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MODELING AND DESIGN METHOD FOR INTERNAL HEAT-INTEGRATED PACKED DISTILLATION COLUMN

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A mathematic model for an internal heat-integrated packed distillation column (IHIPAC) is developed on the basis of interphase mass transfer theory. A new method for design of an IHIPAC is proposed using the newly developed model. The binary system of methanol-water is adopted for the simulation work. The results show that an IHIPAC can be operated at a reflux ratio smaller than the minimum reflux ratio of a conventional distillation column, and that an energy saving of approximate 30% can be achieved under various operating conditions.

Introduction

Distillation is the biggest energy consumer among unit operations in chemical plants. Various methods for energy saving in distillation have been proposed during the last two decades, such as multi-effect method, heat-pump method, intermediate heating-cooling method, and secondary reflux and vaporization [SRV] method. The first three methods have already been applied to actual processes.⁶⁾ In particular, the heat-pump method has been widely used and found effective not only in distillation but also in other heat recovery processes.¹¹⁾ The SRV method, however, has not yet been applied to actual processes.

The SRV method was first proposed by Mah *et al.* (1977).¹⁰⁾ Fitzmorris *et al.* (1980)³⁾ presented a thermodynamic availability analysis of an ethylene-ethane splitter using the SRV method. Nakaiwa *et al.* (1984)^{12,13)} proposed a design method for a plate-to-plate heat-integrated distillation column by modi-

fying the McCabe-Thiele method. Shimizu *et al.* (1985)¹⁴⁻¹⁶⁾ presented a control scheme for binary distillation with SRV, and other techniques have been proposed.^{1,17)}

In this paper we propose a new design method for a packed distillation column in which energy saving is realized by the SRV method. This system is hereafter called an internal heat-integrated packed distillation column (IHIPAC).

1. Structural Description of an IHIPAC

Figure 1 shows a schematic diagram of an IHIPAC. The rectifying section is operated at a higher pressure and a higher temperature than those of the stripping section. For adjusting the pressures, a compressor and a throttling valve are installed between the two sections. A certain amount of heat is differentially transferred from the rectifying section to the stripping section along the heat-integrating section. As a result, flow rates are decreased along the column upward in the rectifying section and downward in the stripping section. The vapor flow rates at the top of the rectifying section and at the bottom of the stripping

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Fig. 1. Schematic diagram of an IHIPAC

section are small. Therefore the heat duties in both condenser and reboiler can be reduced.

Since the flow rates are changed gradually along the column, the operating line becomes a curve and a conventional design method such as the McCabe-Thiele method or that based on the concept of H.T.U. and N.T.U. becomes inapplicable. For a packed or wetted-wall column in particular, the internal heat integration not only causes a change of flow rates but also complicates the mass transfer mechanism in the column.^{4,5)}

In this paper, a new mathematical model is developed on the basis of the interphase mass transfer theory, and a new design method for an IHIPAC is proposed.

2. Modeling

2.1 Definition of mass transfer coefficient

Consider the concentration distribution in vapor and liquid films as shown in Fig. 2. The mass transfer coefficient has been defined in the case of high mass transfer rate, such as in externally heated or cooled wetted-wall distillation, as:¹⁾

$$k_Y^* = \frac{N_A - y_i(N_A + N_B)}{(y_i - y)} \quad (1)$$

and in the case of low mass transfer rate, such as conventional packed distillation, as:

$$k_Y = \frac{\mathcal{N}_A}{(y_i - y)} \quad (2)$$

where N_A , $\mathcal{N}_A$ and N_B , $\mathcal{N}_B$ are the mass fluxes through the interface of components A and B , re-

Fig. 2. Mechanism of mass transfer in heat integrated stripping section

spectively. The subscript i shows the properties at the interface.

2.2 Assumptions

To develop a mathematical model for the design problem of an IHIPAC, the following assumptions are made.

1) The mass transfer mechanism in distillation is vapor phase-controlled.⁷⁻⁹⁾ Then we can replace x_i by x , y_i by y^* and k_Y by K_Y in Eqs. (1) and (2), and we obtain:

$$K_Y^* = \frac{N_A - y^*(N_A + N_B)}{(y^* - y)} \quad (\text{for high mass transfer rate}) \quad (3)$$

$$K_Y = \frac{\mathcal{N}_A}{(y^* - y)} \quad (\text{for low mass transfer rate}) \quad (4)$$

The relation between K_Y^* and K_Y , as an example, was given by the film theory of Bird¹⁾ as:

$$\frac{K_Y^*}{K_Y} = \frac{\phi}{e^\phi - 1} \quad (5)$$

where

$$\phi = \frac{N_A + N_B}{K_Y} \quad (6)$$

2) Heat transfer between vapor phase and liquid phase is neglected.

