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Solutions for Distillation Processes Treating Petroleum Fractions

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The general solution for multicomponent distillation processes is used to obtain solutions for systems treating petroleum fractions. The integral technique is the basis for this adaptation. Example solutions are presented for a conventional distillation column, a distillation column with a side stream, and an absorber. The design capabilities of the general solution are illustrated by an absorber problem in which the composition of the lean vapor is specified. The amount of absorber oil, a petroleum fraction, required to accomplish this degree of separation is found.

Petroleum fractions may be regarded as continua (mixtures of an indefinite number of hydrocarbon components). On the true boiling point (TBP) distillation curve for the mixture, each component is represented by a point, as contrasted with the plateaus that appear for each component on the TBP curve for a mixture containing a finite number of components.

In distillation calculations for finite mixtures, the solutions are obtained by the use of equations involving summations. The heat balance equations and the bubble point equations are summations. For mixtures of an indefinite number of components, these equations naturally become integral in form. Edmister (*1*) has proposed the integral technique for handling such problems.

Taylor and Edmister (*2*) have proposed a method by which it is possible to obtain rigorous solutions for any type of distillation process by means of a single set of equations employed in a single computer program. Distillation columns with multiple feeds and side streams, absorbers, and reboiled absorbers have been solved successfully by this procedure. Both the rating and the design of these processes have been accomplished using the same set of basic equations.

The present work applies the general solution in obtaining rigorous solutions for processes handling petroleum fractions by means of the integral technique. The solutions satisfy all the material balance, heat balance, and equilibrium relationships within the process.

THE INTEGRAL TECHNIQUE IN EQUILIBRIUM FLASH VAPORIZATION CALCULATIONS

The simple distillation process, equilibrium flash vaporization, is used to illustrate the details of the integral technique.

The petroleum fraction must be defined by a molar true boiling point curve consisting of a plot of temperature in degrees Farenheit versus the mole fraction of the original mixture distilled. All calculations are based on this molar true boiling point curve and result in products defined by similar curves. Methods (3) are available for converting standard distillation curves such as the ASTM distillation to molar true boiling point curves.

The material balance in an equilibrium flash vaporization for a component represented by a point on the TBP curve for the feed is

$$F dm_F = V dm_V + L dm_L \tag{1}$$

Since m_F is the total mole fraction distilled up to temperature t on the TBP curve of the feed, the differential dm_F represents the mole fraction in the feed of the component having a true boiling point of t. Analogous meanings are associated with dm_L and dm_V. The equilibrium relationship for this component is

$$dm_V = K dm_L \tag{2}$$

Hence

$$dm_L = \frac{F dm_F}{L + KV} \tag{3}$$

Integration of Equation (3) over the range of all components in the feed gives

$$\int_0^1 dm_L = 1 = \int_0^1 \frac{F dm_F}{L + KV} \tag{4}$$

In one form of the flash problem, the flash pressure and the quantities of liquid and vapor produced are specified. The flash temperature and the characteristics of the liquid and vapor products must be determined. The solution is obtained by trial and error. Values of the flash temperature are assumed until Equation (4) is satisfied.

Any of the standard techniques can be employed for the numerical integration of Equation (4). The well-known Simpson rule (4) is used to illustrate the principles of the integral technique.

The interval of integration includes all the components in the feed. The quantity m_F takes on values from 0 to unity. This corresponds to components with true boiling points ranging from the initial boiling point to the final boiling point of the mixture.

When this interval of integration is broken into n subintervals for the application of Simpson's rule, the integral is approximated by a summation as follows:

$$\int_0^1 \frac{dm_F}{L + KV} \simeq \frac{1/3n}{(L + KV)|_{t_1}} + \sum_{i=2}^{n} \frac{(1/3n)\{3 + (-1)^i\}}{(L + KV)|_{t_i}} + \frac{1/3n}{(L + KV)|_{t_{n+1}}} \tag{5}$$

The notation $(L + KV)|_{t_i}$ indicates that the distribution coefficient K is evaluated for that component whose TBP is t_i. For example, $K|_{t_1}$ is the value of K for the component whose TBP is the initial boiling point of the mixture. If 10 subintervals are employed for approximating the integral, $K|_{t_2}$ is the value of K for the component whose TBP corresponds to a value of 0.1 for m_F on the TBP curve of the feed.

