

Multiple Isotope Cascade Algorithm

GEORGE EMANUEL

*School of Aerospace, Mechanical and Nuclear Engineering,
University of Oklahoma, Norman, Oklahoma 73019*

Received July 29, 1981

An algorithm is described for balancing the steady-state operation of a separative cascade of the up/down type wherein each head stream is fed upward one or more stages, and each tail stream is fed downward one or more stages. The cascade incorporates refluxing and multiple feed points. Of primary importance is the treatment of multiple isotopes, which requires an iterative solution procedure. Only half of the separation factors can be arbitrarily specified, the other half are found as part of the solution process.

1. INTRODUCTION

Cascade theory is well developed for elements with only two dominant isotopes [1], such as uranium or hydrogen. Many elements, however, have three or more isotopes, including both uranium [2, 3] and hydrogen. Newer techniques for waste material management, such as laser processing, may well require small cascades for the separation of elements with many isotopes.

Cascade theory for an arbitrary number of isotopes is scant [2, 3]. Furthermore, published work on multiple isotope cascade algorithms is non-existent. The objective of this work is to bridge this gap. The extension of techniques [1, 4, 5] for the separation of binary mixtures to the multiple isotope situation unexpectedly proved to be far from straightforward.

Section 2 provides the basic equations and the numerical procedures for balancing a steady-state cascade. The cascade's configuration is of the up/down type, recently introduced by Olander [6]. An iterative procedure is used to obtain an accurate, consistent solution. In practice, the algorithm worked well for cascades as large as 20 stages, the largest cascade so far considered. A key issue dealt with in Section 2 is the treatment of the α and β separation factors that incorporate the physics of the process [1]. Section 3 concludes the article with an illustrative example.

2. FORMULATION

2.1. Equations

The mole fraction, or assay, of isotope i entering stage j is denoted by X_{ij} , where the element involved may be in either atomic or molecular form. Similarly, X'_{ij} and

X_{ij}'' respectively represent the heads and tails assay of the material leaving stage j . The amount of isotopic material entering and leaving the stage is L_j , L_j' , and L_j'' . The heads and tails designation is arbitrary, particularly when there are more than two isotopic forms. Nevertheless, this terminology is convenient and well understood [1]. To help fix ideas, a four-stage cascade is shown in Fig. 1, where the head stream from stage 1, L_1' , is fed to stage 3, while the head stream from stage 2 goes to 4. Similarly, the tail stream L_3'' goes to stage 1, etc. In general, u denotes the number of stages a head stream moves upward in the cascade, while d denotes the downward movement of the tail stream. The conventional cascade used for gaseous diffusion has $u = d = 1$. Values of u and/or d greater than one, however, are useful in certain applications, such as enrichment provided by a Becker nozzle [6] or in laser isotope separation [4].

Because $d = 2$ in Fig. 1, the tail streams from stages 1 and 2 constitute the waste stream W with assay X_{iw} , a fraction r'' of which can be refluxed back to stage 1. The product P comes from the head streams of stages 3 and 4, where refluxing into 4 is also considered. Refluxing is useful for making small adjustments to the assay X_{ip} (or X_{iw}). Thus, as r' (r'') increases from zero, X_{ip} will increase (X_{iw} will decrease). Our discussion presumes isotope i is enriched by the cascade starting from stage 1. Otherwise it is depleted; i.e., it is enriched downward from stage 4. Also shown in the

