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Thermodynamically optimal method for separating multicomponent mixtures

A study of thermodynamically reversible multicomponent distillation for the selection of an optimal scheme for separating a multicomponent mixture. Mathematical description for investigation of the features of distillation with reversible mixing of streams.

THE problem of selecting the thermodynamically optimal scheme for separating a multicomponent mixture has as yet remained unsolved, although the problem has been stated by a number of authors (1—6). The approach to selecting the optimal scheme has been purely empirical and only conventional schemes for separating an n-component mixture in (n − 1) two-sectional columns have been considered. Only a study of thermodynamically reversible multicomponent distillation can ensure a scientific approach to the selection of the optimal scheme for separating a multi-component mixture.

A completely reversible process for multicomponent distillation has been described earlier (7, 8). One would naturally expect that a scheme incorporating the features of this process would be thermodynamically optimal. Several cases of such schemes with reversible mixing of the streams are proposed by the authors (see Fig. 1).

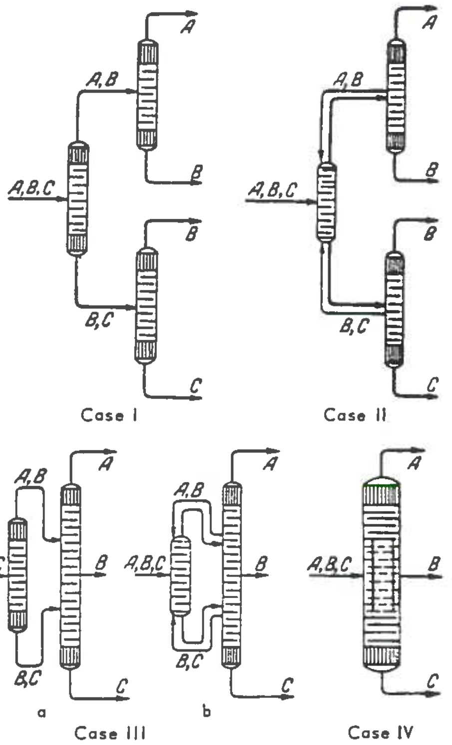

Fig. 1. Variunts of a scheme with a reversible mixing of the streams.

One of the fundamental properties of reversible distillation is characteristic for all of these cases: no more than one component is stripped out (exhausted) in each section (all columns are of the 1st class of fractionation). This guarantees reversibility during mixing of streams at the feed location. In cases II, IIIb, and IV, nonproductive columns do not have their own condensers or reboilers, while reflux streams are formed by selection from adjacent columns. Irreversibility during mixing of streams at ends of columns is also eliminated here. Finally, in cases III and IV, individual two-section columns are combined in such a way that each product is obtained at a single point. This permits the amount of energy supplied to be reduced to a minimum (heating or cooling at the points of juncture of the columns is not required). Case IV illustrates the possibility, in principle, of completely separating a multicomponent mixture in a single complex column, the volume of which is utilized to its maximum extent.

These schemes are distinguished by the following basic features:

the total number of sections required for separating an n-component mixture is equal to n(n − 1) instead of (2n − 1) in the conventional scheme;

it is sufficient to have only one condenser and one reboiler independent of the number of components to be separated (in conventional distillation, as is well known, each column has a reboiler and a condenser);

the key components in each column are not two adjacent ones, as in conventional distillation, but the two components with the extreme volatilities;

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n products of a given purity are obtained in the product column (no more than two products are obtained in each column in the case of conventional distillation).

In a thermodynamic sense the separation technique under consideration is characterized by the occurrence of only "useful" thermodynamic losses during mass transfer on each plate. Thermodynamic losses during mixing of streams at the feed location and at the ends of columns are nonexistent.

In order to investigate the features of distillation with reversible mixing of streams, its mathematical description was developed by the authors.