The equation corresponding to Equation (4) for a finite mixture is

$$\sum_{i=1}^{c} \frac{F X_{Fi}}{L + K_i V} = 1 \tag{6}$$

The quantities $1/3n$, $(1/3n)\{3 + (-1)^i\}$, $1/3n$ appearing in Equation (5) might be considered the compositions of a pseudofeed containing $n + 1$ components. The integral can then be expressed as

$$\int_0^1 \frac{dm_F}{L + KV} \simeq \sum_{i=1}^{n+1} \frac{X'_{Fi}}{(L + KV)|_{t_i}} \tag{7}$$

That is

$$\begin{aligned} X'_{F1} &= X'_{F,n+1} = 1/3n \\ X'_{Fi} &= (1/3n)\{3 + (-1)^i\} \quad \text{for} \quad 2 \leq i \leq n \end{aligned} \tag{8}$$

In this manner the integral technique can be used in standard programs written for finite mixtures. A means of calculating the vapor-liquid equilibrium constants must be provided. Hadden and Grayson (5) present nomographs for K values of petroleum fractions as functions of TBP, temperature, pressure, and convergence pressure. These can be fitted to empirical equations for use in the calculations.

The various points on the TBP curve that characterizes the liquid product of a flash process are obtained by integration of Equation (3) from the initial boiling point where $m_F = 0$ to the value of m_F corresponding to the TBP of the point in question. That is

$$m_L|_t = \int_0^{m_F|_t} \frac{F dm_F}{L + KV} \tag{9}$$

The molar TBP curve of the vapor product is found by a similar operation.

$$m_V|_t = \int_0^{m_F|_t} \frac{K F dm_F}{L + KV} \tag{10}$$

The procedure using a pseudofeed composition in the evaluation of the integral in Equation (4) cannot be used for Equations (9) and (10). Simpson's rule must be reapplied in the appropriate manner over each of the desired intervals.

GENERAL SOLUTION FOR DISTILLATION PROCESSES

The general solution is summarized for systems containing a finite number of components. The equations are then adapted for use in the integral technique as was done for the equilibrium flash vaporization process.

The material balance and heat balance equations as written for the general solution provide for the possibility of vapor and liquid feeds and side draws on each stage. The component-material balance for component i on stage number j is

$$l_{j-1,i} - w^L_{j-1,i} + f^L_{ji} + v_{j+1,i} - w^V_{j+1,i} + f^V_{ji} = v_{ji} = v_{ji} + l_{ji} \tag{11}$$

Stage number j with the various entering and leaving streams is shown in Figure 1.

Use of the equilibrium relationship

$$l_{ji} = \frac{L_j}{V_j K_{ji}} v_{ji} = A_{ji}\, v_{ji} \tag{12}$$

and the side stream ratios

$$w^L_{j-1,i} = \frac{W^L_{j-1}}{L_{j-1}} l_{j-1,i} \tag{13}$$

$$w^V_{j+1,i} = \frac{W^V_{j+1}}{V_{j+1}} v_{j+1,i} \tag{14}$$

gives the following form of the component-material balance equation:

$$- \left[1 - \frac{W^L_{j-1}}{L_{j-1}}\right] A_{j-1,i} v_{j-1,i} + (1 + A_{ji}) v_{ji} - \left[1 - \frac{W^V_{j+1}}{V_{j+1}}\right] v_{j+1,i} = f^V_{ji} + f^L_{ji} \tag{15}$$

The general solution utilizes the linearity of the component-material balance equations with respect to component flow rates. There is an equation of the form of Equation (15) for each stage in the process for each component. For any one component there is a set of N equations where N is the number of stages. These equations can be solved for the N values of v_{ji} for the component in question. The same applies for each component in the system. These results, combined with equations of the form of Equation (12), give each of the quantities l_{ji} and v_{ji} in terms of the absorption factors A_{ji}, the liquid and vapor rates L_j and V_j, and the side stream rates $W^L{}_j$ and $W^V{}_j$.

