The Effects of the Degree of Liquid Mixing on Unsteady Performance Concerning Fluid Concentrations on Fractionating Plates

By

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In order to investigate the effects of the degree of liquid mixing on the dynamics concerning fluid concentrations on a fractionating plate, four kinds of transfer-functions describing the unsteady performance of vapor and liquid concentration were derived and analysed, using two "mixing models" for expressing the liquid mixing phenomena on a plate. Furthmore dynamics concerning unsteady fluid concentration change in the multistage fractionating plates were developed with the results obtained on a plate.

§ 1. Introduction

Dynamic performance of gas-liquid contacting plate columns concerning the concentration changes of flow have been investigated particularly for the distillation operation, assuming that vapor and liquid are perfectly mixed on each plate and that both leave each plate at equilibrium concentrations[1,8-10]. In large columns for industrial use, however, it is doubtful whether these two assumptions are satisfied.

In this paper, the effect of the degree of liquid mixing on the dynamics of a plate is discussed in relation to concentration change. It is most important to express analytically or in formulae the mixing phenomena on a plate so as to obtain the effect of the degree of liquid mixing on static and dynamic performances. It is impossible, however, to express this phenomena faithfully or strictly, because the mixing state on a plate is quite complicated, and an assumed reasonable, "model of mixing", must be introduced to express liquid mixing phenomena.

In this paper, only the problem of liquid mixing in the direction of liquid flow is taken account, while it is assumed that vapor and liquid in the direction of vapor flow are perfectly mixed on a plate where both fluids are cross-currently contacting each other.

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For liquid mixing phenomena in the direction of liquid flow, two familiar models such as the following are adopted, and the transfer-functions which give the unsteady behavior of vapor and liquid concentration changes are considered theoretically.

One mixing model is called the "perfect mixing model[5]", which states that one fractionating plate is equivalent to a series combination of n imaginary perfectly mixed vessels equally divided in the direction of liquid flow, as shown in Fig. 1. In this model, the state of perfect liquid mixing is represented by $n=1$. The state of liquid mixing on a plate becomes apart from the perfect mixing state with the increase of n, and finally the perfect piston flow state is accomplished when n tends to infinity.

The second model of liquid mixing is the well known "diffusional model" which states that "back mixing" occurs in proportion to the liquid concentration gradient in the direction of liquid flow so as to reduce the liquid concentration gradient. The liquid mixing degree is expressed by a "back mixing coefficient" E which is similar to an ordinary diffusion coefficient. The value of E represents the degree of liquid mixing, that is, perfect liquid mixing state is represented by $E=$infinity, and the degree of mixing decreases with the decrease in E. On the other hand, perfect piston flow is represented by $E=0$. The degree of liquid mixing on perforated plates and bubble cap trays[2], and also in multi-tubular heat exchangers[3,7], have been investigated by experimental methods with the diffusional model.

As will be mentioned in a separate paper[*], the expression of the degree of mass-transfer on a plate is to be considered more carefully than the problem of liquid mixing, and only the expression of K_La or K_Ga has been concluded to be reasonable in the consideration of dynamics by authors' investigation, while the Murphree efficiency was introduced by Wood and Armstrong[11]. Therefore in this paper, these overall mass transfer capacity coefficients are used in respect to the expression of the degree of mass transfer.

§2. Transfer-functions for concentration change by "perfect mixing model[4]"

In a fractionating plate in which vapor is in contact with liquid cross-currently and the hold-up of vapor is assumed to be negligibly small in comparison with that of liquid, the transfer-functions which express how a change in concentration of the inlet liquid or vapor, x_I or y_I in mole-fraction, is transferred to those of the outlet liquid or vapor, are considered in this section. With the perfect mixing model, it is supposed that a plate is equivalent to a system combined in series by n
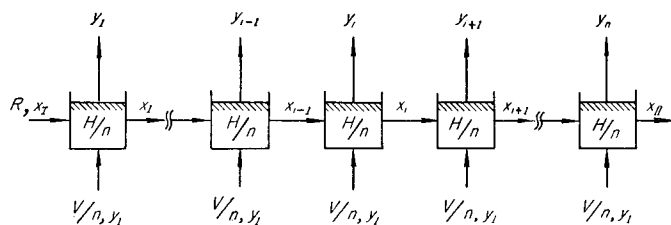
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Fig. 1. Material balance on a plate for "perfect mixing model"

perfectly mixed vessels as shown in Fig. 1, where the flow rates of liquid and vapor R and V (kg-moles/hr), the hold-up of liquid on a plate H (kg-moles) are constant for a given plate. Obviously from Fig. 1, in each vessel, the flow rates of liquid and vapor are R and V/n respectively, and the hold-up of liquid is H/n. In addition, it is assumed that chemical and physical properties, eg., the degree of mass-transfer and the relation of equilibrium between the concentrations of vapor and liquid are constant through all vessels.

In these conditions, the material balance or the equation of continuity for the i-th vessel (in which liquid and vapor are perfectly mixed in all directions) is given by

$$\frac{H}{n} \cdot \frac{dx(i, t)}{dt} = R[x(i-1, t) - x(i, t)] + \frac{V}{n}[y_I(t) - y(i, t)] \qquad (1)$$

Similarly the equation of mass-transfer rate in the liquid phase is given as follows.

$$\frac{H}{n} \cdot \frac{dx(i, t)}{dt} = R[x(i-1, t) - x(i, t)] - K_L a W[x(i, t) - x^*(i, t)] \qquad (2)$$

, where W (m³) is the volume occupied by fluids in a vessel, and $K_L a$ (kg-mole/m³ hr unit driving force) is the overall mass transfer capacity coefficient based on the liquid phase and $x^*(i, t)$ is an imaginary liquid concentration which is in equilibrium with the outlet vapor concentration $y(i, t)$.

Now the relation of equilibrium between the concentrations of vapor and liquid is supposed to be expressed by

$$y(i, t) = mx^*(i, t) + \text{constant} \qquad (3)$$

, where m is always constant.

The fluid concentrations x and y which are dependent on time, may be seperated into the steady-state terms x^0, y^0 which are independent of time and the unsteady-state terms Δx and Δy which are dependent on time, that is,

$$\begin{aligned} x(i, t) &= x^0(i) + \Delta x(i, t) \\ y(i, t) &= y^0(i) + \Delta y(i, t) \end{aligned} \qquad (4)$$

From equations (1), (2), (3) and (4)

$$\frac{\tau}{n}\cdot\frac{d\varDelta x(i,t)}{dt} = \varDelta x(i-1,t)-\left(1+\frac{1}{n}\cdot\frac{\lambda N_{0L}}{\lambda+N_{0L}}\right)\varDelta x(i,t)+\frac{1}{n}\cdot\frac{\lambda N_{0L}}{m(\lambda+N_{0L})}\varDelta y_I(t) \qquad (5)$$

, where τ, λ and N_{0L} are the residence time of liquid on a plate, stripping factor and overall number of transfer units of liquid phase respectively, and given by

$$\tau = \frac{H}{R}[hr], \quad \lambda = \frac{mV}{R}[-], \quad N_{0L} = \frac{nK_L aW}{R}[-]$$

