# THEORY OF CASCADES FOR SEPERATING MULTI-COMPONENT ISOTOPE MIXTURES

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The article discusses a stationary process for the separation of multi-component isotope mixtures in cascades of arbitrary profile. It includes a theoretical investigation of the extreme cases in which the relative rate of isotope exchange has values of zero and infinity. A solution is obtained for a system of transport equations for a rectangular stepped cascade, and a calculation procedure for such cascades is worked out.

The development of nuclear technology today has brought an increased interest in the problem of separating multi-component isotope mixtures. This problem arises in the production of pure isotopes of elements which have more than two stable isotopes and in many cases in which binary isotopes are separated in the form of chemical compounds.

References [1-4] tell of experimental investigations concerned with the separation of multi-component isotope mixtures in cascades. However, the theory of this process has not been sufficiently developed. The authors know of only two theoretical studies [5, 6]: the first deals with a column in which there is no removal of the product, and the second with the development of a theory for an ideal cascade.

The purpose of the present study is to solve a system of transport equations in the stationary case for cascades of arbitrary profile\* which include input and removal of the product, as well as to work out a calculation procedure for practical cascades. When isotopes are separated in the form of chemical compounds, the basic process in the cascade may be accompanied by isotope exchange between the molecules of the components, with the result that the number of molecules of an individual component does not remain the same. The effect of this process on the separation depends on its relative rate. In the present study we consider the two extreme cases—zero and infinite relative rates of isotope exchange—to which most of the practical problems of isotope separation can be reduced.

## 1. Relative Rate of Isotope Exchange

In the separation of multi-component mixtures consisting of nonmonatomic molecules the process in each stage upsets the equilibrium distribution of isotopes among the components. This leads to a process tending to the reestablishment of equilibrium through isotope exchange.

For the sake of simplicity, we shall consider the distribution of a substance consisting of molecules of the type XY. We shall assume that the elements X and Y are different. The components of a mixture, in accordance with the isotopic composition of their molecules  $X^mY^n$ , will be denoted by the indices mn. Let the element X have  $q_1$  isotopes and let the element Y have  $q_2$  isotopes. We shall denote the number of components in the mixture by  $q_1q_2 = k$ . The mole-fraction concentrations of the components are  $[X^mY^n] = N_{mn}$ . The mole fractions of the corresponding isotopes will be denoted by  $x_m$  and  $y_n$ .

Suppose that at time t = 0 this mixture, filling some volume, has concentrations  $N_{mn}$  (0) which are different from the equilibrium concentrations. The decrease in the number of molecules of the component mn per unit volume resulting from isotope exchange in time dt may be written as

<sup>•</sup> By "profile" we shall mean the distribution L(s) of the flow rate of the mixture of isotopes along the cascade, where s is the longitudinal coordinate.

$$d_1 Q_{mn} = -Q_{mn} \sum_{i \neq n, j \neq m} \gamma_{ji}^{mn} Q_{ij} dt, \qquad (1.1)$$

where  $\gamma_{ij}^{mn}$  are kinetic coefficients independent of concentration. Similarly, the increase in the number of molecules is:

$$d_2 Q_{mn} = \sum_{i \neq n, j \neq m} \gamma_{jn}^{mi} Q_{mi} Q_{jn} dt. \tag{1.2}$$

If we assume the coefficients  $\gamma$  to be equal for all components, after some simple transformations, we obtain the differential equation

$$\frac{1}{\gamma} \cdot \frac{d\varrho_{mn}}{dt} = -\varrho_{mn} \sum_{i,j} \varrho_{ij} + \sum_{i} \varrho_{mi} \sum_{j} \varrho_{jn}. \tag{1.3}$$

From the conditions that the number of atoms of the isotopes must remain constant, it follows that  $\sum_{i}^{n} \varrho_{mi}$ ,  $\sum_{i}^{n} \varrho_{jn}$  and  $\sum_{i,j}^{n} \varrho_{ij} = \varrho$  are independent of time. Consequently, after integrating and changing to mole-fraction concentrations, we find

$$N_{mn}(t) - x_m y_n = [N_{mn}(0) - x_m y_n] \exp(-\gamma \varrho t).$$
 (1.4)

Let to be the length of time the mixture remains in the given volume. From (1.4), writing

$$\beta = \exp\left(-\gamma \varrho t_0\right),\tag{1.5}$$

we obtain

$$N_{mn}(t_0) = \beta N_{mn}(0) + (1 - \beta) x_m y_n.$$
(1.6)

