D_H = value of D computed by Hermans (3)
D_L = diffusivity of sulfuric acid in water
D_V = diffusivity of water vapor in air
F = function symbol
G = function symbol
K = empirical equilibrium constant in droplet problem
k = an empirically-determined parameter in the fiber problem
R = dimensional boundary position in fiber
R_o = radius of cylindrical fiber
r = dimensional radial position in fiber
s = dimensionless radial position in fiber
s = dimensional radial position in droplet
T = a parameter corresponding to the time required for penetration to the center of fiber
t = dimensional time in fiber problem or in droplet problem
U = a transformed dependent variable in the droplet problem
u = dimensionless concentration in fiber problem
u = a transformed dependent variable in the droplet problem
v = auxiliary function
W = weight fraction sulfuric acid in water
w = weight fraction water vapor in air
x = dimensionless position in droplet

Greek Letters

β = phase growth parameter in droplet problem
ϵ = density function, $(\rho_V - \rho_L)/\rho_V$
θ = dimensionless time in fiber problem
θ = dimensional time in droplet problem
λ_n = an eigenvalue
ρ_L = liquid phase density
ρ_V = vapor phase density
ϕ = function symbol

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Effect of the Equilibrium Relationship on the Dynamic Characteristics of Distillation Column Sections

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The dependence of the dynamic characteristics of distillation column sections on the shape of the equilibrium relationship and the number of plates comprising the section is discussed. Dynamic characteristics, valid near the steady state operating condition, were obtained for simple column sections separating binary mixtures by a straightforward calculation with a digital computer. Sections with concave downward equilibria respond significantly more slowly than comparable sections with linear equilibria. Sections with concave upward equilibria respond faster. This behavior is explained in terms of the internal recycle within the section. The rate of propagation of a disturbance from end to end in the section is only weakly influenced by the shape of the equilibrium relationship.

A considerable body of literature exists concerning the control of distillation columns. The two major areas of investigation are the behavior of distillation systems during start up and the control of systems operating at steady state and perturbed by small changes in operating conditions. System dynamics and control loop design have been studied in each area. The recent literature has been well summarized by Rosenbrock (*1*, *2*) and by Archer and Rothfuss (*3*). This paper deals with the dynamics of systems operating at a steady state condition.

The complexity of the dynamic behavior of distillation systems is apparent if one considers the number of individual units which are tied together to make up the system. A system consists of one reboiler, one condenser, and one column section, plus one column section for each feed or product stream added or withdrawn between condenser and reboiler. In addition, each column section is composed of a number of individual stages.

The feedback among these units is legion. Within a section, a disturbance introduced on any plate travels toward each end of the section and is partly recycled at each plate owing to the countercurrent flow within the section. The coupling of the basic components provides a gross feedback among these components which further complicates the overall dynamic behavior. Representation of this behavior with Laplace transformation and signal flow diagrams used has been discussed by a number of authors [for example Mickley (*4*), Ceaglske (*5*)].

It would be desirable to be able to predict the dynamic characteristics of a distillation system from the steady state conditions at which the system operates. A correlation of this type, based on the results of computer simulation of distillation systems, was produced by Gilliland and Mohr (*6*). Use of the correlations to predict dynamic characteristics of two-section distillation systems gave good results within the limits of the variables considered. In view of the fact that detailed predictions of column dynamics are not necessary for operation of industrial systems (*7*), it was apparent in the course of the work that such a correlation covering the entire range of all pertinent parameters would be prohibitively difficult and costly to produce because of the interactions among the parameters. The most interesting result of this study was the large effect of the shape of the equilibrium relationship on the time constants of the distillation system. This paper deals specifically with this effect in a simple column section separating a binary mixture.

MODEL OF THE SECTION

Table 1. Assumed Restrictions on Model of System

1. Vapor holdup is insignificant.
2. Equal molal overflow is assumed.
3. Liquid holdup on each plate is the same.
4. Each plate is assumed to be a perfectly efficient, well-mixed equilibrium stage.
5. The time required for fluid to pass between plates is neglected.
6. Only small deviations from steady state are allowed so that the equilibrium relationship can be written in piece-by-piece linear form, $y_n = m_n x_n$.