At minimum reflux, the conditions at the feed point of columns with reversible mixing of streams are the same as in the case of a completely reversible process ($w_1 = 0$, $d_n = 0$) (8):

$$L^F = F\,\frac{\alpha_n^F}{\alpha_1^F - \alpha_n^F}\cdot\frac{\varphi\left(\sum_{i=1}^{n}\alpha_i^F x_i^F\right) + (1-\varphi)\,\alpha_1^F}{\sum_{i=1}^{n}\alpha_i^F x_i^F} \tag{1}$$

$$V^F = F\,\frac{\varphi\left(\sum_{i=1}^{n}\alpha_i^F x_i^F\right) + (1-\varphi)\,\alpha_1^F}{\alpha_1^F - \alpha_n^F} \tag{2}$$

The distribution of the components between the top and bottom products also coincides with the distribution obtained in the case of reversible distillation:

$$d_i = F x_i^F\,\frac{\varphi\left(\sum_{i=1}^{n}\alpha_i^F x_i^F\right) + (1-\varphi)\alpha_1^F}{\sum_{i=1}^{n}\alpha_i^F x_i^F}\cdot\frac{\alpha_i^F - \alpha_n^F}{\alpha_1^F - \alpha_n^F} \tag{3}$$

$$w_i = F x_i^F\,\frac{\varphi\left(\sum_{i=1}^{n}\alpha_i^F x_i^F\right) + (1-\varphi)\alpha_n^F}{\sum_{i=1}^{n}\alpha_i^F x_i^F}\cdot\frac{\alpha_1^F - \alpha_i^F}{\alpha_1^F - \alpha_n^F} \tag{4}$$

Each component is distributed between the top and bottom in proportion to the difference between its volatility and the volatilities of the extreme components.

For cases II, IIIb, and IV:

$$d_i = v_i^d - l_i^d \tag{5}$$

$$w_i = l_i'^w - v_i'^w \tag{6}$$

Proceeding from the material balance and phase equilibrium equations (we assume that equilibrium exists between the descending liquid and the ascending vapor at the end points), we determine the concentrations at the ends of the columns in these cases:

$$x_i^d = \frac{d_i}{V^d\alpha_i^d K_p^d - L^d} \tag{7}$$

$$x_i^w = \frac{w_i}{L'^w - V'^w\alpha_i^w K_p^w} \tag{8}$$

whence:

$$\sum_{i=1}^{n-1}\frac{d_i}{V^d\alpha_i^d K_p^d - L^d} - 1 = 0 \tag{9}$$

$$\sum_{i=2}^{n}\frac{w_i}{L'^w - V'^w\alpha_i^w K_p^w} - 1 = 0 \tag{10}$$

One must first compute K_p^d and K_p^w from (9) and (10) in order to determine x_i^d and x_i^w from Equations (7) and (8).

A program based on Equations (1)—(10) has been written for use with the "Ural-2" and "Minsk-2" computers.

Equations (9) and (10) were solved by means of the numerical method of halving. In the simplest case of separation of a ternary mixture with constant volatilities, constant liquid and vapor flow rates, and a liquid feed, these may be converted to the following quadratic equations in terms of x_1^d and x_3^w ($\alpha_3 = 1$):

$$\begin{aligned}(x_1^d)^2(\alpha_1 - \alpha_2) + x_1^d(\alpha_2 x_1^F + \alpha_1 x_2^F + \alpha_2 - \alpha_1\alpha_2 x_1^F \\ - \alpha_1 - \alpha_1\alpha_2 x_2^F) + (\alpha_1\alpha_2 x_1^F - \alpha_2 x_1^F) = 0\end{aligned} \tag{11}$$

$$\begin{aligned}(x_3^w)^2(\alpha_1\alpha_2 - \alpha_1) + x_3^w(\alpha_1 x_1^F + \alpha_2 x_2^F + \alpha_1 x_3^F + \alpha_2 x_3^F \\ - \alpha_1\alpha_2 - \alpha_1\alpha_2 x_3^F) + (\alpha_1\alpha_2 x_3^F - \alpha_2 x_3^F) = 0\end{aligned} \tag{12}$$

Special programs, based on a tray-to-tray method, for the "Ural-2" and "Minsk-2" computers for simulating distillation with reversible mixing of streams at realistic reflux conditions have also been developed.

