Optimum Design of Step Cascades for Uranium Enrichment by Gaseous Diffusion Process

By

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When the optimization of a step cascade is carried out directly on the basis of an exact formula, the calculus of minimization of a multi-variable function is essential. The numerical calculation is very complex and a brief estimation becomes impossible as the number of steps increases, because the number of variables in the calculus amounts to (2m-1) in all for an m-steps cascade.

In this paper an approximate formula of good feasibility and sufficient accuracy is proposed for the determination of the number of stages and interstage flow rate required for the plant of uranium enrichment by gaseous diffusion process. By using an approximate formula, the minimization of multi-variable function in the optimization of step cascades is able to be avoided. The applicability of the proposed method is verified by the calculation of a typical plant for uranium enrichment.

1. Introduction

At present one is able to analyse the characteristics of various kinds of cascades. The ideal cascade and more general tapered cascade have been discussed by Cohen¹⁾, Higashi²⁾ and others³⁻⁵⁾. Square and step cascades have been investigated by Cohen¹⁾, Benedict⁶⁾, Oliveri⁷⁾ and others. The ideal cascade realizes the smallest size of cascade achieving desired enrichment, but this cascade is not practical in its construction and control because the number of separating elements in each size has to be varied continuously as the isotope ratio of uranium changes. The most practical cascade might be a step cascade, including a square cascade as a possible variation.

When the optimization of a step cascade is carried out directly on the basis of the exact formula by Cohen¹, the calculus of minimization of a multi-variable function is essential. The number of variables in it amounts to (2m-1) in all for an m-steps cascade. Hence, the numerical calculation is very complex and a brief estimation becomes impossible as the number of steps increases.

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Oliveri7) has developed an interesting approximation method for the optimization of step cascade. By means of this method the optimum compositions characterizing each step are rather accurately determined, but it is difficult to discuss the total flow rate in a cascade which is equivalent to the size of a plant. It is often required to analyze how the size of cascade decreases with the number of steps. For instance, it is desired in the programming of a computer code for an economic evaluation of uranium enrichment by gaseous diffusion process to develop such a method as can estimate the size of a step cascade briefly with sufficient accuracy.

In this paper an optimization method for square and step cascades will be proposed and discussed.

2. Approximate Formula

Let us consider an m-step cascade as shown in Fig. 1. P, F and W are the rates of product, feed and waste, respectively. x_P , x_F and x_W denote the mole fractions of lighter isotope in product, feed and waste, respectively. x, denotes the mole

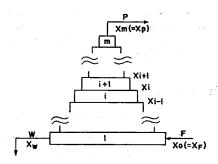


Fig. 1. Enriching Section of a squared-off cascade.

fraction at the top stage in the i-th step. When the separation factor of each stage, α , is close to unity, the total tails flow rate in the i-th step, J_i'' , can be expressed by the following exact formulae by Cohen,

$$\frac{J_{i}''}{P} = \frac{1}{(\alpha - 1)^{2} b_{i} c_{i}} \ln \frac{1 + a_{i}}{1 - a_{i}} \tag{1}$$

where

$$\begin{cases} a_{i} = \frac{b_{i}(x_{i} - x_{i-1})}{(x_{i-1} + x_{i})(1 + c_{i}) - 2x_{i-1}x_{i} - 2c_{i}x_{p}}, \\ b_{i} = \sqrt{1 + 2c_{i}(1 - 2x_{p}) + c_{i}^{2}}, \\ c_{i} = \frac{P}{L_{i}''(\alpha - 1)}. \end{cases}$$

$$(2)$$

$$\begin{cases} b_i = \sqrt{1 + 2c_i(1 - 2x_P) + c_i^2}, \end{cases} \tag{3}$$

$$c_i = \frac{P}{L_i''(\alpha - 1)}. \tag{4}$$

L.'' in Eq. (4) is the talis flow rtae in the *i*-th step.

When both x_{i-1} and x_i are negligibly small compared with 1 or very close to 1, the above equations are reduced to the following expressions.