Considering that the concentrations of the inlet liquid and vapor change around their steady-state $(t<0)$ values x_I^0 and y_I^0 by $\varDelta x_I(t)$ and $\varDelta y_I(t)$ at $t\geq0$, respectively, $\varDelta x(i,t)$ and $\varDelta y(i,t)$ are zero at $t=0$, and hence equation (5) may be rewritten as follows.

$$X(i-1,s)-\left(1+\frac{1}{n}\cdot\frac{\lambda N_{0L}}{\lambda+N_{0L}}+\frac{1}{n}\tau s\right)X(i,s)+\frac{1}{n}\cdot\frac{\lambda N_{0L}}{m(\lambda+N_{0L})}Y_I(s) = 0 \qquad (6)$$

, where $X(i,s)$ and $Y(i,s)$ are Laplace-transforms of $\varDelta x(i,t)$ and $\varDelta y(i,t)$ with time t, respectively, that is,

$$X(i,s) = L[\varDelta x(i,t)]$$
$$Y(i,s) = L[\varDelta y(i,t)]$$

From the difference equation (6) of i, the following equation (7) is obtained by using the boundary condition that $x(i,t)$ at $i=0$ is equal to the inlet liquid concentration $x_I(t)$.

$$X(i,s) = \frac{1}{\left(1+\frac{1}{n}\cdot\frac{\lambda N_{0L}}{\lambda+N_{0L}}+\frac{1}{n}\tau s\right)^i}X_I(s)$$
$$+\frac{\frac{1}{m}\cdot\frac{\lambda N_{0L}}{\lambda+N_{0L}}}{\tau s+\frac{\lambda N_{0L}}{\lambda+N_{0L}}}\left[1-\frac{1}{\left(1+\frac{1}{n}\cdot\frac{\lambda N_{0L}}{\lambda+N_{0L}}+\frac{1}{n}\tau s\right)^i}\right]Y_I(s) \qquad (7)$$

Equation (7) indicates how the outlet liquid concentration of the i-th vessel changes around its steady state value in response to an arbitrary change of the inlet liquid or vapor concentration by the Laplace transform. From equation (7), the response of the outlet liquid concentration is obtained by using the following boundary condition.

$$X_{II}(s) = X(i,s) \qquad \text{at} \quad i = n$$

For the response of the outlet vapor concentration, since the outlet vapor concentration of each vessel is different, the average concentration from a plate must be calculated by the following equation.

$$y_{II}(t) = \frac{1}{n}\sum_{i=1}^{n} y(i,t) \qquad (8)$$

The Laplace transform of the unsteady state term of equation (8) is given as follows.

$$Y_{II}(s) = \frac{1}{n} \sum_{i=1}^{n} Y(i, s) \qquad (9)$$

On the other hand, $Y(i, s)$ is obtained from equations (1), (2) and (3) as follows.

$$Y(i, s) = \frac{\lambda}{\lambda + N_{0L}} Y_I(s) + \frac{mN_{0L}}{\lambda + N_{0L}} X(i, s) \qquad (10)$$

From equations (7), (9) and (10), the equation describing how the outlet vapor concentration changes around its steady state value in response to an arbitrary change of the inlet liquid and vapor concentration at $t \geq 0$ is obtained in the form of the Laplace transform as follows.

$$Y_{II}(s) = \frac{\dfrac{mN_{0L}}{\lambda + N_{0L}}}{\tau s + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}}} \left[1 - \frac{1}{\left(1 + \dfrac{1}{n} \cdot \dfrac{\lambda N_{0L}}{\lambda + N_{0L}} + \dfrac{1}{n} \tau s \right)^{n}} \right] X_I(s)$$

$$+ \left[\frac{\lambda}{\lambda + N_{0L}} + \frac{\lambda \left(\dfrac{N_{b L}}{\lambda + N_{0L}} \right)^2}{\tau s + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}}} - \frac{\lambda \left(\dfrac{N_{0L}}{\lambda + N_{0L}} \right)^2}{\left(\tau s + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}} \right)^2} \left\{ 1 - \frac{1}{\left(1 + \dfrac{1}{n} \cdot \dfrac{\lambda N_{0L}}{\lambda + N_{0L}} + \dfrac{1}{n} \tau s \right)^{n}} \right\} \right] Y_I(s) \quad (11)$$

From equations (7) and (11), four kinds of transfer-functions describing the unsteady performances of fluid concentration are obtained.

$$G_1(s) = \frac{X_{II}(s)}{X_I(s)} = \frac{1}{\left(1 + \dfrac{1}{n} \cdot \dfrac{\lambda N_{0L}}{\lambda + N_{0L}} + \dfrac{1}{n} \tau s \right)^{n}} \qquad (12)$$

$$G_2(s) = \frac{Y_{II}(s)}{X_I(s)} = \frac{\dfrac{mN_{0L}}{\lambda + N_{0L}}}{\tau s + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}}} \left[1 - \frac{1}{\left(1 + \dfrac{1}{n} \cdot \dfrac{\lambda N_{0L}}{\lambda + N_{0L}} + \dfrac{1}{n} \tau s \right)^{n}} \right] \qquad (13)$$

$$G_3(s) = \frac{X_{II}(s)}{Y_I(s)} = \frac{\dfrac{\lambda N_{0L}}{m(\lambda + N_{0L})}}{\tau s + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}}} \left[1 - \frac{1}{\left(1 + \dfrac{1}{n} \cdot \dfrac{\lambda N_{0L}}{\lambda + N_{0L}} + \dfrac{1}{n} \tau s \right)^{n}} \right] \qquad (14)$$

$$G_4(s) = \frac{Y_{II}(s)}{Y_I(s)} = \frac{\lambda}{\lambda + N_{0L}} + \frac{\lambda \left(\dfrac{N_{0L}}{\lambda + N_{0L}} \right)^2}{\tau s + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}}}$$

$$- \frac{\lambda \left(\dfrac{N_{0L}}{\lambda + N_{0L}} \right)^2}{\left(\tau s + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}} \right)^2} \left[1 - \frac{1}{\left(1 + \dfrac{1}{n} \cdot \dfrac{\lambda N_{0L}}{\lambda + N_{0L}} + \dfrac{1}{n} \tau s \right)^{n}} \right] \qquad (15)$$

The effect of the degree of liquid mixing on the dynamics of a fractionating plate can be discussed by analysing the transfer-functions given above, because the degree of liquid mixing is expressed by the value of n.