In the general case of tray-to-tray computation of multicomponent distillation, there arises the difficult problem of ensuring the convergence of successive iterations, which is associated with the problem of accurate determination of the compositions of the separation products. In the design of columns with reversible mixing of streams this problem is simplified, since the key components here, as was mentioned above, are the components with the extreme volatilities. An approximate distribution of the intermediate components may be determined from Equations (3) and (4).

The tray-to-tray method of computation is carried out for the rectification section from the top downward, and for the stripping section from the bottom upward. Given quantities are the reflux ratio and the amounts of the key components in the separated products, while the quantities to be determined are the number of trays in each section for the optimal feed location. The latter is taken to be that point at which the ratio of the concentrations of the key components x_1/x_n is equal to their ratio in the feed mixture (after the final agreement of all the balances, the composition at this point in terms of all of the components is found to be equal to the feed composition).

Disagreements in the concentrations of the intermediate components at the feed point are computed as:

$$\delta_i = \frac{x_i^{\text{rect}} - x_i'}{x_i^{\text{rect}}} \qquad i = 2, 3, \ldots, (n-1) \tag{13}$$

A correction is subsequently applied to the amounts of the intermediate components in the separation products according to the following linear interpolation formula:

$$w_i^k = w_i^{k-1} - \delta_i^{k-1} \frac{w_i^{k-1} - w_i^{k-2}}{\delta_i^{k-1} - \delta_i^{k-2}} \qquad i = 2, 3, \ldots, (n-1) \tag{14}$$

The calculation is completed when the values of δ_i are found to be smaller than the given quantities.

A flow diagram of the calculation program is given in Figure 2.

The method ensures rapid and reliable convergence of the iterations.

The variation of the concentrations along the height of a column, as calculated on a digital computer, during separation of a reference ideal mixture with constant volatilities and constant liquid and vapor flow rates is presented in Figure 3. This figure shows that, under conditions close to the minimum reflux regime and complete separation of the key components, there are two zones of constant concentrations: in the region of the feed and at the point of the maximum concentration of the intermediate component in the stripping section. The recovery of component C would have to be set higher in order to obtain such a zone in the rectification section.

In the general case (for $n > 3$), zones of constant concentrations arise in the region of the feed and at the maximum points of the $(n-1)$th component in the rectification section and of the 2nd component in the stripping section. The analogy with the process of complete separation of a binary mixture at minimum reflux, wherein constant concentration zones also arise in the region of the feed and at the ends of the column, is apparent here.

The maximum points for the concentrations of the $(n-1)$-th and the 2nd components correspond to the ends of a column for operation with the selection of reflux streams from without (see cases II and IIIb of Figure 1). This may be verified directly from Equations (7) and (8).

