

# ANALYTICAL DETERMINATION OF THE NUMBER OF THEORETICAL STAGES IN BINARY RECTIFICATION AND COUNTERCURRENT EXTRACTION FOR NON-LINEAR OPERATING LINES

By

P. RÓZSA and Gy. SÁRKÁNY

Department of Civil Engineering Mathematics, Technical University Budapest

Department of Chemical Unit Operations, Technical University, Budapest

Presented by Prof. K. Tettamanti

Received April 28, 1972

## Introduction

Matrix calculus is known to lend itself for mathematical investigation into systems of discrete units, if the empirical functions describing the behaviour of the systems are linear or linearizable, as shown first by ACRIVOS and AMUNDSON [1]. This method was applied in previous papers of the authors [2, 3] for the determination of the number of theoretical stages of multistage countercurrent separation systems frequently used in the chemical industry.

The basic idea in [2, 3] was that the diagrams of rectification, i.e. enthalpy *vs.* concentration, and that of the solvent content *vs.* solute of countercurrent extraction on a solvent-free basis are similar in structure and may be treated in a similar way, was adapted from the papers of RANDALL and LONGTIN [4].

In the first paper of the authors explicit formulae for the analytical calculation of the number of theoretical stages with respect to the so-called "general" extractor (with two feed solutions) defined in [2] (Fig. 1) were presented for the case where — in addition to the assumption of linearity of the operating line — the equilibrium curve too was approximated by a straight line, or better by a straight sectioned chord polygon for suitably chosen concentration ranges, as a further development of the approximation first used by THORMAN [5] and KREMSER, BROWN and SANDERS [6, 7]. The results for the case of the chord polygon approximation of the equilibrium curve were identical with the formulae and with the sums cited by many authors. Decreasing the distance between the vertex points the chord polygon turns into the equilibrium curve and thus the following formula gives the number of theoretical stages in the concentration range of the double feed countercurrent extractor:

$$N_x = \int_{x_0}^x \frac{f'(x) - g'(x)}{\ln \frac{f'(x)}{g'(x)}} \cdot \frac{1}{f(x) - g(x)} dx.$$

If the operating line  $g(x)$  is linear, this formula is valid for any equilibrium curve  $f(x)$  that may be considered linear within one stage. The above integral is a generalization of the formula given by LEWIS\* [8], valid for a linear operating line and for a parallel linear equilibrium line [9].

In [3] a generalization of [2] is given for the case where the equilibrium curve is approximated by a chord polygon as well, a non-linear operating

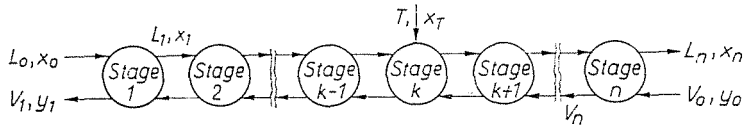


Fig. 1. Diagram of the double feed "general extractor" (concentrations on a solvent-free basis)

line is, however, allowed. The latter was taken into consideration approximating the diagrams of enthalpy *vs.* concentration in rectification, or the solvent content on a solvent-free basis *vs.* concentration in countercurrent extraction by chord polygon.

The idea of approximating the diagrams mentioned above by straight lines to facilitate calculations originates from KIRSCHBAUM who supposed that the equation of the curved operating lines can be written using the difference of the heats of evaporation of the two components of binary mixtures if the heats of solution and the sensible heat can be neglected. Also the calculation of the operating line by BILLET, reported by KIRSCHBAUM [10], is based on this idea. For further details cf. [11].

The assumption of KIRSCHBAUM can be demonstrated to be identical with the idea applied in our work, namely that the enthalpy *vs.* concentration diagram can be replaced by a straight line, or by straight sections.

In [3] the heat of solution was taken into consideration by approximating the enthalpy curve by a chord polygon instead of a single straight line.

### Systems of equations for the calculation of the number of theoretical stages for the general extractor

The basic model chosen for the determination of the number of theoretical stages is the so-called "general" extractor with two feed solutions as defined e. g. by TREYBAL [12] (see Fig. 1). In the extractor the raffinate phase

\* With our symbols the integral of Lewis takes this form:

$$N_x = \int_{x_0}^x \frac{dx}{f(x) - g(x)} \cdot \frac{L}{V}$$

$A + C$  is in countercurrent contact with the extract phase  $B + C$  where  $C$  is the substance to be extracted. The common countercurrent extractor, the extractor with reflux and the rectification column are considered as special cases of the general extractor.

To determine the number of theoretical stages the phase equilibrium curve and the curves defining the phase amounts are given in co-ordinates introduced by JAENECKE and PONCHON—SAVARIT by means of an interpolat-

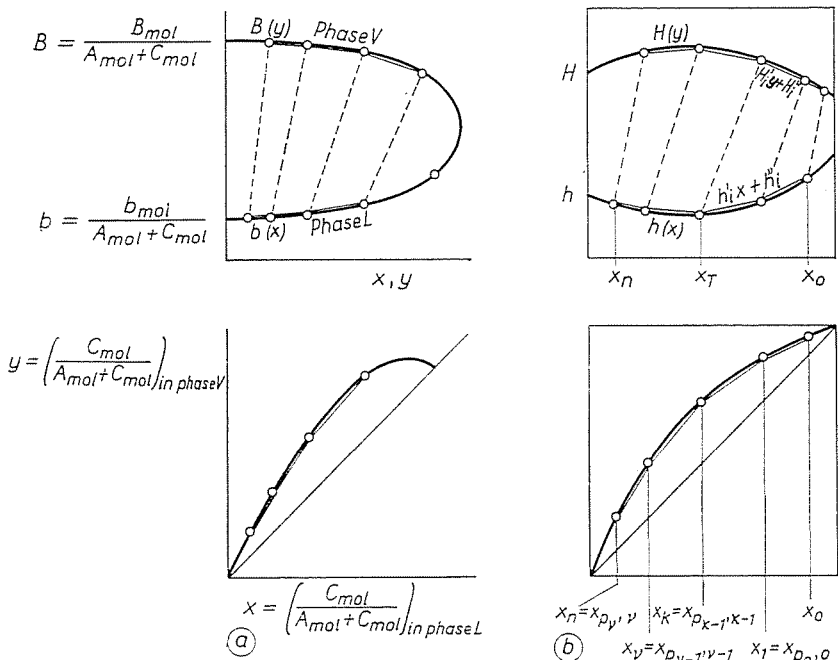


Fig. 2a and 2b. Approximation of the smoothed-out curves by chord polygons

ing formula describing the smoothed out curve passing through the experimental points and these are approximated in the  $(x_0, x_n)$  range by a chord polygon of  $(\nu + 1)$  linear sections connecting the points of abscissae  $x_{0i}$  ( $i = 1, 2, \dots, \nu$ ) chosen adequate close to each other (Fig. 2 a, b).

Hence  $x_{00} \equiv x_0$  and  $x_{0,\nu+1} \equiv x_n$ .

Let

$$y = K_i x + K'_i \quad (i = 0, 1, 2, \dots, \nu)$$

$$x_{0,i+1} \leq x \leq x_{0i} \quad \text{and} \quad K_i \geq 0,$$

be the equation of the linear sections of the phase equilibrium curve obtained in this way and

$$\begin{aligned}
 B &= B'_i y + B''_i & (i = 0, 1, 2, \dots, \nu) \\
 \text{and} \quad b &= b'_i x + b''_i & (i = 0, 1, 2, \dots, \nu) \\
 x_{0,i+1} &\leq x \leq x_{0,i}
 \end{aligned}$$

be the equations of the linear sections of the curves defining the phase ratios of extractors. The number of theoretical stages  $p_i$  ( $i = 0, 1, 2, \dots, \nu$ ) fixed by each section is to be determined. For this purpose the following parameters of the entire apparatus should be given:

$$\begin{array}{lll}
 L_0, & x_0, & b_0 \\
 T, & x_T, & b_T \\
 V_0, & y_0, & B_0 \quad (\text{or } Q'_{B,H}) \\
 L_n, & x_n &
 \end{array}$$

where  $L_n$  is not known, but may be calculated. Symbols are defined in Fig. 1.

Concentrations and material flows referring to each stage are denoted by double subscripts. The subscript pair  $p_{\kappa-1}, \kappa-1$  corresponds to the  $k$ -th stage where the feed proceeds, while the subscript pair  $p_\nu, \nu$  corresponds to the last ( $n$ -th) stage, and the concentration with the subscript pair  $p_i, i$  the last stage of each group referring to a linear section is equal to the concentration marked with zero subscript of the next group:

$$x_{p_i, i} = x_{0, i+1}; \quad y_{p_i, i} = y_{0, i+1}; \quad (i = 0, 1, 2, \dots, \nu).$$

The corresponding material-flow rates — whose values, of course, are not known in advance — are  $L_0 = L_{00}$ ;  $L_{p_i, i} = L_{0, i+1}$ ;  $V_{p_i, i} = V_{0, i+1}$  ( $i = 0, 1, 2, \dots, \nu$ ). Thus, the equations of balances for each group of stages, are as follows.