The model of the column section consists of N component material balances, one around each plate. The restricting assumptions are listed in Table 1. Subject to these restrictions, the material balances are of the form

$$\frac{dx_n}{d\theta} = x_{n-1} - (1 + \phi_n)\, x_n + \phi_{n+1}\, x_{n+1},\ n = 1, N \tag{1}$$

Here x is the difference between the mole fraction of the more volatile component in the liquid at dimensionless time θ and that at the normal steady state condition, and ϕ is the slope of the equilibrium line at the liquid composition of the plate, divided by the liquid-to-vapor rate ratio. Laplace transformation of Equation (1) yields

$$X_{n-1} - \beta_n X_n + \phi_{n+1} X_{n+1} = 0 \tag{2}$$

where X is the Laplace transform of x and

$$\beta_n = 1 + s + \phi_n$$

Equations (2) form a tridiagonal set, as shown in Table 2.

LINEAR CASE

When ϕ_n (and hence β_n) is independent of n, that is when the equilibrium and operating lines are straight, the tridiagonal set of equations can be solved analytically to give the response of each product composition (x_N and y_1) to changes in each feed composition (x_o and y_{N+1}) [see Mickley (*4*)]. For example, a step change in x_c would result in a time dependent change in x_N of the form

$$C_N(\theta) = \frac{x_N(\theta) - x_{NF}}{x_{NI} - x_{NF}} = \sum_{j=1}^{N} A_j e^{(-\theta/\tau_j)} \tag{3}$$

where $C_N(\theta)$ is the normalized product composition. For this case, the N time constants [τ's in Equation (3)] depend only on the value of ϕ, in accordance with

$$\tau_j = \left[\frac{1}{1 + \phi - 2\sqrt{\phi}\cos\dfrac{j\pi}{N+1}}\right]_{j=1,N} \tag{4}$$

A representative plot of $C_N(\theta)$ vs. θ is shown in Figure 1. The response to a step change in x_o is slow initially, gradually becoming a pure single-exponential approach to the final steady state conditions as θ increases.

The N time constant response can be approximated by a single time constant, time lag response. This approximation is frequently made in designing control systems for industrial column and provides a simpler system which retains many of the features of the more complicated response function. The cited approximation yields a response function

$$\hat{C}_N(\theta) = 1, \quad \theta < \tau_L$$
$$\hat{C}_N(\theta) = \exp\left(\frac{-(\theta - \tau_L)}{\tau_1}\right), \quad \theta > \tau_L \tag{5}$$

where τ_1 is the largest of the N time constants appearing in Equation (3) and is obtained from Equation (4) with $j = 1$. The time lag (τ_L) may be associated physically with the finite time required for propagation of a disturbance through the section. Once the disturbance has reached the outlet, the entire section relaxes toward the final equilibrium state at an exponential rate determined by τ_1.

Table 2. Tridiagonal Set of Material Balance Equations

$$\begin{array}{llll}
-\beta_1 X_1 + \phi_2 X_2 & & & = -X_o \\
X_1 - \beta_2 X_2 + \phi_3 X_3 & & & = 0 \\
& \ddots & & \\
& & X_{N-2} - \beta_{N-1} X_{N-1} + \phi_N X_N & = 0 \\
& & X_{N-1} - \beta_N X_N & = -\phi_{N+1} X_{N+1}
\end{array}$$

$\hat{C}_N(\theta)$ is also plotted on Figure 1. τ_L can be evaluated from the intercept of the line with the ordinate 1.0 or from the intercept (A) at $\theta = 0$ and is related to the original time constants as follows:

$$\tau_L = \tau_1 \ln A \tag{6}$$

$$= -\tau_1 \sum_{k=2}^{N} \ln\left(1 - \frac{\tau_k}{\tau_1}\right) \tag{7}$$

The effect of ϕ on the value of τ_1 is shown in Figure 2, in which the major time constant for an eleven-plate section is plotted against ϕ. The maximum value of τ_1 is 14.9 at $\phi = 0.933$, corresponding roughly to parallel equilibrium and operating lines. A physical explanation of this maximum is offered later.

NONLINEAR CASE

For the more interesting case where the equilibrium relationship is not linear, the values of ϕ_n vary with n. The component material balances represented by Equation (1) and shown in Table 2 apply to this situation but cannot be solved analytically. Particular nonlinear systems can be studied by simulation of the response to a particular input disturbance, usually either a step change or sinusoidal fluctuation. Lamb et al. (*8*) describe an analogue system which can handle systems less restricted than the one considered here, and several papers have appeared on efficient digital simulation [See (*1*, *2*)].