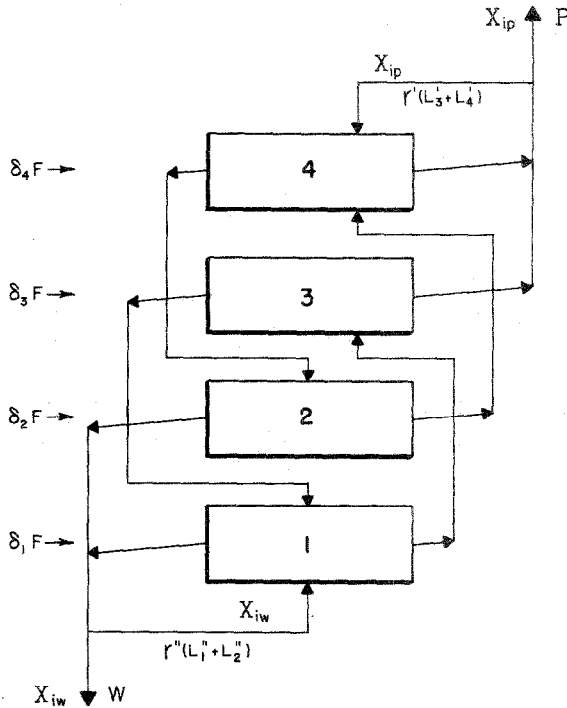


FIG. 1. Schematic for a cascade with $u = 2, d = 2, J = 4$.

figure are the feed streams with assay X_{ij} , which go to each stage according to distribution numbers δ_j . Multiple feed locations are useful for adjusting downward an enriched product assay, or adjusting upward a waste assay. These methods may prove more economical than, say, reducing the assay of the product stream by mixing it with feed material. (This latter method of product assay control is used in economic studies that compare different enrichment techniques.)

The stages in a cascade are numbered sequentially from 1 to J , where we assume $J \geq 2$. The isotopes i are numbered from 1 to I , where $I \geq 2$. Because all X 's are mole fractions, we have

$$\sum_{i=1}^I X_{ij} = \sum_{i=1}^I X'_{ij} = \sum_{i=1}^I X''_{ij} = 1, \quad j = 1, \dots, J. \quad (1)$$

Mass balance for isotope i and for the isotopic mixture, per stage, requires

$$X_{ij}L_j = X'_{ij}L'_j + X''_{ij}L''_j, \quad j = 1, \dots, J, \quad i = 1, \dots, I, \quad (2a)$$

$$L_j = L'_j + L''_j, \quad j = 1, \dots, J. \quad (2b)$$

(The quantities L_j , L'_j , L''_j , F , P , and W must have the same units, which can be either a mass unit or mass per unit time.)

The relationship between the various stages of the cascade are expressed in two sets of equations. The first set controls the passage of isotope i :

$$r'' \sum_{j=1}^d (X_i L)_j'' + (X_i L)_{d+1}'' + \delta_1 X_{ij} F = (X_i L)_1 \quad (3a)$$

$$\vdots$$

$$r' \sum_{j=J-u+1}^J (X_i L)_j' + (X_i L)_{J-u}' + \delta_J X_{ij} F = (X_i L)_J. \quad (3b)$$

Equation (3a) represents mass balance of isotope i for stage 1, where the left (right) side is for incoming (outgoing) material. Intermediate equations would not contain reflux terms. As will be shown, there is a change in form when the $(XL)''$ term is dropped and when the $(XL)'$ term first appears.

The second set of equations is obtained from Eqs. (3) by setting all X 's equal to one, and represents the total material flow. Both sets are for j from 1 to J .

The separation factors α_{ij} and β_{ij} determine the degree of enrichment or depletion of isotope i per stage. They are defined by

$$\alpha_{ij} = \frac{X'_{ij}}{1 - X'_{ij}} \frac{1 - X''_{ij}}{X''_{ij}}, \quad i = 1, \dots, I, \quad j = 1, \dots, J, \quad (4a)$$

$$\beta_{ij} = \frac{X'_{ij}}{1 - X'_{ij}} \frac{1 - X_{ij}}{X_{ij}}, \quad i = 1, \dots, I, \quad j = 1, \dots, J \quad (4b)$$

and provide the connection with the physics of the enrichment process.