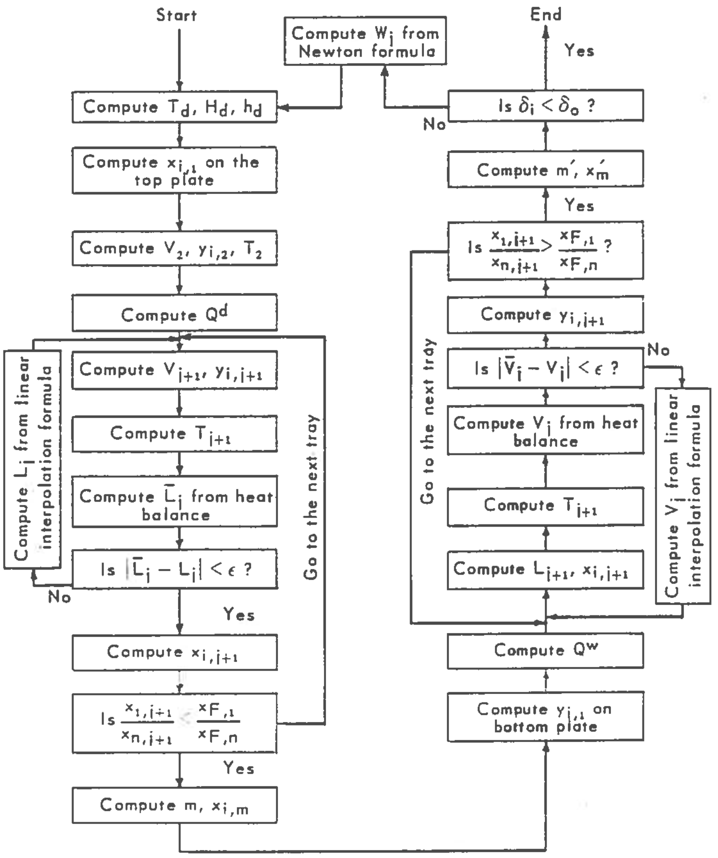


Fig. 2. Block diagram of program for computing distillation with reversible mixing streams.

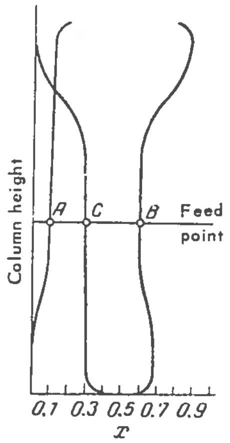


Fig. 3. Variation of component concentrations along the height of a column with reversible mixing of streams (liquid feed, $\alpha_A = 5$, $\alpha_B = 2$, $\alpha_C = 1$, $f_A = 10$, $f_B = 60$, $f_C = 30$, $w_A = 0.01$, $w_C = 29.99$, $L \approx L_{min} = 25$). o) Component concentrations in the feed mixture.

Thus, the calculation on the computer encompasses the operation of a column both with its own condenser and reboiler, as well as with selection of reflux streams from without. A sharp change in the component concentrations, indicating a large thermodynamic irreversibility, takes place in the supplementary terminal sections in the first case (see Figure 3).

Operating and equilibrium lines for the concentrations of each component, based on the computed data for the separation of the reference mixture described above, are constructed in Figure 4. The distances between these lines, corresponding to the driving forces for the process at each point, are characteristic of thermodynamic irreversibility. As the figure shows, the driving forces near the ends of the column for operation with a condenser and a reboiler (cross-hatched areas) are many times greater than the driving forces in its middle section.

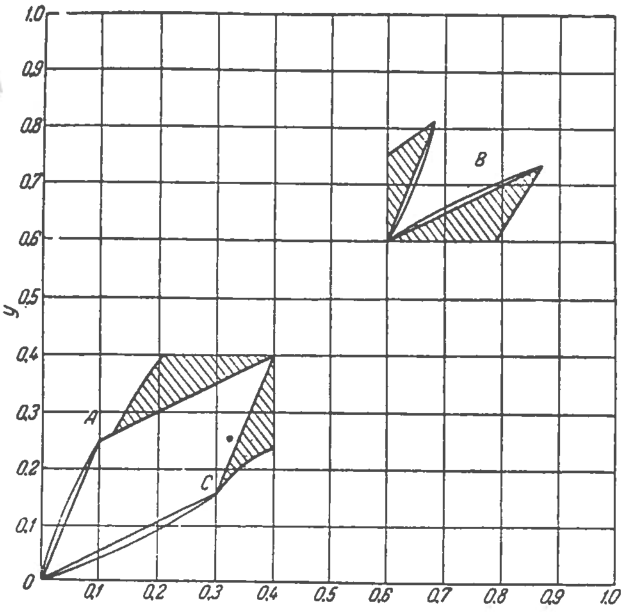


Fig. 4. Operating and equilibrium curves of components for rectification with reversible mixing of streams.

A computer study at minimum reflux conditions was carried out with the aim of comparing the economics of the separation method presented above and conventional separation. Criteria for the comparison were the total (for all columns) specific amount of liquid to be vaporized ($\Sigma V'/F$) and the overall specific work of separation (N/FRT_0). The first criterion reflects the energy expenditures on the separation sufficiently accurately if the temperature levels of the heat-carrying medium in the reboilers and of the cooling agent in the condensers are approximately the same for all cases studied (for example, for the separation of closely boiling mixtures). The overall specific work of separation is a more accurate indicator of energy (power) expenditures in the general case, in particular for low-temperature gas separation units, since its calculation takes into consideration the temperature level of the energy being transferred.