Case I. When $x_{i-1} \ll 1$, $x_i \ll 1$,

$$\frac{J_{i}''}{P} \to \left(\frac{x_{P}g_{Ii}}{x_{i-1}} - 1\right)^{2} \frac{x_{i-1}}{(\alpha - 1)^{2} \lambda_{Ii}g_{Ii}x_{P}} \ln \frac{A_{Ii}g_{Ii} - 1}{g_{Ii} - 1}, \tag{5}$$

Where

$$\begin{cases}
A_{li} = \frac{x_i}{x_{i-1}}, & (6) \\
\lambda_{li} = 1, & (7) \\
g_{li} = \frac{x_{i-1}}{x_P} \left\{ 1 + \frac{(\alpha - 1)\lambda_{li}L_i''}{P} \right\}. & (8)
\end{cases}$$

$$g_{Ii} = \frac{x_{i-1}}{x_{P}} \left\{ 1 + \frac{(\alpha - 1)\lambda_{Ii}L_{i}^{"}}{P} \right\}. \tag{8}$$

Case II. When $(1-x_{i-1}) \ll 1$, $(1-x_i) \ll 1$,

$$\frac{J_{i''}}{P} \to \left\{ \frac{(1-x_{P})g_{hi}}{1-x_{i}} - 1 \right\}^{2} \frac{1-x_{i-1}}{(\alpha-1)^{2} \lambda_{hi} g_{hi}(1-x_{P})} \ln \frac{A_{hi}g_{hi}-1}{g_{hi}-1}, \tag{9}$$

where

$$\begin{cases} A_{hi} = \frac{1 - x_{i-1}}{1 - x_{i}} \\ \lambda_{hi} = -1 \\ g_{hi} = \frac{1 - x_{i}}{1 - x_{P}} \left\{ 1 + \frac{(\alpha - 1)\lambda_{hi}L_{i}''}{P} \right\} \end{cases}$$
(10)

$$\left\{ \begin{array}{l} \lambda_{hi} = -1 \end{array} \right. \tag{11}$$

$$\left\{g_{hi} = \frac{1 - x_i}{1 - x_P} \left\{1 + \frac{(\alpha - 1)\lambda_{hi}L_i^{\prime\prime}}{P}\right\}$$
 (12)

Comparing Eqs. (5)-(8) with Eqs. (9)-(12) and noticing the corresponding relations in them, the following approximate formula may be proposed for J_i'' ,

$$\frac{J_i^{\prime\prime}}{P} \to \frac{(B_i g_i - 1)}{(\alpha - 1)^2 \lambda_i B_i g_i} \ln \frac{A_i g_i - 1}{g_i - 1}, \tag{13}$$

where

$$A_{i} = \frac{x_{i}(1-x_{i-1})}{x_{i-1}(1-x_{i})} \tag{14}$$

$$B_i = \frac{x_P(1-x_P)}{x_{i-1}(1-x_i)} \tag{15}$$

$$\begin{cases}
A_{i} = \frac{x_{i}(1 - x_{i-1})}{x_{i-1}(1 - x_{i})} \\
B_{i} = \frac{x_{P}(1 - x_{P})}{x_{i-1}(1 - x_{i})}
\end{cases}$$

$$\lambda_{i} = \frac{B_{i} - 1}{x_{P}/x_{i-1} - (1 - x_{P})/(1 - x_{i-1})}$$

$$g_{i} = \frac{1}{B_{i}} \left\{ 1 + \frac{(\alpha - 1)\lambda_{i}L_{i}''}{P} \right\}$$
(15)

$$\left(g_{i} = \frac{1}{B_{i}} \left\{ 1 + \frac{(\alpha - 1)\lambda_{i}L_{i}^{\prime\prime}}{P} \right\}$$
 (17)