Material balance of substance C:

$$\begin{array}{lll}
 L_{10}x_{10} & + V_{10}y_{10} - V_{20}y_{20} & = L_0x_0 \\
 -L_{10}x_{10} + L_{20}x_{20} & + V_{20}y_{20} - V_{30}y_{30} & = 0 \\
 -L_{20}x_{20} + L_{30}x_{30} & + V_{30}y_{30} - V_{40}y_{40} & = 0 \\
 \vdots & \vdots & \\
 -L_{p_0-1,0}x_{p_0-1,0} + L_{p_0,0}x_{p_0,0} & + V_{p_0,0}y_{p_0,0} - V_{11}y_{11} & = 0 \\
 \vdots & \vdots & \\
 -L_{p_{\kappa-1}, \kappa-2}x_{p_{\kappa-1}, \kappa-2} + L_{1, \kappa-1}x_{1, \kappa-1} & + V_{1, \kappa-1}y_{1, \kappa-1} - V_{2, \kappa-1}y_{2, \kappa-1} & = 0 \\
 \vdots & \vdots & \\
 -L_{p_{\kappa-1}-1, \kappa-1}x_{p_{\kappa-1}-1, \kappa-1} + L_{p_{\kappa-1}, \kappa-1}x_{p_{\kappa-1}, \kappa-1} & + V_{p_{\kappa-1}, \kappa-1}y_{p_{\kappa-1}, \kappa-1} - V_{1, \kappa}y_{1, \kappa} = Tx_T \\
 \vdots & \vdots & \\
 -L_{p_{\nu-1}, \nu-1}x_{p_{\nu-1}, \nu-1} + L_{1, \nu}x_{1, \nu} & + V_{1, \nu}y_{1, \nu} - V_{2, \nu}y_{2, \nu} & = 0 \\
 \vdots & \vdots & \\
 -L_{p_{\nu-2}, \nu}x_{p_{\nu-2}, \nu} + L_{p_{\nu-1}, \nu}x_{p_{\nu-1}, \nu} & + V_{p_{\nu-1}, \nu}y_{p_{\nu-1}, \nu} - V_{p_{\nu}, \nu}y_{p_{\nu}, \nu} & = 0 \\
 -L_{p_{\nu-1}, \nu}x_{p_{\nu-1}, \nu} & + V_{p_{\nu}, \nu}y_{p_{\nu}, \nu} & = V_0y_0 - \\
 & & -L_nx_n \quad (1)
 \end{array}$$

Material balances of solvent *B*

$$\begin{aligned}
 &L_{10}(b'_0x_{10} + b''_0) + V_{10}(B'_0y_{10} + B''_0) - V_{20}(B'_0y_{20} + B''_0) = L_0(b'_0x_0 + b''_0) \\
 &- L_{10}(b'_0x_{10} + b''_0) + L_{20}(b'_0x_{20} + b''_0) + \\
 &\quad + V_{20}(B'_0y_{20} + B''_0) - V_{30}(B'_0y_{30} + B''_0) = 0 \\
 &- L_{20}(b'_0x_{20} + b''_0) + L_{30}(b'_0x_{30} + b''_0) + \\
 &\quad + V_{30}(B'_0y_{30} + B''_0) - V_{40}(B'_0y_{40} + B''_0) = 0 \\
 &\quad \vdots \\
 &- L_{p_0-1,0}(b'_0x_{p_0-1,0} + b''_0) + L_{p_00}(b'_0x_{p_00} + b''_0) + \\
 &\quad + V_{p_00}(B'_0y_{p_0} + B''_0) - V_{11}(B'_1y_{11} + B''_1) = 0 \\
 &\quad \vdots \\
 &- L_{p_{x-2},x-2}(b'_{x-2}x_{p_{x-2},x-2} + b''_{x-2}) + L_{1,x-1}(b'_{x-1}x_{1,x-1} + b''_{x-1}) + \\
 &\quad + V_{1,x-1}(B'_{x-1}y_{1,x-1} + B''_{x-1}) - V_{2,x-1}(B'_{x-1}y_{2,x-1} + B''_{x-1}) = 0 \\
 &\quad \vdots \\
 &- L_{p_{x-1},x-1}(b'_{x-1}x_{p_{x-1},x-1} + b''_{x-1}) + L_{p_{x-1},x-1}(b'_{x-1}x_{p_{x-1},x-1} + b''_{x-1}) + \\
 &\quad + V_{p_{x-1},x-1}(B'_{x-1}y_{p_{x-1},x-1} + B''_{x-1}) - V_{1,x}(B'_xy_{1,x} + B''_x) = T(b'_{x-1}x_T + b''_{x-1}) \\
 &\quad \vdots \\
 &- L_{p_{v-1},v-1}(b'_{v-1}x_{p_{v-1},v-1} + b''_{v-1}) + L_{1,v}(b'_vx_{1,v} + b''_v) + \\
 &\quad + V_{1,v}(B'_vy_{1,v} + B''_v) - V_{2,v}(B'_vy_{2,v} + B''_v) = 0 \\
 &\quad \vdots \\
 &- L_{p_{v-2},v}(b'_vx_{p_{v-2},v} + b''_v) + L_{p_{v-1},v}(b'_vx_{p_{v-1},v} + b''_v) + \\
 &\quad + V_{p_{v-1},v}(B'_vy_{p_{v-1},v} + B''_v) - V_{p_v,v}(B'_vy_{p_v,v} + B''_v) = 0 \\
 &- L_{p_{v-1},v}(b'_vx_{p_{v-1},v} + b''_v) + V_{p_v,v}(B'_vy_{p_v,v} + B''_v) = Q' - L_n(b'_vx_n + b''_v)
 \end{aligned}
 \tag{2}$$

The quantity  $Q'$  on the right-hand side of the last equation in system (2) refers — in the case of extraction — to the process producing the solvent and — in that of rectification — to the vapour phase. In the case of

a) *general extraction* —  $Q'$  is the amount of solvent required to produce the solvent phase  $V_0$  in equilibrium with the phase  $L_n$ :

$$Q' = V_0B_0,$$

b) *extraction with raffinate-reflux*, —  $Q'$  is the solvent content of the raffinate-reflux  $L_n - R$  in the solvent, i.e. extract phase; and

c) *rectification* —  $Q'$  is the heat quantity transferred to the reboiler. Hence, from the total enthalpy balance of rectification

$$Q' = D(H'_D x_D + H''_0) + M(h'_v x_M + h''_v) - Th_T + L_0[(H'_0 - h'_0)x_D + H''_0 - h''_0].$$

This has to be considered in the case of rectification where  $V_0 = 0$ .

If in the systems of equation (1) and (2), the  $l$ -th equation is replaced by the sum of the  $l$ -th,  $(l + 1)$ -th,  $\dots$ ,  $n$ -th equations ( $l = 1, 2, \dots, n$ ), and if the equations:

$$\begin{aligned} V_l &= L_{l-1} + G_l \\ G_l &= \mathcal{E}_{lk} T + V_0 - L_n, \\ \mathcal{E}_{lk} &= \begin{cases} 1, & \text{if } k - l \geq 0 \\ 0, & \text{if } k - l < 0 \end{cases} \end{aligned}$$

as well as the equations  $y = K_i x + K'_i$ ;  $B = B'_i y + B''_i$  and  $b = b'_i x + b''_i$  are substituted for (1) and (2), then — after rearrangement — the following systems of equations are obtained:

$$\left. \begin{aligned} (L_{0,i} + G_i)K_i x_{1,i} &= L_{0,i} x_{0,i} + T x_T + V_0 y_0 - L_n x_n - K'_i G_i - K'_i L_{0,i} \\ - L_{1,i} x_{1,i} + (L_{1,i} + G_i) K_i x_{2,i} &= \\ &= T x_T + V_0 y_0 - L_n x_n - K'_i G_i - K'_i L_{1,i} \\ &\vdots \\ L_{p_i-1,i} x_{p_i-1,i} + \\ + (L_{p_i-1,i} + G_i) K_i x_{p_i,i} &= T x_T + V_0 y_0 - L_n x_n - K'_i G_i - K'_i L_{p_i-1,i} \end{aligned} \right\} (3)$$

$$\left. \begin{aligned} (L_{0,i} + G_i)B'_i K_i x_{1,i} &= L_{0,i} b'_i x_{0,i} + T b_T + Q' - L_n (b'_i x_n + b''_i) - \\ &\quad - (B'_i K'_i + B''_i) G_i + (b''_i - B'_i K'_i - B''_i) L_{0,i} - \\ - L_{1,i} b'_i x_{1,i} + (L_{1,i} + G_i) B'_i K_i x_{2,i} &= \\ &= T b_T + Q' - L_n (b'_i x_n + b''_i) - (B'_i K'_i + B''_i) G_i + (b''_i - B'_i K'_i - B''_i) L_{1,i} \\ &\vdots \\ - L_{p_i-1,i} b'_i x_{p_i-1,i} + (L_{p_i-1,i} + G_i) B'_i K_i x_{p_i,i} &= \\ &= T b_T + Q' - L_n (b'_i x_n + b''_i) - (B'_i K'_i + B''_i) G_i + (b''_i - B'_i K'_i - B''_i) L_{p_i-1,i} \end{aligned} \right\} (4)$$