Let us now consider the separating cascade. To simplify our discussion, we shall consider a cascade scheme in which isotope exchange takes place in exchangers situated at the outlets of the separating stages. This scheme enables us to write the transport equations correctly in a first approximation using  $\varepsilon$ . The residence time of the mixture in the exchanger is  $t_0 = h/L$ , where h is the exchanger holdup (if the exchange takes place directly in the separating stage, h is the holdup of the stage in the corresponding phase. From this it follows that

$$\beta = \exp\left(-\gamma \varrho \, \frac{h}{L}\right) \,. \tag{1.7}$$

As can be seen from formula (1.6), the effect of the exchange on the separation may be evaluated by means of the expression:

$$\frac{1-\beta}{\beta} = \frac{N_{mn}(0) - N_{mn}(t_0)}{N_{mn}(t_0) - x_m y_n} , \qquad (1.8)$$

which we shall herein after call the relative exchange rate v.

It follows from formula (1.8) that in the extreme case when  $v = \infty$ , we have  $N_{mn}(t_0) = x_m y_n$ , and when v = 0, we have  $N_{mn}(t_0) = N_{mn}(0)$ . It will be shown below that even when  $v \sim 1$ , the isotope exchange may have a considerable influence on the separation. Conversely, in the case when v < 1, the exchange may be neglected. When v = 0, the number of molecules of each component remains constant.

## 2. Derivation of the Transport Equations for the Case of Zero Exchange Rate

Let us consider a separating cascade with an arbitrary profile. At some points of the cascade the mixture is fed in or removed. We shall assume that the cascade consists of a finite number of segments each of which has a continuous profile. At the points of discontinuity of L(s), as well as at the input and output points, there is a discontinuity of the cascade parameters. The part of a cascade included between two adjacent points of discontinuity of the parameters will be called a section. Let B denote the total number of sections in a cascade and let b be the

index number of an individual section. Each section consists of  $\sigma^b$  separating stages connected in series. The serial number of a stage will be denoted by s (for stages within the same section we shall omit the index b).

In a separating section the total flow L and the concentrations of the components N<sub>m</sub> are given by the equations

$$L = L' + L''; (2.1)$$

$$LN_m = L'N'_m + L''N''_m \quad (m = 1, \ldots, k),$$
 (2.2)

where L', and L' are the flows leaving the stages at the beginning and at the end of the section, respectively;  $N_{m}^{*}$  and  $N_{m}^{*}$  are the concentrations of component m in the corresponding flows. We introduce the relative coefficient of separation

$$\alpha_{mn} = \frac{N'_m}{N''_m} \cdot \frac{N''_n}{N'_n} \quad (1 \leqslant m, \ n \leqslant k). \tag{2.3}$$

In the general case the coefficients of separation depend on the concentrations of the components; however, this dependence is so weak that the values  $\alpha_{mn}$  may be regarded as constant. In general the isotopes satisfy the inequality

$$|\alpha_{mn}-1|\ll 1. \tag{2.4}$$

Using this fact, we introduce the small parameter

$$\varepsilon = \frac{1}{k} \sqrt{\sum_{m,n} (\alpha_{mn} - 1)^2}.$$
 (2.5)

In the remainder of this discussion we shall neglect of a smaller order of magnitude. From Eqs. (2.1) and (2.2) we readily obtain

$$\delta'' N_m = \frac{\Theta}{1 - \Theta} \, \delta' N_m, \tag{2.6}$$

where  $\delta$  'N<sub>m</sub> = N<sub>m</sub> - N<sub>m</sub>;  $\delta$  "N<sub>m</sub> = N<sub>m</sub> - N<sub>m</sub> and  $\theta$  = L'/L. If we assume that  $|\theta|(s) - 0.5 \sim |\epsilon|$ , then  $|\delta|$ 'N<sub>m</sub> -  $\delta$ "N<sub>m</sub> | / 2  $\sim \epsilon |\delta|$  N<sub>m</sub> |, where  $\delta|N_m| = \delta$ 'N<sub>m</sub> +  $\delta$ "N<sub>m</sub> = N<sub>m</sub> - N<sub>m</sub>".

Let us estimate the quantity  $\delta N_m$ . From the identity

$$\sum_{n=1}^{h} N_n = 1 \tag{2.7}$$

it follows that

$$\sum_{n} \delta' N_n = \sum_{n} \delta'' N_n = 0. \tag{2.8}$$

Using the definition (2.3), we readily find

$$N_n''(N_m' - N_m'') = N_m''(\alpha_{mn}N_n' - N_n''). \tag{2.9}$$

Summing (2.9) over n and using (2.7) and (2.8), we find

$$N'_m - N''_m = N''_m \sum_{n=1}^k (\alpha_{mn} - 1) N'_n,$$

i.e., the order of magnitude of  $|\delta N_m|$  is no greater than  $\epsilon N_m$ .

