Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

The Use of Powell's Conjugate Gradient Minimization Method for Computing Concentration Profiles in Multicomponent and Multistage Separation Systems

C. Gilath[ab]; R. G. Goodson[abc]; I. Shraga[abd]; D. Wolf[abe]
[a] SOREQ NUCLEAR RESEARCH CENTRE, YAVNE, ISRAEL [b] WEIZMANN INSTITUTE OF SCIENCE, REHOVOT, ISRAEL [c] Institute of Interdisciplinary Engineering Studies, Purdue University, Lafayette, Indiana [d] Department of Applied Mathematics, State University of New York at Stony Brook, Stony Brook, New York [e] Ben Gurion University of the Negev, Beer-Sheva, Israel

To cite this Article Gilath, C. , Goodson, R. G. , Shraga, I. and Wolf, D.(1978) 'The Use of Powell's Conjugate Gradient Minimization Method for Computing Concentration Profiles in Multicomponent and Multistage Separation Systems', Separation Science and Technology, 13: 5, 409 — 428
To link to this Article: DOI: 10.1080/01496397808058291
URL: http://dx.doi.org/10.1080/01496397808058291

SEPARATION SCIENCE AND TECHNOLOGY, 13(5), pp. 409–428, 1978

The Use of Powell's Conjugate Gradient Minimization Method for Computing Concentration Profiles in Multicomponent and Multistage Separation Systems

C. GILATH, R. G. GOODSON,* I. SHRAGA,† and D. WOLF‡

SOREQ NUCLEAR RESEARCH CENTRE
YAVNE, ISRAEL;
WEIZMANN INSTITUTE OF SCIENCE
REHOVOT, ISRAEL

Abstract

Concentration profiles in multicomponent and multistage separation systems can be computed by using a continuous or a stagewise model. The numerical calculations are iterative, and fast convergence is achieved when Powell's conjugate gradient minimization method is used. The procedure developed in the present work was implemented on thermal diffusion separation systems for oxygen and krypton isotopes. Although the technique was developed with special emphasis on isotope separation problems, it is also applicable to other multicomponent and multistage separation processes.

INTRODUCTION

Computations of concentration profiles are important for the design and the operation of multistage separation systems. A typical design problem is the calculation of the required number of stages and/or interconnections in large separating cascades. In cases where existing equip-

*Present address: Institute of Interdisciplinary Engineering Studies, Purdue University, Lafayette, Indiana 47907.

†Present address: Department of Applied Mathematics, State University of New York at Stony Brook, Stony Brook, New York 11794.

‡Present address: Ben Gurion University of the Negev, Beer-Sheva, Israel.

ment having a given number of separating stages is used, the product and waste concentrations must be found under various operating conditions.

Although in some cases only product and waste concentrations must be determined, their computation requires knowledge of the whole concentration profile. This profile is calculated by using stagewise or continuous models. The nonlinear nature of the equilibrium relations makes an analytical solution of the problem difficult or impossible. Thus numerical iterative procedures are used (*1*). The calculations are completed when the total and component mass balances as well as equilibrium conditions at each stage are satisfied. The speed of convergence of these computations is of great practical importance.

The iterative procedures are relatively simple for two-component systems, since by assuming the concentration of one component, the concentration of the other is also defined. Simple procedures, such as the *direct iteration* and the *regula falsi* methods (*1*), can be readily used and convergence is fast. The problem becomes difficult when a multicomponent system is involved. It is further complicated when dealing with very small separation factors, as in isotope separation systems, with several hundreds up to a few thousand stages. Moreover, curved concentration profiles occur for some intermediate components, and make the computations even more difficult. An example of this is the separation of ^{17}O from ^{18}O and ^{16}O (*3*).* Thus there is a need for a converging technique for the computation of the concentration profile and the product concentration.

The present paper describes such an iterative procedure in which Powell's conjugate gradient minimization method (*2*) is used. This procedure was applied to computations on the thermal diffusion separation of oxygen and krypton isotopes in enrichers and systems consisting of enrichers and strippers. Although the technique was developed with special emphasis on isotope separation problems, it is also applicable to other multicomponent and multistage separation processes, such as distillation, extraction, and chemical exchange.

*At certain conditions during the separation of oxygen isotopes by the thermal diffusion of oxygen gas, the concentration of ^{17}O first decreases from the bottom of the cascade upward and then increases again toward the top.