For the restricted nonlinear system considered here, the tridiagonal set of equations shown in Table 2 can be solved by conventional methods [see Forsythe and Wasow (*9*)] for a given size section to yield the response transfer functions in terms of the β's and ϕ's. The general form of the transfer function $G(s)$ is

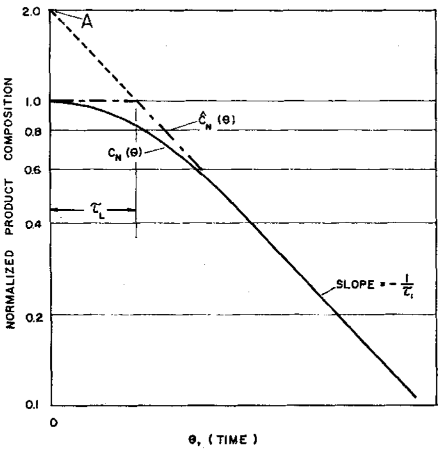

Fig. 1. Representative step response.

$$\frac{X_N}{X_o} = G(s) = \frac{K}{D(\beta_n, \phi_n)} \tag{8}$$

The function $D(\beta_n, \phi_n)$ is composed of N additive terms, one being the product $\prod_{n=1}^{N} (\beta_n)$ and the others being of lesser order in the β's. The N time constants associated with the transfer function are the negative reciprocals of the N poles of Equation (8). A method was developed for calculating the coefficients of the power series representation of $D(\beta_n, \phi_n)$ of the form

$$D(\beta_n, \phi_n) = \sum_{j=0}^{N} a_j s^j \tag{9}$$

directly from the values of ϕ_n. This procedure was incorporated into a digital computer program, which also utilized a general root-finding procedure to solve for the roots of the power series and hence the time constants associated with the column section. The development of the computational procedure and basic operation of the computer program are discussed in the Appendix.

All four column responses are governed by the same N time constants in both the linear and nonlinear cases and therefore give normalized step responses of the form given in Equation (3). However, only the two responses involving end-to-end propagation of the input disturbance behave as shown in Figure 1, and only for these responses is the single time constant, time lag approximation reasonable.

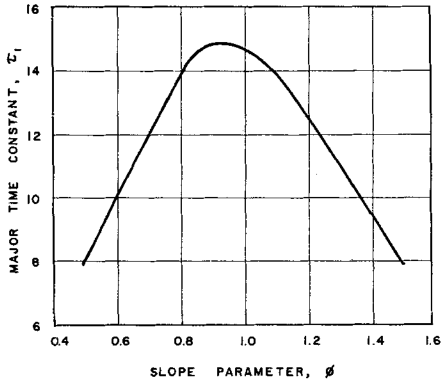

Fig. 2. Effect of the slope parameter on the major time constant.

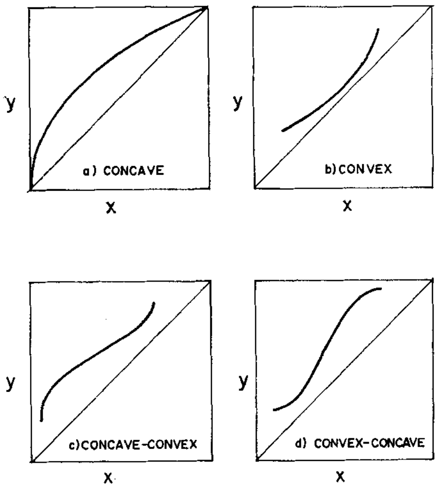

Fig. 3. Types of equilibria considered.

RESULTS AND DISCUSSION

A number of particular distillation systems were studied in order to demonstrate the effect of curvature of the equilibrium line on the major time constant τ_1 and time lag τ_L. The three types of equilibrium relationship considered are shown in Figure 3. The time constants of the system depend only on the values of ϕ_n corresponding to the steady state operating point. The variation of ϕ_n over the column section was taken to be of the form given in Table 3. The parameters which determine the time constants of the system are the values of k_n and some average value of ϕ over the section. The geometric average $\phi_A = \left(\prod_{n=1}^{N} \phi_n\right)^{1/N}$ is used here.