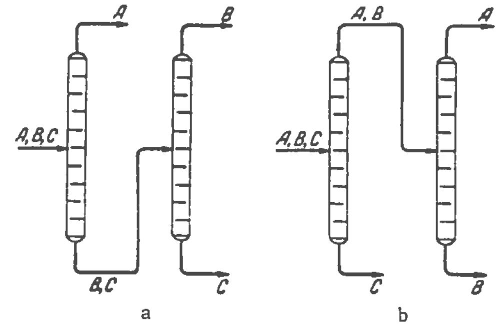
The computation for the conventional method of separation was carried out on "Ural-2" computer using the Underwood (9) method. The overall work of separation was calculated from the following formula:

$$N = T_0\left(\sum\frac{Q_c}{T_c} - \sum\frac{Q_r}{T_r}\right) \qquad (15)$$

(the heat contents of the entering and exit streams were assumed to be equal).

Reference ideal ternary mixtures with symmetrical compositions and symmetrical properties—$\alpha_A = 1.2$, $\alpha_B = 1.1$, $\alpha_C = 1.0$, $x_A^F = x_C^F$—were selected for the comparison. For these mixtures the energy expenditures for both variants of the conventional separation scheme (see Figure 5) are identical.

Values of the total specific amount of liquid vaporized and of the overall specific (unit) work of separa-



a

b

Fig. 5. Variants of the conventional scheme for the separation of a ternary mixture.

x_B^F	Conventional scheme		Variants of the scheme with reversible mixing of streams							
			I		II		IIIa		IIIb and IV	
	$\Sigma V'/F$	N/FRT_0	$\Sigma V'/F$	N/FRT_0	$\Sigma V'/F$	N/FRT_0	$\Sigma V'/F$	N/FRT_0	$\Sigma V'/F$	N/FRT_0
0.1	13.07	1.770	16.05	1.864	9.01	1.317	11.0	1.864	7.49	1.317
0.2	13.68	1.700	16.10	1.772	10.46	1.439	11.0	1.772	8.19	1.439
0.3	15.52	1.850	16.15	1.681	11.52	1.541	11.0	1.681	8.70	1.541
0.4	16.52	1.859	16.20	1.590	12.45	1.629	11.0	1.590	9.14	1.629
0.5	17.55	1.897	16.25	1.499	13.29	1.719	11.0	1.499	9.51	1.719
0.6	18.55	1.918	16.30	1.407	13.93	1.756	11.0	1.407	9.88	1.756
0,7	19.38	1.932	16.35	1.316	14.67	1.834	11.0	1.316	10.18	1.834
0.8	20.33	1.951	16.40	1.225	15.41	1.912	11.0	1.225	10.46	1.912
0.9	21.31	1.973	16.45	1.135	15.83	1.930	11.0	1.135	10.75	1.930

tion for mixtures of various compositions and for different process variants are presented in the table (liquid feed, complete separation).

This table shows that, based on the amount of liquid vaporized, cases II and III are more economical than the conventional scheme for all compositions of the feed mixture, while case I is more economical for $x_B^F > 0.3$. The most economical scheme based on this criterion is case IIIb (the economy achieved in comparison with the conventional scheme is as high as 50 per cent). Based on the overall specific (unit) work of separation, cases II and IIIb are more economical than the conventional scheme for all feed compositions, while cases I and IIIa are more economical for $x_B^F > 0.2$. The economy achieved here using reversible mixing of streams amounts on the average to approximately 20 per cent, and in some cases is as high as 40 per cent.