CONCENTRATION PROFILES IN MULTICOMPONENT MULTISTAGE SEPARATION SYSTEMS

The separation systems treated in this work are enrichers (Fig. 1), strippers (Fig. 2), and cascades consisting of an enricher and a stripper (Fig. 3). Both total reflux operating conditions and conditions of product and waste withdrawal are considered. The systems may consist of discrete separating stages (e.g., plates) or they may be continuous (e.g., packed distillation columns, thermal diffusion columns). The "plate-by-plate" or "stagewise" method for the computation of concentration profiles in binary separating systems is well known in chemical engineering (*1*, *4*). The computation becomes somewhat time consuming when dealing with a large number of stages as is the case in isotope separation. Under these conditions it is questionable whether it is necessary to know the concentrations at every stage. Thus Cohen (*5*) described isotope separation cascades made up of many separating stages, as if they were continuous. This description becomes even more natural when the separating system is in fact continuous. Cohen (*5*) solved his continuous model for the separation of binary mixtures analytically by linearizing the equilibrium relationship. An advantage of the numerical solution of the continuous or stagewise models presented in this paper is that this linearization is not required. These models permit the computation of the concentration profile for given feed, waste, product and internal flow rates, concentration of feed, location of feed, number of stages, and separation factors.

The Continuous Model

Enricher. The countercurrent enricher, shown in Fig. 1, can be either a discrete stage system (e.g., plate column) having N_E theoretical stages or a continuous system with the same number N_E of equivalent theoretical stages and a total height Z. The downflow, upflow, and product flow rates are labeled L_E, V_E, and P, respectively. L_E and V_E are the liquid and vapor flow rates in the case of distillation and the flow rate of the "cold" and "hot" stream, respectively, in thermal diffusion columns (*6*). An m component mixture is separated. The concentrations of component i ($1 \leqslant i \leqslant m$) in the feed, product, and waste streams are X_{Fi}, X_{Pi}, and X_{Wi}, respectively. X_{Ei} and Y_{Ei} are the concentrations in the down and upstream, respectively.

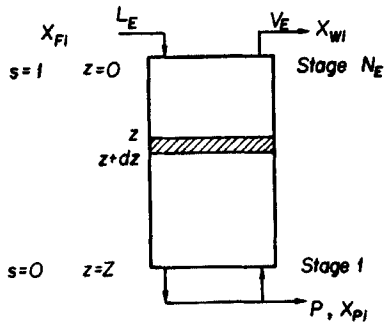

FIG. 1. Enricher.

The concentrations gradients dX_{Ei}/dz and dY_{Ei}/dz are defined by

$$L_E \frac{dX_{Ei}}{dz} = K(X^*_{Ei} - X_{Ei}) \tag{1}$$

$$V_E \frac{dY_{Ei}}{dz} = -K(X^*_{Ei} - X_{Ei}) \tag{2}$$

where z is the height in the column measured from the top and K is the overall mass transfer coefficient. X_{Ei} and Y_{Ei} are the concentrations at height z. X^*_{Ei} is the concentration of component i in the downflow stream, in equilibrium with the upflow stream:

$$X^*_{Ei} = \frac{\alpha_i Y_{Ei}}{\sum_{j=1}^{m} \alpha_j X_{Ej}} \tag{3}$$

α_j is the separation factor for the component j.

It is convenient to convert the height z into the normalized height s, where $s = 0$ at the bottom of the column and $s = 1$ at the top (see Fig. 1). Furthermore, since in isotope separation the operating and equilibrium lines are almost parallel, the number of transfer units and the number of theoretical plates (stages) are in effect the same (*4*). Thus N_E can be substituted for the mass transfer coefficient K by

$$N_E = KZ/\bar{L} \tag{4}$$

where $\bar{L}$ is the downflow rate per unit cross section of the column. X_{Ei} is substituted from Eq. (3) into Eq. (1). Integrating once, the following

equation is obtained:

$$\frac{dY_{Ei}}{ds} - N_E[(1 + R_E)X^*_{Ei} - Y_{Ei}] = C_i \tag{5}$$

where

$$R_E = P/V \tag{6}$$

X_{Ei} and Y_{Ei} are continuous functions of s. The integration constant C_i is found so as to satisfy the prevailing boundary conditions. For isotope separation systems, the reflux ratio is usually very high, R_E is very small, and one can assume that at $s = 0$

$$Y_{Ei}(0) = X_{Ei}(0) \tag{7}$$

Under these conditions, the concentration gradient in the enricher is

$$\frac{dY_{Ei}}{ds} = N_E R_E Y_{Ei}(0) - N_E[(1 + R_E)X^*_{Ei} - Y_{Ei}] \tag{8}$$

Stripper. The stripper shown in Fig. 2 has N_S separation stages. W represents the waste flow rate. By a method similar to that shown in the previous section, the following concentration gradient is obtained for the stripper:

$$\frac{dX_{Si}}{ds} = N_S R_S X_{Si}(1) + N_S[Y^*_{Si} - (1 + R_S)X_{Si}] \tag{9}$$

where

$$R_S = W/V_S \tag{10}$$

$$Y^*_{Si} = \frac{(1/\alpha_i)X_{Si}}{\sum_{j=1}^{m} (1/\alpha_j)X_{Sj}} \tag{11}$$

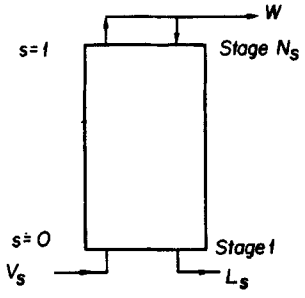

FIG. 2. Stripper.

and X_{Si} and $X_{Si}(1)$ are the concentrations at any height (s) and at $s = 1$, respectively, of the component i in the downflow stream.

