DISTILLATION CALCULATION BY USE OF CONJUGATED GRADIENT METHOD

TAKAITSU IWATA, YUICHI SATO, MITSUHIRO NAKAMURA
AND HIDEO UEMURA

Second Plant Engineering Department of Mitsui
Shipbuilding and Engineering Co., Ltd., Osaka, 559
IKUHO YAMADA*

Department of Industrial Chemistry, Nagoya Institute of
Technology, Nagoya, 466

Distillation calculation by the nonlinear optimization technique is proposed. This method has the following objective function:

$$\phi(t) \equiv \sum w_j (X_j - 1)^2 \longrightarrow \min.$$

where t denotes the temperature vector, w_j the weight factor and $X_j \equiv \sum x_j(i)$, and $x_j(i)$ is obtained by solving the tridiagonal matrix by making the component material balance for a given t . The temperature vector is corrected by the conjugated gradient method suggested by Fletcher and Reeves.

From three numerical examples related to the separation of nC_5 and nC_6 fraction from $nC_5 \sim nC_6$ mixture at atmospheric pressure, that of C_2 fraction from $C_2 \sim C_3$ mixture at high pressure and extractive distillation, it has been confirmed that the proposed method provides a stable solution as compared with the matrix methods by Amundson *et al.* and Saito *et al.*, it retains the simplicity of the matrix method, and the storage required for the main array in this method is obviously less than that for the original gradient method by Naphtali *et al.*

The distillation process is a well-known unit operation to separate liquid mixtures and is widely used in the chemical and petroleum industries. Useful calculation methods to solve the operation problem for distillation may be essentially classified into the following items: matrix method^{1,11)}, θ -method⁶⁾, and relaxation method⁸⁾. Some attempts have recently been made to improve practical use both by combining matrix method with θ -method and by modifying the method itself with the advantage of electronic computer technics. Some attempts have been reviewed by several investigators^{5,12)}. In this paper a new approach using the conjugated gradient method, which provides the stable solution to the mass and heat balance equations, is proposed. This method retains simplicity as well as the Thomas algorithm used in the ordinary matrix method, and may be applicable to similar stagewise operations.

Formulation of Problem

For simplicity an ordinary distillation column with single-feed plate and without any side-cut streams is treated as the model column, and the feed is supplied as saturated liquid. Let us number the plates down from the top of column, so that the total condenser, the feed plate and the reboiler are assigned

the symbols 1, f and N , respectively. The fundamental equations for solving distillation problems are derived by making material and heat balances around j -th plate of the model.

Overall material balance:

$$L_{j-1} - (V_j + L_j) + V_{j+1} = -F_j \quad (1)$$

Component material balance:

$$L_{j-1}x_{j-1}(i) - (V_j y_j(i) + L_j x_j(i)) + V_{j+1} y_{j+1}(i) = -F_j x_{Fj}(i) \quad (2)$$

Heat balance:

$$L_{j-1} h_{j-1} - (V_j H_j + L_j h_j) + V_{j+1} H_{j+1} = -F_j h_j \quad (3)$$

where $F_j x_j = 0$, $F_j h_j = 0$ at $j \neq f$, $F_j x(i) = F x_F(i)$ and $F_j h_j = F h_F$ at $j = f$. On the other hand, the relationship between vapor and liquid compositions can be expressed by

$$y_j(i) = K_j(i) x_j(i) \quad (4)$$

where the assumption of ideal stage is used. Substituting Eq. (4) into Eq. (2), the component material balance is given by the following tridiagonal matrix:

$$Mx = F \quad (5)$$

where $x^T \equiv [x_1(i) \dots x_f(i) \dots x_N(i)]^T$ and $F \equiv [0 \dots -F x_F(i) \dots 0]^T$ and the elements of M are as follows:

$$m_{jj} \equiv -(V_j K_j(i) + L_j)$$

$$m_{11} \equiv -(L_1 + D)$$

Received March 7, 1975.