$i = 0, 1, 2, \dots, v.$

In (3) and (4), the values of  $x_{ji}$  and  $L_{ji}$  are unknown ( $j = 1, 2, \dots, p_i$ ) and the number of stages sought for:  $\Sigma p_i$  in the range  $(x_0, x_n)$  can be calculated as a function of  $T, x_T, b_T, L_0, x_0, V_0, y_0, x_n$ . The parameters  $K_i, K'_i, B'_i, B''_i, b'_i$  and  $b''_i$  ( $i = 0, 1, \dots, v$ ) required for the calculation can be taken from the diagram (or calculated by computer).

The calculation refers to the general extractor fed at the  $k$ -th stage (cf. Fig. 1). The physical state of the feed is now defined by  $b_T$  (in the case of extraction  $b_T$  is the content of substance  $B$  in the feed, in the case of rectification its enthalpy  $h_T$ ). This quantity determines the partition of  $T$  between the phases  $L$  and  $V$ . In [3] this partition was determined by the well-known quantity  $q$ , according to which the following relationship holds between  $q$  and  $b_T$ :

$$q = \frac{B(x_T) - b_T}{B(x_T) - b(x_T)}.$$

It should also be underlined that for the sake of simplicity it has been assumed that  $L_0$  and  $V_0$  are in a state given by the corresponding curve of the solvent and enthalpy diagram, resp., i.e. the solvent content of  $L_0$  is

$$b_0 = b'_0 x_0 + b''_0,$$

and

$$h_0 = h'_0 x_0 + h''_0 \text{ etc.}$$

### Solution of the systems of equations

The systems of equations (3) and (4) are linear in the concentrations. However, the coefficients  $L_{ji}$  of the concentrations are also unknown. The aim is to determine the number of equations in the systems. The solubility of the problem is provided by the structure of these systems, namely they are divided into  $\nu + 1$  groups where the quantities characterizing each section of the functions approximated by chord polygons, i.e.  $K_i, K'_i, B'_i, B''_i, b'_i, b''_i$ , are constant. The principle of the solution is as follows: from the systems of equations (3) and (4) written for each group of stages by eliminating the concentrations  $x_{ji}$ , a system of non-linear equations is obtained for the flow rates  $L_{ji}$ . From this system of equations, a fractional-linear recursive formula is obtained for  $L_{ji}$ . Using this formula and taking into consideration that the concentrations  $x_{0i}$  at the end of the linear sections are known, the number  $p_i$  of the equations in each group can be determined explicitly.

Dividing each equation by the coefficient of concentration  $x_{ji}$  and introducing the symbols

$$s_i = \frac{1}{G_i} (\sum_{ix} T x_T + V_0 y_0 - L_n x_n); \quad S_i = \frac{1}{G_i} (\sum_{ix} T b_T + Q' - L_n b_n), \quad (5)$$

$$(i = 0, 1, 2, \dots, \nu)$$

and the nilpotent matrix  $L_i = [l_{jk}^{(i)}]$  of order  $p_i$  in which all elements but

$$l_{j,j-1}^{(i)} = \frac{L_{j-1,i}}{(L_{j-1,i} + G_i)K_i} \quad \begin{array}{l} j = 2, 3, \dots, p_i \\ i = 0, 1, 2, \dots, \nu; \end{array} \quad (6)$$

are zero; further introducing the vectors

$$\begin{aligned} \mathbf{x}_i &= [x_{ji}]; & \mathbf{f}_i &= [f_{ji}] = \left[ \frac{1}{(L_{j-1,i} + G_i)K_i} \right]; \\ \mathbf{g}_i &= [g_{ji}] = \left[ \frac{L_{j-1,i}}{(L_{j-1,i} + G_i)K_i} \right] & \begin{array}{l} j = 1, 2, \dots, p_i \\ i = 0, 1, \dots, \nu \end{array} \end{aligned} \quad (7)$$

and the unit vector  $\mathbf{e}_1$  whose first element equals 1 and the others are zero, the following matrix equations are obtained for each group of equations in (3) and (4):

$$(\mathbf{E} - \mathbf{L}_i)\mathbf{x}_i = \frac{L_{0i}x_{0i}}{(L_{0i} + G_i)K_i} \mathbf{e}_1 + G_i(s_i - K'_i)\mathbf{f}_i - K'_i\mathbf{g}_i, \quad (8)$$

( $i = 0, 1, 2, \dots, \nu$ )

$$\begin{aligned} \left( \mathbf{E} - \frac{b'_i}{B'_i} \mathbf{L}_i \right) \mathbf{x}_i &= \frac{b'_i}{B'_i} \frac{L_{0i}x_{0i}}{(L_{0i} + G_i)K_i} \mathbf{e}_1 + \frac{G_i}{B'_i} [S_i - (B'_i K'_i + B''_i)] \mathbf{f}_i - \\ &- \left( K'_i + \frac{B''_i - b''_i}{B'_i} \right) \mathbf{g}_i \quad (i = 0, 1, 2, \dots, \nu). \end{aligned} \quad (9)$$

The unknown  $L_{ji}$  values occur — besides in the coefficient matrices of the concentrations — in the components of vectors  $\mathbf{f}_i$  and  $\mathbf{g}_i$  as well.

[Making use of the fact that the molar flow rates above and below the feed point are constant, Eq. (9) is identical with Eq. (8) because the diagrams  $B$  and  $b$  defining the solvent content of the individual phases, and the two enthalpy diagrams are two parallel straight lines, i.e.  $B'_i = b'_i$  and  $H'_i = h'_i$  respectively.]

Premultiplying Eq. (8) with matrix  $(\mathbf{E} - \mathbf{L}_i)^{-1}$ , the expression obtained for  $\mathbf{x}_i$ , and substituting it into Eq. (9) yields a single system of equations (with  $p_i$  unknowns) where the  $L_{ji}$  values are the only unknowns. Taking into consideration that  $\mathbf{L}_i$  is a nilpotent matrix and hence  $(\mathbf{E} - \mathbf{L}_i)^{-1} = \sum_{m=0}^{p_i-1} \mathbf{L}_i^m$ , it is seen that

$$\left( \mathbf{E} - \frac{b'_i}{B'_i} \mathbf{L}_i \right) (\mathbf{E} - \mathbf{L}_i)^{-1} = \frac{b'_i}{B'_i} \mathbf{E} + \frac{B'_i - b'_i}{B'_i} (\mathbf{E} - \mathbf{L}_i)^{-1}$$

and the following system of equations is obtained for the  $L_{ji}$  values:

$$\begin{aligned} \left[ \frac{b'_i}{B'_i} \mathbf{E} + \frac{B'_i - b'_i}{B'_i} (\mathbf{E} - \mathbf{L}_i)^{-1} \right] \left[ \frac{L_{0i}x_{0i}}{(L_{0i} + G_i)K_i} \mathbf{e}_1 + G_i(s_i - K'_i)\mathbf{f}_i - K'_i\mathbf{g}_i \right] &= \\ = \frac{b'_i}{B'_i} \frac{L_{0i}x_{0i}}{(L_{0i} + G_i)K_i} \mathbf{e}_1 + \frac{G_i}{B'_i} [S_i - (B'_i K'_i + B''_i)] \mathbf{f}_i - \left( K'_i + \frac{B''_i - b''_i}{B'_i} \right) \mathbf{g}_i. \end{aligned}$$