We shall call  $\epsilon_{mn}$  = ln  $\alpha_{mn}$  the relative coefficient of enrichment. Let us rewrite (2.3) in the form

$$\varepsilon_{mn} = \ln \frac{N_m'}{N_m''} - \ln \frac{N_n'}{N_n''}. \qquad (2.10)$$

From this it can be seen that the relative coefficients of enrichment are additive and antisymmetric:

$$\begin{array}{l}
\varepsilon_{mn} = \varepsilon_{mr} + \varepsilon_{rn}, \\
\varepsilon_{nm} = -\varepsilon_{mn}.
\end{array}$$
(2.11)

Consider an expression of the type  $\ln (N'/N'') = \ln (N'/N) - \ln (N''/N)$ . Expanding  $\ln (N'/N'') = [1 + \delta'N/N)]$   $-\ln [1 - (\delta'N/N)]$  into a Taylor's series in  $\frac{\delta'N}{N} \sim \epsilon$  and  $\frac{\delta''N}{N} \sim \epsilon$ , we find  $\ln \frac{N'}{N''} = \frac{\delta N}{N}$ . to within terms of the order of  $\epsilon^2$  inclusive. Thus, (2,10) may be written in the form:

$$\mathbf{e}_{mn} = \frac{\delta N_m}{N_m} - \frac{\delta N_n}{N_n} \tag{2.12}$$

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$$N_n \delta N_m = N_m \varepsilon_{mn} N_n + N_m \delta N_n. \tag{2.13}$$

In (2.13) we sum over n and obtain

$$\delta N_m = N_m \sum_{n=1}^k \varepsilon_{mn} N_n. \tag{2.14}$$

From this, using the equations  $\delta N_m = \delta N_m + \delta N_n$ , and (2.6), we find

$$N'_{m} - N_{m} = (1 - \Theta) N_{m} \sum_{n=1}^{k} \varepsilon_{mn} N_{n}.$$
 (2.15)

It should be noted that these relations may also be derived from an analysis of the thermodynamics of the process.

Let us take a hypothetical cross section of the cascade between stages s-1 and s, which belong to section b. The law of conservation of matter is written in the form

$$L'(s-1) + \sum_{r=b}^{B} F^{r} = L''(s) + \sum_{r=b}^{B} P^{r};$$
 (2.16)

$$L'(s-1)N'_{m}(s-1) + \sum_{r=b}^{B} F^{r}\widetilde{N}_{m}(\sigma^{r})$$

$$= L''(s)N'''_{m}(s) + \sum_{r=b}^{B} P^{r}N_{m}(\sigma^{r}).$$
(2.17)

Here  $P^r$  and  $F^r$  are the output and input flows, respectively, at the end of section r;  $N_m(\sigma^r)$  is the concentration of component m at the corresponding removal point;  $\widetilde{N}_m(\sigma^r)$  is the concentration in the corresponding input flow. Passing from difference equations to differential equations and making some simple transformations, we obtain a system of transport equations in section b for a k-component isotope mixture:

$$\frac{dN_m(s)}{ds} = N_m(s) \sum_{n=1}^k \varepsilon_{mn} N_n(s) - \frac{2}{L^b(s)} \sum_{r=b}^B \times \{P^r[N_m(\sigma^r) - N_m(s)] - F^r[\widetilde{N}_m(\sigma^r) - N_m(s)]\}.$$
(2.18)

In this system only (k-1) equations are independent, since the identity (2.7) is its first integral. When k=2, the system (2.18) becomes the well-known transport equation for a binary mixture.

Setting L(s) = const and B = 1, we obtain the system of transport equations given earlier in [5] for thermal diffusion.