First, in the case where the liquid on a plate is perfectly mixed, that is, the liquid mixing parameter n is equal to unity, the following transfer-functions are obtained from equations (12), (13), (14) and (15).

$$G_1(s) = \frac{X_{II}(s)}{X_I(s)} = \frac{1}{1 + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}} + \tau s} \tag{16}$$

$$G_2(s) = \frac{Y_{II}(s)}{X_I(s)} = \frac{\dfrac{mN_{0L}}{\lambda + N_{0L}}}{1 + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}} + \tau s} \tag{17}$$

$$G_3(s) = \frac{X_{II}(s)}{Y_I(s)} = \frac{\dfrac{\lambda N_{0L}}{m(\lambda + N_{0L})}}{1 + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}} + \tau s} \tag{18}$$

$$G_4(s) = \frac{Y_{II}(s)}{Y_I(s)} = \frac{\lambda}{\lambda + N_{0L}} + \frac{\lambda \left(\dfrac{N_{0L}}{\lambda + N_{0L}}\right)^2}{1 + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}} + \tau s} \tag{19}$$

These transfer-functions are all of the first order except that expressed by equation (19), and they have following common time constant.

$$T = \frac{\lambda + N_{0L}}{\lambda + \lambda N_{0L} + N_{0L}} \tau$$

Since only $G_4(s)$ is essentially different from the other three transfer-functions, some discussion will be given in § 4. It is worth while to notice that n, which describes the liquid mixing degree on a plate, is involved only in the following term of the four kinds of transfer-functions.

$$\frac{1}{\left(1 + \dfrac{1}{n} \cdot \dfrac{\lambda N_{0L}}{\lambda + N_{0L}} + \dfrac{1}{n} \tau s\right)^n}$$

Therefore, it may be, first of all, desirable to speculate how $G_1(s)$ is affected by the value of n. $G_1(s)$ is transformed into

$$G_1(s) = \frac{1}{\left(1 + \dfrac{1}{n} \cdot \dfrac{\lambda N_{0L}}{\lambda + N_{0L}}\right)^n} \cdot \frac{1}{(1 + T_n s)^n} \tag{20}$$

, where

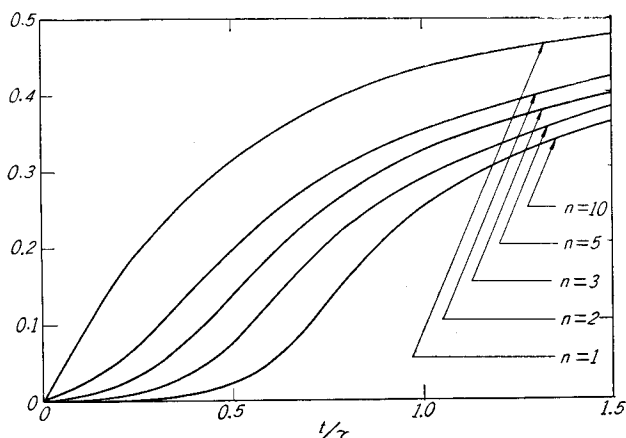$$T_n = \frac{\lambda + N_{0L}}{n(\lambda + N_{0L}) + \lambda N_{0L}} \tau$$

Fig. 2. Transient response of the outlet liquid concentration with unit step change of the inlet liquid concentration with "perfect mixing model" for $\lambda=1$, $1/N_{0L}=0$
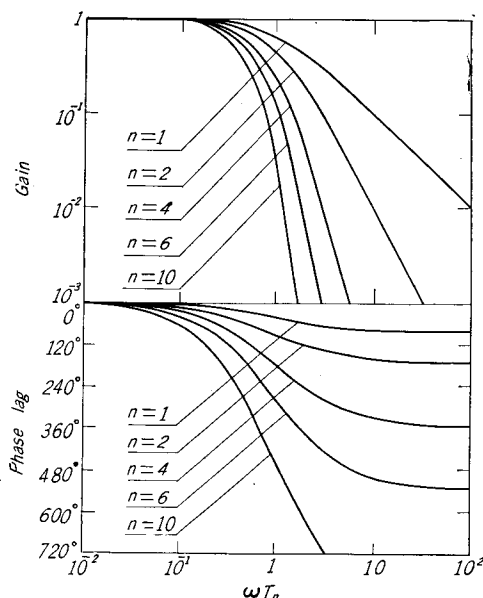

Fig. 3. Bode Diagram of $1/(1+j\omega T_n)^n$

$G_1(s)$ expressed by equation (20) is a transfer-function of the n-th order and its dynamic behavior differs from the transfer-function of the first order shown by equation (16) with increase of n. This fact can be more explicitly seen by the indicial or frequency response curves in Fig. 2 and Fig. 3 calculated from the transfer-function $G_1(s)$. In the perfect piston flow state which is the limiting case,

$$G_1(s) = \exp\left(-\frac{\lambda N_{0L}}{\lambda+N_{0L}}\right)\cdot\exp\left(-\tau s\right) \tag{21}$$

The above equation states that the value of the outlet liquid concentration change for a unit step change of the inlet liquid concentration comes to $\exp\left(-\frac{\lambda N_{0L}}{\lambda+N_{0L}}\right)$ after $t=\tau$ and in frequency response the gain has a constant value of $\exp\left(-\frac{\lambda N_{0L}}{\lambda+N_{0L}}\right)$ and the phase lag increases to infinity from zero with increase of ω.

From the analyses of indicial response or frequency reponse as shown above, $G_1(s)$, the only term involving parameter n in the four kinds of transfer-functions, is seriously affected by the value of n, that is, by the degree of liquid mixing.

On the other hand, the other three kinds of transfer-functions $G_2(s)$, $G_3(s)$ and $G_4(s)$ are not affected so greatly as $G_1(s)$ by the degree of liquid mixing. This explanation will be given in §3 in detail.

§ 3.   Transfer-functions for concentration change with "diffusional model"[4,6]

In this section, the same problems as discussed in §2 are analysed in the
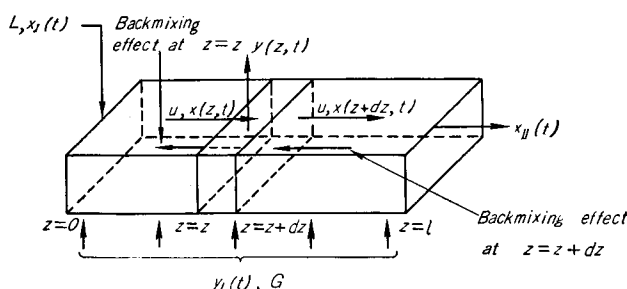
Fig. 4. Material balance on a plate for "diffusional model"

form of a "distributed system" with the assumption of a "diffusional model" for the expression of the degree of liquid mixing. In this case the form of the liquid concentration distribution curve is smooth in the direction of the liquid flow on a plate while the liquid concentration distribution in §2 is assumed to be stepwise. The assumptions used in this section are the same as those of the previous section, except for the method of expressing the degree of liquid mixing, and are as follows;

  a) Vapor hold-up is negligibly small in comparison with that of the liquid on a plate.

  b) Liquid and vapor are perfectly mixed, that is, there is no concentration gradient of liquid or vapor, in the direction of vapor flow.

  c) Liquid stream is continuous and smooth, and the "diffusional model" can be assumed for the concentration gradient of liquid in the direction of liquid flow.

  d) All the physical or chemical properties, eg., the mass transfer capacity coefficient and the relationship of equilibrium between the concentrations of vapor and liquid are always constant despite of changes in fluid concentrations, and are independent of time and position on a plate.

  e) Mass flow rate of liquid and vapor and hold-up of liquid are always constant.

In Fig. 4, L is the molar mass velocity of liquid per unit width in kg-moles/ m·hr, G is the molar mass velocity of vapor per cross sectional area of a plate in kg-moles/m²hr, l is the liquid travelling length on a plate in meters and u is the constant liquid linear velocity in m/hr.