Equation (9) is based on the assumption that at the top of the stripper, where $s = 1$,

$$X_{Si}(1) = Y_{Si}(1) \tag{12}$$

This relation applies to isotope separation systems operating at a high reflux ratio and very small R_S.

Enricher and Stripper. A system consisting of an enricher and a stripper is shown in Fig. 3. Equations (8) and (9) describe the concentration gradients in the enricher and stripper, respectively. Additional equations describe the balances at the feed point and are specific for the given system. For example, it is common to feed a thermal diffusion cascade via a reservoir located between the enricher and the stripper, as shown in Fig. 3. Under conditions of good mixing in this reservoir and equal split of

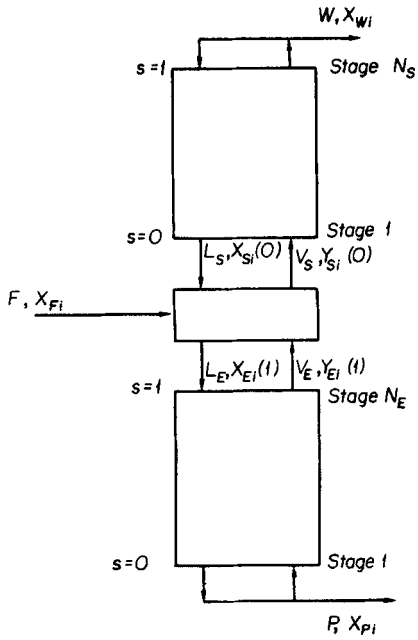

FIG. 3. A system consisting of an enricher (bottom) and a stripper (top) with a reservoir between them.

feed flow rate F to the enricher and stripper, the following relations are obtained:

$$V_S = V_E + F/2 \tag{13}$$

$$L_E = L_S + F/2 \tag{14}$$

$$X_{Ei}(1) = Y_{Si}(0) = [FX_{Fi} + L_S X_{Si}(0) + V_E Y_{Ei}(1)]/(L_E + V_S) \tag{15}$$

The Stagewise Model

This model is based on solving the operating line equation in conjunction with the equilibrium relationships, Eqs. (3) and/or (11). Stages are always counted from the bottom of the enricher and the bottom of the stripper (see Figs. 1–3). The operating line for component i at stage nE of the enricher ($1 \leqslant nE \leqslant N_E$) is

$$X_{nE,i} = (V_E Y_{nE-1,i} + PX_{Pi})/L_E \tag{16}$$

For stage nS of the stripper ($1 \leqslant nS \leqslant N_S$) the operating line is

$$X_{nS,i} = (V_S Y_{nS-1,i} - WX_{wi})/L_S \tag{17}$$

In the equilibrium stage, $X_{nE,i}$ is in equilibrium with $Y_{nE,i}$ and $Y_{nS,i}$ is in equilibrium with $X_{nS,i}$ according to Eqs. (3) and (11), respectively.

To solve for the concentration profile, the operating line equation and the equilibrium relationship must be solved simultaneously at each stage. In the case of an enricher, for example, one starts from the bottom of the system, where $X_{1,i} = X_{Pi}$, and computes $Y_{1,i}$ using Eq. (11), then proceeds to $X_{2,i}$ using the operating line equation Eq. (16) and so on, to the top stage N_E. When the computation converges properly, $Y_{N_E,i} = X_{Wi}$ should satisfy the balance for component i at stage N_E, expressed by

$$L_E X_{Fi} + VY_{N_E-1,i} = L_E X_{N_E,i} + V_E Y_{N_E,i} \tag{18}$$

as well as the overall balance for the whole enricher, expressed by

$$L_E X_{Fi} = PX_{Pi} + V_E X_{Wi} \tag{19}$$

The stagewise and continuous models are different descriptions of the same system. The stagewise model can be considered as resulting from a special discretization in space (column height) of the continuous model according to the height of an equilibrium stage. One takes advantage of the equilibrium relationships given in Eqs. (3) and (11) that hold true for

a height equivalent to an equilibrium stage. One could arrive at different equations similar to Eqs. (16) and (17) by using other discretizations, and in this case the relationship between $X_{nE,i}$ and $Y_{nE,i}$ would be different from Eqs. (3) and (11).

COMPUTATION OF CONCENTRATION PROFILES BY A MINIMIZATION TECHNIQUE

The need for fast converging concentration profile calculations was explained in the introduction. One can define a convergence performance criterion PR and minimize it. Once the minimum PR is reached, the true concentration profile will be available.