## 3. The Extreme Case of an Infinite Exchange Rate

Writing the concentrations at the outlets of a stage or the inlets of the exchangers as  $\overline{N}_{mn}^*$  and  $\overline{N}_{mn}^*$ , respectively, we can rewrite Eqs. (2.15) in the form

$$\overline{N}'_{mn} - N_{mn} = (1 - \Theta) N_{mn} \sum_{i,j} \varepsilon_{ij}^{mn} N_{ij}. \tag{3.1}$$

The relative coefficient of enrichment are determined from the equation of the separating stage

$$\alpha_{ij}^{mn} = \frac{\overline{N}'_{mn}}{\overline{N}''_{mn}} \cdot \frac{\overline{N}''_{ij}}{\overline{N}'_{ij}}$$
(3.2)

by means of the formula

$$\varepsilon_{ij}^{mn} = \ln \alpha_{ij}^{mn}. \tag{3.3}$$

It is evident that the values  $\epsilon_{ij}^{\,mn}$  are additive and antisymmetric.

By the definition of mole-fraction concentrations, we have

$$x_m = \sum_{n=1}^{q_2} N_{mn}, \quad y_n = \sum_{m=1}^{q_1} N_{mn}. \tag{3.4}$$

Since in an exchanger (unlike the case of a separating stage) the isotope concentrations in the mixture do not change, we find from Eqs. (3.1) and (3.4) that the concentrations  $x_m^*$  and  $x_m^*$  at the outlets of the exchangers satisfy the equations

$$x'_{m} - x_{m} = (1 - \Theta) \sum_{n, i, j} \varepsilon_{ij}^{mn} N_{mn} N_{ij}, 
 x_{m} - x''_{m} = \frac{\Theta}{1 - \Theta} (x'_{m} - x_{m}).$$
(3.5)

Writing the law of conservation of matter for the isotopes  $X^{m}$  and passing to differential equations, we obtain the system of transport equations

$$\frac{dx_{m}(s)}{ds} = \sum_{n, i, j} \varepsilon_{ij}^{mn} N_{mn}(s) N_{ij}(s) - \frac{2}{L^{b}(s)} \sum_{r=b}^{B} \times \{P^{r} \left[x_{m}(\sigma^{r}) - x_{m}(s)\right] - F^{r} \left[\widetilde{x}_{m}(\sigma^{r}) - x_{m}(s)\right]\}.$$
(3.6)

In the extreme case of an infinite rate of exchange (see Section 1)

$$N_{mn}(s) = x_m(s) y_n(s).$$
 (3.7)

If we make the substitution  $\varepsilon_{ij}^{mn} = \varepsilon_{mj}^{mn} + \varepsilon_{ij}^{mj}$  in (3.6) and utilize (3.7), noting the identity for  $\sum_{n, i, j} \varepsilon_{mj}^{mn} N_{mn} N_{ij}$ , we find

$$\frac{dx_m}{ds} = x_m \sum_{ij} \varepsilon_{ij}^{mj} x_i y_j - \frac{2}{L^b} \sum_r \times \{ P^r \left[ x_m \left( \sigma^r \right) - x_m \right] - F^r \left[ \tilde{x}_m \left( \sigma^r \right) - x_m \right] \}. \tag{3.8}$$

In a similar manner, we can write the transport equations for the isotopes of element Y.

The system of transport equations, consisting of (3.8) and the equations for the isotopes of the element Y, enables us to carry out the complete calculation for the cascade. It should be noted that the equations of this system contain only coefficients of relative enrichment whose unwritten indices are identical. We may expect that the

dependence of the coefficients of enrichment on the identical indices will become apparent in the second-order terms. This is easily proved in the case when the coefficients of separation depend only on the mass numbers. As a result, we can divide the system of transport equations into two independent systems of the form

$$\frac{dx_{m}(s)}{ds} = x_{m}(s) \sum_{i=1}^{q_{1}} \varepsilon_{i}^{m} x_{i}(s) - \frac{2}{L^{b}(s)} \sum_{r=b}^{B} \times \{P^{r}[x_{m}(\sigma^{r}) - x_{m}(s)] - F^{r}[\widetilde{x}_{m}(\sigma^{r}) - x_{m}(s)]\}. \tag{3.9}$$

Here the coefficients  $\epsilon_i^m$  are obtained from  $\epsilon_{ij}^m$  by fixing the identical indices j. It is readily seen that the values  $\epsilon_i^m$  are additive and antisymmetric.

Thus, in the case of an infinite rate of exchange the transport equations for the isotopes completely coincide with the transport equations for the components of a mixture with a zero rate of exchange.