After arrangement premultiplying with matrix  $(\mathbf{E} - \mathbf{L}_i)$  and introducing the symbols

$$\begin{aligned} \bar{B}_i &= B'_i s_i + B''_i - S_i, \\ \bar{b}_i &= b'_i s_i + b''_i - S_i \end{aligned} \tag{10}$$

and

$$\Delta_{K_i} = (B'_i - b'_i) K_i + (B''_i - b''_i) \tag{11}$$

after rearrangement the following equation is obtained:

$$\begin{aligned} (B'_i - b'_i) \frac{L_{0i} x_{0i}}{(L_{0i} + G_i) K_i} \mathbf{e}_1 + G_i \bar{B}_i \mathbf{f}_i + (B''_i - b''_i) \mathbf{g}_i &= \\ &= \mathbf{L}_i [\Delta_{K_i} \mathbf{g}_i + G_i (\bar{b}_i + \Delta_{K_i}) \mathbf{f}_i]. \end{aligned} \tag{12}$$

Since in the first row of the nilpotent matrix  $\mathbf{L}_i$  all elements are zero and all but the first element of vector  $\mathbf{e}_1$  are zero, too, the first equation of the system (12) becomes:

$$(B'_i - b'_i) L_{0i} x_{0i} + G_i \bar{B}_i + (B''_i - b''_i) L_{0i} = 0 \tag{13}$$

This equation relates the known  $x_{0i}$  and the unknown  $L_{0i}$  values.\*

Taking into consideration the structure of matrix  $\mathbf{L}_i$  (cf. Eq. (6)), a recursive formula is obtained for the  $L_{ji}$  values from the other equations of (12) owing to the fact that the elements of matrix  $\mathbf{L}_i$  are identical with the elements of vector  $\mathbf{g}_i$  if the first element of the latter is omitted. Since the non-zero elements of matrix  $\mathbf{L}_i$  lay immediately below the main diagonal, the vectors  $\mathbf{L}_i \mathbf{g}_i$  and  $\mathbf{L}_i \mathbf{f}_i$  are obtained in the following form:

$$\left. \begin{aligned} \mathbf{L}_i \mathbf{g}_i &= [g_{ji} g_{j-1,i}], (g_{-1,i} = 0) \\ \mathbf{L}_i \mathbf{f}_i &= [g_{ji} f_{j-1,i}], (f_{-1,i} = 0) \end{aligned} \right\} \begin{aligned} j &= 0, 1, 2, \dots, p_i - 1 \\ i &= 0, 1, 2, \dots, \nu. \end{aligned}$$

Thus the other equations in the system (12) are:

$$G_i \bar{B}_i f_{ji} + (B''_i - b''_i) g_{ji} = g_{ji} [\Delta_{K_i} g_{j-1,i} + G_i (\bar{b}_i + \Delta_{K_i}) f_{j-1,i}].$$

Replacing  $f_{ji}$  and  $g_{ji}$  by the  $L_{ji}$  values according to (7), the following relation is obtained:

$$\frac{G_i \bar{B}_i}{L_{ji}} + B''_i - b''_i = \frac{\Delta_{K_i} L_{j-1,i} + G_i (\bar{b}_i + \Delta_{K_i})}{(L_{j-1,i} + G_i) K_i}, \quad j = 1, 2, \dots, p_i - 1.$$

Introducing the symbol

$$A_i = \Delta_{K_i} - K_i (B''_i - b''_i) \tag{14}$$

\* Eq. (13) can be obtained from the material and solvent balance (enthalpy balance) for the column part between the  $i$ -th and  $n$ -th stage, i.e. from the comparison of the corresponding equations in (3) and (4) too.

the following fractional-linear recursive formula is obtained:

$$\frac{1}{\frac{L_{ji}}{G_i \bar{B}_i} + w} = \frac{A_i \frac{L_{j-1,i}}{G_i \bar{B}_i} + \frac{1}{\bar{B}_i} (\bar{b}_i + A_i)}{K_i \frac{L_{j-1,i}}{G_i \bar{B}_i} + \frac{K_i}{\bar{B}_i}}. \quad (15)$$

Since there is a simple explicit relation for solving linear recursive formulae by using appropriate transformation, Eq. (15) should be linearized. This is done by a simple shifting. Introducing the expression

$$\frac{1}{\frac{L_{ji}}{G_i \bar{B}_i} + w} \quad \text{instead of} \quad \frac{1}{\frac{L_{ji}}{G_i \bar{B}_i}}$$

Eq. (15) can be written in the form:

$$\frac{1}{\frac{L_{ji}}{G_i \bar{B}_i} + w} = \frac{A_i}{K_i + wA_i} + \frac{\frac{\bar{b}_i}{\bar{B}_i} K_i}{(K_i + wA_i)^2} \cdot \frac{1}{\frac{L_{j-1,i}}{G_i \bar{B}_i} + \frac{K_i + w(\bar{b}_i + A_i)}{\bar{B}_i(K_i + wA_i)}}.$$

Choosing  $w$  to satisfy the equations:

$$w = \frac{K_i + w(\bar{b}_i + A_i)}{\bar{B}_i(K_i + wA_i)} \quad (16)$$

a linear recursive formula is obtained for the expression

$$\frac{1}{\frac{L_{ji}}{G_i \bar{B}_i} + w}$$

namely

$$\frac{1}{\frac{L_{ji}}{G_i \bar{B}_i} + w} = \frac{A_i}{K_i + wA_i} + \frac{\frac{\bar{b}_i}{\bar{B}_i} K_i}{(K_i + wA_i)^2} \cdot \frac{1}{\frac{L_{j-1,i}}{G_i \bar{B}_i} + w}. \quad (17)$$

From Eq. (17), the value of  $w$  is

$$w^\pm = \frac{1}{2A_i \bar{B}_i} [\bar{b}_i - K_i \bar{B}_i + A_i \pm \sqrt{(\bar{b}_i - K_i \bar{B}_i + A_i)^2 + 4A_i K_i \bar{B}_i}]. \quad (18)$$

The dependence of  $w$  on  $i$  and that of  $u$  and  $v$  in (19) will not be referred to in the following. Eq. (17) is a linear, first-order difference equation with con-

stant coefficients. Its solution can be given by considering some simple matrix relations. Writing the difference equation in the form of system of linear equations and introducing the symbols

$$\begin{aligned} \frac{b_i K_i}{\bar{B}_i(K_i + w^\pm A_i)^2} &= v^\pm, \\ \frac{A_i}{K_i + w^\pm A_i} &= u^\pm(v^\pm - 1), \\ z_j &= \frac{1}{\frac{L_{ji}}{G_i \bar{B}_i} + w^\pm}, \end{aligned} \tag{19}$$

neglecting the superscript  $\pm$  for the time being, the system of equations (17) can be written in the form

$$z_j = u(v - 1) + v z_{j-1}. \tag{17}$$

Introducing the unit matrix  $\mathbf{E}$  of order  $(p_i - 1)$ , the nilpotent matrix  $\mathbf{N} = [n_{\alpha\beta}]$  of the order  $(p_i - 1)$  whose all elements but  $n_{\alpha, \alpha-1} = 1$  ( $\alpha = 2, 3, \dots, p_i - 1$ ) are zero, the unit vector  $\mathbf{e}_1$  of  $(p_i - 1)$  order, the vector  $\mathbf{e}$  with elements solely 1 and the vector  $\mathbf{z} = [z_j]$  ( $j = 1, 2, \dots, p_i - 1$ ), the system of equations (17) takes the form

$$(\mathbf{E} - v\mathbf{N})\mathbf{z} = v z_0 \mathbf{e}_1 + u(v - 1)\mathbf{e}.$$

Hence, since

$$(\mathbf{E} - v\mathbf{N})^{-1} = \sum_{m=0}^{p_i-2} (v\mathbf{N})^m = \begin{bmatrix} 1 & & & & \\ v & 1 & & & \\ v^2 & v & 1 & & \\ \vdots & \vdots & v & 1 & \\ \vdots & \vdots & & & \ddots \\ v^{p_i-2} & v^{p_i-3} & & & & 1 \end{bmatrix}$$

the following expression is obtained for the elements  $z_j$  sought for:

$$z_j = (u + z_0)v^j - u, \quad (j = 1, 2, \dots, p_i - 1). \tag{20}$$

Resubstituting the symbols (19), the unknown values of  $L_{ji}$  as functions of  $L_{0i}$  can be obtained in explicit form, while knowing  $x_{0i}$  values of  $L_{0i}$  are calculated from Eq. (13). The number of stages in each stage group, i.e. the values  $p_i$  are required. The first equation of any group of stages can be written as the last equation of the preceding group, i.e. the points dividing the equilibrium curve and the other diagrams into linear sections may be considered as belonging to the section to their left or to their right. If the system of equations (17) is completed by a  $p_i$ -th equation in which  $L_{p_i} = L_{0,i+1}$  and the

latter is calculated from Eq. (13) by means of  $x_{0,i+1}$ , then the solution of the system of equations (20) is augmented by a  $p_i$ -th relation:

$$z_{p_i} = (u + z_0)v^{p_i} - u. \quad (21)$$

A simple relation existing between  $z_{p_i}$  and  $L_{p_i}$ ,  $p_i$  can be derived from Eq. (21):