3) The heat transfer surfaces in both the rectifying and the stripping sections are completely wetted by the downward flow of liquid.

2.3 Mathematical model derivation

Consider a differential height dz of a packed distillation column having heat flux into or out of the system as shown in Fig. 2. The rate of heat transfer

from the rectifying section to the stripping section can be obtained by the following equation:

$$q_w = U_w[T_R(P_R, x_R) - T_S(P_S, x_S)] \quad (7)$$

where $T_R(P_R, x_R)$ and $T_S(P_S, x_S)$ are the temperatures of liquids in the rectifying section and the stripping section, respectively. q_w is hereafter called the internal heat integration rate. A heat continuity equation with the help of our assumption 2) gives:

$$q_w = \lambda_A N_A + \lambda_B N_B \quad (8)$$

For the *heat-integrated stripping section*, heat is added to the system differentially. The changes of flow rates over dz can be expressed as:

$$dV = dL = -(N_A + N_B)Adz \quad (9)$$

where A is the heat transfer surface per unit column height. The changes of compositions over the same dz can be obtained from the sum of the rates of mass transfer of component A on the surfaces disengaged and engaged to the heat integration as:

$$d(yV) = d(xL) = -(N_A aS + N_A A)dz \quad (10)$$

$$= - \begin{bmatrix} \text{change of component} & \text{change of component} \\ A \text{ due to mass} & A \text{ due to mass} \\ \text{transfer on the} & + \text{transfer on the} \\ \text{surface \textbf{disengaged}} & \text{surface \textbf{engaged}} \text{ with} \\ \text{from heat integration} & \text{heat integration} \end{bmatrix}$$

where aS is regarded as the specific surface of the packing disengaged from heat integration. From Eqs. (3), (4) and (10), we obtain:

$$\begin{aligned} d(yV) &= d(xL) \\ &= -\{(K_Y aS + K_Y^* A)(y^* - y) \\ &\quad + y^*(N_A + N_B)A\}dz \end{aligned} \quad (11)$$

If we let

$$\alpha = (N_A + N_B)A \quad (12)$$

$$\beta = K_Y aS + K_Y^* A \quad (13)$$

and

$$\kappa = \beta/\alpha \quad (14)$$

then from Eq. (9) we have

$$dV = dL = -\alpha dz \quad (15)$$

Rearranging Eq. (11), we obtain:

$$\frac{dx}{(y^* - x) + \kappa(y^* - y)} = \frac{dL}{L} \quad (16)$$

In the same way, for the *heat-integrated rectifying section*, in place of Eqs. (15) and (16) we have Eqs. (17) and (18).

$$dV = dL = \alpha dz \quad (17)$$

Fig. 3. Application of an approximation method

$$\frac{dx}{(y^* - x) - \kappa(y^* - y)} = \frac{dL}{L} \quad (18)$$

Equations (15) and (17) express the changes of flow rates, while Eqs. (16) and (18) express the changes of liquid concentration along the column in the heat-integrated rectifying and stripping sections, respectively.

3. Design Method for an IHIPAC

Because an IHIPAC is by its nature more complicated than a conventional packed distillation column, to design an IHIPAC the following matters must be prescribed.

3.1 Introduction of the approximation method

Since the heat integration rate, q_w , is not explicitly known, we cannot analytically integrate the above equations. We must divide the column into many small parts Δz as shown in Fig. 3, and q_w is assumed to be constant within each Δz . Now, we can integrate Eqs. (15), (16), (17) and (18) to obtain Eqs. (20), (22), (24) and (25) as shown in Table 1. Then the concentrations and the flow rate distributions along the heat-integrating sections can be found step by step using these equations. For non-heat integrating sections H_1 , H_3 , the conventional model of Eqs. (26), (27) is used.