Let us estimate the effect of the isotope exchange on the separation. Taking account of the equation

$$dN_{mn} = x_m dy_n + y_n dx_m,$$

we find

$$\frac{dN_{mn}}{ds} = N_{mn} \sum_{ij} \left( \varepsilon_{ij}^{mj} + \varepsilon_{ij}^{in} \right) N_{ij} - \frac{2}{L^b} \sum_{r} \times \left\{ P^r \left[ x_m y_n \, \sigma^r \right) + y_n x_m \left( \sigma^r \right) - 2N_{mn} \right] - F^r \left[ x_m \widetilde{y_n} \left( \sigma^r \right) + y_n x_m \left( \sigma^r \right) - 2N_{mn} \right] \right\}.$$
(3.10)

From a comparison of (3.10) and (2.18) it follows that in total-reflux operation the effect of the isotope exchange is manifested as a second-order effect, since we generally have  $|\epsilon_{ij}^{mj} + \epsilon_{ij}^{in} - \epsilon_{ij}^{mn}| \propto \epsilon^2$ , whereas when there is removal, it is a first-order effect.

It is readily shown that in the cases when the kinetic coefficients  $\gamma$  are considered identical for all components of the mixture, the transport equations for mixtures consisting of arbitrary molecules may be represented in the form of independent systems of the type (3.9).

## 4. Solutions of the System of Transport Equations

Since the written forms of the transport equations in the extreme cases of high and low rates of isotope exchange are identical we shall concern ourselves with solving the system (2.18). Let us write

$$2\sum_{r=b}^{B} [P^{r}N_{m}(\sigma^{r}) - F^{2}\widetilde{N}_{m}(\sigma^{r})] = g_{m}^{b}.$$
(4.1)

Using (2.7), we obtain

$$2\sum_{r=b}^{B} (P^{r} - F^{r}) = \sum_{m=1}^{b} g_{m}^{b}.$$
(4.2)

If we use this notation, the system (2.18) becomes

$$L^{b}(s) \frac{dN_{m}(s)}{ds} = L^{b}(s) N_{m}(s) \sum_{n=1}^{k} \varepsilon_{mn} N_{n}(s) - g_{m}^{b} + N_{m}(s) \sum_{n=1}^{k} g_{n}^{b}.$$
(4.3)

From the equation for component m we successively subtract the equations for the remaining components, so that we obtain (k-1) equations of the type

$$\frac{d \ln N_m(s)}{ds} - \frac{d \ln N_n(s)}{ds} = \varepsilon_{mn} - \frac{g_m^b}{L^b(s) N_m(s)} + \frac{g_n^b}{L^b(s) N_n(s)}. \tag{4.4}$$

Equations (4.4) and the identity (2.7) together form a complete system for determining the concentrations of the components. It is readily shown that the systems of differential Eqs. (4.3) and (4.4) are equivalent if the condition (2.7) is satisfied.

In the simplest case, when the cascade operates without removal  $(g_m^b = 0)$ , the system (4.4) is readily integrable, and the solution is of the form

$$N_{m}(s) = \frac{N_{m}(0)}{\sum_{n=1}^{k} N_{n}(0) \exp \varepsilon_{nm}s}$$
 (4.5)

Thus, the distribution of concentrations of components in such operation is independent of the shape of the cascade.

In order to find a solution in the general case, let us consider an individual section. We shall temporarily omit the index b and introduce a numbering system for the stages within the section,  $0 \le s \le \sigma$ . If we write

$$L(s) N_m(s) = x_m(s), (4.6)$$

then

$$\sum_{m=1}^{k} x_m(s) = L(s) \tag{4.7}$$

and Eqs. (4.4) become

$$\frac{d \ln x_m}{ds} - \frac{d \ln x_n}{ds} = \varepsilon_{mn} - \frac{g_m}{x_m} + \frac{g_n}{x_n} . \tag{4.8}$$

Making the substitution

$$\frac{d\ln x_m}{ds} = \frac{g_m}{x_m} = \frac{d\ln x_m}{ds} , \qquad (4.9)$$

we reduce Eqs. (4.8) to the form

$$\frac{d}{ds}\ln\frac{\bar{x}_m}{\bar{x}_n} = \varepsilon_{mn}.$$
 (4.10)

As a result, we obtain

$$\frac{\bar{x}_{m}(s)}{\bar{x}_{m}(0)} = \frac{\bar{x}_{n}(s)}{\bar{x}_{n}(0)} \exp \varepsilon_{mn} s. \tag{4.11}$$

The solution of the linear differential Eq. (4.9) with the boundary condition  $x_m(0) = x_{m0}$  is of the form

$$x_{m}(s) = \frac{\bar{x}_{m}(s)}{\bar{x}_{m}(0)} \left\{ x_{m0} - g_{m} \int_{0}^{s} \frac{\bar{x}_{m}(0)}{\bar{x}_{m}(t)} dt \right\}, \qquad (4.12)$$

