



ON THE VALUE FUNCTION FOR MULTI-ISOTOPE MIXTURES SEPARATION

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ABSTRACT

One of the possible approaches to the introduction of the value function in the case of multi-isotope mixtures is presented. It is based on a simplified model of separation processes in cascades and elements with multiplication of elementary effects of separation. A differential equation, being a necessary condition of the determination of the value function, is derived from the analysis of this model. Its general solution contains arbitrary constants which should be obtained considering boundary conditions and specific features of the separation process. Additional conditions concerning use of the found solutions in cascade criteria of an estimation of separation efficiency are stipulated. The applicability of various kinds of the value function forming at different conditions is analyzed. The forms of the value function which reflect best the optimal cascade with assigned external concentrations of the desired isotope and the matched abundance ratio cascade (MARC) are established.

Keywords: Isotope separation; Value function; Multi-isotope mixtures

INTRODUCTION

K. Cohen, D. Dirac and R. Peierls developed the principles of the value function theory for a binary isotope mixture [1, 2]. Their theoretical provisions allowed determining the value function for the main instances of

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binary separation. However, their application to a multicomponent isotope mixture leads to an inconsistency of the obtained results and initial assumptions [3].

Attempts were made to negotiate the contradictions by changing Dirac's axioms [3-6] and constructing the value function and the separation power on the basis of a matched abundance ratio cascade (MARC) [7, 8]. All of them restrict, somehow or other, the use of the obtained results in practice. For example, the approach changing Dirac's axioms preassigns certain properties to the value function and the separation power, which may not be realized in a specific separation process. Oppositely, the approach of MARC reduces the problem of determining this function to particular case. This study develops one of the possible axiomatic approaches which was used for substantiation of the value function of a binary isotope mixture [9] and a multi-isotope mixture [10]. This approach makes it possible to derive the most general expression for the value function. It includes arbitrary constants depending on specific features of the separation process.

PROBLEM STATEMENT

The isotopes mixture value is characterized by the function U , which is the product of the mixture quantity and the value function V . The value function depends on the isotope component concentrations C_1, C_2, \dots, C_m , which imply molar or mass fractions. We shall assume, to make certain, that the components are numbered in the increasing order of their molecular masses $M_1 < M_2 < \dots < M_m$. The sum of the concentrations equals unity, i.e.

$$\sum_{i=1}^m C_i = 1. \text{ Therefore, } m-1 \text{ concentrations are independent.}$$

Let us label the concentration vector as $\mathbf{C} = (C_1, C_2, \dots, C_m)$. Assume that the variation (increment) rate of the mixture value in any separation device (an element, a stage, a cascade, etc.) can be written in the form

$$\Delta U = G^+V(\mathbf{C}^+) + G^-V(\mathbf{C}^-) - G_0V(\mathbf{C}_0) \quad (1)$$

where G^+ , G^- , and G_0 are the product, waste and feed flows respectively (see Fig.1.); \mathbf{C}^+ , \mathbf{C}^- , and \mathbf{C}_0 are the corresponding concentration vectors. The ΔU value determines the separation power of the device, i.e. the work done by the device per unit of time.

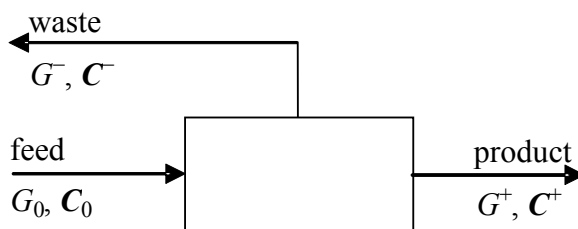


Fig. 1. Diagram of a separation device.

Considering the matter balance equation $G_0 = G^+ + G^-$, the expression (1) can be transformed to $\Delta U = G^+ [V(C^+) - V(C_0)] + G^- [V(C^-) - V(C_0)]$ and, assuming the presence of continuous derivatives for $V(C)$, to the relationship

$$\Delta U = G^+ \int_{C_0}^{C^+} dV + G^- \int_{C_0}^{C^-} dV. \tag{2}$$

Here $dV = \sum_{i=1}^m \frac{\partial V}{\partial C_i} dC_i$ is the total differential of the function $V(C)$, while

integration is taken along the curvilinear path. The expression (2) can be interpreted as follows.