$$p_i = \frac{\ln \frac{u + z_{p_i}}{u + z_0}}{\ln v}. \quad (22)$$

Expressing  $L_{0i}$  from Eq. (13) and substituting this into the formulae for  $z_{p_i}$  and  $z_0$ , then, introducing the symbol

$$\Delta_i = (B'_i - b'_i)x_{0i} + (B''_i - b''_i), \quad (23)$$

the expressions

$$z_0 = \frac{1}{w^\pm - \frac{1}{\Delta_i}}$$

and

$$z_{p_i} = \frac{1}{w^\pm - \frac{1}{\Delta_{i+1}} \cdot \frac{\bar{B}_{i+1}}{\bar{B}_i} \cdot \frac{G_{i+1}}{G_i}}$$

are obtained. Substituting them into Eq. (22), using the symbol

$$r = \frac{\bar{B}_i}{\bar{B}_{i+1}} \cdot \frac{G_i}{G_{i+1}}$$

and performing some transformations, the following equation is obtained:

$$p_i = \frac{\ln \frac{\left[ \left( w^\pm + \frac{1}{u^\pm} \right) \cdot r \Delta_{i+1} - 1 \right] (w^\pm \Delta_i - 1)}{\left[ \left( w^\pm + \frac{1}{u^\pm} \right) \Delta_i - 1 \right] (w^\pm r \Delta_{i+1} - 1)}}{\ln v^\pm}. \quad (24)$$

Consider that from Eq. (18)

$$K_i + w^\pm A_i = \frac{1}{2\bar{B}_i} [\bar{b}_i + K_i \bar{B}_i + A_i \pm \sqrt{(\bar{b}_i + K_i \bar{B}_i + A_i)^2 - 4K_i \bar{B}_i \bar{b}_i}]$$

and

$$\frac{1}{K_i + w^\pm A_i} = \frac{1}{2K_i \bar{b}_i} [\bar{b}_i + K_i \bar{B}_i + A_i \mp \sqrt{(\bar{b}_i + K_i \bar{B}_i + A_i)^2 - 4K_i \bar{B}_i \bar{b}_i}],$$

hence:

$$\frac{1}{K_i + w^\pm A_i} = \frac{\bar{B}_i}{K_i \bar{b}_i} (K_i + w^\pm A_i); \tag{25}$$

then, substituting these into Eq. (19):

$$v^\pm = \frac{\bar{b}_i + K_i \bar{B}_i + A_i \mp \sqrt{(\bar{b}_i + K_i \bar{B}_i + A_i)^2 - 4K_i \bar{B}_i \bar{b}_i}}{\bar{b}_i + K_i \bar{B}_i + A_i \pm \sqrt{(\bar{b}_i + K_i \bar{B}_i + A_i)^2 - 4K_i \bar{B}_i \bar{b}_i}} \tag{26}$$

and since — also from (19) —

$$\frac{1}{u^\pm} = \frac{\frac{\bar{b}_i}{\bar{B}_i} \frac{K_i}{(K_i + w^\pm A_i)^2} - 1}{\frac{A_i}{K_i + w^\pm A_i}},$$

utilizing Eq. (25), we obtain

$$\frac{1}{u^\pm} = w^\mp - w^\pm.$$

Substituting this into the expression (24) for  $p_i$ ,

$$p_i = \frac{\ln \frac{(w^\pm \Delta_i - 1)(w^\mp r \Delta_{i+1} - 1)}{(w^\pm r \Delta_{i+1} - 1)(w^\mp \Delta_i - 1)}}{\ln v^\pm} \tag{27}$$

is obtained. The expressions of  $v^\pm$  and  $w^\pm$  as well as  $\Delta_i$  are given in the formulae (26), (18) and (23). Further symbols occurring in these formulae are given in (11), (10) and (5). From Eq. (27), the uniqueness of the result is evident since it is identical irrespective of the value of the roots in expressions for  $v^\pm$  and  $w^\pm$  being chosen positive or negative.

The number of stages in the column section above the feed point is given by

$$k = \sum_{i=0}^{\alpha-1} p_i, \tag{28}$$

the number of stages in the column section below the feed point by

$$n - k = \sum_{i=\alpha}^v p_i. \tag{29}$$

### Applications of the results for numerical calculations

Introducing the symbols

$$\varphi_i = (\bar{b}_i + K_i \bar{B}_i + A_i)^2 - 4K_i \bar{B}_i \bar{b}_i \tag{30}$$

and

$$q_i = \bar{b}_i + K_i \bar{B}_i + A_i \tag{31}$$

Eqs (18) and (26) can be written in the form:

$$w^\pm = \frac{1}{2A_i \bar{B}_i} (\bar{b}_i - K_i \bar{B}_i + A_i \pm \sqrt{\varphi_i})$$

$$v^\pm = \frac{q_i \mp \sqrt{\varphi_i}}{q_i \pm \sqrt{\varphi_i}},$$

noting that Eq. (27) can only be used for  $\varphi_i > 0$ , but neither for  $\varphi_i \leq 0$  not for  $B'_i - b'_i \rightarrow 0$ .

If  $\varphi_i < 0$ , Eq. (27) can be written as follows:

$$p_i = \frac{\operatorname{ar\,th} \frac{\sqrt{\varphi_i}}{\bar{b}_i - K_i \bar{B}_i + A_i - \frac{2A_i \bar{B}_{i+1}}{\Delta_{i+1}}} - \operatorname{ar\,th} \frac{\sqrt{\varphi_i}}{\bar{b}_i - K_i \bar{B}_i + A_i - \frac{2A_i \bar{B}_i}{\Delta_i}}}{\operatorname{ar\,th} \frac{\sqrt{\varphi_i}}{\bar{b}_i + K_i \bar{B}_i + A_i}} \tag{32}$$

It is obvious that for  $\varphi_i < 0$  the function  $\operatorname{ar\,th}$  becomes  $\operatorname{arc\,tan}$ , leading to the expression

$$p_i = \frac{\operatorname{arc\,tan} \frac{\sqrt{-\varphi_i}}{\bar{b}_i - K_i \bar{B}_i + A_i - \frac{2A_i \bar{B}_{i+1}}{\Delta_{i+1}}} - \operatorname{arc\,tan} \frac{\sqrt{-\varphi_i}}{\bar{b}_i - K_i \bar{B}_i + A_i - \frac{2A_i \bar{B}_i}{\Delta_i}}}{\operatorname{arc\,tan} \frac{\sqrt{-\varphi_i}}{\bar{b}_i + K_i \bar{B}_i + A_i}} \tag{33}$$

In this expression it is effective to calculate the sum  $\Sigma p_i$  by increasing the number of chords to a value where the difference between two successive sums is less than one theoretical stage. At the beginning of the calculation, the number of chords has to be chosen so that no chord intersects, or is tangential to the operating line, since this would yield infinite number of theoretical stages.

Considering the case where the difference  $B'_i - b'_i$  is zero and substituting (18) and (23) into (27) we obtain

$$p_i = \frac{\ln \left[ \frac{\frac{q_i \pm \sqrt{\varphi_i}}{2\bar{B}_i} - 1 + (B'_i - b'_i)\Phi_i^\pm}{\frac{q_i \pm \sqrt{\varphi_i}}{2\bar{B}_i} - 1 + (B'_i - b'_i)\Phi_{i+1}^\pm} \cdot \frac{\frac{q_i \mp \sqrt{\varphi_i}}{2\bar{B}_i} - 1 + (B'_i - b'_i)\Psi_{i+1}^\pm}{\frac{q_i \mp \sqrt{\varphi_i}}{2\bar{B}_i} - 1 + (B'_i - b'_i)\Psi_i^\pm} \right]}{\ln v_i^\pm} \tag{34}$$

where

$$\begin{aligned} \Phi_i^\pm &= \frac{1}{B_i'' - b_i''} \left[ \left( \frac{\varrho_i \pm \sqrt{\varphi_i}}{2\bar{B}_i} - K_i \right) x_i - K_i' \right] \\ \Psi_i^\pm &= \frac{1}{B_i'' - b_i''} \left[ \left( \frac{\varrho_i \mp \sqrt{\varphi_i}}{2\bar{B}_i} - K_i \right) x_i - K_i' \right]. \end{aligned} \tag{35}$$