2.2. Numerical Formulation

A convenient starting point is the unit transfer equations [5] that can be derived from Eqs. (2) and (4):

$$X'_{ij} = \frac{\beta_{ij} X_{ij}}{1 + (\beta_{ij} - 1) X_{ij}}, \tag{5a}$$

$$X''_{ij} = \frac{\beta_{ij} X_{ij}}{\alpha_{ij}(1 - X_{ij}) + \beta_{ij} X_{ij}}, \tag{5b}$$

$$L'_j = \frac{1}{\beta_{ij}} \frac{\alpha_{ij} - \beta_{ij}}{\alpha_{ij} - 1} [1 + (\beta_{ij} - 1) X_{ij}] L_j, \tag{5c}$$

$$L''_j = \frac{1}{\beta_{ij}} \frac{\beta_{ij} - 1}{\alpha_{ij} - 1} [\alpha_{ij}(1 - X_{ij}) + \beta_{ij} X_{ij}] L_j. \tag{5d}$$

Equations (5) are used to eliminate head and tail quantities in Eqs. (3), thereby obtaining

$$\begin{aligned} (aY)_{i,d+1} & - Y_{i,1} + r'' \sum_{j=1}^d (aY)_{ij} &= - Y_{i,f} \\ (aY)_{i,d+2} & - Y_{i,2} &= - Y_{i,2f} \\ & \vdots & \\ (aY)_{i,d+u+1} + (bY)_{i,1} & - Y_{i,u+1} &= - Y_{i,u+1f} \\ & \vdots & \\ (aY)_{ij} & + (bY)_{i,j-d-u} - Y_{i,j-d} &= - Y_{i,j-df} \\ & (bY)_{i,j-d-u-1} - Y_{i,j-d+1} &= - Y_{i,j-d+1f} \\ & \vdots & \\ & (bY)_{i,j-u-1} - Y_{i,j-1} &= - Y_{i,j-1f} \\ & (bY)_{i,j-u} - Y_{ij} + r' \sum_{j=j-u+1}^J (bY)_{ij} &= - Y_{ijf} \end{aligned} \tag{6}$$

where

$$a_{ij} = \frac{\beta_{ij} - 1}{\alpha_{ij} - 1}, \quad b_{ij} = \frac{\alpha_{ij} - \beta_{ij}}{\alpha_{ij} - 1}, \tag{7}$$

$$Y_{ij} = X_{ij} L_j, \quad Y_{ijf} = \delta_j X_{ij} F. \tag{8a,b}$$

Equations (6) are called the Y equations. They are linear in the Y_{ij} and are solved by matrix inversion, providing the α_{ij} and β_{ij} are known. The change in form, mentioned

after Eqs. (3), can be seen in the $u + 1$ and $J - d + 1$ rows. It stems from the up/down structure of the cascade. Because of this structure, it is possible to have a cascade where a given stage has no entering material. For example, suppose u is changed to 3 and $\delta_3 = 0$ in Fig. 1 then stage 3 has no incoming material. To rule out this possibility, each row of (6) is tested with $i = 1$:

$$E_j = Y_{1jf} + \sum_k (\text{coefficients of all } Y_{1k} \text{ in row } j), \quad j = 1, \dots, J. \quad (9a)$$

If

$$E_j > 0, \quad j = 1, \dots, J, \quad (9b)$$

then material enters each stage, otherwise the cascade's configuration is discarded.

A second matrix equation is formed starting from Eqs. (3) with all X 's equal to one. This equation is generated from (6) by the transformation:

$$\begin{aligned} a_{ij} &\rightarrow c_j, \\ b_{ij} &\rightarrow d_j, \\ Y_{ij} &\rightarrow L_j, \\ Y_{ijf} &\rightarrow L_{jff}, \end{aligned} \quad (10)$$