The internal vapor rates can be eliminated by means of total material balances. If the side stream rates are given, the quantities l_{ji} and v_{ji} are functions only of the stage temperatures T_j and the internal liquid rates L_j.

The bubble point equation is used to describe the equilibrium requirement of a stage. The preferred form is

$$\sum_{i=1}^{c} K_{ji}\, l_{ji} = L_j \tag{16}$$

The heat balance equation for stage number j is

$$\sum_{i=1}^{c} (l_{j+1,i} - w^L_{j-1,i}) h_{j-1,j} + \sum_{i=1}^{c} f^L_{ji} h^F_{ji} + \sum_{i=1}^{c} (v_{j+1,i} - w^V_{j+1,i}) H_{j+1,i} + \sum_{i=1}^{c} F^V_{ji} H^F_{ji} + Q_j = \sum_{i=1}^{c} l_{ji} h_{ji} + \sum_{i=1}^{c} v_{ji} H_{ji} \tag{17}$$

The solutions for l_{ji} and v_{ji} obtained from the component-material balances are substituted into the bubble point and heat balance equations. After the equilibrium constants and the enthalpies have been expressed in terms of the stage temperatures, the final equations contain only the stage temperatures, the condenser duty, the reboiler duty, and the internal liquid rates as unknowns. As above, the vapor rates V_j are eliminated by means of total material balances. If the product and reflux rates are given, there are N unknown temperatures and $N - 2$ unknown liquid rates. The two heat duties Q_C and Q_R give a total of $2N$ unknowns. Since there is a bubble point equation and a heat balance for each stage, there is a total of $2N$ equations. The number of equations and unknowns is small enough that simultaneous solutions can be obtained by means of the Newton-Raphson technique. The size of the column that can be solved is limited only by the size of computer available.

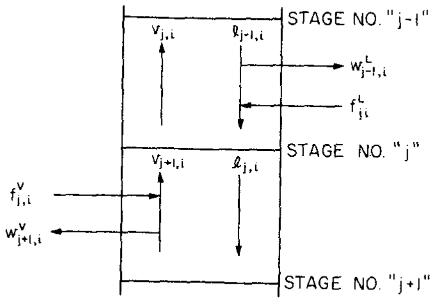

Fig. 1. Scheme for the component-material balance for stage number *j*.

The partial derivatives required for a Newton-Raphson solution must be obtained numerically. Care must be taken in the solution of the component-material balance equations. Light and heavy components must be handled in different ways because of the magnitudes of the numbers in the calculations. Both of these procedures are described in reference 2.

An important characteristic of the general solution is its ability to solve design problems directly. Other techniques often have poor convergence properties in this type of solution. The basic procedure in the general solution remains unchanged for handling design problems. The specification of the compositions of two key components adds two equations. It is therefore possible to find the reflux rate and the distillate rate required to give the desired separation.

APPLICATION OF THE GENERAL SOLUTION TO PETROLEUM FRACTIONS

The general solution is readily adapted for solving problems involving an indefinite number of components by means of the integral technique. This adaptation is accomplished by the use of a pseudofeed that has the proper composition for use in the summations which appear in the application of Simpson's rule.

Consider the bubble point equation for stage number j. Equation (16) is the proper form for a finite mixture. For an integral mixture, the equation becomes

$$\int_0^1 K_j L_j dm_{L_j} = L_j \tag{18}$$

The set of component-material balance equations for component i represented by Equation (15) gives a solution for v_{ji} that depends only on the absorption factors A_{1i}, A_{2i}, . . ., A_{Ni} for that component and the feed quantities $f^V{}_{ji}$ and $f^L{}_{ji}$. For the special case of only one bubble point liquid feed on stage number k, this solution for v_{ji} might be represented by

$$v_{ji} = \Phi_i\, f^L{}_{ki} \tag{19}$$

where Φ_i is the proper function of all the absorption factors for component i. The procedure for calculating v_{ji} is