We introduce the notation

$$\varphi_m(s) = \frac{\bar{x}_m(0)}{\bar{x}_m(s)} , \qquad (4.13)$$

by means of which we shall write Eqs. (4.12) and (4.11), respectively, as

$$x_{m}(s) \varphi_{m}(s) = x_{m_{0}} - g_{m} \int_{0}^{s} \varphi_{m}(t) dt;$$
 (4.14)

$$\varphi_m(s) = \varphi_n(s) \exp \varepsilon_{nm} s. \tag{4.15}$$

In (4.14) we sum over m, and using (4.7) and (4.15), we find

$$\varphi_n(s) + \frac{1}{L(s)} \int_0^s \varphi_n(t) \sum_{m=1}^k g_m \exp \varepsilon_{mn}(s-t) \cdot dt = \frac{1}{L(s)} \sum_{m=1}^k x_{m0} \exp \varepsilon_{mn} s.$$

$$\tag{4.16}$$

Thus, the system (2.18) has been reduced to (4.16), a Volteva integral equation with a degenerate kernel, and k-1 algebraic relations for the functions  $\varphi_m(s)$ . For a given form of L(s) the Eq. (4.16) has a unique solution, which may be constructed, for example, by the method of successive approximation. After this, using formulas (4.14) and (4.6), we find the concentrations of the components of the mixture (within the given section).

Let us consider the case which is of greatest practical interest: a rectangular stepped cascade. Within one section we have L(s) = const, and Eq. (4.6) may be written as

$$\varphi_n(s) + \int_0^s \varphi_n(t) \sum_{m=1}^k c_m \exp \varepsilon_{mn}(s-t) dt = \sum_{m=1}^k N_{m0} \exp \varepsilon_{mn} s, \qquad (4.17)$$

where

$$c_m = \frac{g_m}{L} . (4.18)$$

The integral Eq. (4.17) is solved by the Laplace transform method. It should be noted that the kernel and the free term of the equation are analytic functions in the entire open plane of the complex argument z. Consequently  $\varphi_n(z)$  can be analytically continued for all  $|z| < \infty$ , and we shall denote the continuation for all real values of the argument z = s by  $\psi_n(s)$ .

Applying the Laplace transform to  $\psi_{\Pi}(s)$ , we find

$$F_n(q)\left\{1+\sum_{m=1}^k \frac{c_m}{q-\varepsilon_{mn}}\right\} = \sum_{m=1}^k \frac{N_{m0}}{q-\varepsilon_{mn}}, \qquad (4.19)$$

where

$$F_n(q) = \int_0^\infty \exp(-qs) \,\psi_n(s) \,ds. \tag{4.20}$$

By the inversion formula we have

$$\psi_n(s) = \frac{1}{2\pi i} \int_{q_0 - ic.}^{q_0 + ic.} \exp(qs) F_n(q) dq. \tag{4.21}$$

As can be seen from formula (4.19), when  $q \to \infty$ , the function  $F_n(q)$  approaches 0 like  $q^{-1}$ . Consequently, in (4.21) we integrate along a contour closed at the left, and, applying the residue theorem, we obtain

$$\psi_n(s) = \sum \operatorname{res} \left[ F_n(q) \exp qs \right] \quad (0 \leqslant s < \infty). \tag{4.22}$$

Since  $\varphi_n(s)$  is identical with  $\psi_n(s)$  in the interval  $0 \le s \le \delta$ , we can substitute into (4.22) the value of  $F_n(q)$  from (4.19) and obtain

$$\varphi_{n}(s) = \sum_{k} \operatorname{res} \left[ \frac{\sum_{m=1}^{k} N_{m0} (q - \varepsilon_{mn})^{-1}}{1 + \sum_{m=1}^{k} c_{m} (q - \varepsilon_{mn})^{-1}} \exp qs \right].$$
(4.23)

In particular, for a cascade with no removal ( $c_m = 0$ ;  $m = 1, \ldots, k$ ), we have

$$\varphi_n(s) = \sum_{m=1}^h N_{m0} \exp \varepsilon_{mn} s,$$

which leads to the formula (4.5) for the distribution of concentrations in such a cascade.