One should keep in mind that, if one takes into consideration thermodynamic losses during heat exchange in condensers and reboilers, the thermodynamic gain in using the separation techniqud presented above is even greater (these losses are proportional to the amount of energy transferred), and that this advantage increases as the number of components to be separated increases. Thus, for $n = 4$, $\alpha_A = 1.3$, $\alpha_B = 1.2$, $\alpha_C = 1.1$, $\alpha_D = 1.0$, $x_A = x_B = x_C = x_D = 0.25$, minimum reflux and complete separation, the total specific amount of liquid vaporized during conventional distillation is equal to 19.72, as compared with $\Sigma V'/F = 7.84$ for distillation with reversible mixing of streams.

A calculation of the minimum reflux ratio for the conventional separation scheme, using the approximate method of Robinson and Gilliland (3, 10), permits one to make a more complete comparison of it with case I of the scheme with reversible mixing of streams, using the $\Sigma V'/F$ criterion.

For the scheme of Figure 5a:

$$\frac{\Sigma V'}{F} = \frac{\alpha_A}{\alpha_A - \alpha_B} x_A + \left(\frac{\alpha_B}{\alpha_A - \alpha_B} + \frac{\alpha_B}{\alpha_B - \alpha_C}\right) x_B$$

$$+ \left(\frac{\alpha_C}{\alpha_A - \alpha_C} + \frac{\alpha_C}{\alpha_B - \alpha_C}\right) x_C \tag{16}$$

For the scheme of Figure 5b:

$$\frac{\Sigma V'}{F} = \left(\frac{\alpha_A}{\alpha_A - \alpha_C} + \frac{\alpha_B}{\alpha_A - \alpha_B}\right) x_A$$

$$+ \left(\frac{\alpha_B}{\alpha_B - \alpha_C} + \frac{\alpha_B}{\alpha_A - \alpha_B}\right) x_B + \frac{\alpha_C}{\alpha_B - \alpha_C} x_C \tag{17}$$

And for case I (see Figure 1):

$$\frac{\Sigma V'}{F} = \left(\frac{\alpha_A}{\alpha_A - \alpha_C} + \frac{\alpha_B}{\alpha_A - \alpha_B}\right) x_A$$

$$+ \left[\frac{\alpha_B}{\alpha_A - \alpha_C} + \frac{\alpha_B(\alpha_B - \alpha_C)}{(\alpha_A - \alpha_B)(\alpha_A - \alpha_C)} + \frac{\alpha_B(\alpha_A - \alpha_B)}{(\alpha_A - \alpha_C)(\alpha_B - \alpha_C)}\right] x_B$$

$$+ \left(\frac{\alpha_C}{\alpha_A - \alpha_C} + \frac{\alpha_C}{\alpha_B - \alpha_C}\right) x_C \tag{18}$$

Equations (16)—(18) were obtained for a liquid feed and complete separation. It follows from these equations that case I is more economical than both variants of the conventional scheme for $x_B > (1/\alpha_B)x_A$ and $x_B > (1/\alpha_B)x_C$.

The optimality regions for the three schemes considered are presented in Fig. 6. This figure shows that case I of the technique proposed for separation

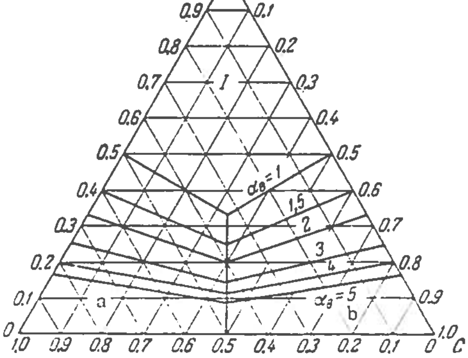


Fig. 6. Optimality regions for the two variants of the conventional scheme for separating ternary mixtures (a and b) and case I of the scheme with reversible mixing of streams (based on the total specific amount of liquid vaporized).

of mixtures, which is the least favorable in terms of the $\Sigma V'/F$ criterion of all the cases, is, for a majority of feed compositions, more economical than both variations of the conventional scheme.

The maximum thermodynamic effect may be achieved by combining schemes with reversible mixing of streams and nonadiabatic distillation.