The computation of a concentration profile involves the following steps:

(1) The formulation of a model describing the change of concentration along the system, as shown in the previous section.
(2) Definition of an adequate performance criterion.
(3) Computation of an initial guess regarding the concentration at either end, product or waste, of the system.
(4) Computation of the concentration profile by an iterative procedure until a minimum of the performance criterion is achieved. A minimization technique "drives" this computation by providing the guesses for subsequent iterations.

The flow chart shown in Fig. 4 illustrates the above-mentioned procedure for the computation of the product concentration and concentration profile in an enricher (see Fig. 1) used for the separation of m components. The initial guess on which the computation is based is labeled $\bar{X}_{Pj}$ $(j = 1, \ldots, m)$.

The minimization method used in the present work is the conjugate gradient method, described by Powell (*6*), who also gave the algorithm for its implementation. However, other minimization techniques may also give satisfactory results.

Performance Criterion

A quadratic performance criterion was used throughout the present work:

$$PR = \sum_{i=1}^{m} f_i^2 \tag{20}$$

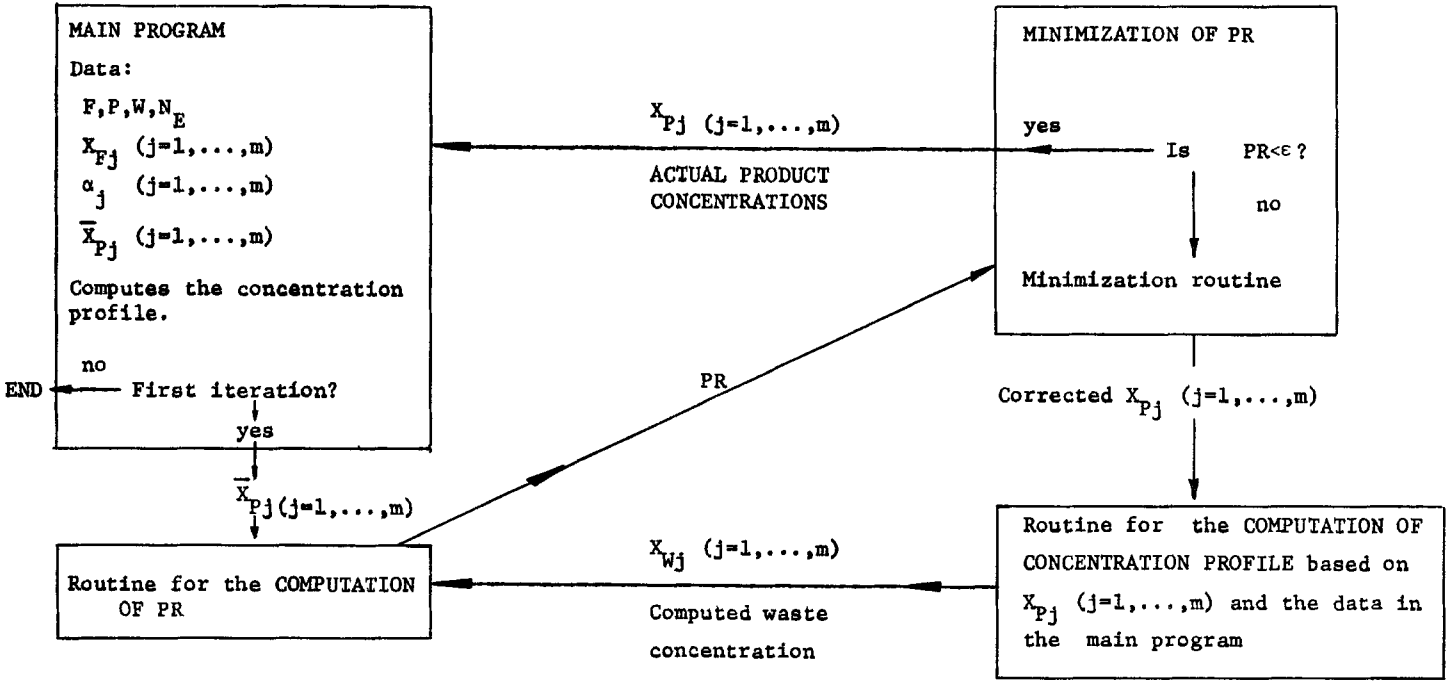

FIG. 4. Flow chart for the computation of the product concentration in an enricher by using the minimization technique.

f_i are functions defined for each of the m components. For the enricher (Fig. 1), f_i expresses the overall balance for $(m - 1)$ components:

$$f_i = L_E X_{Fi} - P X_{Pi} - V_E X_{Wi} \tag{21}$$

when $1 \leqslant i \leqslant m - 1$, while for $i = m$:

$$f_m = \sum_{i=1}^{m} X_{Pi} - 1 \tag{22}$$

This last equation is based on the requirement that the sum of the product concentrations X_{Pi} should be equal to 1.

f_i can be defined similarly for the stripper (Fig. 2). For the enricher and stripper (Fig. 3), the f_i for $m - 1$ components is based on the material balance at the feed reservoir:

$$f_i = L_S X_{Si}(0) + F X_{Fi} + V_E Y_{Ei}(1) - L_E X_{Ei}(1) - V_S Y_{Si}(0) \tag{23}$$

f_m remains defined by Eq. (22).