 A_i is the over-all separation factor in the *i*-th step. λ_i approaches unity when x_{i-1} , x_i and x_P tend to zero, and approaches to -1 when x_{i-1} and x_i tend to unity. Therefore, Eq. (13) is reduced to Eq. (5) at low concentrations, and to Eq. (9) at high concentrations. In order that Eq. (13) may converge to the formula of ideal cascade with increasing m, λ_i should be expressed as Eq. (16). In a special case, g_i is in accord with g defined by Benedict and Pigford⁶. By making the g_i of Eq. (17) equal to unity one obtains the wellknown condition for minimum reflux ratio in square and step cascades,

$$\left(\frac{L_{i}^{"}}{P}\right)_{s_{i}=1} = \frac{1}{\alpha - 1} \left(\frac{x_{P}}{x_{i-1}} - \frac{1 - x_{P}}{1 - x_{i-1}}\right),\tag{18}$$

3. The Condition of Minimum Square and Step Cascades

Now, let us consider the i-th step in an m-steps cascade. Differentiating Eq. (13) with respect to g_i and putting the derivative equal to zero, we obtain

$$\ln \frac{A_i g_i - 1}{g_i - 1} = \frac{(A_i - 1) g_i (B_i g_i - 1)}{(A_i g_i - 1) (g_i - 1) (B_i g_i - 1)}$$
(19)

or

$$B_i = \frac{1}{g_i} \cdot \frac{1 + D_i}{1 - D_i} \tag{20}$$

where

$$D_{i} = \frac{(A_{i}g_{i}-1)(g_{i}-1)}{(A_{i}-1)g_{i}} \ln \frac{A_{i}g_{i}-1}{g_{i}-1}$$

Substituting Eq. (19) into Eq. (13) we get the minimum value of (J_i''/P) , $(J_i''/P)_{\min}$, for a given set of x_i 's,

$$\left(\frac{J_i''}{P}\right)_{\min} = \frac{(A_i - 1)(B_i g_i - 1)^3}{(\alpha - 1)^2 B_i \lambda_i (g_i - 1)(A_i g_i - 1)(B_i g_i + 1)},\tag{21}$$

Eq. (21) should be used in relation to Eq. (19). The relation between A_i , B_i and g_i given by Eq. (19) is shown in Fig. 2. In this figure two branches can be seen for a single value of A_i . One branch starting from the point $(B_i=1, g_i=1)$ corresponds mainly to the enriching section, and the other in the region of $0 < B_i < 1$ corresponds mainly to the analysis of stripping section. It should be noticed that Fig. 2 or Eq. (19) is available independently of x_P , x_i 's, m and α . One may thus determine $(J_i''/P)_{\min}$ by the following successive procedure.

First, A_i , B_i and λ_i are calculated from Eqs. (14)-(16). Then, g_i for the values of A_i and B_i is obtained from Eq. (19) or Fig. 2. Finally, substituting

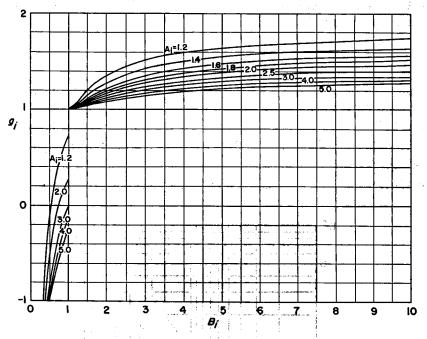


Fig. 2. The relation between A_i , B_i and g_i , which is obtained from Eq. (20).

these values into Eq. (21), $(J_{i}"/P)_{\min}$ is determined.

The interstage flow rate of each step corresponding to $(J_i''/P)_{\min}$, L_i''/P , and the total number of stages, n, are also easily calculated as follows:

$$\frac{L_i''}{P} = \frac{B_i g_i - 1}{(\alpha - 1)\lambda_i},\tag{22}$$

from Eq. (17), and

$$n = \sum_{i=1}^{m} n_i = \sum_{i=1}^{m} (J_i''/P)/(L_i''/P)$$
 (23)

from Eqs. (21) and (22).