Considering that

$$\frac{\varrho_i - \sqrt{\varphi_i}}{2\bar{B}_i} - 1 = \frac{2(\bar{B}_i + K_i \bar{b}_i - \varrho_i)}{\varrho_i - 2\bar{B}_i + \sqrt{\varphi_i}}, \tag{36}$$

and

$$\bar{B}_i + K_i \bar{b}_i - \varrho_i = (B_i' - b_i')(s_i - K_i s_i - K_i'), \tag{37}$$

the expression in the numerator can be reduced by the difference  $(B_i' - b_i')$ , to lower terms and hence, using the symbols

$$U_i^\pm = \frac{\varrho_i \pm \sqrt{\varphi_i}}{2\bar{B}_i} - 1$$

and

$$W_i^\pm = \frac{2(s_i - K_i s_i - K_i')}{\varrho_i \pm \sqrt{\varphi_i} - 2\bar{B}_i} \tag{38}$$

the final result is written in the following form:

$$P_i = \frac{\ln \left[ \frac{U_i^\pm + (B_i' - b_i') \Phi_i^\pm}{U_i^\pm + (B_i' - b_i') \Phi_{i+1}^\pm} \cdot \frac{W_i^\pm + \Psi_{i+1}^\pm}{W_i^\pm + \Psi_i^\pm} \right]}{\ln \frac{\varrho_i \mp \sqrt{\varphi_i}}{\varrho_i \pm \sqrt{\varphi_i}}}. \tag{39}$$

The advantage of Eq. (39) is that it is suitable for numerical calculation, however small the difference  $B_i' - b_i'$  be. Obviously for  $B_i' - b_i' = 0$ , the final result will be

$$P_i = \frac{\ln \frac{W_i^\pm + \Psi_{i+1}^\pm}{W_i^\pm + \Psi_i^\pm}}{\ln v_i^\pm}. \tag{40}$$

This expression is identical with the result published in a previous paper [2] of the authors.

### Substitution of the approximating sum by integral

The sums (28) and (29) obtained for the number of theoretical stages can be substituted — under certain conditions — by definite integrals, this being an advantage for the numerical calculations. When the number of stages is large enough to consider the phase equilibrium curves and the curves determining the phase conditions i.e. the operating lines as straight lines within one stage, it is sufficient to consider the first term alone when expanding in series the above logarithmic functions. The expression  $\Sigma p_i$  may be regarded as an integral approximating sum and replaced by the integral.

In the foregoing the sign  $\pm$  referring to the ambiguity of the square root has been indicated. This ambiguity — as already pointed out — is, however, ostensible only. In the following none the less the minus sign is used only since then the limit of the integral approximating sums can easily be obtained.

Thus, from the numerator of (39) leads to the approximative formulae

$$\ln \left[ 1 + \frac{(B'_i - b'_i)(\Phi_i^{(-)} - \Phi_{i+1}^{(-)})}{U_i^{(-)} + (B'_i - b'_i)\Phi_{i+1}^{(-)}} \right] \approx + \frac{(B'_i - b'_i)}{U_i^{(-)} + (B'_i - b'_i)\Phi_{i+1}^{(-)}} (\Phi_i^{(-)} - \Phi_{i+1}^{(-)}) > 0 \quad (41)$$

and

$$\ln \left[ 1 - \frac{\Psi_i^{(-)} - \Psi_{i+1}^{(-)}}{W_i^{(-)} + \Psi_i^{(-)}} \right] \approx - \frac{1}{W_i^{(-)} + \Psi_i^{(-)}} (\Psi_i^{(-)} - \Psi_{i+1}^{(-)}) < 0. \quad (42)$$

Introducing the symbol  $x_i - x_{i+1} = \Delta x$

$$\frac{(B'_i - b'_i)(\Phi_i^{(-)} - \Phi_{i+1}^{(-)})}{U_i^{(-)} + (B'_i - b'_i)\Phi_{i+1}^{(-)}} = \frac{\frac{B'_i - b'_i}{B''_i - b''_i} [U_i^{(-)}\Delta x + (1 - K_i)\Delta x]}{U_i^{(-)} + \frac{B'_i - b'_i}{B''_i - b''_i} [U_i^{(-)}x_{i+1} + (1 - K_i)x_{i+1} - K_i]} \quad (43)$$

and since  $W_i^{(-)} = \frac{1}{B'_i - b'_i} \cdot \bar{U}_i^{(-)}$ , where  $\bar{U}_i^{(-)}$  is the conjugate of the expression  $U_i^{(-)}$ , hence

$$\frac{\Psi_i^{(-)} - \Psi_{i+1}^{(-)}}{W_i^{(-)} + \Psi_i^{(-)}} = \frac{\frac{B'_i - b'_i}{B''_i - b''_i} [\bar{U}_i^{(-)}\Delta x + (1 - K_i)\Delta x]}{\bar{U}_i^{(-)} + \frac{B'_i - b'_i}{B''_i - b''_i} [\bar{U}_i^{(-)} \cdot x_i + (1 - K_i)x_i - K_i]} \quad (44)$$

(43) and (44) being each other's conjugates if  $x = x_i = x_{i+1}$ . Therefore summing over  $i$  in (34) and taking the limit of it, the integrand obtained is a fraction whose numerator is the difference of complex conjugate expressions for



$\varphi < 0$ , i.e. a pure imaginary one and its denominator is a pure imaginary one as well, hence the integrand is a real one.

The imaginary part of the right-hand side of Eq. (43) is:

$$\text{Im } U_i = - \frac{\sqrt{\varphi_i}}{2\bar{B}_i}$$

introducing

$$a_i = \frac{B'_i - b'_i}{B''_i - b''_i}$$

the following expression is obtained for the difference of (43) and (44):

$$\frac{a_i \cdot \frac{\sqrt{\varphi_i}}{\bar{B}_i} (1 - K_i + a_i K'_i) \Delta x}{\frac{\bar{B}_i + K_i \bar{b}_i - \varrho_i}{\bar{B}_i} (1 + a_i x)^2 + \frac{a_i (1 + a_i x) [(1 - K_i)x - K'_i]}{\bar{B}_i} (\varrho_i - 2\bar{B}_i) + a_i^2 [(1 - K_i)x - K'_i]^2} \cdot i \quad (45)$$

Since the denominator of (39) for  $\varphi < 0$  is

$$\ln \frac{\varrho_i + \sqrt{\varphi_i}}{\varrho_i - \sqrt{\varphi_i}} = 2i \text{ arc tg } \frac{\sqrt{-\varphi_i}}{\varrho_i}, \quad (46)$$

reducing by  $i$  it yields the following form for the integral approximating sum:

$$\sum \frac{(1 - K_i + a_i K'_i) \sqrt{-\varphi_i} \cdot \Delta x}{\text{arc tg } \frac{\sqrt{-\varphi_i}}{\varrho_i} \{ (s - K_i s - K'_i)(B''_i - b''_i)(1 + a_i x)^2 - \varrho_i (1 + a_i x)(K_i x + K'_i - x) + \bar{B}_i (K_i x + K'_i - x) [a_i (K_i x + K'_i + x) + 2] \}} \quad (47)$$

For  $\varphi_i > 0$ , a similar formula is obtained:

$$\sum \frac{(1 - K_i + a_i K'_i) \sqrt{\varphi_i} \cdot \Delta x}{\ln \frac{\varrho_i + \sqrt{\varphi_i}}{\varrho_i - \sqrt{\varphi_i}} \{ (s - K_i s - K'_i)(B''_i - b''_i)(1 + a_i x)^2 - \varrho_i (1 + a_i x)(K_i x + K'_i - x) + \bar{B}_i (K_i x + K'_i - x) [a_i (K_i x + K'_i + x) + 2] \}} \quad (48)$$

Taking the limit of (47) and (48) for  $\Delta x \rightarrow 0$ , the quantities occurring in these integrals can be interpreted on the basis of Fig. 3 as follows. Increasing the number  $x_i$  of the dividing points, the limit position of the chords of the curves  $y = f(x)$ ,  $B = B(y)$  and  $b = b(x)$  become the tangents of the curves at the corresponding points

and

$$\left. \begin{aligned} \bar{B} &= B(y) - (y - s)(B') - S \\ \bar{b} &= b(x) - (x - s)(b') - S \\ y &= f(x) = K \cdot x + K'; \quad y' = K; \quad K' = y - y' \cdot x \\ \varrho &= y'(b - S) + (B - S) - b'(y - s) - y' B'(x - s) \\ \varphi &= \varrho^2 - 4y' \bar{B} \bar{b} \end{aligned} \right\} \quad (49)$$

can be substituted. Following integrals are obtained from (47) and (48):

$$\frac{1}{2} \int_{x_n}^{x_0} \frac{\frac{\sqrt{-\varphi}}{\varrho}}{\operatorname{arc\,tg} \frac{\sqrt{-\varphi}}{\varrho}} \frac{\varrho}{(b - S)(y - s) - (B - S)(x - s)} dx, \quad (50)$$

and

$$\int_{x_n}^{x_0} \frac{\frac{\sqrt{\varphi}}{\varrho}}{\ln \frac{\varrho + \sqrt{\varphi}}{\varrho - \sqrt{\varphi}}} \frac{\varrho}{(b - S)(y - s) - (B - S)(x - s)} dx. \quad (51)$$

If  $B' = b'$  then the expressions for  $\varrho$  and for  $\varphi = \varrho^2 - 4K\bar{B}\bar{b}$  take the form:

$$\begin{aligned} \varrho &= \bar{B} + y' \bar{b} + (B' - b') [y'(s - x) - (s - y)] \quad \text{and} \\ \varphi &= \varrho^2 - 4y' \bar{B} \bar{b} = (\bar{B} - y' \bar{b})^2 + 2(\bar{B} + y' \bar{b}) [y'(s - x) - (s - y)] (B' - b') + (B' - b')^2 [y'(s - x) - (s - y)]^2. \end{aligned} \quad (52)$$

Substituting these into (50) and (51), the generalized Lewis integral applied in (2) and (3) is obtained.