where

$$\begin{aligned} c_j &= \frac{\alpha_{1j} \beta_{1j} - 1}{\beta_{1j} \alpha_{1j} - 1}, \\ d_j &= \frac{\alpha_{1j} - \beta_{1j}}{\beta_{1j}(\alpha_{1j} - 1)}, \\ L_{1f} &= Y_{11f} - [(\beta_1 - 1) dY_1]_{d+1} - r'' \sum_{j=1}^d [(\beta_1 - 1) dY_j] \\ L_{2f} &= Y_{12f} - [(\beta_1 - 1) dY_1]_{d+2} \\ &\vdots \\ L_{u+1,f} &= Y_{1,u+1,f} - [(\beta_1 - 1) dY_1]_{d+u+1} + [(\beta_1 - 1) dY_1]_1 \\ &\vdots \\ L_{J-d,f} &= Y_{1,J-d,f} - [(\beta_1 - 1) dY_1]_J + [(\beta_1 - 1) dY_1]_{J-d-u} \\ L_{J-d+1,f} &= Y_{1,J-d+1,f} + [(\beta_1 - 1) dY_1]_{J-d-u+1} \\ &\vdots \\ L_{Jf} &= Y_{1Jf} + [(\beta_1 - 1) dY_1]_{J-u} + r' \sum_{j=J-u+1}^J [(\beta_1 - 1) dY_j] \end{aligned} \quad (11)$$

We refer to this equation as the L equation; its derivation also requires Eqs. (5), written in the form

$$\begin{aligned} L'_j &= d_j L_j + (\beta_{1j} - 1) d_j Y_{1j}, \\ L''_j &= c_j L_j - (\beta_{1j} - 1) d_j Y_{1j}. \end{aligned} \tag{13}$$

If all the α_{ij} and β_{ij} could be specified as well as the cascade parameters, such as J, I, u, \dots , the solution would be straightforward. The Y matrix equation would be inverted I times and the L equation once. Mole fractions X_{ij} are then given by Eq. (8a), while the heads and tails mole fractions are obtained from Eqs. (5). This procedure, unfortunately, is incorrect, since only half of the $2IJ$ α and β parameters can be arbitrarily specified. The other half must be determined as part of the solution process. The reason for this can be seen by writing Eq. (5c) as

$$\frac{L'_j}{L_j} = \frac{1}{\beta_{ij}} \frac{\alpha_{ij} - \beta_{ij}}{\alpha_{ij} - 1} [1 + (\beta_{ij} - 1) X_{ij}], \quad i = 1, \dots, I, \quad j = 1, \dots, J. \tag{14}$$

The left-hand side is independent of i . If all the α and β 's could be prescribed, then the X 's are readily determined from (5c) and by solving the L equation, without invoking the Y equation, or its antecedent, Eq. (3). The only recourse, therefore, is for either α_{ij} or β_{ij} to be unspecified. Thus, only half of the α and β 's can be arbitrarily specified. We assume as given, or prescribed

$$\begin{aligned} \alpha_{ij}, \quad & i = 1, \dots, I - 1, \quad j = 1, \dots, J, \\ \beta_{1j}, \quad & j = 1, \dots, J. \end{aligned} \tag{15}$$

This choice is somewhat arbitrary, but proved computationally convenient.

The steps involved in iteratively obtaining a solution follow the sequence:

- (1) A first estimate $\beta_{ij}^{(1)}$ for the unknown β_{ij} is

$$\beta_{ij}^{(1)} = 1 + 0.1(\alpha_{ij} - 1), \quad i = 2, \dots, I - 1, \quad j = 1, \dots, J. \tag{16}$$

- (2) The $a_{ij}, b_{ij}, c_j,$ and $d_j,$ exclusive of $i = I,$ are computed.

- (3) The Y matrix equation is solved for $i = 1, \dots, I - 1$ by any standard matrix inversion routine. (We use Gaussian elimination, but sparse routines would be advantageous for large cascades.)

- (4) Using only the Y_{1j} result from step 3, the L matrix equation is solved. Because the α_{1j} and β_{1j} are prescribed, the solutions for Y_{1j} and L_j are exact and are not recomputed in subsequent iterations.