The integrals in (2) are curvilinear integrals of the second type. These integrals are used for calculation of the work (mechanical, thermodynamic, etc.) done to convert a physical object from one state to another [11]. In this model the conversion process is assumed to be continuous with an infinitely small change of the state parameters. In the case of a separator, this model provides a simplified description of real processes, which are characterized by a one-parametric change of the concentrations. This representation corresponds most to linear cascades and elements with analogous multiplication of elementary effects of separation (for example, a gas centrifuge). Since the integration operation is additive, the work and, consequently, the ΔU value in (2) is calculated without losses irrespective of the form of the integrands. It means that the work is exactly equal to the sum of a large number of all single made works. If the integrands are total differentials as in (2), the work value is independent of the curvilinear path, but is determined by the initial and final states of the object.

In this paper, the problem of finding the explicit value function which is not connected with properties of the concrete separator is reduced to the introduction of a standard unit of the separation power. This unit should not be related to the flow measurement units. Since ΔU is the sum of a large number of small changes in flow concentrations G^+ and G^- , an infinitely small δU value can be reasonably taken as the standard. Let it correspond to an elementary effect of separation (EES) of a small feed flow $\delta \tilde{g}$. Assume that EES is characterized by the total enrichment factors $\varepsilon_i \ll 1$ and $i = \overline{1, m}$ at each point of the curvilinear path determined by the vector C . The ε_i values will be calculated relative to the m -th base component on the assumption of their concentration independence. In this case, δU can be written as (1) with C^+ , C^- , and C_0 replaced by C' , C'' , and C . Expanding the obtained expression as a Taylor series about C and limiting ourselves to terms of the second order of smallness, we have, by analogy to relationships

for element of small separation effects (see, e.g., [12, 13]), the following relationship:

$$\delta U = 0.5 \delta g \theta (1 - \theta) \sum_{i,j=1}^m \frac{\partial^2 V}{\partial C_i \partial C_j} (C_i' - C_i'') (C_j' - C_j'') \quad (3)$$

where θ is the cut of the flow δg ; $(C_i' - C_i'')$ and $(C_j' - C_j'')$ stand for the total enrichment of the i -th and j -th components calculated from the formulas [3]

$$C_i' - C_i'' = C_i \left(\varepsilon_i - \sum_{n=1}^m \varepsilon_n C_n \right), \quad i = \overline{1, m} \quad (4)$$

The estimation of ΔU in the EES units is equivalent to calculation of the corresponding number of elementary effects of separation N . As implied by the model with respect to a cascade, N is a minimal required number of separation elements having infinitely small effects of separation. Mathematically, the N value is found by integration of analogous differential values of $n(t)$ over curvilinear path which are assigned by parametric equations $C_i = C_i(t)$, $i = \overline{1, m}$. The $n(t)$ values are determined from the formula $n(t) = \rho_{\Delta U}(t) / \delta U$ assuming the absence of the separation work loss. The function $\rho_{\Delta U}(t)$ is the density of the ΔU distribution over points of the curvilinear path corresponding to (2) reduced to the form

$$\Delta U = \int_{t_0}^{t_1} \rho_{\Delta U}(t) dt + \int_{t_0}^{t_2} \rho_{\Delta U}(t) dt, \quad \text{where } t_0, t_1, \text{ and } t_2 \text{ are values of the}$$

parameter t at the points C_0 , C^+ , and C^- . Since δU contains effects of the second order of enrichment smallness, $\rho_{\Delta U}(t)$ should be expressed through (2) as the second derivatives of the value function $V(C)$. To this end, the expression (2) is rearranged using the formula of integration by parts.

