2, \dots, N_c. \end{aligned} \quad (3)$$

The total flow in the cascade is calculated by the expression

$$\begin{aligned} \sum_{n=1}^{N_S} G_n &= F \sum_{i=1}^{N_c} C_{Fi} \frac{\beta_i + 1}{(\beta_i - 1)(1 - \beta_i^{N_S+1})} \\ &\quad \left[ -(N_S - N_F + 1)\beta_i^{N_S+1} + (N_S + 1)\beta_i^{N_S-N_F+1} - N_F \right] \end{aligned} \quad (4)$$

where  $G_n$  is the entering flow at the stage  $n$ ;  $F$  is the cascade feed flow;  $N_S$  is the total number of stages in the cascade;  $N_F$  is the number of the stage where the feed flow enters;  $\beta_i = \gamma'_{ik}(\gamma''_{ik} - 1)/(\gamma'_{ik} - 1)$ ,  $i \neq k$ ,  $\beta_k = (\gamma''_{ik} - 1)/(\gamma'_{ik} - 1)\gamma''_{ik}$ .

For one particular case, a quasi-ideal cascade may have symmetric separation for the  $m$  and  $k$  components at all separation stages. This condition leads to the following equality for the stage head and tail separation factors:

$$\gamma'_{mk} = \gamma''_{mk} = \sqrt{\gamma_{mk}}, \quad \gamma'_{ik} \neq \gamma''_{ik}, \quad i \neq m. \quad (5)$$

where  $\gamma_{mk}$  is an overall separation factor.

In this case, the abundance ratio between the  $n$ -th and  $k$ -th components is matched wherever two streams come together: i.e., at all interstage connections and feed points. Such a cascade is termed as a matched abundance ratio cascade, or MARC. It may also be referred to in Russian language papers as the “*R*-cascade”.

De La Garza et al. are credited with the initial theory of a MARC for overall separation factors close to unity (5,6). Later the theory was broadened for the case of arbitrary

separation factors (7,8,10–12). In the case of isotope separation by molecular-kinetic methods (gas centrifuge, gas diffusion, thermal diffusion, mass diffusion, etc.), the stage separation factor for the pair of components can be expressed as a function of the difference between their mass numbers in the following equation:

$$\gamma_{ik} = \gamma_0^{M_k - M_i}, \quad (6)$$

where  $M_i$  and  $M_k$  are the mass numbers of the  $i$ th and  $k$ th component and  $\gamma_0$  is the overall separation factor for unit mass numbers difference. In this case  $\beta_i = \gamma_0^{M^* - M_i}$ , where  $M^* = (M_m + M_k)/2$ .

The case when the overall separation factors at separation stages are close to unity, the parameter  $M^*$  that is equal to the arithmetic mean of the mass numbers of the key (target) component  $m$  and the supporting component  $k$  appeared for the first time in (9). The approach suggested that it is possible to introduce components, whose concentrations are negligible ( $C_{Fm} \rightarrow 0$  and/or  $C_{Fk} \rightarrow 0$ ), with mass number  $M^*$  lying in the range  $M_1 < M^* < M_{N_c}$  (8). In this case it is not necessary that either the  $m$ th or the  $k$ th component be present in the multicomponent mixture. As a result, the value of  $M^*$  can vary continuously in a range from the lightest to the heaviest masses of a separating isotope mixture and therefore may be used as a free parameter. The practical effect of this outcome is that  $M^*$  may assume any real value regardless of the actual mass numbers of the components present. So in practice,  $M^*$  is assigned the value that optimizes the performance of a cascade. Note that sometimes the  $M^*$  parameter is referred to as the “key” molar mass (10). The parameter  $M^*$ , as a special optimization variable applied in (11), allows adjustment and analysis of quasi-ideal cascade properties together with integer type stage number and feed location. These have been used as the optimization variables to minimize the total flow of cascades in (12).

## CALCULATION

Firstly, we review the process of finding the optimum parameters for a MARC. There are only 3 variables in this optimization problem using the minimum total flow in a MARC as an efficiency criterion. Two of them are integer variables: the number of stages  $N_S$  and the feed location  $N_F$ , while the key mass number  $M^*$  is a real variable. In this case the optimization problem can be summarized as “Problem 1”:

$$\begin{aligned} \min \sum G(N_F, N_S, M^*)/F, \\ s.t. \quad C_{Pt}(N_F, N_S, M^*) \geq C_{Pt}^*, \\ C_{Wt}(N_F, N_S, M^*) \leq C_{Wt}^*, \\ N_F \in \mathbb{Z}, N_S \in \mathbb{Z}, M^* \in R. \end{aligned} \quad (7)$$

where  $C_{Pt}^*$  and  $C_{Wt}^*$  are the given concentrations of the target component in the product and waste flows, respectively;  $Z$  establishes the integer set of parameters and  $R$  sets up the real one.