Rewriting (4.23) in the form

$$\varphi_{n}\left(s\right) = \sum \operatorname{res} \times \left[ \frac{\sum\limits_{m} N_{m0} \prod\limits_{j \neq m} \left(q - \varepsilon_{jn}\right)}{\prod\limits_{m} \left(q - \varepsilon_{mn}\right) + \sum\limits_{m} c_{m} \prod\limits_{j \neq m} \left(q - \varepsilon_{jn}\right)} \exp qs \right],$$

we find that the number of poles is k, since all the poles are determined by the zeros of the denominator. The latter are found from an algebraic equation of degree k:

$$1 + \sum_{m=1}^{k} \frac{c_m}{q - \varepsilon_{mn}} = 0. \tag{4.24}$$

Since the components are different, the roots of Eq. (4.24) are simple roots in the general case. If the mixture contains two components with very similar properties, for which  $|\varepsilon_{mn}| \sim \varepsilon^{\beta}$ , where B > 1, we may assume that this mixture consists of one component whose concentration is  $N_m(s) + N_n(s)$ . An analysis of the roots for the case k = 2 shows that they are always real. It can be shown that the roots of Eq. (4.24) are real for any number of components.

If we denote the roots of (4.24) by  $q_{nj}$ , we can write

$$\varphi_n(s) = -\sum_{j=1}^k \left\{ \sum_{m=1}^k \frac{N_{m0}}{q_{nj} - \varepsilon_{mn}} \times \left[ \sum_{m=1}^k \frac{c_m}{(q_{nj} - \varepsilon_{mn})^2} \right]^{-1} \right\} \exp q_{nj} s. \tag{4.25}$$

Now we introduce the index of the section B, and write (4.25) in the form

$$\varphi_n(s) = \sum_{j=1}^k a_{nj}^b \exp q_{nj}^b s, \tag{4.26}$$

where  $a_{nj}^b$  denotes the quantity in curly brackets in (4.25). Multiplying (4.26) by exp  $\epsilon_{nm}$ s and taking account of (4.15), we find that  $a_{nj}^b$  is independent of n and

$$q_{nj}^b = q_{mj}^b + \varepsilon_{mn}. \tag{4.27}$$

The last formula may also be obtained easily from Eq. (4.24). Thus,

$$\varphi_n^b(s) = \sum_{j=1}^h a_j^b \exp q_{nj}^b s. \tag{4.28}$$

Here

$$a_{j}^{b} = -\sum_{m=1}^{h} \frac{N_{m0}^{b}}{q_{mj}^{b}} \left[ \sum_{m=1}^{h} \frac{c_{m}^{b}}{(q_{mj}^{b})^{2}} \right]^{-1}, \tag{4.29}$$

where  $N_{m0}^{b}$  is the concentration of component m at the beginning of section b. We substitute (4.28) into (4.12). Taking account of (4.6) and (4.18), after integrating, we obtain

$$N_m(s) = \frac{\left(N_{m0}^b + c_m^b \sum_{j=1}^k \frac{a_j^b}{q_{mj}^b}\right) - c_m^b \sum_{j=1}^k \frac{a_j^b}{q_{mj}^b} \exp q_{mj}^b s}{\sum_{j=1}^k a_j^b \exp q_{mj}^b s} \qquad \sum_{j=1}^k a_j^b \exp q_{mj}^b s$$
 (4.30)

Now let us consider Eq. (4.24). For a cascade with no removal this equation is homogeneous and has a zero root  $q_{nn} = 0$ . In the general case  $c_n^b \neq 0$  for every root, since the cascade does not yield an infinite degree of separation. Consequently, Eq. (4.24), which is inhomogeneous in the general case, does not have any zero roots, and formula (4.28) is the sum of a finite number of exponential terms. Since a finite number of exponents do not form a complete system of functions, it follows from the integral Eq. (4.17), written in the form

$$\varphi_{n}^{b}\left(s\right)=\sum_{m=1}^{h}\left(N_{m0}^{b}-c_{m}^{b}\int_{0}^{s}dt\times\ \varphi_{n}^{b}\left(t\right)\exp\ \epsilon_{nm}t\right)\exp\epsilon_{nm}s,$$

that the value of the primitive function at the lower limit of integration s = 0 for all m = 1, . . . , k is equal to  $-N_{m0}^b/c_m^b$ . Carrying out the integration, we obtain

$$c_m^b \sum_{j=1}^k \frac{a_j^b}{q_{mj}^b} + N_{m0}^b = 0; (4.31)$$

$$N_m(s) = -c_m^b \sum_{j=1}^k \frac{a_j^b}{q_{mj}^b} \exp q_{mj}^b s \times \left( \sum_{j=1}^k a_j^b \exp q_{mj}^b s \right)^{-1}.$$
 (4.32)