The analysis of the derived expressions shows that the density $n(t)$ for a multicomponent mixture ($m \geq 3$) depends, as distinct from the binary separation (see [9]), on the choice of the value function. If the function $V(C)$ is chosen arbitrarily, N depends on the integration path, i.e. on intermediate stages of the mixture value variation. The N value is invariant, and ΔU is defined unambiguously in EES units by the initial and final concentrations if the $n(t)$ differs from $\rho_{\Delta U}(t)$ by a constant factor. This is the case only when δU is independent of the concentration, i.e.

$$\delta U = \text{const} \quad (5)$$

The condition (5) limits the choice of the value function $V(C)$. The relationship (5) is written for a separator with an arbitrary separation effect. Its physical essence reduces to the necessary requirement of a unique determination of the separation power of any element. In the case of infinitely small effect of an element, it is analogous to the differential equation considered by Smorodinsky [12]. It should be noted also that the solution of the equation (5) for a binary mixture and small separation effects

leads to Dirac's value function $V(C) = (2C - 1) \ln [C/(1 - C)]$ (C being the concentration of a light isotope).

The fulfillment of the condition (5) means that the separation power of a separator is proportional to the number of elementary separation effects performed, i.e.

$$\Delta U = G^+V(C^+) + G^-V(C^-) - G_0V(C_0) = N\delta U \quad (6)$$

The formula (6) represents an idealized relationship. It can be used to estimate the lower limit of the number of separation elements in a multicomponent cascade with assigned external parameters.

SOLUTION OF THE PROBLEM

The constant in (5) can be chosen differently. If it is taken equal to the factor $0.5 \delta g \theta (1 - \theta)$ before the summation signs in (3), the differential equation

$$\sum_{i,j=1}^m \frac{\partial^2 V}{\partial C_i \partial C_j} C_i C_j \left(\varepsilon_i - \sum_{n=1}^m \varepsilon_n C_n \right) \left(\varepsilon_j - \sum_{n=1}^m \varepsilon_n C_n \right) = 1 \quad (7)$$

should be solved to determine $V(C)$.

The solution of (7) depends on boundary conditions. When the mixture value is assumed as infinitely large after removal of a component, boundary conditions are written in the form [13]:

$$V(0, C_2, \dots, C_m) = V(C_1, 0, C_3, \dots, C_m) = \dots = V(C_1, C_2, \dots, C_{m-1}, 0) = \infty. \quad (8)$$

These conditions are met by Smorodinsky's solution [12]

$$V(C) = \sum_{i,j=1}^m A_{ij} (C_i - C_j) \ln C_i \quad \text{at} \quad A_{ij} = 1 / \left[(m-1)(\varepsilon_i - \varepsilon_j)^2 \right], \quad i \neq j.$$

In the case of small separation effects, $\varepsilon_i = \varepsilon_0 (M_m - M_i) = \varepsilon_0 \Delta M_{mi}$ and $\varepsilon_j = \varepsilon_0 \Delta M_{mj}$, where ε_0 is the enrichment factor per unit of the mass difference. Therefore, the value function was multiplied by ε_1^2 :

$$V(C) = \frac{1}{m-1} \sum_{\substack{i,j=1 \\ i < j}}^m (\Delta M_{mi} / \Delta M_{ji})^2 (C_i - C_j) \ln [C_i / C_j] \quad (9)$$

The formula (9) corresponds to Dirac's value function for binary mixtures and the substitution of the constant in (5) for $\delta U = 0.5 \delta g \theta (1 - \theta) \varepsilon_1^2$. One more form of Smorodinsky's value function [13] is found analogously. It is derived by substituting the factor ε_0^2 for ε_1^2 . Notice that the value function may be supplemented with linear and power functions of component concentrations which represent solutions of the corresponding homogeneous differential equation (7) [7, 13]. However, they are not necessarily taken into account if the separation power is concerned.

One more form of the value function is known in addition to Smorodinsky's value function. It was obtained by De La Garza for a three-component mixture [7]. A specific feature of the solution is that it corresponds to the matched abundance ratio cascade (MARC). The determined value function need to be verified to apply for other cascades.

The analysis shows that Smorodinsky's value function is a particular case of the general solution which can be found from the consideration of the function $V_{kl} = a_k C_k \ln [C_k / C_l]$ at $k \neq l$ and $k, l = \overline{1, m}$. The second concentration derivatives V_{kl} are $\partial^2 V_{kl} / \partial C_k^2 = a_k / C_k$, $\partial^2 V_{kl} / \partial C_l^2 = a_k C_k / C_l^2$, $\partial^2 V_{kl} / \partial C_k \partial C_l = -a_k / C_l$ and $\partial^2 V_{kl} / \partial C_i \partial C_j = 0$ at $i, j \neq k, l$. Hence, substituting V_{kl} as $V(\mathbf{C})$ to the left-hand side of the equation (7), we have

$$\sum_{i,j=1}^m \frac{\partial^2 V_{kl}}{\partial C_i \partial C_j} C_i C_j \left(\varepsilon_i - \sum_{n=1}^m \varepsilon_n C_n \right) \left(\varepsilon_j - \sum_{n=1}^m \varepsilon_n C_n \right) = a_k (\varepsilon_k - \varepsilon_l)^2 C_k.$$