When $k = 1$, the equilibrium line is straight and ϕ_n is independent of n. The major time for the linear system, denoted τ_1^*, and the $N - 1$ lesser time constants can be obtained analytically from Equation (4). From the lesser time constants, the value of τ_L, denoted τ_L^*, can be calculated from Equation (7).

MAJOR TIME CONSTANT

Figure 4 shows the time constant ratio τ_1/τ_1^* as a function of k and ϕ_A for a five-plate section. Only results corresponding to convex and concave equilibria (k constant over each section) are shown. For a fixed value of ϕ_A, the major time constant varies inversely with k. The more concave the equilibrium curve, the slower is the response of the column section. It is interesting to note that the greatest effect occurs when ϕ_A is near unity, the same condition which produces a maximum time constant in linear systems.

TABLE 3. STATEMENT OF EQUILIBRIA CONSIDERED

I	Concave equilibria	
	$\phi_{n+1} = k\phi_n$	$k < 1, n = 1, N - 1$
II	Convex equilibria	
	$\phi_{n+1} = k\phi_n$	$k > 1, n = 1, N - 1$
III	S shaped equilibria	
	$\phi_{n+1} = k_n\phi_n$	$n = 1, N - 1$
	where $k_n = k^*$	$n = 1, \dfrac{N-1}{2}$
	$k_n = \dfrac{1}{k^*}$	$n = \dfrac{N+1}{2}, N - 1$

Values of $k^* < 1$ give the concave-convex curve (Figure 3c) and $k^* > 1$ gives the convex-concave curve (Figure 3d).

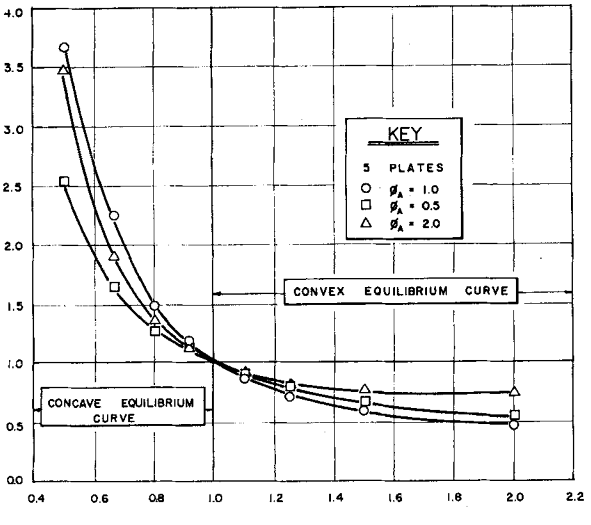

Fig. 4. Effect of equilibrium curvature on the major time constant.

The variation of τ_1 with k can be explained in terms of the relative rates of internal reflux. Consider a three-plate section as shown in Figure 5, and assume that a composition disturbance is introduced on the middle plate. In order for the section to return to its steady state condition, the disturbance must flow out of the ends of the sections causing changes in y_1 and x_3.

As x_2 changes, the compositions leaving plate 3 are affected. Any change in x_3 allows some of the disturbance to escape from the section, while a change in y_3 causes a recycle of the disturbance back to plate 2. The changes in y_3 and x_3 are related by the slope of the equilibrium line at the condition of plate 3, namely $\Delta y_3 = m_3 \Delta x_3$. Therefore, the relative magnitude of disturbance escape to disturbance recycle on plate 3 is

$$R_3 = \frac{L\Delta x_3}{V\Delta y_3} = \frac{L}{Vm_3} = \frac{1}{\phi_3}$$

Similar consideration of the top plate shows that the escape to recycle ratio is

$$R_1 = \frac{V\Delta y_1}{L\Delta x_1} = \frac{Vm_1}{L} = \phi_1$$

The disturbance will dissipate rapidly if both R_1 and R_3 are high, corresponding to a system in which the equilibrium curve is convex. Conversely, the response will be relatively slow if R_1 and R_3 are low, as is the case when the equilibrium curve is concave.