In some cases, cascades are operated at total reflux. This is a particular case of the enricher and stripper discussed above, with $R_E = 0$ and $R_S = 0$. The f_i for the stagewise model are defined for $1 \leqslant i \leqslant m$ as

$$f_i = X_{Fi} - \sum_{n=1}^{N_E+N_S} X_{ni}/(N_E + N_S) \tag{24}$$

If a continuous model is used, f_i becomes

$$f_i = X_{Fi} - \int_0^1 X \, ds \tag{25}$$

Initial Guess

As mentioned above, an initial guess $\bar{X}_{Pj}$ $(j = 1, \ldots, m)$ is needed for the computation of the product concentration and concentration profile in an enricher. Similarly, $\bar{X}_{Wj}$ $(j = 1, \ldots, m)$ is needed for computations on a stripper. The initial guess should be as close as possible to the expected concentrations in order to facilitate the calculation and reduce the number of iterations necessary to reach convergence. Several ways to obtain the initial guess are described below.

Linearization of Equilibrium Relationship. Linearizing the equilibrium relationship expressed by Eq. (3) yields

$$X_{Ei}^* = \alpha_i Y_{Ei} \tag{26}$$

Substituting Eq. (26) into Eq. (8) for an enricher, the following equation

is obtained:

$$Y_{Ei}(s) = U_i Y_{Ei}(1)[1 + q_i e^{N_E \varepsilon_i (1-s)}] \tag{27}$$

where

$$U_i = \frac{T_i e^{N_E \varepsilon_i}}{T_i e^{N_E \varepsilon_i} + 1} \tag{28}$$

$$T_i = \frac{r}{1 + r} \frac{1}{\alpha_i - 1} \tag{29}$$

$$r = P/V_E \tag{30}$$

$$\varepsilon_i = (1 + r)\alpha_i - 1 \tag{31}$$

$$q_i = 1/T_i \tag{32}$$

At the bottom of the enricher, $X_{Pi} = Y_{Ei}(0)$. $Y_{Ei}(1)$ is determined from a material balance over the enricher (Fig. 1) and substituted into Eq. (27). Thus the following relation is obtained:

$$Y_{Ei}(0) = \frac{L_E X_{Fi}(1 + q_i)}{PU_i(1 + q_i) + W} \tag{33}$$

This expression can serve as an initial guess which will be closer to the actual concentration, the lower the concentrations are and the closer the separation factors are to 1. Under these conditions the error introduced by linearization becomes relatively less significant.

Lehrer (7) considered the case of an enricher separating a binary mixture in which the isotope of interest is at a low concentration and has a separation factor very close to 1. Under these conditions the enrichment E_i for component i was found by him to be

$$E_i = \frac{Y_{Ei}(0)}{X_{Fi}} = \frac{E_{\infty i} e^{\Omega_i}}{E_{\infty i} + e^{\Omega_i} - 1} \tag{34}$$

where

$$\Omega_i = \Omega_{oi} \frac{E_{\infty i}}{E_{\infty i} - 1} \tag{35}$$

$$E_{\infty i} = 1 + \rho/\alpha_i^* \tag{36}$$

$$\Omega_{oi} = N_E(\alpha_i - 1) \tag{37}$$

$$\rho = L_E/P \tag{38}$$

$$\alpha_i^* = \alpha_i/(\alpha_i - 1) \tag{39}$$

$Y_{Ei}(0)$ can be found by solving Eq. (34). Although originally the above expressions were developed (*7*) for binary mixtures, they can be expanded to multicomponent mixtures, provided the component i is at low concentration. For example, if the natural abundancies of the oxygen isotopes is considered, i.e., 0.037% ^{17}O and 0.204% ^{18}O, the enrichment of ^{18}O vs ^{16}O can be computed as if there were no ^{17}O. That is, the system is reduced to a "quasi-binary" one.

Treating an enricher and stripper (Fig. 3), the initial guess, X_{Pi}, is obtained by Eq. (33), disregarding the stripper. The initial guess for the stripper, X_{Wi}, is then obtained by performing a material balance over the whole system using the X_{Pi} found above.

Weighted Separation Factors. The above Eqs. (33) and (34) are useful mainly at relatively low concentrations. At higher concentrations it is usual to use a separation factor α_i related to one of the components and disregard the other components. However, this introduces significant error. For example, in a 6-component system (the separation of krypton isotopes) an enrichment of some components was obtained in spite of the fact that stripping was expected for those components. An improvement can be achieved by using a weighted separation factor $\bar{\alpha}_j$:

$$\bar{\alpha}_j = \sum_{i=1}^{m} \alpha_i X_i \tag{40}$$

where α_i is the separation factor of component i from j.