Therefore, if $a_k = 1/(\varepsilon_k - \varepsilon_l)^2$ is chosen, this expression is equal to C_k . Significantly, this result is independent of the l -th component at hand.

Therefore, any linear combination $V_k = \sum_{l=1}^m b_{kl} V_{kl}$ at $\sum_{l=1}^m b_{kl} = 1$ and $l \neq k$ gives an analogous C_k value. If all functions V_k are summed up, i.e. the value function is taken as $V(\mathbf{C}) = \sum_{\substack{k,l=1 \\ k \neq l}}^m b_{kl} \left[1/(\varepsilon_k - \varepsilon_l)^2 \right] C_k \ln [C_k / C_l]$, the

equation (7) is identically satisfied:

$$\sum_{i,j=1}^m \frac{\partial^2 V}{\partial C_i \partial C_j} C_i C_j \left(\varepsilon_i - \sum_{n=1}^m \varepsilon_n C_n \right) \left(\varepsilon_j - \sum_{n=1}^m \varepsilon_n C_n \right) = \sum_{k=1}^m C_k \equiv 1.$$

This value function may be written in the form analogous to (9):

$$V(\mathbf{C}) = \sum_{\substack{i,j=1 \\ i < j}}^m \left(\Delta M_{m1} / \Delta M_{ji} \right)^2 \left(b_{ij} C_i - b_{ji} C_j \right) \ln [C_i / C_j] \quad (10)$$

The coefficients b_{ij} should be chosen considering assigned boundary conditions and the requirement $\sum_{j=1}^m b_{ij} = 1$ at $i \neq j$. It is easy to see that the

same "weight" $b_{ij} = 1/(m-1)$ of all pairs of components leads to Smorodinsky's formula (9) corresponding to the conditions (8). The value function (10) at arbitrary $b_{ij} \neq 0$ satisfies these conditions, too. This is an indication that the boundary conditions (8) are not exhaustive. Specific features of the separation process at hand should be itemized for the proper selection of the constants b_{ij} .

One of the most frequent practical problems is separation of two main components r and s (having extremely opposite molecular masses as a rule) at small concentrations of other isotopes. In this case, most of the separation work is done for these components. Therefore, it is reasonable not to assume the mixture value in the absence of other isotopes as infinite, but to find it from the binary value function $V_0(C_r, C_s)$ of the r -th and s -th components, i.e.

$$V(\mathbf{C}) = V_0(C_r, C_s) = (\Delta M_{m1} / \Delta M_{sr})^2 (C_r - C_s) \ln [C_r / C_s] \quad (11)$$

at $C_i = 0$, $i = 1, m$, and $i \neq r, s$.

It is easy to see that (11) is fulfilled if $b_{ri} = b_{si} = 0$ is taken in (10) for all $i \neq r, s$. Hence, $b_{rs} = b_{sr} = 1$ and the terms with the indices r and s are equal to $V_0(C_r, C_s)$. Moreover, finiteness of the value function when concentrations of individual minor components turn to zero requires that $b_{ij} = 0$ at $i, j \neq r, s$. Therefore, the value function can be written in the form

$$V(\mathbf{C}) = V_0(C_r, C_s) + \sum_{\substack{i=1 \\ i \neq r, s}}^m (V_{ir}^* + V_{is}^*) \quad (12)$$

where $V_{ir}^* = (\Delta M_{m1} / \Delta M_{ri})^2 b_{ir} C_i \ln [C_i / C_r]$ and V_{is}^* is an analogous expression, in which the index r is replaced by s .