To simplify the solution of the mixed (integer and real) optimization, two integer variables can be substituted for the real variables and the simplified problem is now identified as "Problem 2":

$$\begin{aligned} \min & \sum G(N_F, N_S, M^*)/F, \\ \text{s.t.} & C_{Pt}(N_F, N_S, M^*) \geq C_{Pt}^*, \\ & C_{Wt}(N_F, N_S, M^*) \leq C_{Wt}^*, \\ & N_F, N_S, M^* \in R. \end{aligned} \quad (8)$$

One further step to facilitate solving the problem under investigation is to transform the constraints for specified concentrations into a penalty function. In this way, the problem becomes a nonlinear optimization without constraints.

In the common case for the system of equations describing the process of mass transfer in the multicomponent separating cascade (1,2), the problem of finding its optimum, by using the minimum total flow as the efficiency criterion, is formulated as "Problem 3", which was solved for the first time in (1):

$$\begin{aligned} \min & \sum G(\mathbf{C}, N_F, N_S)/P, \\ \text{s.t.} & C_{Pt}(\mathbf{C}, N_F, N_S) \geq C_{Pt}^*, \\ & C_{Wt}(\mathbf{C}, N_F, N_S) \leq C_{Wt}^*, \\ & \mathbf{C} = \{C''_2, \dots, C''_{N_S-1}\} \in (0, 1)^{N_S-2}, N_F \in Z, N_S \in Z, \end{aligned} \quad (9)$$

where  $G''_n$  is the concentration of the target component in the depleted flow of the  $n$ th stage.

The analysis demonstrates that if the parameters of the cascade feed ( $F, C_{Fj}$ ), the values of concentrations of a target component in the product  $C_{Pt}$  and waste  $C_{Wt}$  flows, as well as the  $N_S$  and  $N_F$  parameters are fixed, the quantity of free parameters of the cascade will be equal to  $N_{S-2}$ . For the minimum total flow in a cascade as the efficiency criterion, it has been suggested to use the waste concentrations of the lightest component  $C''_1(n)(n = 2, N_S - 1)$  in the separating mixture at the  $N_{S-2}$  stages as the free  $N_{S-2}$  parameters.

Hence, "Problem 3" can be rewritten in the problem statement and renamed as "Problem 4" as in the following:

$$\begin{aligned} \min & \sum G(\Theta, N_F, N_S)/P, \\ \text{s.t.} & C_{Pt}(\Theta, N_F, N_S) \geq C_{Pt}^*, \\ & C_{Wt}(\Theta, N_F, N_S) \leq C_{Wt}^*, \\ & \Theta = \{\theta_2, \dots, \theta_{N_S-1}\} \in (0, 1)^{N_S-2}, N_F \in Z, N_S \in Z, \end{aligned} \quad (10)$$

where  $\theta_n$  is the cut at the  $n$ th stage. Note that searching for the optimum parameters in the above problem statement is much more complicated than for "Problem 1" or "Problem 2".

For sorting of concentration  $G''_1(2), \dots, G''_{N_S-1}$  in the optimization procedure, it is expedient to utilize different methods of nonlinear programming and, if it is necessary, to combine these with a random searching method (13). As a whole, the problem of optimization appears to be relatively labor-intensive. In addition, the associated difficulties increase with the increase of the number of stages  $N_S$  in a cascade.

Researchers at MEPhI have followed the optimization algorithm described in (1). In addition, they have applied a special method approximating the value of a stage separation factor to calculate the parameters of a cascade (14).

With respect to "Problem 4" researchers at Tsinghua University have carried out independently the direct numerical optimization of the optimum cascade without the above constraints of a MARC using simulated annealing (15,16) and the Hooke-Jeeves method (17). In addition, the  $Q$ -iteration method (18) was applied to calculate the concentration distributions and the optimization variables  $N_S, N_F$  as well as the flow cuts at the  $N_{S-2}$  intermediate stages.

## RESULTS AND DISCUSSION

The mixture of krypton isotopes of the natural abundance (see Table 1) has been chosen as the inlet concentrations.

The overall separation factor for the unit mass number difference was fixed at  $\gamma_0 = 1.1$  and did not vary over the stages of cascades.

In Case 1, the research was devoted to the optimization of the cascade, enriching the lightest component of the mixture with the concentrations of  $^{78}\text{Kr}$  in the product and waste flows and satisfying the conditions as follows:  $C_{Pt} \geq 20\%$ ,  $C_{Wt} \leq 0.12\%$ . The results of optimization by  $M^*$  of the dimensionless total flow in the MARC performed at Tsinghua, where the number of stages is given as real or integer, are shown in Fig.1.