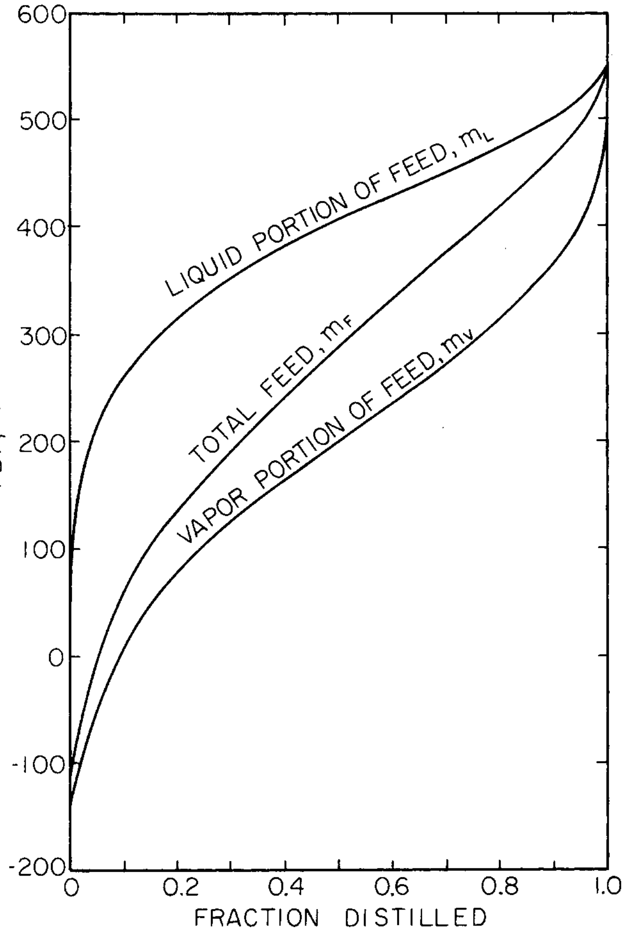

Fig. 2. TBP curves for the total feed and the liquid and vapor portions of the flashed feed for example 1.

given in reference 2. Equation (15) for the other components gives a similar solution for the other component vapor rates. From Equation (12)

$$l_{ji} = A_{ji}\,\Phi_i\,f^L{}_{ki} \tag{20}$$

The form of this equation for a mixture of an indefinite number of components is

$$L_j dm_{Lj} = A_j \Phi F dm_F \tag{21}$$

Substitution in Equation (18) gives

$$\int_0^1 K_j A_j \Phi F\, dm_F = L_j \tag{22}$$

The procedure for performing this integration with respect to m_F by Simpson's rule is the same as the procedure used to evaluate the integral in Equation (7). Equation (22) illustrates the principle involved when there is only one feed. For multiple feeds, Equation (22) would have the form of a sum of integrals, one for each feed. Otherwise the solution for such a case would be identical. Evaluation of each of the terms in the heat balance, Equation (17), is analogous.

The correct pseudofeed compositions to be used when breaking the interval $0 \leq m_F \leq 1$ into 10 subintervals are given in Table 1. This table also gives the proper point on the TBP curve for identifying each component by its normal boiling point. Once the boiling point of each of the pseudocomponents is found from the TBP curve of the feed, it is possible to evaluate the vapor-liquid equilibrium constants (5) and the enthalpies (6) at the stage temperatures and pressures. In this manner all of the equations and all of the operations of the general solution can be applied to the integral technique. The procedure for obtaining the solution is the same as for mixtures with a finite number of components.

The TBP curves for each of the products can be determined from values obtained in the final solution. Consider a conventional column with a single feed having pseudocomponent rates $f^L{}_{ki}$ and producing only distillate and bottom products. The solution gives values of d_i and b_i for the components of the pseudofeed. From these, the quantities $d_i/f^L{}_{ki}$ and $b_i/f^L{}_{ki}$ can be calculated. The corresponding quantities for a petroleum fraction are Ddm_D/Fdm_F and Bdm_B/Fdm_F, respectively, at the various points

TABLE 1. PSEUDOFEED COMPOSITIONS USED IN NUMERICAL INTEGRATION BY SIMPSON'S RULE

Component No.	Composition of pseudofeed, mole fraction	Value of m_F on the TBP curve to find the boiling point of the component
1	1/30	0.0
2	4/30	0.1
3	2/30	0.2
4	4/30	0.3
5	2/30	0.4
6	4/30	0.5
7	2/30	0.6
8	4/30	0.7
9	2/30	0.8
10	4/30	0.9
11	1/30	1.0