Fig. 1 Flow chart of calculation procedure

$$m_{NN} \equiv -(V_N K_N(i) + W) \quad (6)$$

$$m_{j,j-1} \equiv L_{j-1}$$

$$m_{j,j+1} \equiv V_{j+1} K_{j+1}(i)$$

Eq. (5) is a nonlinear simultaneous equation with respect to $x_j(i)$ and t_j , but this equation can be regarded as a linear equation if the element $m_{j,k}$ ($k=j-1, j, j+1$) is given by a constant, which can be solved by the Thomas method. This is the first characteristic of the original matrix method¹. However, the elements are affected by the temperature profile in the column. Therefore, the relationships $\sum x_j(i)=1$ and $0 < x_j < 1$ for all j are unsatisfied unless the profile is given exactly. Then, $x_j(i)$ is corrected by the treatment of normalization, and the elements $m_{j,k}$ are modified by calculating the bubble point with normalized $x_j(i)$ and by calculating Eq. (3) with new values of H_j^V and h_j^L obtained from the normalized $x_j(i)$. This procedure is the second characteristic of the original matrix method. But it can be shown that when the liquid mixture has nonideality this method will fail to converge in general. Saito *et al.*⁹ improved this method by using the concept of perturbation theory to correct the component activity coefficient. Tomich¹⁰ and Naphtali *et al.*⁷ proposed the matrix method with Newton-Raphson method to solve the material and the heat balances simultaneously.

To obtain a stable solution for the distillation problem, the conjugated gradient method by Fletcher and Reeves³ is herein applied.

Let us now define an objective function $\phi(\mathbf{t})$ as follows:

$$\phi(\mathbf{t}) = \sum w_j (X_j - 1)^2 \quad (7)$$

where $X_j \equiv \sum_i x_j(i)$, $\mathbf{t}$ is the liquid temperature vector and w_j denotes a weight factor for plate j . It is considered that the value of $\phi(\mathbf{t})$ must be zero when the calculation has converged with an infinite number of significant figures. Since the actual calculation is carried out by several number of significant figures, $\phi(\mathbf{t})$ has a finite value which depends upon the accuracy of calculation. Generally, $\phi(\mathbf{t})$ has a single peak around the solution.

Based on the consideration mentioned above, it will be concluded that to solve the distillation problem can be reduced to solving the nonlinear optimization problem with the following objective function:

$$\begin{cases} \phi(\mathbf{t}) - \min. \\ \text{Subject to Eqs. (3) and (5)} \end{cases} \quad (8)$$

Calculation Procedure

The calculation procedure of the present method is summarized by the flow chart shown in Fig. 1. The algorithm of the conjugated gradient method for correcting the temperature vector, $\mathbf{t}$, can be shown as follows:

For ν -th calculation step, the temperature gradient vector with respect to $\phi(\mathbf{t})$ is written as

$$\mathbf{g}^{(\nu)} = \frac{\partial \phi(\mathbf{t})}{\partial \mathbf{t}} = \begin{bmatrix} \frac{\partial \phi}{\partial t_1} & \frac{\partial \phi}{\partial t_2} & \dots & \frac{\partial \phi}{\partial t_N} \end{bmatrix} \quad (9)$$

Hence, the temperature vector is corrected by

$$\mathbf{t}^{(\nu)} = \mathbf{t}^{(\nu-1)} + \lambda_{\text{opt}}^{(\nu-1)} \mathbf{d}^{(\nu)} \quad (10)$$

where $\mathbf{d}^{(\nu)}$ is calculated by the following equations.

For initial step:

$$\mathbf{d}^{(1)} = -\mathbf{g}^{(1)} \quad (11)$$

where this equation implies that $\mathbf{t}$ is corrected in the direction of the maximum gradient

For general routine:

$$\mathbf{d}^{(\nu)} = -\mathbf{g}^{(\nu)} + \frac{\mathbf{g}^{(\nu)T} \mathbf{g}^{(\nu-1)}}{\mathbf{g}^{(\nu-1)T} \mathbf{g}^{(\nu-1)}} \mathbf{d}^{(\nu-1)} \quad (12)$$

where this equation shows that $\mathbf{t}$ is corrected in the direction of the conjugated gradient.