Setting s = 0 in (4:32) and taking account of (4.31), we find

$$\sum_{j=1}^{h} a_j^b = 1. {(4.33)}$$

As can be seen from (4.29),  $a_j^b = a_j$  ( $c_1^b, \ldots, c_k^b$ ). It is readily shown that

$$\lim a_j^b = N_{j0}^b,$$

$$(c_1^b, \ldots, c_h^b) \longrightarrow 0.$$

$$(4.34)$$

## 5. Calculation of Rectangular Stepped Cascades

Let us carry out the calculation for a cascade with the given parameters  $\varepsilon_{mn}$ ,  $L^b$ ,  $P^b$ ,  $F^b$ ,  $\sigma^b$ , B,  $N_m(\sigma^b)$ . Consider the results shown in section 4. We assume that the quantities  $c_m^b$  are known. Using the algebraic Eq. (4.24) and the relations (4.27), we find all the roots  $q_{mn}^b$ . From the system of Eqs. (4.32) with  $s = \sigma^b$ , taking account of Eq. (4.33), we find the coefficient  $a_j^b$ . Then, using the relations (4.31), we find  $N_{m0}^b$  and, consequently,  $c_m^{b=1}$ . It is therefore desirable to use the following method for the calculation of the cascade.

Let the concentrations at one end of the cascade be specified in an arbitrary manner. By the method indicated, we find the concentrations at all the points of the cascade at which material is removed. Varying the concentrations at the end of the cascade, we can see to it that the system satisfies the conditions

$$\sum_{b=1}^{B} P^{b} N_{m} \left( \sigma^{b} \right) = \sum_{b=1}^{B} F^{b} \widetilde{N}_{m} \left( \sigma^{b} \right), \tag{5.1}$$

which correspond to the law of conservation of matter for the cascade as a whole. In the calculation of specific problems of isotope separation it has been found that the number of such intervals required for obtaining a result with satisfactory accuracy is proportional to k-1. This becomes almost obvious if we write the system (4.3) in the form:

$$L^{b}(s) \frac{d \ln N_{m}(s)}{ds} = L^{b}(s) \sum_{n=1}^{l} \varepsilon_{mn} N_{n}(s) - \frac{g_{m}^{b}}{N_{m}(s)} + \sum_{n=1}^{k} g_{n}^{b}.$$

Furthermore, in practical calculations we must first of all use a correct numbering system for the roots of Eq. (4.24). Let us write

$$1 + \sum_{m=1}^{k} \frac{c_m t}{q - \varepsilon_{mn}} = 0, \tag{5.2}$$

where t varies from 0 to 1. When t=0, we have  $q_{nrn}=\varepsilon_{mn}$ . For small values of t ( $c_m t << \varepsilon$ ;  $m=1,\ldots,k$ ), by expanding into a MacLaurin's series, we find  $q_{mn}\approx\varepsilon_{mn}-c_m t$  (there are no zero roots). From this it follows that as t increases, the signs of the roots do not change and the order in which the roots are arranged remains the same, since otherwise at least one of the elements of the  $q_{mn}$  matrix would have to pass through 0. Thus,

$$\frac{q_{ni} - q_{nj}}{\varepsilon_{ij}} > 0 \tag{5.3}$$

Taking account of Eq. (4.27), we can write formulas (4.32), as

$$N_{m}(s) = -c_{m}^{b} \sum_{i} \frac{a_{j}^{b}}{q_{mj}^{b}} \exp q_{nj}^{b} \times \left(\sum_{i} a_{j}^{b} \exp q_{nj}^{b} s\right)^{-1}.$$
 (5.4)

We introduce

$$x_{j}^{b}(s) = a_{j}^{b} \exp q_{nj}^{b} s.$$
 (5.5)

As a result, (5.4) becomes

$$N_{m}(s) = -c_{m}^{b} \sum_{i} \frac{x_{j}^{b}(s)}{q_{mj}^{b}} \left(\sum_{i} x_{j}^{b}(s)\right)^{-1}.$$
 (5.6)

If we express all the values  $x_j^b(\sigma^b)$  by one of them [the system (5.6) is homogeneous], by using (5.5) and (5.33), we obtain the coefficients  $a_j^b$ . The value of  $N_{m0}^b$  is determined from the formulas (4.31). After satisfying the conditions (5.1), we find  $N_m(s)$  from (5.6).

In conclusion, it should be mentioned that this method was used on the Ural-1 electronic computer for calculating a cascade for the separation of oxygen isotopes by low-temperature distillation of nitric oxide.

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