When we define $(J''/P)_{\min}$ as

$$(J''/P)_{\min} = \sum_{i=1}^{m} (J_{i}''/P)_{\min},$$
 (24)

this gives the minimum total tails flow rate of an m-steps cascade with a given set of x_F , x_P and intermediate compositions x_1 , x_2 , ..., x_{m-1} . Therefore, $(J''/P)_{\min}$ depends on x_2 , x_2 , ..., x_{m-1} . In order to optimize a step cascade one should find a set of x_i 's which makes $(J''/P)_{\min}$ minimum. For the discussions below, let us

define $(J''/P)_{\text{opt}}$ and n_{opt} as the minimum value of $(J''/P)_{\text{min}}$ and the number of stages corresponding to it.

4. Application to Square Cascade

Square cascade is a kind of step cascade with m=1. Then Eq. (21) can be made applicable to square cascade by putting $x_0=x_F$ and $x_1=x_F$. As an example, the calculations below were carried out on the enrichment of natural uranium.

The results are shown in Fig. 3. Curve (1) represents the required size of square cascade calculated by Eq. (21). Curve (2) is obtained by means of the trial-and-error method from Cohen's exact formula Eq. (1). Curve (3) shows

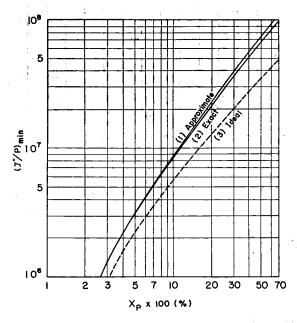


Fig. 3. The minimum size of a square cascade. Curves (1) and (2) are calculated by means of the approximate formula (21) and the exact solution (1), respectively. Curve (3) shows the size of the ideal cascade. ($\alpha = 1.0028$, $x_F = 0.00714$)

the size of the ideal cascade. From Fig. 3 we may conclude that the approximate formula Eq. (13) is precisely applicable within a rather wide range of x_F and x_P . A slight deviation of curve (1) from curve (2) for $x_P > 0.1$ may be of little importance practically, because it is unusual to adopt a square cascade up to $x_P > 0.1$ in natural uranium enrichment.

A single step in actual step cascade has a rather small overall separation

factor. Therefore, it may be expected that Eq. (21) will also provide a sufficiently accurate estimation for a step cascade.

5. Application to Step Cascade

We shall now consider a 3-steps cascade. $(J''/P)_{\min}$ is calculated directly from the exact formula Eq. (1) for various sets of (x_1, x_2) under the same value of x_F and x_P , as shown in Fig. 4. In this example $(J''/P)_{\text{opt}}$ appears at point Q $(x_1=0.0124, x_2=0.0236)$. The parameter in Fig. 4 denotes the value of $(J''/P)_{\min}$ divided by $(J''/P)_{\text{opt}}$ at point Q. The loops drawn in this figure are contourlines corresponding to the same value of the parameter.

As shown in Fig. 4 the value of $(J''/P)_{\min}$ changes very slowly around the optimum point Q. For instance, let us compare the three cascades Q, R and S in Fig. 5 with one another. These cascades correspond to points Q, R and S in Fig. 4, respectively. Though they are much different in shape, as seen in Fig. 5,

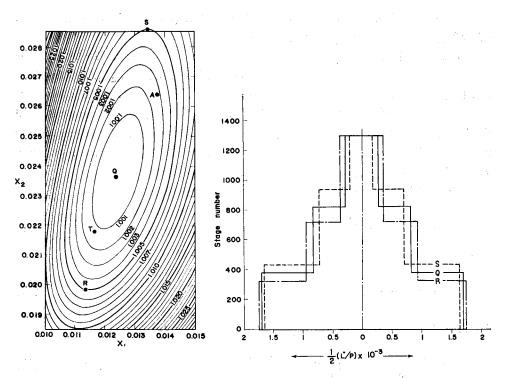


Fig. 4. The minimum size of 3-steps squared-off cascade expressed as a function of (x_1, x_2) . The optimum cascade is realized at point Q in this example. The parameter denotes the value of $(J''/P)_{\min}/(J''/P)_{\text{opt}}$. $(\alpha=1.0028, x_F=0.00714, x_P=0.05)$

Fig. 5. The shapes of 3-steps squared-off cascades corresponding to the points Q, R, and S in Fig. 4. (α =1.0028, x_F =0, 00714, x_P =0.05).

all of them achieve the same job of enrichment and both the cascades R and S are larger in size by only 0.5% than the cascade Q. One should remark the rather broad flexibility existing in the design of a real plant.