By expanding in series the first factor of the integrands in (50) and (51) for  $\varphi \rightarrow 0$ , formulae suitable for calculation are obtained.

### Application to countercurrent extraction, refluxed extraction and rectification

(Specifying conditions)

In the *simple extractor* (Fig. 3a) the molar flow rate of phase E (extract phase) on a solvent-free basis is  $V_0$ . This flow consists of pure solvent, when  $B(y) = \infty$ , or of recovered solvent containing A and C, but in the latter case — according to the assumption made — the point representing is lying on the

curve  $B(y)$ . This is described by

$$Q' = V_0 B_0 = V_0(B'_v y_0 + B''_v),$$

i.e. the solvent contains the substance  $C$  to be extracted at a concentration  $y_0$ . Applying pure solvent  $Q'$  may be substituted into the definition formula of  $S_i$ .

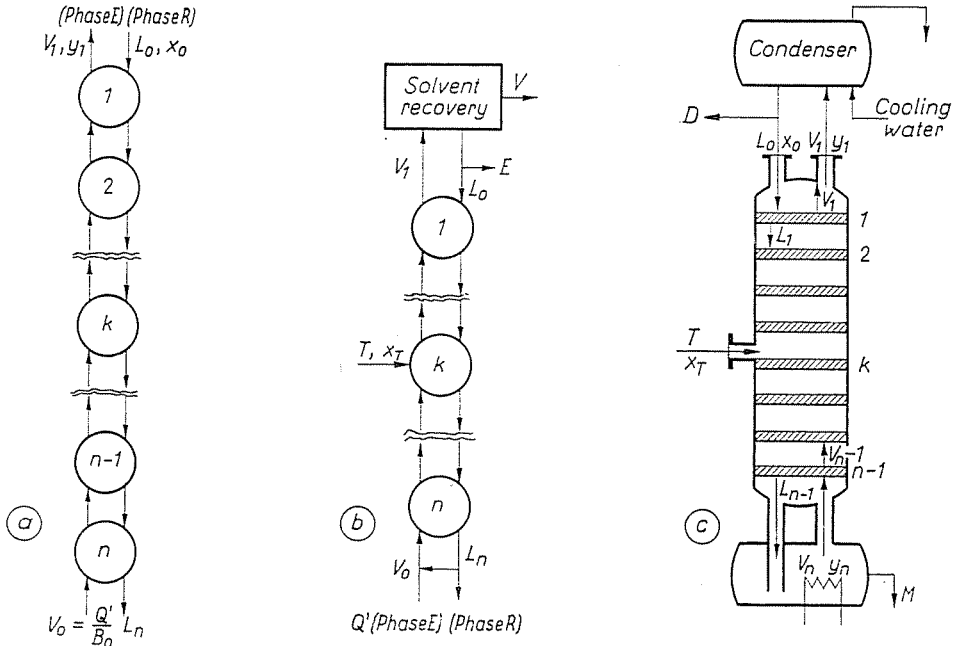


Fig. 3a. Simple countercurrent extractor  
 Fig. 3b. Extractor operating with extract and raffinate reflux  
 Fig. 3c. Rectification column

Following conditions have to be considered for the calculation of the theoretical stage number:

$$T = 0; \quad Q' = V_0(B'_v y_0 + B''_v);$$

$$s = \frac{V_0 y_0 - L_n x_n}{V_0 - L_n}; \quad S = \frac{Q' - L_n(b'_v x_n + b''_v)}{V_0 - L_n};$$

$$A_i = (B'_i - b'_i)K'_i + (1 - K_i)(B''_i - b''_i);$$

$$\bar{b}_i = b'_i s + b''_i - S; \quad \bar{B}_i = B'_i s + B''_i - S.$$

Calculating with the integral,  $A$  vanishes;  $b(x)$ ,  $B(y)$ ,  $y = f(x)$  and their derivatives, as well, are obtained by interpolation formulae fitting the experimental data as closely as possible. Hence  $\varphi$ ,  $q$ ,  $\bar{B}(y)$  and  $\bar{b}(x)$  can be calculated from Eq. (49).

Since  $L_n$ , encountered in the formulæ for  $s$  and  $S$ , is usually not given, its value has to be calculated from the material balances referring to the total extractor.

### Extractor operating with raffinate reflux and extract reflux (Fig. 3b)

Conditions:

$$\begin{aligned}x_n (= y_0) &= x_R; & V_0 &= L_n - R; \\x_0 = y_{10} &= x_E; & V_{10} &= L_0 + E; \\Q' &= V_0(B'_v x_R + B''_v).\end{aligned}$$

The value of  $L_n$  being unknown,  $V_0$  is unknown as well, therefore no direct substitution of the above expressions can be applied. Calculations can, however, be performed making use of the material balance of substance  $B$  over the entire apparatus, using the relationship:

$$Q'_B = V_0(B'_v y_0 + B''_v) = V_{10}(B'_0 y_{10} + B''_0) + L_n(b'_v x_n + b''_v) - L_0(b'_0 x_0 + b''_0) - T b_T$$

and the definitions of  $s$  and  $S$ :

$$\begin{aligned}s &= \frac{\mathcal{E}_{ix} T x_T + (L_n - R)x_R - L_n x_R}{\mathcal{E}_{ix} T + (L_n - R) - L_n} = \begin{cases} x_E & \text{above the feed point} \\ x_R & \text{below the feed point} \end{cases} \\S &= \frac{\mathcal{E}_{ix} T b_T + V_{10}(B'_0 x_E + B''_0)}{\mathcal{E}_{ix} T + (L_n - R) - L_n} + \\&+ \frac{L_n(b'_v x_R + b''_v) - T b_T - L_0(b'_0 x_E + b''_0) - L_n(b'_v x_R + b''_v)}{\mathcal{E}_{ix} T + (L_n - R) - L_n} = \\&= \frac{(\mathcal{E}_{ix} - 1)T b_T + (L_0 - E)A_{x_E} + E(b'_0 x_E + b''_0)}{T - R}.\end{aligned}$$

The values of  $A_i$ ,  $\bar{b}_i$  and  $\bar{B}_i$  are obtained from Eqs (14) and (10).

Using integral (50), the remark on page 353. is valid for the calculation.

### Rectification

Conditions:

$$\begin{aligned}x_n &= x_M; & V_0 &= 0; & L_n &= M; \\x_0 = y_{10} &= x_D; & V_{10} &= L_0 + D.\end{aligned}$$

The heat  $Q'_H$  transferred in the reboiler is calculated from the enthalpy balance (see page 340.):

$$Q'_H = (L_0 + D)A_{x_D} + D(h'_0 x_D + h''_0) + M(h'_v x_M + h''_v) - T h_T.$$

Thereby the values of  $s$  and  $S$  corresponding to the expressions for the refluxed extractor are obtained in the form:

$$s = \begin{cases} x_D & \text{above the feed point} \\ x_M & \text{below the feed point} \end{cases}$$

$$S = \frac{(\mathcal{S}_{ix} - 1)Th_T + (L_0 + D)\Delta_{x_D} + D(h'_0x_D + h''_0)}{\mathcal{S}_{ix} \cdot T - M}$$

The values of  $A_i$ ,  $\bar{b}_i$  and  $\bar{B}_i$  are obtained from Eqs (14) and (10).

The remark concerning the calculation with the integral is valid here, too.