According to the original conjugated gradient method³, the calculation of Eq. (12) is repeated until $\nu = N'$, where N' shows number of variables for general problem. In this work, however, as suggested in Fig. 1, maximum value of ν is given by a finite number ρ which is less than N' , to eliminate the truncation error in Eq. (12), then, for $\nu = \rho + 1$ the step returns at $\nu = 1$. Furthermore, λ_{opt} in Eq. (10) to accelerate and stabilize the convergence is obtained by minimum values of the equation

$$\Omega(\lambda)=\phi(\overset{(\nu-1)}{t}+\overset{(\nu-1)}{\lambda d})\rightarrow\min. \quad (13)$$

where the golden section method is useful to obtain λ_{opt} . The graphical meaning of the presentation of λ_{opt} is shown in Fig. 2 for two variables of t_j .

This procedure is repeated until the convergence function used in the distillation calculation has converged within a certain accuracy.

The procedure to obtain $\partial\phi/\partial t_k$ is as follows: The differentiation of Eq. (7) gives

$$\frac{\partial\phi}{\partial t_k}=\frac{\partial}{\partial t_k}[\sum_j w_j(X_j-1)^2]=2\sum_j\left\{w_j(X_j-1)\sum_i\frac{\partial x_j(i)}{\partial t_k}\right\} \quad (14)$$

Furthermore, the calculation of $\partial x_j(i)/\partial t_k$ proceeds below. The differentiation of Eq. (5) leads to the following tridiagonal matrix.

$$M\dot{x}=z \quad (15)$$

where the element in M is already in Eq. (6). x, z and the elements in z are given as follows:

$$\dot{x}\equiv[\partial t_k(i)/\partial t_k\partial x_2(i)/\partial t_k\cdots\partial x_N(i)/\partial t_k]^T \quad (16)$$

$$z\equiv[z_{1k}z_{2k}\cdots z_{Nk}]^T \quad (17)$$

$$z_{11}=0$$

$$z_{k,k}=V_kx_k(i)\frac{dk_k(i)}{dt_k}$$

$$z_{k-1,k}=-V_kx_k(i)\frac{dk_k(i)}{dt_k} \quad (18)$$

$$z_{j,k}=0 \text{ for } j < k-2, j > k+1$$

where $dk_k(i)/dt_k$ can be calculated by the differentiation of Cox-Antoine's equation if the concept of perturbation theory⁹⁾ is applied to the component activity coefficient. For a given set value of t_j , V_j and L_j are calculated from Eq. (3) with the value of H^V and h^L obtained by the previous step, and $\partial x_j(i)/\partial t_k$ can be obtained by solving Eq. (15). Then, g is calculated by Eq. (9). Thus, the temperature vector is corrected by Eq. (10).

Discussion and Conclusion

To examine the proposed method, three numerical examples for calculating $x_D(i)$ and $x_W(i)$ under the given operating condition are illustrated.

The first example given in Table 1 is related to the separation of nC_5 and nC_6 fractions from $nC_5 \sim nC_9$ mixture at atmospheric pressure. Under the given condition of convergence, $|X_j-1|_{\max} < 10^{-3}$, the solution, number of iteration and C.P.U. time required by use of I.B.M. 370-168 are given in Table 2, where w_j is unity for all j . For this example much more C.P.U. time for the proposed method is required as compared with the primitive matrix method¹⁾.

The second example shown in Table 3 is related to the separation of C_2 fraction from $C_2 \sim C_3$ mixture at

Fig. 2 Graphical presentation of λ_{opt} for two variables of t_j

Table 1 Statement of numerical example 1

Component	<i>i</i>	$x_F(i)$	$A(i)$	$B(i)$	$C(i)$
nC_5	(1)	0.25	2.15752	221.383	65.755
nC_6	(2)	0.25	2.06306	234.633	51.669
nC_7	(3)	0.20	2.18817	338.723	67.551
nC_8	(4)	0.15	2.34773	462.208	82.981
nC_9	(5)	0.15	2.53973	610.673	99.707

$F=1.0 \quad D=0.5 \quad R=2.0 \quad N=12 \quad f=6 \quad q=1.0 \quad \Pi=1.0$

Table 2 Solution of numerical example 1

<i>i</i>	$x_D(i)$	$x_W(i)$	This method	Mat. method
(1)	0.5000	—		
(2)	0.4909	0.0093	No. of iterations 17	35
(3)	0.0091	0.3907		
(4)	—	0.3000	C.P.U. Time 2.81 sec	1.15 sec
(5)	—	0.3000	required	