The second term in (12) is negative. It decreases the mixture value as compared to the binary estimation depending on the presence and the combination of minor components with r -th and s -th isotopes. Therefore, b_{ir} and b_{is} ($b_{ir} + b_{is} = 1$) may be chosen considering the priority of the loss of the mixture value. For example, if the separation work done for the r -th isotope is assumed to be most important, one can take $b_{ir} = 1$ and $b_{is} = 0$. If the r -th and s -th components are equally significant, then $b_{ir} = b_{is} = 0.5$.

An analogous approach may be used in more complicated cases. A large number of choice options for the constants b_{ij} and b_{ji} in (10) suggests a host of idealized separation processes without the loss of the separation work (according to (6)). It is necessary to note that the value function of MARC is received if the coefficients are equal: $b_{rs} = b_{sr} = 1$, $b_{ri} = b_{si} = b_{ij} = 0$ for all $i, j \neq r, s$ and $b_{ir} = -(\Delta M_{ir})^2 / a_i$, $b_{is} = (\Delta M_{is})^2 / a_i$, where $a_i = \Delta M_{sr} (M_r + M_s - 2M_i) \neq 0$. In this case, the value function is written in the form

$$V(\mathbf{C}) = \Delta M_{m1}^2 \sum_{i=1}^m (1/a_i) C_i \ln \left(\frac{C_r}{C_s} \right) \quad (13)$$

It corresponds to the expression founded by De La Garza for a three – component mixture of isotopes in the case when the key components r and s

are neighboring by molecular weight, i.e. $s = r + 1$. The general efficiency criterion of such cascades was received in [8]. In this case, the value function satisfies boundary condition (9) and equalities of the abundance ratio between r -th and s -th components $R = C_r / C_s$ for the feed flows of the each stage of MARC.

ANALYSIS OF SOLUTIONS

Specific features of the obtained value functions were analyzed by calculating the separation power δU_e of separating elements at different concentrations of the feed flow C_0 . The δU_e value was calculated from the formula (1) considering assigned total separation factors q_i , $i = \overline{1, m}$ (determined from the relation to the m -th isotope) and the cut $\theta = G^+ / G_0$. The concentrations C^+ and C^- were calculated using known relationships of the theory of separating elements [3]. It was found that if separation effects were small and characterized by $q_i \leq 3$ and $\theta = 0.5$, the separation power weakly depended on C_0 . The δU_e value changed by ~ 1 -2% for different forms of the value function. The variation was much larger at $\theta \neq 0.5$. If $\theta = 0.3$ or $\theta = 0.7$, the difference of the δU_e values was as large as 20% at different C_0 and $q_1 = 3$.

This behavior of the dependence of δU_e on the mixture composition is analogous to the one obtained for a binary mixture of isotopes (see [9]). However, the invariance of θ at different concentrations of the feed flow does not reflect optimal operating conditions of elements in a cascade. In this connection, calculation was made for counter-flow symmetric cascades with assigned external concentrations of the desired isotope. Calculation was performed by the method [14] using the criterion of the minimum of the total feed flow. It was found that when C_0 and θ changed considerably in stages of an optimal cascade, the specific separation power of stages $\Delta U_{st} / G_0$ changed insignificantly.

By way of example, the Table 1 gives $\Delta U_{st} / G_0$ in stages of a silicon cascade [10]. The initial mixture of silicon isotopes corresponded to the working gas SiF_4 having the molecular masses of 104, 105, and 106, and the concentrations of 92.21%, 4.70%, and 3.09%. The desired isotope concentration of the product flow was assigned at 99.99% and that of the waste flow at 0.01%. The product flow of the cascade was taken as 1 g/s and the separation factors of the stages were $q_1 = 3$ and $q_2 = \sqrt{3}$. The optimization procedure gave an optimal cascade comprising 62 stages with the feed flow fed to the 40th stage. At considerable changes ΔU_{st} and G_0 of

the stages, the $\Delta U_{st} / G_0$ value, which was determined from (1) and the value function (12) at $r = 1$, changed little and was equal to $\sim 0.147-0.150$.