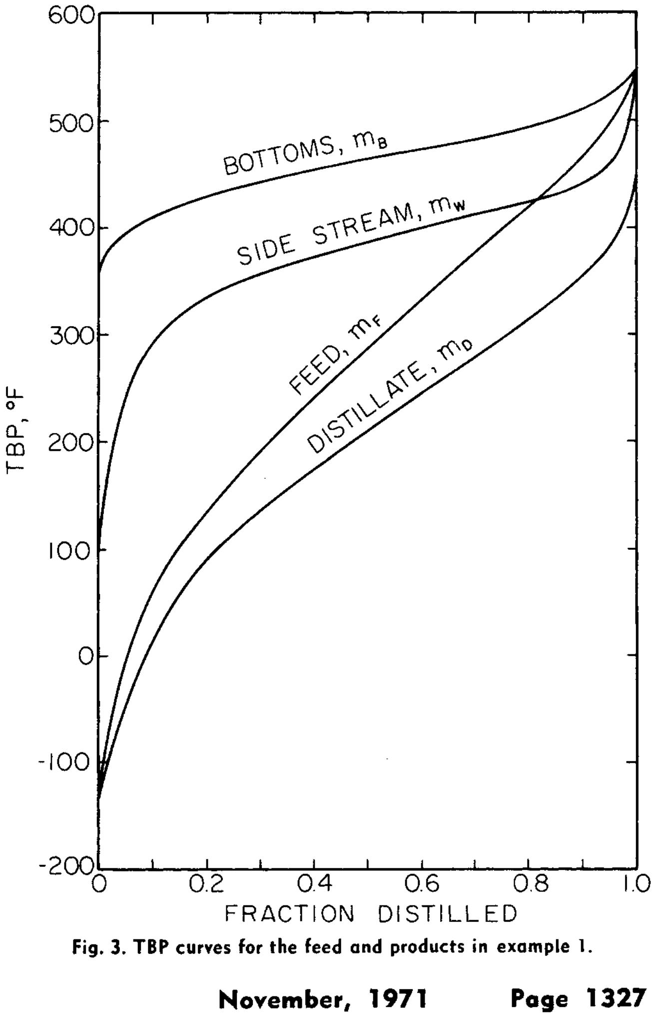

Fig. 3. TBP curves for the feed and products in example 1.

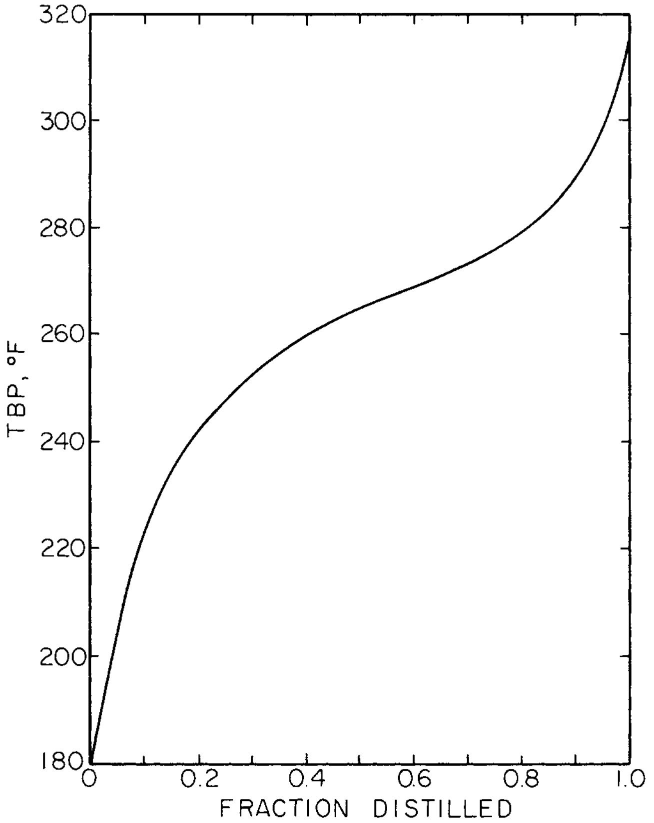

Fig. 4. TBP curve of the lean oil feed to the absorber in example 2.

on the distillation curve.