- (5) All L'_j and L''_j are computed using Eqs. (13). (The calculation is also exact and is not recomputed.) Next determine for $i = 1, \dots, I - 1, j = 1, \dots, J$

$$\begin{aligned} X_{ij} &= Y_{ij}/L_j, \\ X'_{ij} &= b_{ij} Y_{ij}/L'_j, \\ X''_{ij} &= a_{ij} Y_{ij}/L''_j. \end{aligned} \tag{17}$$

(6) A set of α 's, denoted here as $\tilde{\alpha}_{ij}^{(m)}$, are computed using Eq. (4a) and the X 's from step 5. If

$$\left| \frac{\alpha_{ij} - \tilde{\alpha}_{ij}^{(m)}}{\alpha_{ij}} \right| \leq \varepsilon, \quad i = 2, \dots, I-1, j = 1, \dots, J, \quad (18)$$

where ε is typically 10^{-8} , a converged solution is obtained, and step 8 is then performed.

(7) A second β_{ij} estimate is used

$$\beta_{ij}^{(2)} = \alpha_{ij} - 0.1(\alpha_{ij} - 1) \quad (19)$$

and steps 2, 3, 5, and 6 are repeated. After the first two iterations, the new β 's are chosen by

$$\beta_{ij}^{(m+1)} = \beta_{ij}^{(m-1)} + (\beta_{ij}^{(m)} - \beta_{ij}^{(m-1)}) \frac{\alpha_{ij} - \tilde{\alpha}_{ij}^{(m-1)}}{\tilde{\alpha}_{ij}^{(m)} - \tilde{\alpha}_{ij}^{(m-1)}}, \quad i = 2, \dots, I-1, j = 1, \dots, J \quad (20)$$

and the above steps are repeated.

(8) After convergence, the remaining part of the solution is given by

$$\begin{aligned} Y_{ij} &= L_j - \sum_{i=1}^{I-1} Y_{ij}, \\ Y'_{ij} &= L'_j - \sum_{i=1}^{I-1} Y'_{ij}, \\ Y''_{ij} &= L''_j - \sum_{i=1}^{I-1} Y''_{ij}, \end{aligned} \quad (21)$$

where $j = 1, \dots, J$, and where these equations satisfy Eqs. (1). With these Y 's, the X_{ij} , α_{ij} , and β_{ij} 's are readily obtained. The cascade parameters of physical interest [5] are given by

$$T = \text{throughput} = \sum_{j=1}^J L_j, \quad (22a)$$

$$P = \text{product} = (1 - r') \sum_{j=J-u+1}^J L'_j, \quad (22b)$$

$$W = \text{waste} = (1 - r'') \sum_{j=1}^d L''_j, \quad (22c)$$

$$\theta = \text{cascade cut} = P/T, \quad (22d)$$

$$X_{ip} = \text{product assay} = \frac{(1 - r')}{P} \sum_{j=J-u+1}^J Y'_{ij}, \tag{22e}$$

$$X_{iw} = \text{waste assay} = \frac{(1 - r'')}{W} \sum_{j=1}^d Y''_{ij}. \tag{22f}$$

This procedure is not presumed to be optimum, nevertheless, in practice convergence is rapid. For cascades with $J = 20$ and $I = 5$, convergence occurred by $m = 7$ when ϵ is 10^{-8} .

2.3. $I = 2$

On the surface, the algorithm may appear to be incompatible for a cascade where $I = 2$. However, this is not the case. When $I = 2$, α_{1j} and β_{1j} are prescribed, while α_{2j} and β_{2j} are unknown. Thus, half of the α and β 's are unknown. In fact, the standard theory does not define or utilize α_{2j} and β_{2j} . Their values, however, are given by the algorithm, or directly from Eqs. (4), as

$$\alpha_{2j} = \frac{1}{\alpha_{1j}}, \quad \beta_{2j} = \frac{1}{\beta_{1j}} \tag{23}$$

regardless of the cascade's configuration. When enriching U^{235} the α_{1j} and β_{1j} are chosen, by convention, to be greater than one. Consequently, for the U^{238} the α_{2j} and β_{2j} are less than unity, which, of course, means the U^{238} is depleted by the cascade.