Table 1. Separation efficiency of a silicon cascade

Value function		Stages		Cascade		
Formula	Features	$\Delta U_{st}/G_0$	$\eta_{st}, \%^{*)}$	$\mu, \%$	$\eta, \%$	
(12)	$r=1, s=2$	$b_{31}=1$	0.148-0.150	96.1-100.0	99.4	99.6
		$b_{31}=0.5$	0.147-0.150	89.8-100.0	98.7	98.9
		$b_{31}=0$	0.147-0.150	83.4-100.0	98.0	98.2
	$r=1, s=3$	$b_{21}=1$	0.148-0.149	1.8-99.4	77.6	78.5
	$r=2, s=3$	$b_{12}=1$	0.142-0.150	3.5-100.0	89.5	90.0
Smorodinsky's formula (9)		0.145-0.149	37.7-99.1	83.5	84.3	

*) Without extreme stages.

One of the indicators of the stage performance is the coefficient of efficiency $\eta_{st} = 1 - \Delta U_{mix} / \Delta U_{st}$ [12]. It takes into account the loss of the separation power ΔU_{mix} caused by mixing of feed flows with different concentrations. The loss ΔU_{mix} is equal numerically to the separation power, which is required to separate a total feed flow G_0 with concentrations C_0 into individual feed flows: $\Delta U_{mix} = \sum_i G_{0i} [V(C_{0i}) - V(C_0)]$. In this formula, G_{0i} and C_{0i} denote the flow and the concentration of the i -th feed flow while the sum is taken over all flows fed to the stage. From the Table, it is seen that η_{st} of the stages was high, $\eta_{st} \approx 96-100\%$, for the value function (12) at $r = 1, s = 2$ and $b_{31} = 1$: $V(C) = (\Delta M_{31} / \Delta M_{21})^2 (C_1 - C_2) \ln [C_1 / C_2] + C_3 \ln [C_3 / C_1]$. This form of the value function is intended to estimate the mixture value with respect to the first two components with the priority of the desired light isotope. In terms of other value functions which do not distinguish this isotope, the aforementioned optimal cascade is less efficient.

This difference of the value functions shows up at the level of cascade parameters, too. By way of example, the Table gives estimates of coefficients η and μ of the cascade. These coefficients were calculated from the formulas: $\eta = \Delta U_{casc} / \Delta U_{tot}$, $\mu = \sum G_{cal} / \sum G$. Here, ΔU_{casc} is the separation power of the cascade with respect to external flows and concentrations (according to (1)), and ΔU_{tot} is the total separation power of the stages. Accordingly, $\sum G_{cal}$ is the calculated total feed flow of the stages, and $\sum G$ is the actual one. To determine $\sum G_{cal}$, the value ΔU_{casc} was divided

by the maximum specific separation power of a stage $(\Delta U_{st}/G_0)_{max}$. This quantity was taken equal to 0.150 for all the value functions. The best cascade parameters were calculated from the value function (13) at $r = 1$, $s = 2$ and $b_{31} = 1$: $\eta = 99.6\%$ and $\mu = 99.4\%$. The corresponding values were $\eta \approx 78.5-98.9\%$ and $\mu \approx 77.6-98.7\%$ for the other value functions considered above.

These data show that a large difference of the calculated and actual values of the total flow in an optimal cascade is due to low coefficient of efficiency. This is because each of the value functions is "adjusted" to its main components and any deviation of the process from the "ideal" is evaluated as a large loss of the mixture value. This loss occurs in a cascade mostly when flows with different concentrations are mixed.

This example and other calculation results show that the performance of stages and cascades should be estimated from the value function (12) with the appropriate choice of main components and the coefficients b_{ij} . If the value function is selected properly, the criteria of an optimal cascade are the large values of $\Delta U_{st}/G_0$, η and μ . It should be noted that unlike binary mixtures, external concentrations of undesired components cannot be assigned arbitrarily for a cascade intended for separation of a multicomponent mixture. They can be changed within a certain interval corresponding to the permissible variation of the number of stages in the depletion and enrichment sections of a cascade. Therefore, the separation power of a cascade and its other characteristics can be estimated mainly after relevant calculation has been made for the cascade.

CONCLUSION

The general equation (10) for the value function contains $m(m - 2)$ arbitrary constants whose values can be chosen differently. This fact suggests a multitude of idealized processes of multicomponent separation which are not accompanied by the loss of the separation work. From the standpoint of practical applications, it is reasonable to perform further studies and establish the correspondence between different forms of the value function (10) and solutions of the equation (7), and particular efficiency functions of the cascade optimization.

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