In the infinitesimal interval between $z=z$ and $z=z+dz$ in the direction of liquid flow in Fig. 4, the material balance (the equation of continuity) and the equation of mass transfer rate are given under the assumptions mentioned above as follows.

$$E \frac{\partial^2 x}{\partial z^2} - u \frac{\partial x}{\partial z} - \frac{\partial x}{\partial t} - \frac{u}{H_{0L}}(x - x^*) = 0 \tag{22}$$

and

$$y = y_I - \frac{L}{G} \cdot \frac{\partial x}{\partial z} - \frac{1}{u} \cdot \frac{L}{G} \cdot \frac{\partial x}{\partial t} + \frac{E}{u} \cdot \frac{L}{G} \cdot \frac{\partial^2 x}{\partial z^2} \tag{23}$$

As in §2, the relation of equilibrium between the concentrations of vapor and liquid is given by the following equation.

$$y = mx^* + \text{constant} \tag{24}$$

In equations (22) to (24), E is the "back mixing coefficient" in m²/hr and H_{0L} is the length of an overall liquid phase transfer unit in meters. H_{0L} is essentially the same expression for the degree of mass transfer as an overall mass transfer capacity coefficient $K_L a$ and is equivalent to l/N_{0L}. x and y, of course, are functions of time t and position z. The boundary conditions for equations (22) to (24) are given from the material balances at the inlet and outlet positions of the plate as follows;

$$E \left[\frac{\partial x}{\partial z} \right]_{z=0} = u \left(\underset{z=0}{x} - x_I \right) \tag{25}$$

and

$$E \left[\frac{\partial x}{\partial z} \right]_{z=l} = 0 \tag{26}$$

Using the boundary conditions (25) and (26), the following equation for the liquid concentration distribution on a plate is obtained from equations (22) to (24) in the Laplace transforms of fluid concentration changes Δx and Δy from their steady state values x^0 and y^0.

$$X(z, s) = \frac{P_e (b_2 e^{b_2 + b_1(z/l)} - b_1 e^{b_1 + b_2(z/l)})}{b_2^2 e^{b_2} - b_1^2 e^{b_1}} \cdot X_I(s)$$

$$+ \frac{\dfrac{\lambda N_{0L}}{m(\lambda + N_{0L})}}{\tau s + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}}} \left[1 - \frac{P_e (b_2 e^{b_2 + b_1(z/l)} - b_1 e^{b_1 + b_2(z/l)})}{b_2^2 e^{b_2} - b_1^2 e^{b_1}} \right] Y_I(s) \tag{27}$$

, where

$$b_1, b_2 = \frac{1}{2} \left(P_e \mp \sqrt{P_e^2 + 4 P_e \left(\tau s + \frac{\lambda N_{0L}}{\lambda + N_{0L}} \right)} \right)$$

In the above equation, λ and N_{0L} are defined as follows;

$$\lambda = \frac{mGl}{L}, \quad N_{0L} = \frac{l}{H_{0L}}$$

These parameters λ and N_{0L} are exactly the same as those in the previous section.

From equation (27), the liquid concentration distribution on a plate is easily obtained, where P_e is called the "Peclet number" which gives the degree of liquid mixing on a plate in the direction of liquid flow, and is a function not only of the back mixing coefficient but also of the liquid travelling length l and liquid linear velocity u. That is,

$$P_e = \frac{ul}{E}$$

From the fact mentioned above it may be found that the degree of liquid mixing on a plate can be accounted for more reasonably by P_e than by the back mixing coefficient E alone. Therefore P_e will be used to describe the degree of liquid mixing on a plate, hereafter.

Similarly, the concentration distribution of the outlet vapor may be obtained. The concentration of "local" outlet vapor from the infinitesimal section in the direction of liquid flow is a function of position z and time t, and therefore the average concentration of all outlet vapor rising from a plate must be calculated by

$$y_{II}(t) = \frac{1}{l} \int_0^l y(z, t)dz \tag{28}$$

From equations (23), (27) and (28), the following equation describing the average concentration distribution of the outlet vapor on a plate in Laplace transform is obtained.

$$Y_{II}(s) = \frac{\dfrac{mN_{0L}}{\lambda + N_{0L}}}{\tau s + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}}} \left[1 - \frac{P_e\, e^{P_e}(b_2 - b_1)}{b_2^2 e^{b_2} - b_1^2 e^{b_1}} \right] X_I(s)$$

$$+ \left[\frac{\lambda}{\lambda + N_{0L}} + \frac{\lambda \left(\dfrac{N_{0L}}{\lambda + N_{0L}} \right)^2}{\tau s + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}}} - \frac{\lambda \left(\dfrac{N_{0L}}{\lambda + N_{0L}} \right)^2}{\left(\tau s + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}} \right)^2} \left\{ 1 - \frac{P_e\, e^{P_e}(b_2 - b_1)}{b_2^2 e^{b_2} - b_1^2 e^{b_1}} \right\} \right] Y_I(s) \tag{29}$$

From equations (27) and (29), the following four kinds of transfer-functions are obtained as well as in the case of §2.

$$G_1(s) = \frac{X_{II}(s)}{X_I(s)} = \frac{P_e\, e^{P_e}(b_2 - b_1)}{b_2^2 e^{b_2} - b_1^2 e^{b_1}} \tag{30}$$

$$G_2(s) = \frac{Y_{II}(s)}{X_I(s)} = \frac{\dfrac{mN_{0L}}{\lambda + N_{0L}}}{\tau s + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}}} \left(1 - \frac{P_e\, e^{P_e}(b_2 - b_1)}{b_2^2 e^{b_2} - b_1^2 e^{b_1}} \right) \tag{31}$$

$$G_3(s) = \frac{X_{II}(s)}{Y_I(s)} = \frac{\dfrac{\lambda N_{0L}}{m(\lambda + N_{0L})}}{\tau s + \dfrac{\lambda N_{0L}}{\lambda + N_{0L}}} \left(1 - \frac{P_e\, e^{P_e}(b_2 - b_1)}{b_2^2 e^{b_2} - b_1^2 e^{b_1}} \right) \tag{32}$$

$$G_4(s) = \frac{Y_{II}(s)}{Y_I(s)} = \frac{\lambda}{\lambda+N_{0L}} + \frac{\lambda\left(\dfrac{N_{0L}}{\lambda+N_{0L}}\right)^2}{\tau s + \dfrac{\lambda N_{0L}}{\lambda+N_{0L}}}$$

$$- \frac{\lambda\left(\dfrac{N_{0L}}{\lambda+N_{0L}}\right)^2}{\left(\tau s + \dfrac{\lambda N_{0L}}{\lambda+N_{0L}}\right)^2}\left[1 - \frac{P_e\, e^{Pe}(b_2-b_1)}{b_2^2\, e^{b_2} - b_1^2\, e^{b_1}}\right] \qquad (33)$$

In both extreme conditions, that is, in the state of perfect liquid mixing and of perfect liquid piston flow, these transfer-functions expressed by equations (30) to (33) are found to be equivalent to the corresponding transfer functions obtained in §2.