TABLE 1  
Natural composition of the krypton isotopes

Mass number	Natural abundance, molar concentration
78	0.0035
80	0.0228
82	0.1158
83	0.1149
84	0.5700
86	0.1730

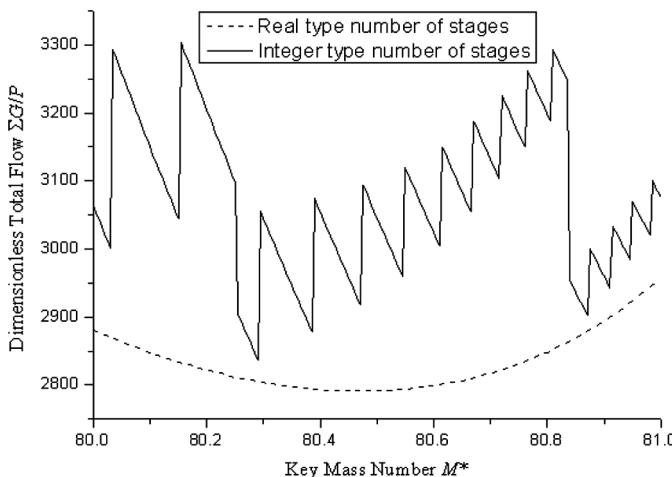


FIG. 1. Dependence of the dimensionless total flow  $\Sigma G/P$  versus the key mass number  $M^*$  in case 1. ( $\Delta M^* = 0.005$  for integer type of  $N_S$  and  $N_F$ ).

The zigzag line demonstrates the process of optimization of the dimensionless total flow in the cascades with the integer type of  $N_S$  and  $N_F$  (“Problem 1”). At these sudden jumps there is a change of  $N_S$  or  $N_F$ . The dashed line corresponds to the real type of  $N_S$  and  $N_F$  (“Problem 2”). As shown in Fig. 1, the procedure of optimization led to the lower minimum in the second case. However, the two minima in both cases are in relatively close agreement. It is obvious that the results for the optimization of a MARC with the real stage numbers can be used directly in numerical optimization of the optimum cascade parameters. Since evaluation of the total flow can be performed relatively easily by using formulas (2)–(6) for any given  $N_S$  and  $N_F$  (either integer or real) values, the time consumed for optimization becomes negligibly small.

The comparison of the parameters obtained by two independent research groups in the optimization of a MARC (“Problem 1” and “Problem 2”) and the optimum cascade (“Problem 3” and “Problem 4”) for the end component is presented in Table 2.

In Table 2, the withdrawal concentrations in a MARC with integer stage number  $N_S$  (i.e., the first case) are

$C_{Pt} \approx 20.0\%$  and  $C_{Wt} \approx 0.117\% < 0.12\%$ , while the withdrawal concentrations of the following three cases are equal to the predefined values. It is reasonable that the estimation of the total flow in the first case is larger. The best (in the sense of the minimum total flow) case, i.e., the fourth one, was obtained in the following way. The set of parameters from the optimized MARC was used as the initial guess in the optimization procedure for the optimum cascade. In order to avoid dealing with the mixed variables, the optimization was carried out separately by using  $\Theta$  as variable but specifying the number of stages and the feed location around the values obtained from the optimization of MARC. Table 3 gives the parameters  $N_S$  and  $N_F$  of those optimum cascades and the corresponding total flows for the fixed product and waste concentrations of the target component, which have been obtained by means of the numerical optimization at Tsinghua.

The optimization resulted in the minimum total flow of about 2785, the corresponding number of stages is 24, and the feed location is 4. So as one can see, the set of the parameters for the optimized MARC gives a good prediction for that of the optimum cascade.

In Case 2, the optimization was carried out for the intermediate target component  $^{83}\text{Kr}$  where its concentrations in the withdrawal flows are required to satisfy the following conditions:  $C_{Pt} \geq 20\%$ ,  $C_{Wt} \leq 5\%$ . The concentration of the  $^{84}\text{Kr}$  isotope in the inlet is high. Therefore, in order to enrich the intermediate component  $^{83}\text{Kr}$ , it must be separated from the  $^{84}\text{Kr}$  isotope. For this purpose the target and supporting components for the match in a MARC should be  $^{83}\text{Kr}$  and  $^{84}\text{Kr}$ , and so the key mass number should be equal to  $M^* = 83.5$ . The effect of varying  $M^*$  and seeking for the best  $N_S$  and  $N_F$  to find the minimum total flow is seen in Fig. 2. Again, the zigzag and dashed lines correspond in the optimization to the integer type of  $N_S$  and  $N_F$  and real type, respectively.