A decrease in the work of separation may be simply achieved by the incorporation of intermediate condensers and reboilers into the scheme. If one condenser and one reboiler are introduced into case IIIb, the work of separation for a three-component mixture can, in a number of instances, be diminished by a factor of two or three. The number of levels of energy input will be the same as for the conventional scheme without intermediate heat exchangers.

A determination of the optimal intermediate point for energy input is of considerable significance. An analytical solution to this problem for the binary sections of infinite distillation columns is presented below.

It is assumed that conditions of phase equilibrium between the liquid flowing down and the rising vapor (minimum reflux conditions) exist both at the feed point and at the intermediate point of energy input.

Under these conditions and for complete binary distillation, we obtain for the rectification section:

$$\min\left(-\sum\frac{Q}{T}\right)=\min\left[\frac{z}{\alpha-1}\cdot\frac{1+(\alpha-1)x}{x}R\ln\alpha+\frac{z}{\alpha-1}\left(\frac{1}{x^F}-\frac{1}{x}\right)R\ln(1+\alpha x-x)\right] \tag{19}$$

where x is the concentration at the intermediate point of energy input.

Differentiating (19) with respect to x and setting the derivative equal to zero, we obtain:

$$-\frac{\ln\alpha}{x_{\text{opt}}^2}+\frac{\alpha-1}{1+(\alpha-1)x_{\text{opt}}}\left(\frac{1}{x^F}-\frac{1}{x_{\text{opt}}}\right)+\frac{1}{x_{\text{opt}}^2}\ln(1+\alpha x_{\text{opt}}-x_{\text{opt}})=0 \tag{20}$$

For small values of α, the quantity $\ln(1+\alpha x-x)\approx(\alpha-1)x$ and Equation (20) becomes quadratic.

Finally, we have for the rectification section:

$$x_{\text{opt}}^{\text{rect.}}=\frac{x^F(\alpha-1)+\sqrt{(x^F)^2(\alpha-1)^2+4x^F(1+\alpha x^F-x^F)}}{2(1+\alpha x^F-x^F)} \tag{21}$$

An analogous derivation for the stripping section yields:

$$x'_{\text{opt}}=\frac{-\alpha x^F(\alpha-1)+\sqrt{(x^F)^2(\alpha-1)^2+4x^F(\alpha-\alpha x^F+x^F)}}{2(\alpha-\alpha x^F+x^F)} \tag{22}$$

Calculations based on Equations (21) and (22) showed that, specifically, for the reference mixture described above with $x_B^F = 0.9$, the work of separation with one intermediate condenser and reboiler in case IIIb is three times less than that for the conventional separation scheme.

An even greater effect may be achieved if the principle of a condensation-vaporization unit (11, 12) is employed, that is, the combination of pairs of rectification and stripping sections such that heat exchange between them along their complete height is effected. Here, in the cases of schemes with reversible mixing of streams, the effect achieved is particularly great, since, for example, the rectification section of column ABC* and the stripping section of column AB, and also the stripping section of column ABC and the rectification section of column BC (see Figure 1), operate approximately in the same temperature ranges. Using this principle, one can obtain a work of separation which is as close as one pleases to the thermodynamic minimum.

However, even in conventional adiabatic distillation, the economy may be significant. The application of schemes with reversible mixing of streams does not require the development of any new specialized equipment.

Somewhat more complex is the problem of automatic control for cases II and III, since all of the columns in these represent a single interconnected system. If the separation is effected at atmospheric pressure, the thermodynamic efficiency of these cases may be reduced as a result of the hydraulic reisstances of the system. Thus, in case IIIb, column BC operates at an increased pressure, which leads to a decrease in the relative volatility of the components and to an increase of the temperature in the reboiler (this is of some importance for a large number of trays). In order to fully utilize the thermodynamic advantages of these schemes, trays with low hydraulic resistances should be used. However, this disadvantage appears only if the mixture of the components B and C is more closely boiling than the mixture of A and B. In the remaining cases, column AB, which operates under conditions which are the same as those for conventional distillation, determines the heat expenditures. A pressure increase in the lower column does not lead to any increase in the heat expenditures, and reveals itself thermodynamically only in an increase in the reboiler temperature.