As in §2, the meaning of the transfer functions given by equations (30) to (33) can be analysed by indicial response and frequency response methods. The response of the outlet liquid concentration with a step change of the inlet liquid concentration, as one example of indicial response, is given by the following equation.

$$\frac{\Delta x_{II}(t)}{\Delta x_I} = L^{-1}\left[\frac{G_1(s)}{s}\right] = R_0 - \sum_{k=2}^{\infty} R_k \qquad (34)$$

, where

$$R_0 = \left[\frac{P_e\, e^{Pe}(b_2-b_1)}{b_2^2\, e^{b_2} - b_1^2\, e^{b_1}}\right]_{s=0}$$

$$R_k = \frac{2P_e\, e^{Pe/2}\,\theta_k^2\left(\theta_k^2 - \dfrac{P_e^2}{4}\right)e^{s_k t}}{\cos\theta_k\left(\theta_k^2 + \dfrac{P_e^2}{4}\right)\left(\theta_k^2 + \dfrac{P_e^2}{4} + P_e\right)\left(\theta_k^2 + \dfrac{P_e^2}{4} + \dfrac{\lambda N_{0L}}{\lambda+N_{0L}}P_e\right)}$$

$$s_k = -\frac{\theta_k^2 + \dfrac{P_e^2}{4} + \dfrac{\lambda N_{0L}}{\lambda+N_{0L}}P_e}{P_e\cdot\tau}$$

and θ_k is the k-th positive root of

$$\tan\theta = \frac{4P_e\theta}{4\theta^2 - P_e^2}$$

Numerical solutions of equation (34), as one example, with $\lambda=1$ and $1/N_{0L}=0$, is shown in Fig. 5. It is seen from Fig. 5, that the response of the outlet liquid concentration deviates rather markedly from that of the first order system and its dead time increases with
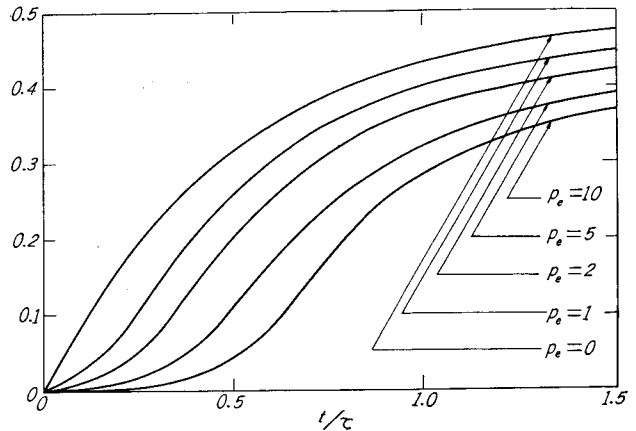

Fig. 5. Transient response of the outlet liquid concentration with unit step change of the inlet liquid concentration with "diffusional model" for $\lambda=1$, $1/N_{0L}=0$

increase in P_e. This fact is more obvious from the Bode Diagram showing the frequency response of $G_1(s)$ as shown in Fig. 6. The response at $P_e=0$ is essentially different to those at $P_e \neq 0$, particularly in the region of higher frequency.

Similar numerical calculations of the frequency response of $G_2(s)$ or $G_3(s)$ and $G_4(s)$ expressed by equations (31) or (32) and (33), are shown in Fig. 7 and 8.
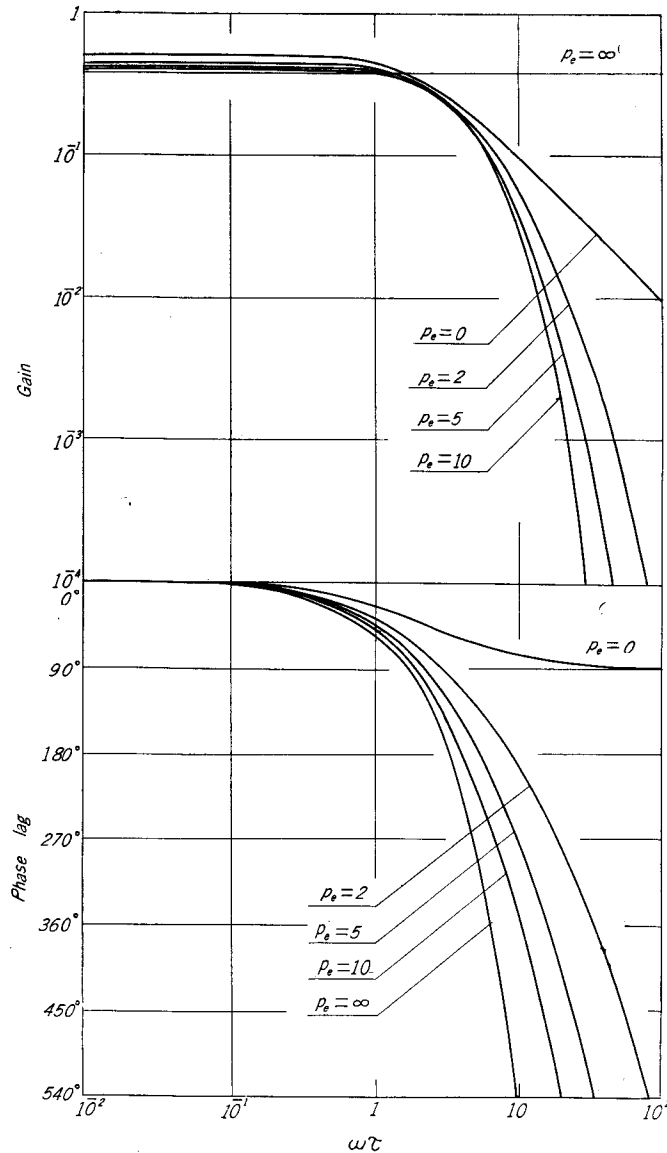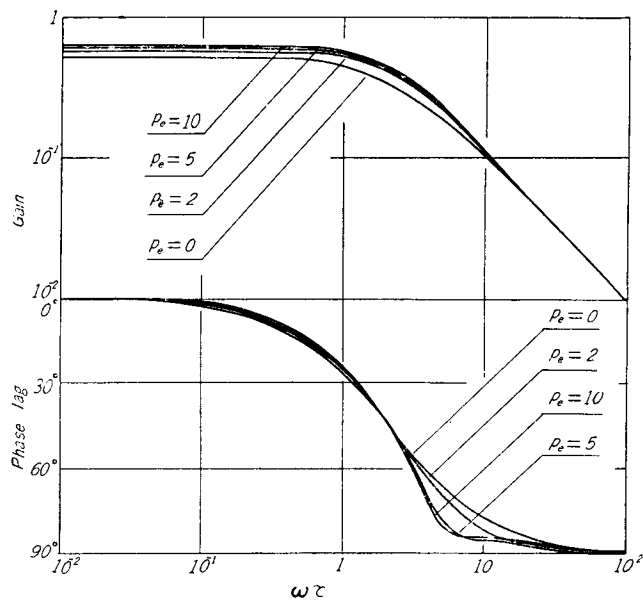