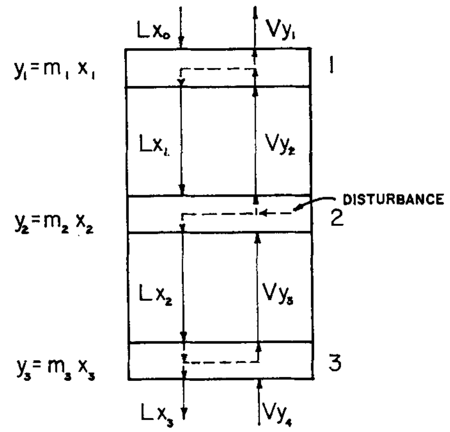

Fig. 5. Three-plate section.

This condition also explains the maximum value of τ_1 obtained near $\phi = 1$ for linear systems. R_1 increases with increasing ϕ, while R_3 decreases. The net R for the section can be thought of as some weighted sum of R_1 and R_3. R therefore must go through a minimum at an intermediate value of ϕ, corresponding to the condition of slowest response.

Figure 6 shows the effect on τ_1/τ_1^* of k for larger column sections. Results are given for five-, eleven-, and fifteen-plate sections, all systems having ϕ_A equal to unity. The magnitude of the variation with k increases with in-

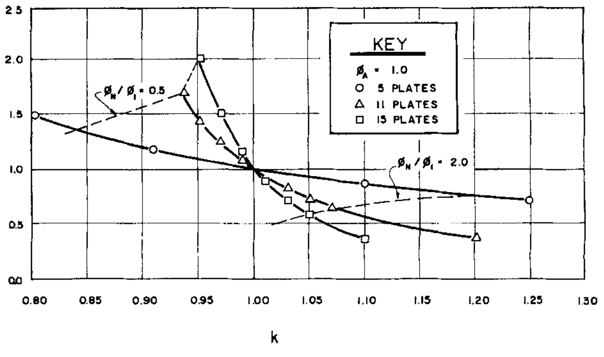

Fig. 6. Effect of section size on the major time constant.

creasing column size. Part of this increase in effect is due to the fact that total change in curvature of the equilibrium line for a given k is larger for larger columns. The dotted lines on Figure 6 connect points of equal ϕ_N/ϕ_1, which correspond to systems with the same total change in curvature. The figure shows that the major time constant can differ from that calculated with an overall average ϕ by as much as 50 or 60% in a fifteen-plate section with modest curvature of the equilibrium relationship.

Several runs were made with S shaped equilibrium relationships. The results are shown in Table 4. For the concave-convex relationship ($k^* < 1$), τ_1/τ_1^* is slightly less than unity. For convex-concave relationship ($k^* > 1$), τ_1/τ_1^* is greater than unity. However, even with quite severe departure from linearity, τ_1 was within ± 7% of the gross-linear approximation. The effects of the opposite curvatures tend to cancel. A system of this kind can be pictured as two smaller systems in series, one operating on a convex equilibrium and having a relatively rapid response, the other operating on a concave equilibrium and having a relatively slow response. In the limit, the response of the series would be that of the slower half. This response would approximate the response of the larger system which has, on the average, more favorable equilibrium curvature.

TIME LAGS

Baber et al. (*10*) compared experimental step response data obtained on a five-plate laboratory column with a simulated response based on the N time constant model used here. Although the response agreed well at long times following the step change, the short time responses were less consistent. This comparison indicates that the lesser time constants calculated from the model do a relatively poor job of defining the short time response, probably because the effect of transit time between plates is not included in the model.

Therefore, the values of τ_L obtained in this study show only the effect of the distributed nature of the system and do not include the important effect of fluid flow lags. The absolute values of τ_L are therefore of questionable value. However, the variation of the calculated values of τ_L with the curvature of the equilibrium relationship should be significant. Table 5 gives values of τ_L/τ_L^* as a function of k and ϕ_A. Over wide ranges of k, τ_L/τ_L^* varies relatively little, the maximum change being approximately 15%. It appears that the dependence of τ_L on the distributed nature of the system is relatively insensitive to the curvature of the equilibrium relationship.

TABLE 4. EFFECT ON MAJOR TIME CONSTANT OF S SHAPED EQUILIBRIUM CURVE

k^*	τ_1/τ_1^*
0.769	0.980
0.833	0.982
0.909	0.989
1.000	1.000
1.100	1.022
1.200	1.045
1.300	1.072

CONCLUSIONS

The shape of the equilibrium relationship was shown to have a strong effect on the major time constant governing the dynamic response of a distillation column section. The effect can be explained in terms of the relative rates of propagation and recycle of a disturbance at each plate within the section. The dependence of the lesser time constants on the shape of the equilibrium relationship is less clear. Combination of the lesser time constants into a time lag parameter allowed the overall effect to be considered in a straightforward manner. This effect arising from the distributed nature of the system was shown to be relatively insensitive to the shape of the equilibrium curve.