Generally, we may conclude from this fact that a rough estimation without any complicated procedures will suffice in determining the set of intermediate compositions which optimizes a step cascade.

Then, we may simply determine x_i 's with little error in the size of optimum cascade by putting all the over-all separation factors in each step equal to a constant value, that is

$$A_{i} = A = \left\{ \frac{x_{P}(1-x_{F})}{x_{F}(1-x_{P})} \right\}^{1/m} \quad (i = 1, 2, \dots m-1),$$
 (25)

hence

$$x_{i} = \frac{x_{F}A^{i}}{1 - x_{F}} / \left(1 + \frac{x_{F}A^{i}}{1 - x_{F}}\right). \tag{26}$$

Point A in Fig. 4 was plotted according to Eq. (26). It is remarkable that $(J''/P)_{\min}$ at point A is only 0.3% larger than at point Q.

Step cascade should approach to ideal cascade as m increases. In the extreme case where m is equal to the number of stages of ideal cascade, n_{ideal} , A defined by Eq. (25) is in accordance with the so-called heads separation factor of each stage in ideal cascade, $\beta(=\sqrt{\alpha})$, and x_i obtained from Eq. (26) gives the composition at the i-th stage of ideal cascade. Thus, it is expected that a gradually more accurate value of optimum composition will be calculated by Eq. (26) with the increase of m. This is also one reason we adopt Eq. (26) to determine x_i 's.

When A is equal to β and very close to unity, we get

$$\ln \frac{A_i g_i - 1}{g_i - 1} \simeq \frac{(\beta - 1) g_i}{g_i - 1},\tag{27}$$

and then

$$g_i \simeq 2 - \frac{1}{B_i} \quad \text{(when } A = \beta \simeq 1\text{)},$$
 (28)

from Eq. (19). Substituting Eq. (28) into Eqs. (21) and (23), we obtain

$$\left(\frac{J_{i}^{"}}{P}\right)_{\text{opt}} = \left(\frac{L_{i}^{"}}{P}\right)_{\text{opt}} \cong \frac{2}{\alpha - 1} \left(\frac{x_{P}}{x_{i-1}} - \frac{1 - x_{P}}{1 - x_{i-1}}\right) \quad \text{(when } A = \beta \cong 1\text{)}$$
 (29)

This is twice the minimum reflux ratio (Eq. (18)) and equal to the tails flow rate in the *i*-th stage of ideal cascade, $(L_i''/P)_{ideal}$. In this extreme case the number of stages in each step, $n_i = J_i''/L_i''$, is reduced to unity as seen in Eq. (29). There-

fore, n is equal to m, that is,

$$n_{\text{opt}} = m = \ln \left\{ \frac{x_P(1 - x_F)}{x_F(1 - x_P)} \right\} / \ln \beta.$$
 (30)

This is in accord with the expression for n_{ideal} . Hence, it is obvious that $(J''/P)_{\text{opt}}$ calculated by using Eqs. (19), (21) and (26) approaches $(J''/P)_{\text{ideal}}$ with the increase of m.

It is important in a design of an uranium enrichment plant to investigate how the size of a step cascade approaches that of an ideal cascade. $(J''/P)_{opt}/(J''/P)_{ideal}$ and n_{opt}/n_{ideal} are calculated by the proposed method and shown in Figs. 6 and 7 as a function of m.

Fig. 6 shows the value of $(J''/P)_{opt}$ decreases rapidly with the increase of m. The larger the value of m is, the more slowly a cascade decreases in size and the less simple it becomes. Therefore, we may conclude that in most cases the number of steps fewer than 5 will be sufficient in an enriching section as well as in a stripping section.