Fig. 6. Bode Diagram of $G_1(j\omega)$ for $\lambda=1$, $1/N_{0L}=0$

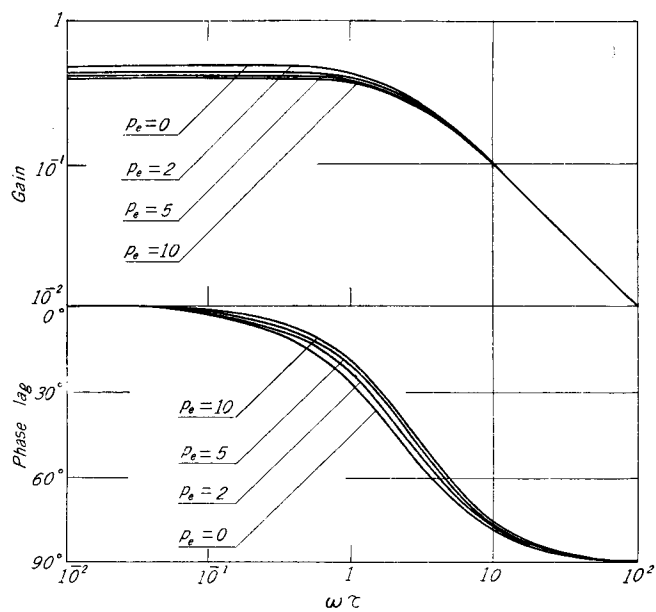Fig. 7.   Bode Diagram of $G_2(j\omega)/m$ or $mG_3(j\omega)$ for $\lambda=1$, $1/N_{0L}=0$

Fig. 8.   Bode Diagram of $G_4(j\omega)$ for $\lambda=1$, $1/N_{0L}=0$

From these diagrams it can be seen that the transfer-functions of $G_2(s)$ or $G_3(s)$ and $G_4(s)$ are slightly affected by the value of P_e.

From these frequency response analyses, involving results for several conditions in addition to the above, it may be concluded finally that the effect of the degree of liquid mixing on unsteady performance with respect to the fluid concentration on a plate is large only in the case of the signal transfer given by the transfer-function $G_1(s)$ and is negligibly small for those given by the transfer-functions $G_2(s)$, $G_3(s)$ and $G_4(s)$.

Therefore these transfer-functions may be substituted for with first order functions without large error. This fact also may be recognized in §2.

§4. Discussion of the expressions for the degree of liquid mixing and other problems from the view point of "dynamics"

1. On the expressions for the degree of "liquid mixing"

From the discussion of the transfer-functions given in §2 and 3, it has been concluded that the unsteady performances expressed by the transfer-functions $G_2(s)$, $G_3(s)$ and $G_4(s)$ are affected but slightly by the degree of liquid mixing and in practice may be replaced with fair accuracy by the corresponding transfer-functions at $P_e = 0$, and that $G_1(s)$ is remarkably affected by the value of n or P_e which is the parameter of the liquid mixing degree. In this section, the differences between the transfer-functions expressed by the two "liquid mixing models" are discussed. By comparing the four kinds of transfer-functions $G_1(s)$, $G_2(s)$, $G_3(s)$ and $G_4(s)$ given by equations (12) to (15) for the case of the "perfect mixing model" with those in the "diffusional model" given by equations (30) to (33), it is easily recognized that the difference between both models is only in $G_1(s)$, that is, in the following term showing the effect of the liquid mixing degree.

$$P_1 = \frac{1}{\left(1 + \frac{1}{n} \cdot \frac{\lambda N_{0L}}{\lambda + N_{0L}} + \frac{1}{n} \tau s\right)^n} \quad \text{(for perfect mixing model)}$$

$$P_2 = \frac{P_e e^{Pe}(b_2 - b_1)}{b_2^2 e^{b_2} - b_1^2 e^{b_1}} \quad \text{(for diffusional model)}$$

In the above equations, it is worth noticing that when P_1 is always equal to P_2 analytically, the two expressions for the liquid mixing degree are equivalent to each other in the sense of "dynamics". However, P_1 is generally not equivalent to P_2 except for the two extreme cases when the state on a plate is one of perfect liquid mixing and perfect liquid piston flow.

In the Bode Diagram of $G_1(j\omega)$ based on the "diffusional model", the phase lag changes from zero to infinity corresponding to values of ω from zero to

infinity, and the rate of decrease in gain becomes large with increase of ω. On the other hand, in the Bode Diagram of $G_1(j\omega)$ expressed by the "perfect mixing model", the phase lag approaches $90 n^0$ and the gain decreases linearly with a slope of $20 n$ decibels per decade in the region of large ω.

From the considerations mentioned above, it may be concluded that when P_1 and P_2 are not equivalent to each other, both expressions for liquid mixing degree are not essentially equivalent to each other in the sense of "dynamics".

These essential differences caused by the expression of the liquid mixing model are not found in analyses of steady state performances, for example the effect of liquid mixing degree on plate efficiency.

2. On the effect on the transfer-functions of neglecting the vapor hold-up on a plate.

In the discussions given above, it was assumed that the hold-up of vapor is negligibly small in comparison with that of the liquid. It must be noticed that in the transfer-function $G_4(s)$ expressed by equation (15) or (33), the phase lag and the gain for very large values of ω come to zero and constant value $\lambda/(\lambda+N_{oL})$ respectively *if the vapor is not completely in equilibrium with the liquid on a plate.* This fact, for example, is shown in Fig. 9 for $\lambda=1$, $N_{oL}=3$. And furthmore the transfer-function $G_4(s)$ given by equation (15) or (33) expresses that the outlet vapor concentration change $\varDelta y_{II}(t)$ is not zero at $t=0$, for example, with an unit step change of the inlet vapor concentration. These phenomena, however, can not occur in actual practice and therefore these irrationalities are due to neglecting vapor hold-up. This discussion will be given in detail in another paper[*].
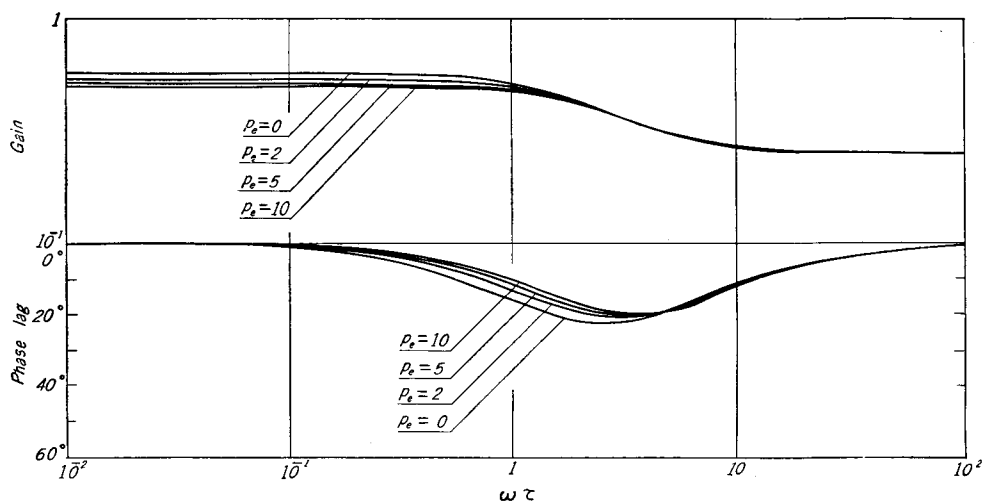

Fig. 9. Bode Diagram of $G_4(j\omega)$ for $\lambda=1$, $N_{oL}=3$

[*] Unpublished.

§5. Development to multistage fractionating plates

In §2 and 3, the transfer-functions concerning fluid concentration change on a plate have been considered for the case where the liquid on a plate is not perfectly mixed. This information obtained from §2 and 3 can be used to procure the "dynamics" of fractionating plates column.