TABLE 5. RELATIVE VALUES OF TIME LAG τ_L/τ_L^*, FIVE-PLATE SECTION

k \ ϕ_A	0.5	1.0	2.0
0.5	0.96	0.88	0.86
0.8	0.99	0.96	0.97
1.00	1.00	1.00	1.00
1.25	0.99	1.00	0.98

NOTATION

a_n or $a_{n,j}$ = coefficient in power series [see Equations (9) and (A1)]
$C(\theta)$ = normalized composition response [see Equation (3)]
$\hat{C}(\theta)$ = approximate response of normalized composition [see Equation (5)]
$G(s)$ = transfer function of Laplace transform variable s
H = liquid holdup per plate, moles
k, k_n, k^* = ratio of slope parameters at adjacent plates (see Table 3)
L = liquid rate in moles per unit time
m_n = slope of the equilibrium curve at n^{th} plate
N = number of equilibruim stages in column section
R_n = rate of disturbance escape rate to disturbance recycle at the n^{th} plate
s = Laplace transform variable
S_n = power series representing the denominator of $G(s)$ [see Equation (A1)]

TABLE 6. SERIES EXPANSION OF $D(\beta_n, \phi_n)$

No. of plates (N)	$G(s) = \dfrac{K}{D(\beta_n,\phi_n)}$	$D(\beta_n,\phi_n)$ expanded into power series in s, denoted S_N
1	$\dfrac{1}{\beta_1}$	$S_1 = s + (1 + \phi_1)$
2	$\dfrac{1}{\beta_1\beta_2 - \phi_2}$	$S_2 = s^2 + (2 + \phi_1 + \phi_2)s + (1 + \phi_1 + \phi_1\phi_2)$
3	$\dfrac{1}{\beta_1\beta_2\beta_3 - \phi_2\beta_3 - \phi_3\beta_1}$	$S_3 = s^3 + (3 + \phi_1 + \phi_2 + \phi_3)s^2 + (3 + 2\phi_1 + \phi_2 + \phi_3 + \phi_1\phi_3 + \phi_2\phi_3 + \phi_1\phi_2)s + (1 + \phi_1 + \phi_1\phi_2 + \phi_1\phi_2\phi_3)$

t = time
V = vapor rate in moles per unit time
x_n = difference between the liquid composition on plate during transient period and that at the initial steady state prior to introduction of the disturbance
X_n = Laplace transform of x_n
y_n = difference in vapor composition; $y_n = m_n x_n$

Greek Letters

β_n = $1 + s + \phi_n$
θ = dimensionless time, Ht/L
τ = time constant in terms of dimensionless time θ
τ_1 = largest time constant associated with a column section
τ_1^* = largest time constant associated with a linear ($k = 1$) column section
τ_L = time lag in terms of dimensionless time θ
τ_L^* = time lag associated with a linear ($k = 1$) column section
ϕ_n = slope parameter at the n^{th} plate, Vm_n/L
ϕ_A = geometric average of ϕ_n over the column section

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APPENDIX

A general method for determining the time constants associated with a particular column section was developed by the following procedure:

1. The tridiagonal set of equations (Table 2) representing the Laplace transformed material balance equations was solved for one-, two-, and three-plate sections yielding the transfer functions $G(s) = X_N/X_o$ of the form shown in Equation (8):

$$G(s) = \frac{K}{D(\beta_n, \phi_n)} \tag{8}$$

2. The denominator of each transfer function was expanded into a power series in the Laplace variables.

The results obtained in parts 1 and 2 are shown in Table 6.