When $I > 2$, no simple equations exist, such as (23). In contrast to (23), the non-prescribed α and β 's now depend on many factors including the cascade's configuration (i.e., u, d, r', r''). This situation means care should be exercised when $I > 2$ in the physical, or experimental, derivation of the α_{ij} and β_{ij} . Over- or under-specification must be avoided. The unknown α_{ij} and β_{ij} , which constitute half of the total, are not solely dependent on the physics of the process, but also depend on the cascade's configuration. Only when $I = 2$, does the situation simplify to where these caveats are unnecessary.

2.4. Constraints, Assumptions and Input

One constraint, Eqs. (9), has already been dealt with. It is easy to show, using Eq. (4), that if $\alpha_{ij} < 1$, then

$$0 < \alpha_{ij} < \beta_{ij} < 1, \tag{24a}$$

whereas if $\alpha_{ij} > 1$, then

$$1 < \beta_{ij} < \alpha_{ij}. \tag{24b}$$

These inequalities are checked and enforced during the iterative solution process.

For ease of presentation, we have delayed mentioning a number of assumptions. A

TABLE I
Input Parameters

Parameter	Comments
I	$I \geq 2$
J	$J \geq 2$
u	$J \geq u \geq 1$
d	$J \geq d \geq 1$
r'	$1 > r' \geq 0$
r''	$1 > r'' \geq 0$
X_{ij}	$1 > X_{ij} > 0; \sum_{i=1}^I X_{ij} = 1$
δ_j	$1 \geq \delta_j \geq 0; \sum_{j=1}^J \delta_j = 1$
F	$F > 0$
α_{ij}	$\alpha_{ij} \neq 0, 1$; prescribed for $j = 1, \dots, J$ and $i = 1, \dots, I - 1$
β_{ij}	$\beta_{ij} \neq 0, 1$; prescribed for $j = 1, \dots, J$ and $i = 1$

principal one is that the algorithm requires steady-state, or a cyclic batch, mode of operation. A single assay for the feed material is assumed, and refluxing is restricted to stages 1 and J . Both of these assumptions, however, are readily removable.

Required input for a given cascade is shown in Table I, along with bounds or constraints that are self-evident.

3. ILLUSTRATIVE EXAMPLE

We illustrate the algorithm with a 5-isotope, 20-stage case, described by the parameters

$$\begin{aligned}
 I = 5, \quad J = 20, \quad u = 2, \quad d = 1, \quad r' = r'' = 0, \quad F = 1, \\
 X_{1j} = 0.6, \quad X_{5j} = 0.1 \quad (j = 2, \dots, 5), \\
 \delta_{12} = 1, \quad \delta_j = 0 \quad (\text{all other } j), \\
 \alpha_{1j} = 3, \quad \alpha_{ij} = 0.3 \quad (i = 2, 3, 4, \text{ all } j), \\
 \beta_{1j} = 2.5 \quad (\text{all } j).
 \end{aligned}$$

Figure 2 shows the variation with stage number of the unspecified separation factors.