Fractionating column has multistage fractionating plates as shown in Fig. 10, in which the total number of plates is N. In fractionating plates as shown in Fig. 10, assuming that the transfer-functions of all plates are the same througout a column, the following equations are obtained around the k-th stage.

$$X_k(s) = G_1(s) X_{k+1}(s) + G_3(s) Y_{k-1}(s) \qquad (35)$$

$$Y_k(s) = G_2(s) X_{k+1}(s) + G_4(s) Y_{k-1}(s) \qquad (36)$$

and hence,

Fig. 10. Schematic diagram of multi-stages fractionating plates

$$G_1(s) X_{k+1}(s) - (1 + G_1(s)G_4(s) - G_2(s)G_3(s)) X_k(s) + G_4(s) X_{k-1}(s) = 0 \qquad (37)$$

From the difference equation (37), the following four kinds of transfer-functions for multistage fractionating plates are obtained.

$$g_1^N(s) = \frac{X_1(s)}{X_{N+1}(s)} = \frac{G_1(s)(A_1 - A_2)}{(A_1^N - A_2^N) - G_4(s)(A_1^{N-1} - A_2^{N-1})} \qquad (38)$$

$$g_2^N(s) = \frac{Y_N(s)}{X_{N+1}(s)} = \frac{G_2(s)(A_1^N - A_2^N)}{(A_1^N - A_2^N) - G_4(s)(A_1^{N-1} - A_2^{N-1})} \qquad (39)$$

$$g_3^N(s) = \frac{X_1(s)}{Y_0(s)} = \frac{G_3(s)(A_1^N - A_2^N)}{(A_1^N - A_2^N) - G_4(s)(A_1^{N-1} - A_2^{N-1})} \qquad (40)$$

$$g_4^N(s) = \frac{Y_N(s)}{Y_0(s)} = \frac{G_1(s)(A_1 A_2)^N(A_1 - A_2)}{(A_1^N - A_2^N) - G_4(s)(A_1^{N-1} - A_2^{N-1})} \qquad (41)$$

, where

$$A_1, A_2 = \frac{1}{2G_1(s)} \{1 + G_1(s)G_4(s) - G_2(s)G_3(s)$$

$$\pm \sqrt{(1 + G_1(s)G_4(s) - G_2(s)G_3(s))^2 - 4G_1(s)G_4(s)}\}$$

The transfer-functions as shown above can be analysed analtically in the form of frequency response, but it is rather difficult to obtain the indicial response from equations (38) to (41), except for the extreme case calculated by
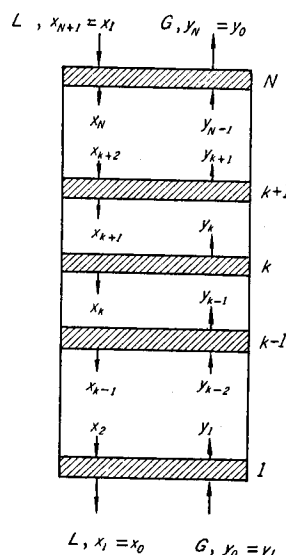
Voetter[8]. From the frequency response analyses of $g_1^N(s)$, $g_2^N(s)$, $g_3^N(s)$ and $g_4^N(s)$ by using the NO. 1 digital computer of Kyoto University (KDC-1) for several conditions of λ, P_e and N for $1/N_{0L}=0$, the following conclusions have been made.

1. The transfer-functions $g_1^N(s)$ is seriously affected by the value of P_e and the effect of P_e on the transfer-function $g_1^N(s)$ becomes large with increase in the total number of plates N, and $g_1^N(s)$ can not be replaced easily by a simple equation.

2. The transfer-functions $g_2^N(s)$ or $g_3^N(s)$ is almost not affected by the value of N and P_e, that is, $G_2(s)$ and $G_3(s)$ at $P_e=0$ may be substituted for $g_2^N(s)$ and $g_3^N(s)$ respectively with considerable accuracy.

3. The transfer-function $g_4^N(s)$ is affected slightly by the value of P_e, and in practice $g_4^N(s)$ at $P_e=0$ may be substituted. In Fig. 11, the Bode Diagram of $g_4^N(s)$ is shown, as one example, for $\lambda=1$, $1/N_{0L}=0$ with several values of N and P_e as parameters.

4. From the conclusions mentioned above, the only remaining problem

Fig. 11. Bode Diagram of $g_4^{(N)}(j\omega)$ for $\lambda=1$, $1/N_{0L}=0$

for expressing simply and accurately the complicated transfer-functions of multistage fractionating plates is how to express the transfer-function $g_1^N(s)$ simply.

In the region of large ω in frequency response,

$$|A_1(j\omega)| \gg 1 \gg |A_2(j\omega)| \quad \text{and} \quad A_1(j\omega) \to 1/G_1(j\omega)$$

and hence,

$$g_1^N(j\omega) \to G_1(j\omega)^N$$

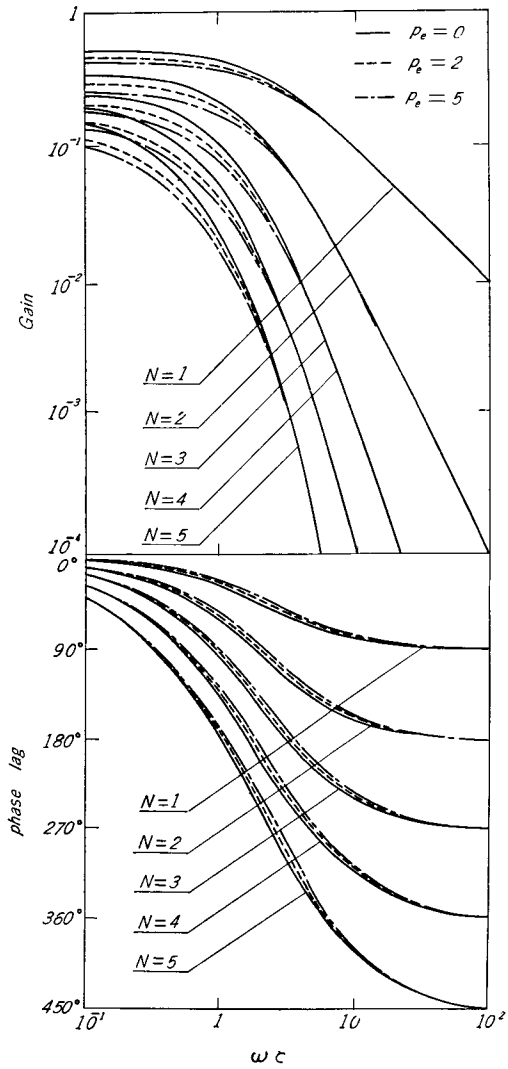That is, $g_1^N(s)$ approaches the transfer-function of the system which is combined of n elements, the transfer-function of which is equivalent to $G_1(s)$. From the point of view mentioned above, it may be worth while to define the following two factors.