Another possibility exists for thermal diffusion systems where the computation of the separation factors is based on the molecular weight of the components. In this case an average molecular weight M_{av} of all the components in the system except j is computed:

$$M_{av} = \sum_{\substack{i=1 \\ i \neq j}}^{m} M_i X_i \Big/ \sum_{\substack{i=1 \\ i \neq j}}^{m} X_i \tag{41}$$

The weighted separation factor $\bar{\alpha}_j$ is then computed from the molecular weight of component j and M_{av}, and the computation of the initial guess $\bar{X}_{Pi}$ proceeds using Eqs. (33) and (34). However, the values $\bar{X}_{Pj}$ obtained have to be normalized in order to preserve

$$\sum_{j=1}^{m} \bar{X}_{Pj} = 1$$

Other Methods. In an enricher (Fig. 1) having a large number of stages, X_{Wi} can be computed by assuming it to be in equilibrium with the feed X_{Fi}. $\bar{X}_{Pi}$ can then be calculated from the material balance on the entire enricher. Similarly, in the case of an enricher and stripper (Fig. 3), it can be assumed that $X_{Ei}(1) = X_{Fi}$. Then $Y_{Ei}(1)$ is computed as if it is in equilibrium with $X_{Ei}(1)$. $\bar{X}_{Pi}$ is readily obtained from the material balance on the enricher.

In some cases the initial guess does not lead, within a limited number of iterations, to a convergence of the computation to the required accuracy. It is then useful to start the minimization procedure again, using the results from the previous step as the new initial guess. Similarly, the use of initial guesses based on previous plant operation experience is found to be very useful.

Use of Normalized Concentrations

In some multicomponent calculations, when concentrations are spread over a wide range, the performance criterion as defined in Eq. (20) becomes weighted in favor of components at higher concentrations, while those occurring at low and very low concentrations are not sufficiently represented. The solution converges for the components at medium and high concentrations, while for the other components the convergence is very slow. In order to avoid this inconvenience, one could either weigh properly the different values of f_i (Eqs. 21–25) in the performance criterion or normalize the concentrations vs the initial guess. The second approach is more attractive because it is more general. For example, in the case of the enricher (Fig. 1), the normalized concentration of component i at height s, $W_{Ei}(s)$, is defined as

$$W_{Ei}(s) = Y_{Ei}(s)/\bar{Y}_{Ei}(0) \tag{42}$$

After replacing $Y_{Ei}(s)$ by the normalized concentrations $W_{Ei}(s)$, Eq. (8) becomes

$$\bar{Y}_{Ei}(0)\frac{dW_{Ei}(s)}{ds} = N_E R_E \bar{Y}_{Ei}(0) W_{Ei}(0) - N_E\left[(1 + R_E)\frac{\alpha_i \bar{Y}_{Ei}(0) W_{Ei}(s)}{\sum_{j=1}^{m} \alpha_j \bar{Y}_{Ej}(0) W_{Ej}(s)} - \bar{Y}_{Ei}(0) W_{Ei}(s)\right] \tag{43}$$

Substituting β_j for $\alpha_j Y_{Ej}(0)$, the above equation can be simplified

$$\frac{dW_{Ei}(s)}{ds} = N_E R_E W_{Ei}(0) - N_E \left[(1 + R_E) \frac{\alpha_i W_{Ei}(s)}{\sum_{j=1}^{m} \beta_j W_{Ej}(s)} - W_{Ei}(s) \right] \quad (44)$$

The minimization technique is used with a performance factor based on the normalized concentrations, W_{Ei}. At the end of the computation these are reconverted into the actual concentrations $Y_{Ei}(s)$. This method was successfully used in computations performed for thermal diffusion cascades separating isotopes of krypton.

RESULTS AND DISCUSSION

The methodology described in the previous section was implemented in many computations. A few typical cases are described below.

Separation of Oxygen Isotopes by Thermal Diffusion

Enricher. The enricher, shown in Fig. 1, has 300 theoretical stages and is fed with oxygen having natural abundancies of ^{17}O (0.037%) and ^{18}O (0.204%). Flow rates are: $L_E = 35000$ cc (STP)/day and $P = 50$ cc (STP)/day. Separation factors are $\alpha_{17/16} = 1.0066$ and $\alpha_{18/16} = 1.0133$. The following initial guesses were used for the product concentrations:

(a) 0.18% ^{17}O and 2% ^{18}O, based on previous experience.
(b) 0.1529% ^{17}O and 1.8702% ^{18}O, based on Eq. (34).
(c) 0.1536% ^{17}O and 1.8923% ^{18}O, based on Eq. (33).

The product concentrations found by using the minimization technique are 0.1512% ^{17}O and 1.8612% ^{18}O. The computations were ended for $PR \leqslant 10^{-20}$.