The TBP curves for the distillate and bottom products can then be obtained by integration of the equations

$$m_D|_t = \int_0^{m_F|_t} (dm_D/dm_F)\,dm_F \qquad (23)$$

$$m_B|_t = \int_0^{m_F|_t} (dm_B/dm_F)\,dm_F \qquad (24)$$

EXAMPLE PROBLEMS

Example 1 is a gasoline rerun column with a side stream. The TBP curve for the feed is given in Figure 2. The molecular weights and gravities of the various pseudocomponents were determined from Smith and Watson (*6*) using a characterization factor of 11.8. The vapor-liquid equilibrium constants were taken from Hadden and Grayson (*5*) using a convergence pressure of 5,000. Enthalpy data are from Bauer and Middleton (*7*).

The feed is 60% flashed on entering the column. A flash calculation of the type described after Equation (4) gives a flash temperature of 315°F. The column pressure and hence the flash pressure is 22 lb./sq.in.abs. Figure 2 shows the TBP curves of the feed and the liquid and vapor portions of the flashed feed. These curves are the result of the flash calculations described above.

The feed rate, overhead, sidestream, and bottom product rates are specified as 806, 540, 110, and 156 moles, respectively. The reflux rate is 240 moles. The overhead product is a vapor, and the sidestream is a liquid withdrawn below stage 3. The initial assumptions for stage temperatures were values that were linear with stage number from 250° to 500°F. The initial liquid rates were 240 moles above the side stream, 130 moles between the side stream and the feed, and 452 moles below the feed.

The solution of Example 1 is presented in Table 2 and Figure 3. Three complete trials were required to obtain these results.

Example 2 illustrates the use of the solution in absorber design. One feed, the vapor, contains a finite number of components with given compositions. These are listed in Table 3. The other feed, the absorber oil, is a petroleum fraction characterized by the TBP curve given in Figure 4. The absorber has eight ideal stages and operates at a pressure of 300 lb./sq.in.abs. There are 100 moles of rich vapor feed at 100°F. The lean oil is at 90°F. The principal objective is to determine the amount of absorber oil required to give 0.5% n-C_4 in the overhead vapor. The initial assumptions for temperatures were linear with stage number from 90° to 100°F. The initial liquid rates were also linear with stage number from 25 to 35 moles.

The final solution for the lean vapor composition is given in Table 3. The final temperatures and liquid rates are given in Table 4. This process requires 29.4 moles of absorber oil to produce 0.5% n-C_4 in the overhead vapor.

The equilibrium data for C_1, C_2, C_3, C_4, C_5, and C_7 were calculated for 300 lb./sq.in.abs. with equations from Holland (*8*). The values of K for the petroleum fraction were calculated using values for b taken from Hadden and Grayson (*5*). The quantity b is employed in the equation

$$K|_t = \frac{K_{n\text{-heptane}}}{(K_{\text{ethane}}/K_{n\text{-heptane}})^b} \qquad (25)$$

Enthalpy data for the components in the vapor feed were calculated from information given by Holland (*8*). Enthalpies of the components in the absorber oil were taken from Bauer and Middleton (*7*), again using a characterization factor of 11.8. Enthalpy values were fitted to equations in temperature for convenient use in the calculations.

Table 2. Solution of Example 1 Column with a Side Stream

Stage No.	Temp., °F.	Liquid rate, moles
1 (Condenser)	282.7	240.0
2	320.9	237.0
3	335.0	219.9
4	346.0	99.9
5	351.4	93.4
6	354.7	90.5
7	406.9	525.0
8	440.9	567.1
9	464.0	578.6
10 (Reboiler)	487.5	156.0

Reboiler duty = 9.34 × 10^6 B.t.u.; condenser duty = 5.89 × 10^6 B.t.u.