3.2 Necessity of giving additional operating conditions

To make heat integration possible, the operating pressures of rectifying and stripping sections, P_R and

Table 1. Mathematical model of an IHIPAC

Heat integration rate	$q_{w,i} = U_w(\Delta T)_i$	(19)
Mass balances	$V_{R,i+1} = V_{R,i} + \alpha_i(z_{i+1} - z_i)$	(20)
	$L_{R,i+1} = V_{R,i+1} - D$	(21)
	$V_{S,i+1} = V_{S,i} - \alpha_i(z_{i+1} - z_i)$	(22)
	$L_{S,i+1} = V_{S,i+1} + W$	(23)
Component balances	$\int_{x_{R,i}}^{x_{R,i+1}} \frac{dx}{(y^* - x) - \kappa_i(y^* - y)} = \ln \left[\frac{L_{R,i+1}}{L_{R,i}} \right]$	(24)
	$\int_{x_{S,i}}^{x_{S,i+1}} \frac{dx}{(y^* - x) + \kappa_i(y^* - y)} = \ln \left[\frac{L_{S,i+1}}{L_{S,i}} \right]$	(25)
Non-heat integrating sections	$H_1 = \frac{V_{R,0}}{K_y a_R S_R} \int_{y_{R,0}}^{y_D} \frac{dy}{(y^* - y)}$	(26)
	$H_3 = \frac{V_{S,N}}{K_y a_S S_S} \int_{y_{S,N}}^{y_{S,0}} \frac{dy}{(y^* - y)}$	(27)

P_S , the overall heat transfer coefficient, U_w , and the heat transfer area per unit height in the heat-integrating section, A , must be given. Furthermore, because of the increase in degree of freedom between the external reflux ratio and column height, $H_1 + 2H_2 + H_3$, affected by heat integration, another operating condition must be given in addition. Here, the height of the non-heat integrated rectifying section, H_1 , was selected.

3.3 Feed position

In designing a conventional distillation column, the feed position is usually located at a point where the rectifying operating line intersects the q -line in order to achieve minimum column height. For the same reason, let us assume that the above concept is also applicable to the design of an IHIPAC.

On the basis of the above considerations, a calculation algorithm for design of the IHIPAC shown in **Fig. 4** is proposed. The computations for $N_A + N_B$ and for $x_{R,i+1}$, $x_{S,i+1}$ indicated in the calculation algorithm are explained in detail in **Appendix A** and **Appendix B**, respectively.

4. Calculation Example

4.1 Outline of simulation framework

The binary system of methanol–water was adopted. Wilson's method was used in the equilibrium calculation. The operating specifications, the operating conditions and the additional design parameters are summarized in **Table 2**. The physical data of the system are listed in **Table 3**.

In simulation, the overall heat transfer coefficient, U_w , reflux ratio, R , and feed condition parameter, q , were varied while other parameters were kept constant. First, U_w was manipulated to determine how

Fig. 4. Calculation algorithm**Table 2.** Given conditions for calculation example of methanol–water system

Operating specifications		
F	$= 1.39$	[mol/s]
z_F	$= 0.45$	[—]
x_D	$= 0.85$	[—]
x_w	$= 0.05$	[—]
q	$= 0.5/0.7/0.9$	[—]
Operating conditions		
P_S	$= 1.013 \times 10^5$	[Pa]
P_R	$= 2.026 \times 10^5$	[Pa]
R	$= 0.10\text{--}0.22$	[—]
Given design parameters		
H_1	$= 0.0$	[m]
U_w	$= 800\text{--}1200$	[W/(K·m ²)]
A	$= 0.72$	[m ² /m]
a_S, a_R	$= 50, 200$	[m ² /m ³]

Table 3. Physical data used in simulation

	MeOH	Water
Antoine's constants:	A	7.8763
	B	1473.11
	C	230
Wilson's parameters:	at 101.3 [kPa]	.428560
	at 202.6 [kPa]	.524796
Latent heat of vaporization:		
λ [J/mol]	33494	40612
Overall mass transfer coefficient:		
K_Y [mol/(s·m ²)]	Predicted by the correlation given by Brid ⁽¹⁾ (pp. 411, 647).	

much the minimum reflux ratio can be decreased by heat integration. Next, the effects of U_w and R on column height and energy saving were investigated.