In this example, low concentrations were involved. Therefore, the initial guesses based on the linearization of the equilibrium relationship (above, b and c) were within about 2% of the actual product concentrations.

Enricher and Stripper. A cascade (6) consisting of 6 columns (ZA1–ZA6) of 100 theoretical stages each (see Fig. 5) was fed at its middle (i.e., between the top of column ZA4 and the bottom of column ZA3) with oxygen gas at a flow rate of 470 cc (STP)/day. The cascade was operated

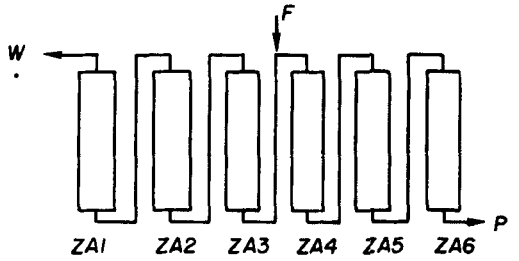

FIG. 5. A thermal diffusion cascade for the separation of oxygen isotopes. The columns are labeled ZA1–ZA6.

at $P = W = 235$ cc (STP)/day and $L_S = V_E = 26{,}000$ cc (STP)/day. The enricher (columns ZA4, 5, and 6) and the stripper (columns ZA1, 2, and 3) have 300 theoretical stages each, i.e., $N_E = N_S = 300$. The feed composition was 12.4% ^{17}O, 49.9% ^{18}O, and 37.7% ^{16}O. Separation factors are as in the previous example.

Two sets of initial guess concentrations, $\bar{X}_{Pi}$, were used:

(1) 11.29% ^{17}O, 81.985% ^{18}O, and 6.725% ^{16}O. These concentrations were obtained, as shown in the section entitled "Other Methods," i.e., assuming that $Y_{Ei}(1)$ is in equilibrium with $X_{Ei}(1) = X_{Fi}$.
(2) 10.9% ^{17}O, 75.6% ^{18}O, and 13.5% ^{16}O, based on previous experience.

With both initial guesses, at convergence ($PR < 10^{-14}$), the product concentrations are 10.946% ^{17}O, 78.411% ^{18}O, and 10.643% ^{16}O. The experimental and computed ^{17}O and ^{18}O concentration profiles along this cascade are shown in Fig. 6. ZA1T represents the top of column ZA1 and therefore the top of the entire cascade. ZA1–ZA6 are concentrations at the bottom of columns ZA1–ZA6, respectively.

The curved concentration profile for ^{17}O as mentioned in the introduction was found both experimentally and by computation. Good agreement was found between the computed and experimental ^{17}O and ^{18}O profiles.

Separation of Krypton Isotopes by Thermal Diffusion

An enricher consisting of thermal diffusion columns with a total of 600 separation stages was fed at its top with krypton. The feed con-

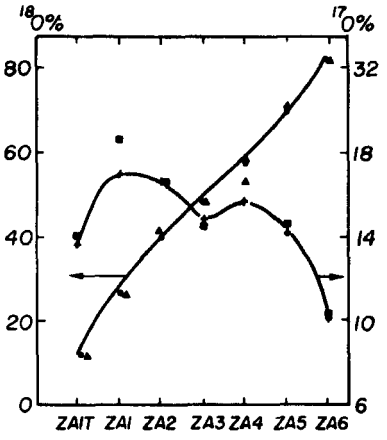

FIG. 6. Concentration profiles in the ZA1–ZA6 thermal diffusion cascade (Fig. 5) for the separation of oxygen isotopes: (■) ^{17}O measured concentration, (+) ^{17}O calculated concentration, (●) ^{18}O measured concentration, and (▲) ^{18}O calculated concentration.

sisted of 6 components, i.e., ^{78}Kr, ^{80}Kr, ^{82}Kr, ^{83}Kr, ^{84}Kr, and ^{86}Kr, at their natural abundancies, X_{Fi}, as shown in Table 1. The cascade was operated at $L_E = 15{,}000$ cc (STP)/day and $P = 50$ cc (STP)/day. Separation factors of the various isotopes from ^{86}Kr, as well as weighted separation factors, are given in the same table. Five different initial guesses, IG1–IG5, as shown in Table 1, were used:

(1) IG1 is based on the linearization of the equilibrium relationship and Eq. (34).
(2) IG2 is also based on the linearization of the equilibrium relationship, but uses Eq. (33). Neither IG1 nor IG2 satisfy the condition $\sum_{j=1}^{m} \bar{X}_{Pj} = 1$.
(3) IG3 was obtained by normalizing the concentrations found in IG2, thus satisfying $\sum_{j=1}^{m} \bar{X}_{Pj} = 1$.
(4) IG4 was found by assuming that X_{Wj} is in equilibrium with X_{Fj} (see the section antitled "Other Methods"). It can be seen in the table that this assumption lead to negative product concentrations.
(5) In IG5, the negative concentration obtained with IG4 were replaced by an arbitrary value of 0.1 %.