Table 3. Feed and Product Compositions for Absorber Example

Component	Feed vapor	Product vapor
CH_4	0.65	0.787
C_2H_6	0.13	0.135
C_3H_8	0.10	0.070
n-C_4	0.08	0.005
n-C_5	0.04	0.20×10^{-5}
Absorber oil	——	0.003

Table 4. Solution of Example 2 Absorber Design

Stage No.	Temp., °F.	Liquid rate, moles
Lean oil feed	90.0	29.4
1	117.9	37.4
2	129.7	39.4
3	137.7	40.5
4	144.0	41.3
5	149.2	41.9
6	153.1	42.7
8	154.2	44.1
7	147.4	49.7

CONCLUSIONS

The proposed method for handling complex petroleum mixtures in distillation processes provides a mathematically consistent technique for selecting the pseudocomponents and performing each numerical integration. The usual manner of choosing pseudocomponents (9) by roughly equalizing areas on the TBP curve has little meaning in the integration of the bubble point and heat balance equations. In the new technique the lightest and heaviest components in the mixture are pseudocomponents. These components may have considerable influence on the values obtained for the equilibrium temperatures, depending on the shape of the distillation curve.

The proposed method can be employed in existing programs for mixtures of a finite number of components. The only addition required is the determination of the distillation curves for the products. This is not needed until the final solution has been obtained for temperatures and flow rates throughout the process.

NOTATION

A_{ji} = absorption factor for component i on stage j, defined by $A_{ji} = L_j/(V_j K_{ji})$

c = total number of components in the system

D, B = total molal rate of the distillate and bottom products, respectively

d_i, b_i = molal rate of component i in the distillate and bottoms, respectively

F = total molal feed rate

$f^L{}_{ji}$ = molal rate at which component i enters the column in the liquid part of the feed above stage j

$f^V{}_{ji}$ = molal rate at which component i enters the column in the vapor part of the feed below stage j

h_{ji}, H_{ji} = enthalpy of one mole of a pure component i in the liquid and vapor states, respectively, at the temperature and pressure of stage j

H^F_{ji} = enthalpy of one mole of component i in the vapor feed below stage j

h^F_{ji} = enthalpy of one mole of component i in the liquid feed above stage j

K = vapor-liquid equilibrium constant

K_{ji} = the value of K for component i on stage j

$K|_t$ = the value of K for the component whose TBP = t

L = molal liquid rate

L_j = the total rate of flow of the liquid from stage j

l_{ji} = molal rate at which component i in the liquid state leaves stage j

m_B, m_D, m_W = mole fraction distilled on a molar TBP distillation curve of the bottom product, the distillate, and the side stream product, respectively, of a distillation process

m_F = mole fraction distilled on a molar TBP distillation curve of the feed stream

m_L, m_V = mole fraction distilled on a molar TBP distillation curve of the liquid and vapor products, respectively, of an equilibrium flash vaporization

n = number of subintervals employed in the numerical approximation of an integral by Simpson's rule

Q_C = condenser duty

Q_j = heat added to stage j from an external source

Q_R = reboiler duty

V = molal vapor rate

V_j = the total rate of flow of the vapor from stage j

v_{ji} = molal rate at which component i in the vapor state leaves stage j

$W_j{}^L, W_j{}^V$ = total molal rate of flow of the liquid side stream below stage j and the vapor side stream above stage j, respectively

w^L_{ji}, w^V_{ji} = molal rate at which component i leaves the column in the liquid side stream below stage j and the vapor side stream above stage j, respectively

x = mole fraction of a liquid phase

x_{ji} = the composition of component i in the liquid stream leaving stage number j

X_{Fi} = mole fraction of component i in the feed stream

X'_{Fi} = the composition of a pseudofeed used in the integral technique. See Equation (8)

Greek Letter

Φ = function of the absorption factors. See Equation (19) and the following text

Subscripts

i = component number, $i = 1$ to $i = c$

j = stage number from top to bottom. When they are included in the process, the overhead condenser is stage 1 and the reboiler is stage N

t indicates that the associated variable is evaluated for the component whose TBP is t

Superscripts

F specifies that the associated enthalpy is for a feed stream

L, V specifies that the associated quantity is for a liquid and a vapor stream, respectively

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