The values of the dimensionless total flow in the optimum cascade and the corresponding optimized MARC are shown in Tables 4 and 5 as follows:

Similar to the previous case, the total flow in a model cascade for the separation of the intermediate component

TABLE 2  
The optimum parameters for enrichment of the  $^{78}\text{Kr}$  isotope

The cascade type	$(M^*)_{\text{opt}}$	$N_S$	$N_F$	$(\sum SG/P)_{\text{min}}$
MARC (Problem 1, integer $N_S$ ) (Tsinghua)	80.2946 ( $\approx 80.3$ )	24	5	2828.5
MARC (Problem 2, real $N_S$ ) (MEPhI, Tianjin institute)	80.5	25.576 ( $\approx 26$ )	4.5268 ( $\approx 5$ )	2791.2
Optimum cascade (Problem 3, MEPhI)	–	24	4	2786.4
Optimum cascade (Problem 4, Tsinghua)	–	24	4	2784.7

TABLE 3

The dimensionless total flow in the optimum cascades with various numbers of stages and feed locations

$N_S \backslash N_F$	3	4	5
22	2881.4	2795.1	2828.4
23	2876.9	2785.7	2808.8
24	2880.8	<b>2784.7</b>	2822.2
25	2895.0	2790.8	2815.9

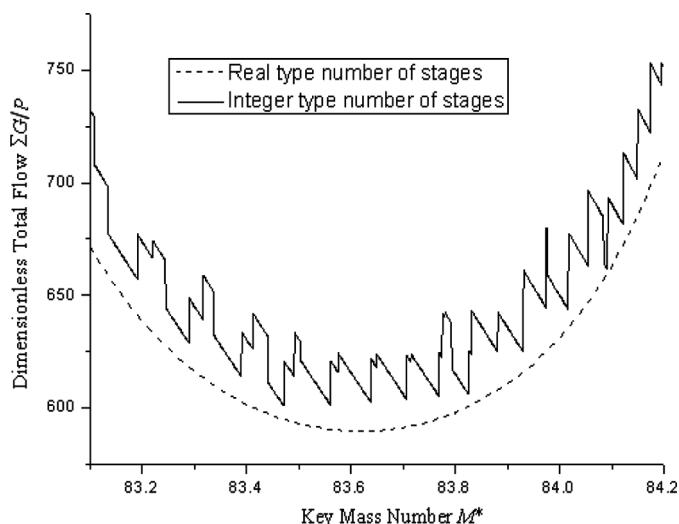


FIG. 2. Dependence of the dimensionless total flow  $\Sigma G/P$  versus a key mass number  $M^*$  in case 2. ( $\Delta M^* = 0.005$  for integer type of  $N_S$  and  $N_F$ ).

optimized by varying the parameter  $M^*$  is also very close to the value in the optimum cascade. The results of numerical optimization show that the number of stages and the feed location resulting from the optimization of the corresponding MARC are also close to that of the cascades with the minimum total flow.

TABLE 4

The dimensionless total flow of the optimum cascades with various number of stages and feed locations

$N_S \backslash N_F$	17	18	19	20
33	590.0	589.9	599.0	—
34	593.8	589.5	590.1	592.8
35	—	598.4	589.9	591.2
36	—	—	600.9	600.9

TABLE 5

The parameters of the optimized MARC and the optimum cascade

Type of a cascade	$(M^*)_{\text{opt}}$	$N_S$	$N_F$	$(\sum G/P)_{\min}$
MARC (integer $N_S$ ) (Problem 1, Tsinghua)	83.47	35	21	600.57
MARC(real $N_S$ ) (Problem 2, MEPhI, Tianjin institute)	83.65	34.119	17.780 (≈ 34) (≈ 18)	589.57
Optimum cascade (Problem 3, MEPhI)	—	34	18	589.4
Optimum cascade (Problem 4, Tsinghua)	—	34	18	589.5

## CONCLUSION

Finding the optimum parameters of long cascades for separation of multicomponent isotope mixtures by iterative mixed real-integer optimization is a difficult and time-consuming problem, especially when a good initial guess and proper bounds for optimization variables are unknown.

Two independent research groups, using different approaches to calculate and to optimize cascade parameters, verified that the optimized parameters of a MARC provide needed feedback in terms of the minimum total flow, the number of stages, and the feed location in the optimum cascade. It is therefore suggested that the method provides an easier approach to evaluate optimum cascade parameters and to find a good initial guess for its further direct optimization.

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