$$C_N = \frac{|g_1^N(j\omega)|}{|G_1(j\omega)|^N} \quad \text{and} \quad \phi_N = (-\angle g_1^N(j\omega)) - N(-\angle G_1(j\omega))$$
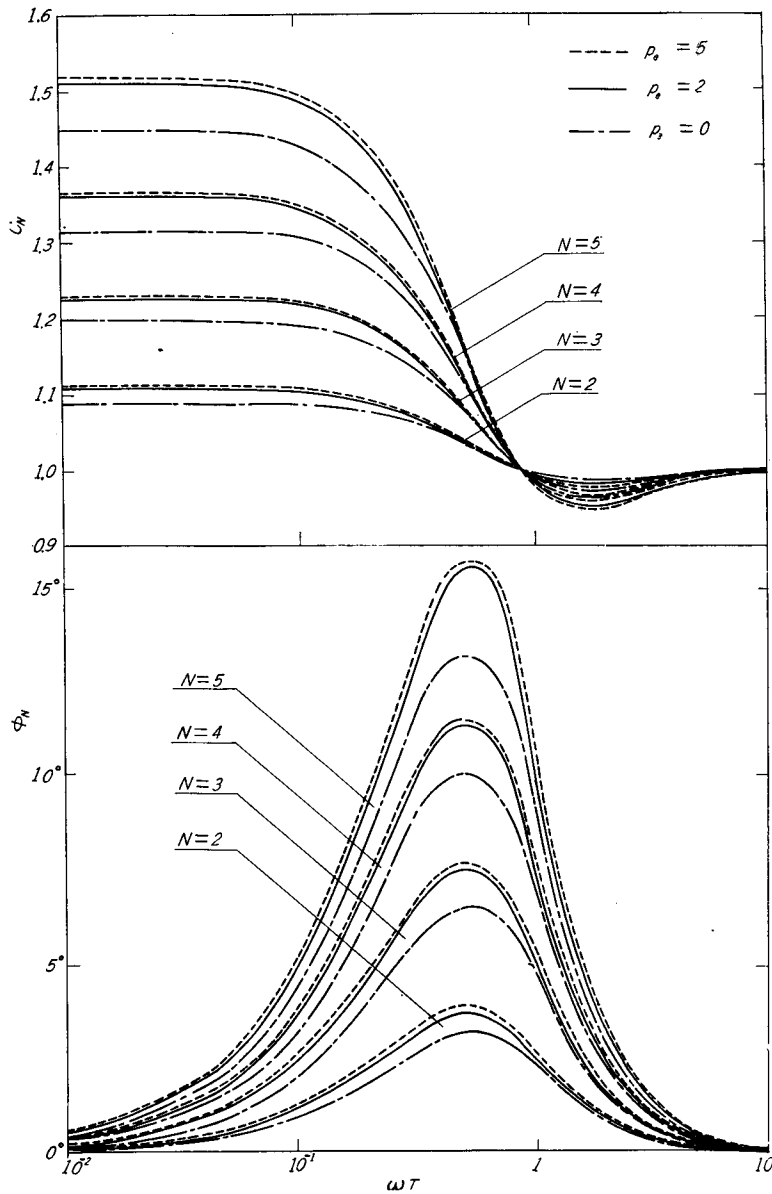

Fig. 12. Diagram of C_N and ϕ_N for $\lambda=10$, $1/N_{0L}=0$

In the region of large ω, C_N and ϕ_N approach to unity and zero respectively.

$$C_N \to 1, \quad \phi_N \to 0 \quad \text{for} \quad \omega \gg 1 \tag{42}$$

On the other hand, in the region of very small ω,

$$C_N = \lim_{\omega \to 0} \frac{|g_1^N(j\omega)|}{|G_1(j\omega)|^N} = \lim_{s \to 0} \frac{|g_1^N(s)|}{|G_1(s)|^N} = \text{constant}, \quad \phi_N \to 0 \quad \text{for} \quad \omega \ll 1 \tag{43}$$

C_N and ϕ_N are shown in Fig. 12, as one example, with $\lambda = 10$ and $1/N_{0L} = 0$.

From the results in the case of $1/N_{0L} = 0$, the conditions given by equations (42) and (43) may be satisfied only in the region of $\omega T > 10$ and $\omega T < 10^{-2}$ respectively. In Fig. 12, T is defined by

$$T = \tau/(1+\lambda)$$

That is, $g_1^N(s)$ may be estimated rather accurately only in the region of $\omega T < 10^{-2}$ or $\omega T > 10$.

Nomenclatures

E ; Back mixing coefficient (m²/hr)

G ; vapor flow rate per unit cross-sectional area (kg-moles/m²hr)

H ; liquid hold-up (kg-moles)

H_{0L} ; overall length of transfer unit based on liquid phase (m)

$K_L a$; overall mass transfer capacity coefficient based on liquid phase (kg-mole/m³hr unit driving force of liquid phase)

L ; liquid flow rate per unit width (kg-moles/m hr)

L ; operator of Laplace transformation

L^{-1} ; operator of Laplace inverse transformation

l ; liquid travelling length (m)

N ; number of plates in the fractionating column (—)

N_{0L} ; overall number of transfer units based on liquid phase (—)

P_e ; Peclet's number defined by $P_e = \dfrac{ul}{E}$ (—)

R ; liquid flow rate (kg-moles/hr)

T ; modified time constant defined by $T = \dfrac{\tau}{1+\lambda}$ (hr)

T_n ; modified time constant defined by $T_n = \dfrac{\lambda + N_{0L}}{n(\lambda + N_{0L}) + \lambda N_{0L}} \tau$ (hr)

u ; liquid linear velocity on a plate (m/hr)

V ; vapor flow rate (kg-moles/hr)

W ; volume occupied by fluids in a vessel (m³)

X ; Laplace transformation of Δx with time t

x ; liquid concentration (mole fraction)

Y ; Laplace transformation of Δy with time t

y ; vapor concentration (mole fraction)

z ; distance from liquid inlet point (m)

λ ; stripping factor defined by $\lambda = \dfrac{mV}{R}$ or $\lambda = \dfrac{mGl}{L}$ $(-)$

τ ; residence time of liquid on a plate given by $\tau = \dfrac{H}{R}$ or $\tau = \dfrac{l}{u}$ (hr)

ω ; angular frequency (rad/hr)

Literatures cited

1) Armstrong, W. D. and W. L. Wilkinson; Trans. Inst. Chem. Engrs. (London), **35**, 352 (1957).
2) Gilbert, T. J.; Chem. Eng. Sci., **10**, 243 (1959).
3) Hougen, J. O. and R. A. Walsh; Chem. Eng. Progr., **57**, NO. 3, 69 (1961).
4) Mizushina, T., T. Takamatsu and E. Nakanishi; Control Eng. (Japan), **3**, 342 (1959).
5) O'connell, H. E.; Trans. A. I. Ch. E., **42**, 741 (1946).
6) Takamatsu, T. and E. Nakanishi; Control Eng. (Japan), **5**, 217 (1961).
7) Vincent, G. C. and J. O. Hougen; Chem. Eng. Progr., **57**, NO. 7, 48 (1961).
8) Voetter, H.; "Plant & Process Dynamic Characteristics", Butterworth Scientific Publication, London, pp 73 (1957).
9) Wilkinson, W. L. and W. D. Armstrong; ibid., pp 56
10) Wilkinson, W. L. and W. D. Armstrong; Chem. Eng. Sci., **7**, 1 (1957).
11) Wood, R. M. and W. D. Armstrong; ibid., **10**, 272 (1960).