Comparative calculations for the separation of a mixture of xylenes, according to the conventional scheme presently in use and according to the scheme with reversible mixing of streams and with a high recovery of the product components, were carried out by the authors. As is well known the separation of xylenes is a high-tonnage operation and requires large energy (power) and capital expenditures. The projected extensive growth in the production of xylenes makes the problem of decreasing expenditures on

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*Throughout this paper columns are named according to the components being separated in them.

their separation a particularly pressing one.

In the existing separation scheme the minimum total amount of liquid to be vaporized amounts to 1829 moles per 100 moles of initial mixture. In the new scheme the minimum vapor flow for the lower section of the product column is equal to 677 moles per 100 moles of starting mixture, while for the upper section it is 1279 moles. It is obvious that the conditions in the upper section will be the governing ones, and a pressure increase in the lower section will not lead to any increase in the heat expenditures. Thus, the use of a scheme with reversible mixing of streams results in a decrease in the energy expenditure of 31 per cent. The required number of trays for both schemes is approximately the same, while the number of reboilers and condensers for the new scheme is less than that for the existing one.

Nomenclature

A, B, C and D .... components
H and h .... heat content of the vapor and liquid, respectively
L and V .... amounts of liquid and vapor
$\overline{L}$ and $\overline{V}$ .... amounts of liquid and vapor, as obtained from a heat balance calculation
l and v .... amount of a component in the liquid and in the vapor
F .... amount of feed mixture
f .... amount of a component in the feed mixture
d .... amount of a component in the distillate
w .... amount of a component in the bottom (still) product
x .... concentration of the component in the liquid phase
φ .... fraction of liquid in the feed mixture
α .... relative volatility
K_p .... phase equilibrium constant for the principal component
δ .... material balance error
δ_0 and ϵ .... fixed constants
T .... temperature
Q .... amount of heat
N .... work of separation
m and m′ .... number of trays in the upper and lower sections
z .... overall mole fraction of a component in the feed mixture

Subscripts and superscripts

F .... feed
d .... distillate
w .... bottoms (still) product
i .... component number ($i = 1, 2, \ldots, n$; components are arranged in the order of decreasing volatility)
j .... tray number ($j = 1, 2, \ldots, m$)
k .... iteration number
o .... ambient conditions
c .... condenser
r .... reboiler
opt .... optimal
′ (prime) .... stripping section

Literature cited

1. Lockhart, F. J., *Petrol. Eng.*, No. 8, p. 111 (1947).
2. Harbert, W. D., *Petrol. Refiner* 36, No. 3, p. 169 (1957).
3. Rod, V. and Marek, J., *Coll. Czech. Chem. Comm.* 24, p. 3240 (1959).
4. Duhen, P., *Genie Chem.* 83, p. 125 (1960).
5. L'vov, S. V., "Nekotorye Voprosy Rektifikatsii Binarnykh i Mnogokomponentnykh Smesei" (Some Problems in the Distillation of Binary and Multicomponent Mixtures), Izd. AN SSSR (Publishing House of the Academy of Sciences of the USSR), 1960.
6. Kondrat'ev, A. A., Candidate's Dissertation, MIKhM (Moscow Institute of Chemical Engineering (1963).
7. Grunberg, J. F., "Advances in Cryogenic Engineering," Vol. II, New York (1960).
8. Petlyuk, F. B., and Platonov, V. M., *Khim. Prom.* No. 10, p. 723 (1964).
9. Underwood, A. I. V., *Chem. Eng. Prog.* 44, p. 603 (1948).
10. Robinson, C. S. and Gilliland, E. R., "Elements of Fractional Distillation," New York (1950).
11. Haselden, G. G., *Trans. Inst. Chem. Eng.* 36, No. 3, p. 123 (1958).
12. Bergo, B. G., *Trudy NIISS* (Trans. Sci. Res. Inst. for Synthetic Resins) No. 3, p. 44 (1961).