Table 4. Energy saving of some specified cases

Case No.	Specified conditions				$R_{\min}^*$ & column heights			Energy duties [kW]		
	$U_w A$ [W/(K·m)]	H_1 [m]	q [—]	R [—]	$R_{\min}^*$ [—]*	H_2 [m]	H_3 [m]	Reboiler + feed preheat	Comp. work	Energy conservation**
1	625	0.0	0.5	0.180	.169 (.517)	2.83	0.04	5.30 + 26.00	.64	29.9%
2	1000	0.0	0.5	0.145	.134 (.517)	2.30	0.01	4.18 + 26.00	.68	33.2%
3	625	0.0	0.7	0.140	.133 (.392)	2.34	0.11	14.24 + 15.60	.57	25.8%
4	1000	0.0	0.7	0.115	.107 (.392)	2.05	0.05	12.61 + 15.60	.65	29.6%
5	625	0.0	0.9	0.120	.111 (.308)	2.13	0.25	23.80 + 5.20	.53	21.1%
6	1000	0.0	0.9	0.095	.092 (.308)	2.15	0.06	21.73 + 5.20	.63	26.3%

* The value in parentheses is the minimum reflux ratio in case of conventional distillation.

** Compared with the minimum reflux ratio operation of conventional distillation.

4.2 Simplification of calculation

In practical design, a precise value of the overall mass transfer coefficient, K_Y , is required. Normally, it is measured from experiments on a system of the design target. But in our simulation work we avoid that work by using a K_Y value predicted by a correlation given by Bird.¹⁾ Therefore, it is reasonable that the variation of K_Y affected by the bulk mass flow through the interface, described by Eq. (5), can be neglected. Then we let $K_Y^* = K_Y$ throughout our calculation work.

Also, we introduce the bulk latent heat of vaporization defined as

$$\bar{\lambda} = \bar{H}_V - \bar{h}_L \quad (28)$$

where

$$\bar{H}_V = y_A H_A + y_B H_B \quad (29)$$

$$\bar{h}_L = x_A h_A + x_B h_B \quad (30)$$

are the bulk enthalpies of vapor and liquid, respectively. Then we apply

$$\bar{\lambda}(N_A + N_B) \simeq \lambda_A N_A + \lambda_B N_B \quad (31)$$

to Eq. (8).

These simplifications reduce the time required for computation of $N_A + N_B$.

4.3 Calculation results and discussion

Figure 5 shows the operating line of an IHIPAC for a specified case. A continuous curve expresses the differential change of the internal reflux ratio along the column. The external reflux ratio is smaller than the minimum reflux ratio in the conventional distillation. The total energy consumption in some cases, as shown in Table 4, are about 70% of those of the conventional distillation operated at minimum reflux, assuming that the compression efficiencies are 100%. Here, the two separate equilibrium lines result from the difference of operating pressures in the rectifying and stripping sections.

Figure 6 shows the variations of minimum reflux

Fig. 5. Operating line of an IHIPAC

Fig. 6. Dependence of minimum reflux ratio on $U_w A$

ratio against $U_w A$. It appears that minimum reflux ratios decrease when $U_w A$ increases. Here, it can be concluded that the higher the heat integration rate, the smaller the reflux ratio that can be specified.

Figures 7, 8 and 9 show the contour lines of the constant heat-integrating section height, H_2 , on the $U_w A$ vs. R plane. It is found that, given the same operating specifications, operation at lower heat integration requires a longer column while higher heat integration allows use of a shorter column.

Fig. 7. Height of heat-integrating section on $U_w A$ & R plane, $q=0.5$

Fig. 8. Height of heat-integrating section on $U_w A$ & R plane, $q=0.7$

Fig. 9. Height of heat-integrating section on $U_w A$ & R plane, $q=0.9$

From these results, the following problems concerning the IHIPAC should be noted.

First, let us consider a constant H_2 line in Figs. 7, 8 and 9. When an IHIPAC has been operated for a long time, the apparent $U_w A$ tends to become smaller than the initially designed value, so the reflux ratio must be

increased to keep the product concentrations at the former values. As a result, energy consumption increases. Therefore, like a heat exchanger, an IHIPAC needs a suitable periodic cleaning.

Secondly, in an occasional case, it appears in Fig. 9 that the $U_w A$ around the region of 792–864 W/(K·m²) gives a stationary column height. This is because the improved separation performance of the stripping section and the degraded separation performance of the rectifying section due to the effect of heat integration cancel out. In this situation, it should be kept in mind if one is going to design an IHIPAC that: 1) it is not necessary to set $U_w A$ beyond this region; and 2) design of an IHIPAC using the $U_w A$ within this region will achieve the least sensitivity of the product concentrations on heat integration rate. Therefore, the $U_w A$ around this region is recommended in practice.