The computations were performed by using normalized concentrations,

TABLE 1

Separation of Krypton Isotopes

Isotope	^{78}Kr	^{80}Kr	^{82}Kr	^{83}Kr	^{84}Kr	^{86}K
Feed concentration, X_{Fi} (%)	0.35	2.27	11.56	11.55	56.90	17.37
Separation factor from ^{86}Kr:						
α_i	1.0195	1.0144	1.0094	1.00706	1.0046	1.000
$\bar{\alpha}_j$ (based on M_{av}, Eq. 41)	1.0195	1.00935	1.0075	1.00205	0.9987	0.9936
Initial guess:						
IG1	0.000022	0.0413	0.55	6.20	76.90	49.80
IG2	0.000022	0.041	0.55	6.20	76.90	49.80
IG3	0.000022	0.031	0.41	4.65	57.6	37.30
IG4	−1.7	−4	−3.9	4.0	61.0	43.0
IG5	0.1	0.1	0.1	4.3	61.8	43.0
Product concentration, X_{Pi}:						
Continuous model	0.000239	0.02641	1.71805	4.84151	52.74379	40.67001
Stagewise model	0.000229	0.026051	1.716424	4.843634	52.7524	40.65942

as shown in the section entitled "Use of Normalized Concentrations." The X_{Pi} product concentrations were computed by using the various initial guesses and the minimization technique. The computation was completed when a performance factor of less than 10^{-16} was achieved. The results were found to be independent of the different initial guesses used. However, the convergence was very slow when using the initial guess IG5 because, in this case, the guessed ^{78}Kr, ^{80}Kr, and ^{82}Kr concentrations were very different from the actual values. The results obtained by using the continuous model were slightly different from those computed by the stagewise model. However, these minor differences have no practical importance.

CONCLUSIONS

Concentration profiles in multicomponent and multistage separation systems can be computed by using a continuous or a stagewise model. It was shown that both models lead to essentially the same results. Numerical solutions of these models are iterative. Therefore, initial guesses of concentrations at one end (usually the product) of the system are needed. Several methods for obtaining such initial guesses were presented. The use of the minimization technique permitted the fast convergence of concentration profile computations. The thermal diffusion separation of krypton isotopes was given as an example to illustrate the potential of the procedure developed in this work. This is a difficult separation of 6-component mixture involving widely varying concentrations over 600 separation stages. Although developed with special emphasis on isotope separation problems, the procedure is also applicable to other multicomponent and multistage separation processes.

SYMBOLS

C	integration constant
E	enrichment
f	defined in Eqs. (21)–(25)
K	mass transfer coefficient
L	downflow rate
$\bar{L}$	downflow rate/unit cross section
M_{av}	average molecular weight, Eq. (41)
m	number of components

N	total number of stages
P	product flow rate
PR	performance factor defined in Eq. (20)
q	defined in Eq. (32)
R_E	defined in Eq. (5)
R_S	defined in Eq. (10)
r	defined in Eq. (30)
s	normalized height
T	defined in Eq. (29)
U	defined in Eq. (28)
V	upflow rate
W	normalized concentration, defined in Eq. (42)
X	concentration in the downflow stream
X^*_{Ei}	defined in Eq. (3)
$\bar{X}_P$	initial guess of product concentration
Y	concentration in the upflow stream
Z	total height
z	height

Subscripts

E	enricher
F	feed
i	component
nE	number of the stage in the enricher
nS	number of the stage in the stripper
P	product
S	stripper
W	waste
∞	infinite column

Greek Letters

α	separation factor
α^*_i	defined in Eq. (39)
$\bar{\alpha}_j$	defined in Eq. (40)
ε	defined in Eq. (31)
Ω	defined in Eq. (35)
ρ	defined in Eq. (38)

REFERENCES

1. Ch. D. Holland, *Multicomponent Distillation*, Prentice-Hall, Englewood Cliffs, New Jersey, 1963.
2. M. D. Powell, in *Numerical Methods for Nonlinear Algebraic Equations* (Ph. Rabinowitz, ed.), Gordon and Breach, London, 1970.
3. D. Wolf and H. Cohen, *Can. J. Chem. Eng.*, *50*, 621 (1972).
4. H. R. C. Pratt, *Countercurrent Separation Processes*, Elsevier, Amsterdam, 1967.
5. K. Cohen, *The Theory of Isotope Separation as Applied to the Large-Scale Production of* ^{235}U, McGraw-Hill, New York, 1951.
6. Ch. Gilath, "Dynamics of Packed Distillation and Thermal Diffusion Columns Used in Isotope Separation," Ph.D. Thesis, Weizmann Institute of Science, Rehovot, Israel, 1973.
7. I. Lehrer, *Separation of Isotopes by Distillation*, Israel AEC Report IA 624, 1961.

Received by editor July 14, 1977