Table 3 Statement of numerical example 2

Component	<i>i</i>	$x_F(i)$	$A(i)$	$B(i)$	$C(i)$
C_2H_4	(1)	0.64339	2.48956	173.497	181.642
C_3H_6	(2)	0.12805	2.48604	187.010	173.162
C_3H_4	(3)	0.00141	2.93288	455.269	215.704
C_3H_8	(4)	0.21634	2.47325	226.453	155.943
C_2H_8	(5)	0.01031	2.45346	234.968	158.263

$F=1.0 \quad D=0.7725 \quad R=3.0 \quad N=18 \quad f=9 \quad q=1.0 \quad \Pi=27.7 \text{ atm}$

Table 4 Solution of numerical example 2

<i>i</i>	$x_D(i)$	$x_W(i)$	This method	Mat. Method
(1)	0.8333	0.0008		
(2)	0.1651	0.0024	No. of iteration 17	122
(3)	—	0.0062		
(4)	0.0016	0.9454	C.P.U. Time 4.68 sec	4.87 sec
(5)	—	0.0452	required	

high pressure, where the Cox-Antoine constants, $A(i)$, $B(i)$ and $C(i)$ are correlated by using the K -value estimated from the method proposed by Chao and Seader²⁾. Under the same condition of convergence and w_j as in the first example, the solution is given in Table 4. The C.P.U. time required for the proposed method is almost the same as that for the primitive matrix method¹⁾.

The third example is related to the extractive distillation shown in Table 5, where constant molar rates of vapor and liquid are assumed. In this ex-

Table 5 Statement of numerical example 3

Component	<i>i</i>	$x_{F1}(i)$	$x_{F2}(i)$			
Acetone	(1)	0.000	0.50	4.41365	1161.00	224.000
Methanol	(2)	0.00	0.50	4.99782	1473.11	230.000
Water	(3)	1.00	0.00	5.08600	1668.21	228.000

Wilson's constants:

$$A_{12}=0.6924 \quad A_{13}=0.1652 \quad A_{23}=0.3852$$

$$A_{21}=0.7498 \quad A_{31}=0.4181 \quad A_{32}=1.0850$$

$$F_1=40 \quad F_2=20 \quad D=9.0 \quad R=2.0 \quad \Pi=1.0 \text{ atm}$$

$$N=16 \quad f_1=5 \quad f_2=12 \quad q_{F1}=1.0 \quad q_{F2}=1.0$$

Table 6 Solution of numerical example 3

<i>i</i>	$x_D(i)$	$x_W(i)$		This method	Mat. method
(1)	0.9260	0.0327	No. of iteration	33	not
(2)	0.0084	0.1946			converged
(3)	0.0656	0.7727	C.P.U. Time required	9.26 sec	—

ample, Eqs. (1)~(3) are corrected as follows:

$$F_j \neq 0 \quad \text{at } j=f_1, f_2$$

$$\begin{cases} F_j x_j(i) = F_{f1} x_{Ff1}(i), & F_{f2} x_{Ff2}(i) \\ F_j h_j = F_{f1} h_{f1}, & F_{f2} h_{f2} \end{cases} \quad \text{at } f=f_1, f_2$$

where suffix f_1 denotes the main feed and f_2 the solvent feed. For the same condition of convergence as in the examples 1 and 2, the solution is given in **Table 6**, where the iteration method by use of the concept of perturbation theory⁹⁾ has been applied to correct the component activity coefficient. Also, the variation of molar rate of liquid throughout the column has been considered to evaluate w_j in each section. In this example the solution by use of the matrix method⁹⁾ has not converged.

As observed in the solutions mentioned in Tables 2, 4 and 6, it can be shown that the proposed method has the remarkable advantage of a stable solution with a small number of iteration steps as compared with other matrix methods^{1,9)}.