Conclusion

A new mathematical model of an IHIPAC was developed on the basis of the interphase mass transfer theory. A calculation algorithm for the design of an IHIPAC was proposed.

In the calculation example, a methanol-water system was adopted. We simplified the calculation by using a predicted overall mass transfer coefficient, K_y , neglecting the variation of K_y due to the high bulk mass transfer effect and using a bulk latent heat of vaporization in the calculation for $N_A + N_B$. However, the calculation algorithm presented in Fig. 4 is applicable without any help from the above simplification.

From the calculation results, the energy saving is about 30% when compared with conventional distillation operated at minimum reflux ratio. The greater the heat integration, the less the energy consumption becomes. In general, the increase of heat integration improves separation performance, but in some cases it should not be increased beyond a certain point in order to avoid an adverse effect.

Note that we confirmed the validity of the proposed model using experimental data obtained by Honda *et al.*,^{4,5)} but they are omitted here since they are too far from our purpose.

Appendix A: Computation of $N_A + N_B$

- 1) Assume $N_A + N_B$.
- 2) Find K'_y by Eqs. (6) and (5).
Find N_A by Eq. (3), N_B by Eq. (8).
- 3) Use the new $N_A + N_B$ and repeat step 2 till $N_A + N_B$ converges.

Appendix B: Computations of $x_{R,i+1}$ and $x_{S,i+1}$

Consider Fig. A-1 in the case of the rectifying section; a point $(x_{R,i}, y_{R,i})$, a slope $L_{R,i}/V_{R,i}$ and a slope $L_{R,i+1}/V_{R,i+1}$ are known.

- 1) Calculate the RHS of Eq. (24).
- 2) Assume a point $(x_{R,i+1}, y_{R,i+1})$ on the line of slope $L_{R,i+1}/V_{R,i+1}$.

Fig. A-1. Computation for $x_{R,i+1}$, $y_{R,i+1}$ in Eq. (24)

- 3) Integrate the LHS of Eq. (24) numerically. The values of $(y^* - x)$ and $(y^* - y)$ are as indicated in Fig. A-1.
- 4) Use trial points $(x_{R,i+1}, y_{R,i+1})$ and repeat step 3 till the LHS equals the RHS.

The same procedure is applicable to the computation of $x_{S,i+1}$ in the stripping section.

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Nomenclature

A	= heat transfer area per unit height	[m ² /m]
a	= specific surface area of packing	[m ² /m ³]
D	= distillate flow rate	[mol/s]
F	= feed flow rate	[mol/s]
H_1, H_2, H_3	= heights of non-heat-integrated rectifying, heat-integrating, and non-heat-integrated stripping sections	[m]
H	= enthalpy of saturated vapor	[J/mol]
h	= enthalpy of saturated liquid	[J/mol]
K_y	= overall mass transfer coefficient	[mol/(s·m ²)]
K_y^*	= overall mass transfer coefficient at high mass transfer rate	[mol/(s·m ²)]
k_y	= vapor phase mass transfer coefficient	[mol/(s·m ²)]
L	= liquid flow rate	[mol/s]
N	= mass flux at high bulk mass transfer rate	[mol/(s·m ²)]
$\mathcal{N}$	= mass flux at low bulk mass transfer rate	[mol/(s·m ²)]
P	= operating pressure	[Pa]
q_w	= heat integration rate	[W/m ²]
q	= feed condition parameter	[—]
R	= reflux ratio	[—]
$R_{\min}$	= minimum reflux ratio	[—]

S	= sectional area of column	[m ²]
T	= temperature of liquid	[K]
U_w	= overall heat transfer coefficient	[W/(K·m ²)]
V	= vapor flow rate	[mol/s]
W	= bottoms flow rate	[mol/s]
x	= mole fraction in liquid phase	[—]
y	= mole fraction in vapor phase	[—]
y^*	= equilibrium mole fraction	[—]
z	= height of column	[m]
z_F	= mol fraction of A in feed	[—]
α	= convective mass transfer parameter, Eq. (12)	[mol/(s·m)]
β	= diffusive mass transfer parameter, Eq. (13)	[mol/(s·m)]
κ	= dimensionless parameter, β/α	[—]
λ	= latent heat of vaporization	[J/mol]

<Subscripts>

A	= component A
B	= component B
D	= distillate
i	= interface
F	= feed
R	= rectifying section
S	= stripping section
W	= bottoms

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