So, serious technical problems, we think, will not be arised in squaring-off a cascade into several steps. The separative work ratio¹⁰ of a step cascade with

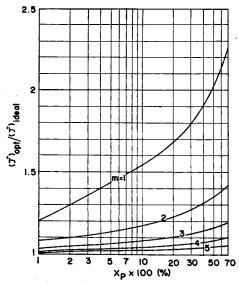


Fig. 6. The convergence of the size of a squared-off cascade to that of an ideal cascade with the increase of m. The parameter m denotes the number of steps of a squared-off cascade. ($\alpha = 1.0028$, $x_F = 0.00714$)

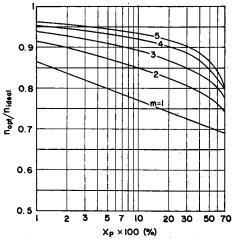


Fig. 7. Comparison between the total number of the stages of the optimum m-steps squared-off cascade and that of the corresponding ideal cascade. Parameter m denotes the number of steps of the squared-off cascade. ($\alpha=1.0028$, $x_F=0.00714$)

several steps to a corresponding ideal cascade is close to 1, as seen in Fig. 6. Thus it may be more realistic in the economics of large scale separation of uranium-235 to evaluate enriched uranium on the basis of an ideal cascade. The optimum waste composition may be determined in like manner.

The point T in Fig. 4 is determined by the approximatation developed by Oliveri. This point T is rather close to the optimum point Q, and it shows the superiority of Oliveri's method in the determination of the set of x_i 's. The value of $(J''/P)_{opt}$ obtained, however, increases with m and does not approach the value of $(J''/P)_{ideal}$.

Though the above discussions are concerned with the enriching section of uranium enrichment plant, one may be able to extend the analysis for the optimization of the stripping section. It is briefly shown below how to calculate the minimum value of J_i'' in the stripping section.

Steps in the stripping section of the cascade are numbered consecutively from 1 at the waste end of the plant to the feed step. A_i and B_i of a given step are obtained from Eqs. (14) and (15). The corresponding g_i is determined by the relation of Eq. (19) or Fig. 2. When the uranium is enriched from natural uranium (x_F =0.0072), the value of λ_i in stripping section may be approximated to 1. g_i in stripping section should be defined as follows,

$$g_{i} = \frac{1}{B_{i}} \left\{ 1 - \frac{(\alpha - 1) L_{i}^{"}}{W} \right\} \quad \text{stripping section} . \tag{31}$$

Then W/L_i'' can be obtained by using Eq. (31). The absolute value of the right hand side of Eq. (21) substituting A_i , B_i , and g_i is equal to $(J_i''/W)_{\min}$ in a step in the stripping section. The number of stages of the step can be obtained by multiplying of (J_i''/W) and (W/L_i'') . The relation of

$$\frac{W}{P} = \frac{x_P - x_F}{x_F - x_W} \tag{32}$$

is available to convert $(J^{\prime\prime}/W)_{\rm min}$ to $(J^{\prime\prime}/P)_{\rm min}$

The preceding results may be summarized as follows:

- 1. The approximate formula Eq. (13) was proposed as against the exact solution Eq. (1).
- 2. It was pointed out that there exists in practice a rather broad flexibility in the determination of intermediate compositions x_i 's. On the basis of this fact Eq. (26) was proposed for x_i .
- 3. The size of the optimum m-steps cascade, $(J''/P)_{opt}$, is obtained by the following successive procedures:

- a) x_i 's are calculated from Eq. (26).
- b) A, B_i and g_i are obtained from Eqs. (14), (15) and (19) (or Fig. 2), respectively.
- c) $(J''/P)_{opt}$ is determined by means of Eqs. (21) and (22).
- 4. The behavior of $(J''/P)_{opt}$ and that of n_{opt} are investigated for the variation of m and x_P by using the method described above. The results are shown in Figs. 6 and 7.
- 5. We have concluded from Fig. 6 that the number of steps fewer than 5 may be sufficient in most cases.
 - 6. It is shown briefly how to optimize the stripping section of a plant.

Most of the calculctions required in this report were carried out by using the Kyoto University Digital Computer II.

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