### Example for the application of the integral

Calculating the number of theoretical stages of a simple countercurrent extractor using the integrals (50) and (51), the system  $A, B, C$  is not real, the diagrams characterizing the system are arbitrarily chosen typical curves. The calculation refers exclusively to the "geometry" of the problem, i.e. in Fig. 4 — in addition to the curves — the value of  $S$  and its abscissa was

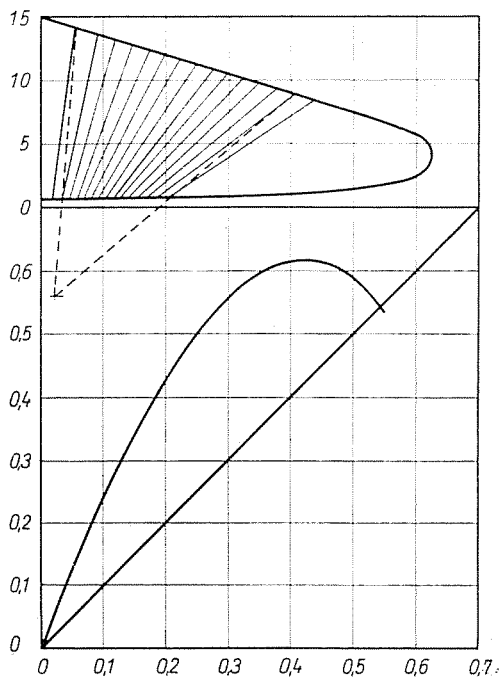


Fig. 4. Graphical solution of the example

chosen arbitrarily as well, to eliminate using values for the initial flow rates. Specifying the data on countercurrent extractor on page 352. as follows:

Initial concentration of the raffinate (on the binodal curve)	$x_0 = 0.200,$
Final concentration of the raffinate	$x_n = 0.020,$
Ratio of flow rates characterized by	$S = -7.00$
Extraction with pure solvent, i.e.	$s = x_n$

the interpolation functions of the curves in Fig. 4 are (for explanation, cf. Fig. 2a):

$$B = B(y) = -15.00 y + 15.00 \text{ (straight line within the range calculated)}$$

$$b = b(x) = 0.363 e^{0.315e^{2.13102x}}$$

$$y = y(x) = 1.993 x^{0.908} - 3.450 x^{2.871}$$

The detailed data of calculations are presented in the following table:

$x$	$e$	$\varphi$	$\frac{\frac{\sqrt{1-\varphi}}{e}}{\arctan \frac{\sqrt{1-\varphi}}{e}}$	$\frac{(b-S)(y-s) - (B-S)(x-s)}{(B-S)(x-s)}$	Integrand
$x_0 = 0.200$	31.810	-30.130	1.0098	0.31338	51.25
0.190	32.387	-29.484	1.0093	0.29839	54.77
0.180	32.939	-28.828	1.0088	0.28340	58.63
0.170	33.468	-28.200	1.0083	0.26879	62.78
0.160	33.975	-27.634	1.0079	0.25490	67.17
0.150	34.461	-27.157	1.0076	0.24207	71.72
0.140	34.927	-26.796	1.0073	0.23062	76.28
0.130	35.376	-26.571	1.0070	0.22085	80.65
0.120	35.809	-26.501	1.0069	0.21304	84.62
0.110	36.229	-26.604	1.0067	0.20746	87.90
0.100	36.637	-26.897	1.0066	0.20433	90.25
0.090	37.039	-27.397	1.0066	0.20387	91.44
0.080	37.438	-28.129	1.0067	0.20624	91.37
0.070	37.840	-29.124	1.0067	0.21154	90.04
0.060	38.255	-30.431	1.0069	0.21984	87.60
0.050	38.696	-32.130	1.0071	0.23108	84.32
0.040	39.186	-34.358	1.0074	0.24502	80.56
0.030	39.770	-37.371	1.0078	0.26117	76.73
0.020	40.547	-41.703	1.0084	0.27838	73.44

Integral calculated using the method of Simpson, yields the number of theoretical stages

$$n = 13.997.$$

A value between 14 and 15 is obtained graphically.

### Summary

Following a definition of the double fed "general extractor" given in [2] the multistage countercurrent extractor, the countercurrent extractor operating with raffinate and extract reflux and the binary mixture rectification apparatus are considered as special cases of the general extractor. For the numerical calculation of the number of theoretical stages or plates an integral approximating sum is derived. In the present paper the restriction that the operation line is a straight one, is omitted and a general analytical method of calculation is given for the determination of the number of theoretical stages of the countercurrent separation methods mentioned above. The assumption is made that in the case of extraction the equilibrium diagrams and the diagrams describing quantitative phase conditions on a solvent-free basis, or in the case of rectification, the equilibrium and enthalpy diagrams are known. An example was given for a suitable interpolation formula describing the diagrams.

### Symbols

$A$	Solvent carrying the substance investigated
$B$	Extracting solvent
$C$	Substance to be extracted
$A_{\text{mol}}, B_{\text{mol}}, C_{\text{mol}}$	Quantities of $A$ , $B$ and $C$ in mols
$B$	Molar ratio of substance $B$ in phase $V$ on a solvent-free basis
$b$	Molar ratio of substance $B$ in phase $L$ on a solvent-free basis
$B(y)$ and $b(x)$	Interpolation functions describing the quantities of substance $B$ in the extract phase and raffinate phase, respectively
$H(y)$ and $h(x)$	Interpolation functions describing the enthalpies of the vapour phase and liquid phase, respectively
$b_T$	Molar ratio of substance $B$ in the feed $T$ on a solvent-free basis
$D$	Molar flow rate of the distillate
$E$	Molar flow rate of the extract
$H$	Molar enthalpy of phase $V$
$h$	Molar enthalpy of phase $L$
$k$	Number of feed stages
$L$	Molar flow rate of the phase where the concentration of the substance of interest decreases (raffinate phase expressed by $A+C$ )
$M$	Molar flow rate of the distillation residue
$n$	Number of theoretical stages
$Q'$	Amount of substance $B$ required to produce phase $V$ (in the case of rectification, amount of heat transferred in the reboiler)
$q$	Fraction of feed joining $L$
$R$	Molar flow rate of raffinate at outlet
$T$	Molar flow rate of feed
$V$	Molar flow rate of the phase where the concentration of the substance investigated increases (extract phase expressed by $A+C$ )
$x$	Mol fraction of the substance investigated in $L$
$y$	Mol fraction of the substance investigated in $V$
$y = f(x)$	Interpolation function describing equilibrium conditions
$y = g(x)$	Operating line
$\mathcal{E}_{i,k} =$	$\begin{cases} 1 & \text{for } k - l \geq 0 \\ 0 & \text{for } k - l < 0 \end{cases}$ function of unit jump
$E$	Unit matrix
$e_1$	First unit vector

## List of indirect symbols

	Page	No. of formula
$A_i$	343	(14), (11)
$\bar{B}_i, \bar{b}_i$	343	(10)
$\Delta_i$	346	(23)
$G_i$	340	—
$p_i$	346	(24)
$S_i, s_i$	341	(5)
$v_i$	345, 347	(19), (26)
$w_i$	344	(18)
$K_i, K'_i$	} 337, 338	—
$B_i, B_i$		—
$b_i, b'_i$		—
$U_i, \bar{W}_i$	} 349	(38)
$\Phi_i, \Psi_i$		(35)
$a_i$	351	—
$\varphi_i$	347	(30)
$\varrho_i$	348	(31)

## References

1. ACRIVOS, A., AMUNDSON, N. R.: *Ind. Eng. Chem.* **47**, 1533 (1955)
2. JUNG, G., RÓZSA, P., SÁRKÁNY, GY.: *Publ. Math. Inst. Hung. Acad. Sci.*, *II*, 227 (1957)
3. RÓZSA, P., SÁRKÁNY, GY.: *Publ. Math. Inst. Hung. Acad. Sci.*, *IV*, 277 (1959)
4. RANDALL, M., LONGTIN, B.: *Ind. Eng. Chem.* **30**, 1063, 1188, 1311 (1938)
5. THORMAN, K.: *Destillieren und Rektifizieren*. Leipzig, 1928. pp. 68–69.
6. KREMSEK, A.: *Nat. Petr. News*, **22**, 21 (1930)
7. SOUDERS, M., BROWN, G. G.: *Ind. Eng. Chem.* **24**, 519 (1932)
8. LEWIS, W. K.: *Ind. Eng. Chem.* **14**, 492 (1922)
9. SÁRKÁNY, GY., RÓZSA, P., TETTAMANTI, K.: *Per. Polytechn. Chem. Eng.*, **14**, 321 (1970)
10. KIRSCHBAUM, E.: *Destillier- und Rektifizierertechnik*. Springer, Berlin, 1950. (2. Aufl.)
11. MOLE, P. D. A.: *Chem. Eng. Sci.*, **3**, 236 (1958)
12. TREYBAL, R. E.: *Liquid Extraction*. McGraw-Hill, 1963. (2nd Edition) p. 241

Prof. Dr. Pál RÓZSA }  
 Dr. György SÁRKÁNY } 1502. Budapest, P. O. B. 91. Hungary