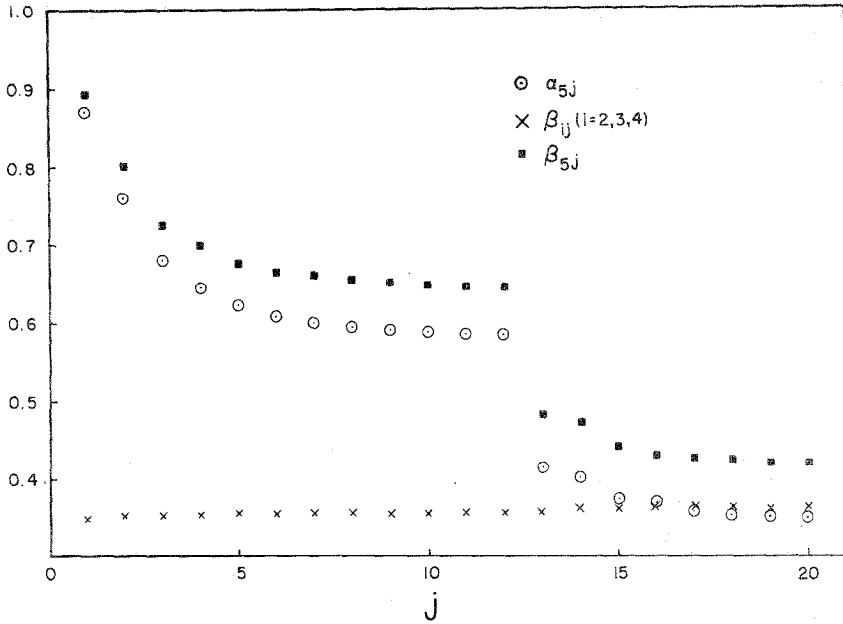


FIG. 2. Variation with stage j of computed α and β .

All of them are below unity, and the jump that occurs at $j = 13$ is caused by stage 12 being the feed point. Even though all of the specified α_{ij} and β_{ij} are constant with j , the computed factors vary from stage to stage. These include the β_{ij} in Fig. 2 that vary from 0.3505 at $j = 1$ to 0.3634 at $j = 20$. Note that

$$\alpha_{5j} \neq \alpha_{ij}, \quad \beta_{5j} \neq \beta_{ij}$$

for $i = 2, 3, 4$, even though, for instance, the X_{ij} are equal for $i \geq 2$. This is a direct consequence of Eqs. (1), which are utilized in step 8 of the algorithm. The large decrease of α_{5j} and β_{5j} with increasing j means the top of the cascade is more efficient in depleting isotope 5 than is the bottom. Because of the low α_{ij} and β_{ij} throughout

TABLE II
Product and Waste Assays

i	X_{ip}	X_{iw}
1	0.99864	0.57949
2	0.14910(-3)	0.10514
3	0.14910(-3)	0.10514
4	0.14910(-3)	0.10514
5	0.90930(-3)	0.10510

the cascade for $i = 2, 3, 4$, these isotopes are more rapidly depleted than is isotope 5. Only isotope 1, where $\alpha_{1j} > 1$, is enriched by the cascade.

The cascade's overall performance with $F = 1$ is summarized by

$$T = 31.49, \quad P = \theta = 0.04893, \quad W = 0.95107$$

and by Table II for X_{ip} and X_{iw} . Almost 5% of the feed leaves stage 1 in the form of 99.86% pure isotope 1, whereas the waste material, despite the number of stripping stages, is still rich in this isotope.

ACKNOWLEDGMENTS

This investigation began when the author was at Los Alamos Scientific Laboratory. He gratefully acknowledges the programming assistance of K. Witte and J. Hafer.

REFERENCES

1. M. BENEDICT, T. PIGFORD, AND H. LEVI, "Nuclear Chemical Engineering," 2nd ed., Chap. 12, McGraw-Hill, New York, 1981.
2. A. DE LA GARZA, G. A. GARRETT, AND J. E. MURPHY, *Chem. Eng. Sci.* **15** (1961), 188.
3. A. DE LA GARZA, *Chem. Eng. Sci.* **18** (1963), 73.
4. G. EMANUEL, *Nucl. Tech.* **51** (1980), 238.
5. G. EMANUEL, *Nucl. Tech.* **43** (1979), 314.
6. D. R. OLANDER, *Nucl. Tech.* **29** (1976), 108.