3. From the results shown in Table 6, a general recursion formula, with which the series coefficients for an N-plate section could be calculated from the coefficients corresponding to an $N - 1$ plate section, was established by induction. When one defines

$$S_n = \sum_{j=0}^{N} a_{n,j} s^j \tag{A1}$$

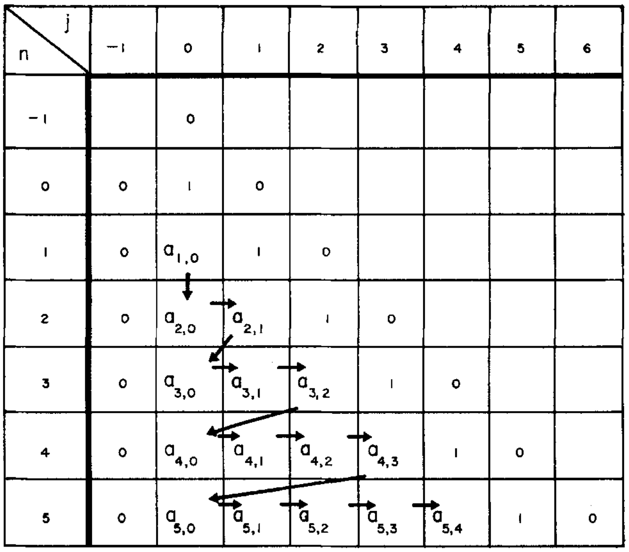
the recursion formula is

$$a_{n,j} = a_{n-1,j}(1 + \phi_n) + a_{n-1,\,j-1} - \phi_n a_{n-2,j} \tag{A2}$$
$$j = 0,\ n - 1$$

subject to the restrictions that

$$\left.\begin{aligned} a_{l,l} &= 1 \\ a_{l,-1} &= 0 \\ a_{l,-1,l} &= 0 \end{aligned}\right\} \quad l = 0,\ n \tag{A3}$$

The calculation procedure was programmed for a digital computer. The required input is the values of ϕ_n for $n = 1, N$ A coefficient matrix is built up in the computer, as shown in Figure 7 for a five-plate section. The ones and zeros are a permanent part of the matrix as required by Equation (A3), and the coefficients are generated in the order indicated. The final set of coefficients ($a_{5,j}$) are those required to specify the denominator of the transfer function.



n \ j	−1	0	1	2	3	4	5	6
−1		0						
0	0	1	0					
1	0	$a_{1,0}$	1	0				
2	0	$a_{2,0}$	$a_{2,1}$	1	0			
3	0	$a_{3,0}$	$a_{3,1}$	$a_{3,2}$	1	0		
4	0	$a_{4,0}$	$a_{4,1}$	$a_{4,2}$	$a_{4,3}$	1	0	
5	0	$a_{5,0}$	$a_{5,1}$	$a_{5,2}$	$a_{5,3}$	$a_{5,4}$	1	0

Fig. 7. Coefficient matrix for five-plate example.

Example

Consider a three-plate column section, characterized by ϕ_1, ϕ_2, and ϕ_3. The series representing the denominator of the transfer function is calculated as follows in accordance with Equation (A2):

$$n = 1 \quad a_{1,0} = 1(1 + \phi_1) + 0 - \phi_1(0) = 1 + \phi_1$$
$$n = 2 \quad a_{2,0} = (1 + \phi_1)(1 + \phi_2) + 0 - \phi_2(1)$$
$$= 1 + \phi_1 + \phi_1\phi_2$$
$$a_{2,1} = 1(1 + \phi_2) + (1 + \phi_1) - \phi_2(0)$$
$$= 2 + \phi_1 + \phi_2$$
$$n = 3 \quad a_{3,0} = (1 + \phi_1 + \phi_1\phi_2)(1 + \phi_3) + 0 - \phi_3(1 + \phi_1)$$
$$= 1 + \phi_1 + \phi_1\phi_2 + \phi_1\phi_2\phi_3$$
$$a_{3,1} = (2 + \phi_1 + \phi_2)(1 + \phi_3) + (1 + \phi_1 + \phi_1\phi_2) - \phi_3(1)$$
$$= 3 + 2\phi_1 + \phi_2 + \phi_3 + \phi_1\phi_3 + \phi_2\phi_3 + \phi_1\phi_2$$
$$a_{3,2} = 1(1 + \phi_3) + (2 + \phi_1 + \phi_2) - \phi_3(0)$$
$$= 3 + \phi_1 + \phi_2 + \phi_3$$

and

$$S_3 = s^3 + a_{3,2}\,s^2 + a_{3,1}\,s + a_{3,0}$$

Having evaluated the series coefficients, the computer program then finds the N roots of the series, using a general root-finding subroutine (*11*), and calculates the time constants as the negative reciprocals of the roots.

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