It is well-known that the matrix method with Newton-Raphson method is useful to solve nonlinear simultaneous equations. Therefore, several investigators^{7,10)} have proposed the so-called gradient method to solve Eq. (5). This method, however, is not certain to obtain a stable solution in general when the variable increases. So some attempts⁴⁾ have been made to improve the original gradient method, whereas the present method provides a stable solution even when the number of components and theoretical plates increase. Moreover, in this work the storage required for the main array can be shown as follows:

h_j	N
H_j	N
$y_j(i)$	$N \times C_n$
$x_j(i)$	$N \times C_n$
$\gamma_j(i)$	$N \times C_n$
$K_j(i)$	$N \times C_n$
$P_j(i)$	$N \times C_n$

t_j	N
V_j	N
L_j	N
$\partial\phi/\partial t_k, (d, g, g)$	$3N$
X_j	N
Total	$N(5C_n+9)$

where C_n denotes the number of components. Thus, the total storage required in this work is obviously less than $N(2C_n^2+9C_n+6)$, which is required in the original gradient method⁷⁾.

Nomenclature

$A(i), B(i), C(i)$	= constants of component <i>i</i> in Cox-Antoine's equation, $\log P(i) = A(i) - B(i)/(t + C(i))$
D	= molar rate of distillate
$d^{(\nu)}$	= direction vector for correction of temperature given by Eqs. (11) and (12) at ν -th convergence step
F	= vector of molar rate of feed
$F^{(\nu)}$	= molar rate of feed
g	= temperature gradient vector for ϕ given by Eq. (9) at ν -th convergence step
H	= enthalpy of vapor phase
h	= enthalpy of liquid phase
$K(i)$	= <i>K</i> -value of component <i>i</i>
L	= liquid molar rate
M	= tridiagonal matrix of Eq. (5) and (15) with elements given by Eq. (6)
m_{ij}	= elements given by Eq. (6)
N	= total number of plates including total condenser and reboiler
$P(i)$	= vapor pressure of component <i>i</i>
q	= thermal factor of feed
R	= reflux ratio
t	= temperature vector
t	= temperature of liquid
V	= molar rate of vapor
W	= molar rate of bottoms
w	= weight factor in Eq. (7)
X	= $\sum x(i)$
$\dot{x}$	= vector of temperature gradient for $x(i)$ with the elements given by Eq. (16)
x	= vector of liquid composition
$x(i)$	= molar fraction of liquid of component <i>i</i>
$y(i)$	= molar fraction of vapor of component <i>i</i>
z	= vector in Eq. (15)
z_{ij}	= elements given by Eq. (18)
$\gamma(i)$	= activity coefficient of component <i>i</i>
A	= Wilson's constant
λ	= parameter in Eq. (13)
ν	= number of iterations
Π	= total pressure
ρ	= parameter to eliminate the truncation error in Eq. (12)
ϕ	= objective function given by Eq. (7)
Ω	= function given by Eq. (13)
<Subscripts>	
F	= feed
f	= feed plate
j, k	= <i>j</i> -th and <i>k</i> -th plates

Literature Cited

- 1) Amundson, N. R. and A. J. Pontinen: *Ind. Eng. Chem.*, **50**, 730 (1955).
- 2) Chao, K. C. and J. P. Seader: *AIChE J.*, **7**, 98 (1961).
- 3) Fletcher, R. and C. M. Reeves: *Computer Journal*, **7**, 149 (1964).
- 4) Hirose, Y. and M. Kudo: Preprint for 8th Autumn Meeting of The Soc. of Chem. Engrs. Japan, B-215, Tokyo (1974).
- 5) Holland, C. D.: "Multicomponent Distillation" Prentice-Hall Inc. (1963).
- 6) Lyster, W. N. and C. D. Holland: *Petrol Refiner*, **41**, 206 (1959).
- 7) Naphtali, L. M. and D. P. Sandholm: *AIChE J.*, **17**, 148 (1971).
- 8) Rose, A.: *Ind. Eng. Chem.*, **50**, 737 (1958).
- 9) Saito, H., H. Wada, S. Kanno and A. H. Sugie: *J. Chem. Eng. Japan*, **7**, 267 (1974).
- 10) Tomich, J. J. F.: *AIChE J.*, **16**, 229 (1970).
- 11) Wang, J. C. and C. E. Henk: *Hydrocarbon Processing*, **45**, 155 (1966).
- 12) Yamada I.: "Advanced Chemical Engineering", Vol. 7, p. 95, Maki Shyoten (1973).