1. Introduction

Distillation is the most frequently-used separation process in the chemical industry. Distillation columns account for about 40% of the total energy consumption in chemical plants, so energy savings in distillation processes are highly desirable. Internally heat integrated distillation columns (HIDiC) are one of the most promising alternatives to conventional distillation columns. Our present design of HIDiC has a similar structure to a heat exchanger (Fig. 1). In the HIDiC, the distillation column is divided into two parts, rectifying and stripping sections. The pressure in the rectifying section is raised by compressing the vapor flow from the top of the stripping section. Physical contact between all or part of the rectifying section with the stripping section then allows internal heat exchange. If the temperature of the rectifying section is appropriately higher than that of the stripping section, the residual heat in the rectifying section can be used as a heat source for the stripping section, resulting in lower requirement for reboiler heat. At the same time, the vapor and the liquid flow rates in the rectifying section are decreased towards the top due to the condensation caused by the internal heat exchange, resulting in reduction of the external reflux ratio. The idea of the HIDiC was first proposed by Haselden for energy savings in cryogenic air separation1). Later the idea was reintroduced by Mah et al.2), and the outline of the present design of the HIDiC was established. In Japan, the fundamentals and practical application of the HIDiC have been studied by Takamatsu et al.3) since the 1980’s. Their great efforts have lead to construction of prototypes and pilot plants of the HIDiC. In particular, the HIDiC pilot plant in Maruzen Petrochemical Co., Ltd. has succeeded in...
operation without both condenser and reboiler, the so-called ideal HIDiC \(^{(4,5)}\) with the maximum energy conserving conditions. In the separation of a 12-component hydrocarbon mixture, the operation results of the pilot HIDiC achieved energy savings of more than 60 % compared to the conventional distillation column \(^{(6)}\).

Introduction of the HIDiC to other separation applications is the main subject of the next phase of HIDiC development. To estimate the energy saving potential of the HIDiC in other separation problems, the characteristics of the internal heat exchange in the HIDiC must be analyzed by both experiments and simulations. In our HIDiC research project, Kimura Chemical Plants Co., Ltd., Kansai Chemical Engineering Co. and Taiyo Nippon Sanso Corp. have developed their own experimental setups and measured the overall heat transfer coefficients of the internal heat exchange in the HIDiC have been. By utilizing these overall heat transfer coefficients, simulation studies have been carried out using both in-house software and commercial process simulators such as Pro/II \(^{®}\) from Invensys PLC and Aspen Plus \(^{®}\) from Aspen Technology, and mainly targeted to predict the energy saving performance \(^{(3,11)}\).

If the equilibrium stage model is applied to the simulation of HIDiCs, the energy flow rate of the internal heat exchange, \(q_{\text{HEX}}\), of a stage is usually expressed as follows \(^{(6,7,9,10)}\):

\[
q_{\text{HEX}} = UA\Delta T
\]

where \(U\) is the overall heat transfer coefficient, \(A\) is the heat transfer area and \(\Delta T\) is the temperature difference between the rectifying and stripping sections. Since \(q_{\text{HEX}}\) can be treated as the heat duty of the pseudo side cooler/heater to the stage, the resultant values significantly affect the energy saving performance of the HIDiC. In this equation, the heat transfer area \(A\) is a design variable and can be determined by the size of the column. \(\Delta T\) is also an operation variable and can be controlled by changing the compression ratio. Therefore, these two variables can be controlled in general.

In contrast, the overall heat transfer coefficient \(U\) has been usually and used in the simulation. Consequently, simulation results based on experimental values measured in small experimental setups might not be predictive if the HIDiC is applied to the distillation of mixtures without available experimental data for the overall heat transfer coefficient of the internal heat exchange. In addition, since condensation and evaporation due to the internal heat exchange occur in the HIDiC, the flow regime and the mass and heat transfer phenomena would be more complicated than for conventional distillation columns. The observed numbers of transfer units or tray efficiencies have shown irregular scattering and over and/or under estimation of separation performance, possibly due to the effects of interactions between heat and mass transfer and interactions between heat integrated through the walls as well as the effect of liquid and vapor flow rates in the HIDiC system. Therefore, accurate estimation of the separation performance with overall heat transfer coefficient is extremely important for further introduction of HIDiCs.

In this study, a HIDiC simulator with the rate based model was developed. To concisely estimate the heat transfer coefficient, the Chilton-Colburn analogy was applied to the simulator, and the effects of operation conditions of the HIDiC on the overall heat transfer coefficients were investigated.

2. Rate-based Model for the HIDiC

The rate-based model for the HIDiC was developed in our previous studies \(^{(12)}\). Mass and heat transfer in the vicinity of the vapor-liquid interface on the packing, and the typical temperature profile around the wall between the rectifying and stripping sections for the HIDiC are schematically shown in Fig. 2. Our model includes internal heat exchange through the wall of the HIDiC as well as heat and mass transfer between the bulk phases and the interface. Since most basic equations for the model are described elsewhere \(^{(12)}\), only the equations modified in this study are shown below.
2.1. Correlations of Vapor and Liquid Phase Mass Transfer Coefficients for the Structured Packing

Correlations for the mass transfer coefficients for random packings were used in our previous model. However, since a sheet-type metal structured packing MCPACK350 is used in the pilot plant of the HIDiC, correlations for structured packings are employed in the present model. The hydraulic and mass transfer models developed by Bravo et al. and Rocha et al. were used. The approach of Taylor and Krishna was followed to calculate the mass transfer coefficient matrices $\kappa^v$ and $\kappa^l$.

\[
\frac{k^v}{D^v} = 0.0338 \times 10^3 \times \frac{k_{08}}{S_c^{1/3}} \quad (2)
\]

\[
k^l = 2 \left[ \frac{D_L \mu_L}{\pi S} \right]^{0.5} \quad (3)
\]

\[
\frac{a_v}{a_p} = 0.6 + \frac{0.0058 \mu_L}{(\pi S)^{0.5}} \left( \frac{\mu_L}{(\pi S)^{0.5}} \right) < 0.85 \quad (4)
\]

\[
a_v = a_p \left( \frac{\mu_L}{(\pi S)^{0.5}} \right) \geq 0.85 \quad (5)
\]

2.1.2. Heat Transfer Coefficients for the Internal Heat Exchange

The driving force of internal heat exchange of the HIDiC is the temperature difference between fluids on the surface of the packings and the surface of the wall. In our model, the actual condensation and evaporation due to the internal heat exchange are assumed to occur on the surface of the packings. Therefore, the amount of the heat removed from the $j$-th segment of the rectifying section in the HIDiC can be expressed by the following equations.

\[
q_{\text{HEX},j}^V = h^V \times A_{\text{cross},j} \times T_{\text{wall}} - T_{\text{rec},j} \quad (6)
\]

\[
q_{\text{HEX},j}^L = h^L \times A_{\text{cross},j} \times \left( T_{\text{wall}} - T_{\text{rec},j} \right) \quad (7)
\]

\[
q_{\text{wall},j} = \frac{T_{\text{wall},j} - T_{\text{rec},j}}{\Delta T_{\text{wall},j}} \quad (8)
\]

The heat exchange area is not defined by the surface area of the wall but by the interfacial area of the packings in the above equation.

2.1.3. Simulation of the HIDiC on gPROMS®

The HIDiC simulator with the rate-based model was constructed using the commercial process simulation software gPROMS® ver. 2.3.10 with AML: GLC (Advanced Model Library of gPROMS® for Gas-Liquid Contactors) by Process Systems Enterprise Ltd. (PSE), and was further modified in this study. The model structure is shown in Fig. 3. The simulation conditions are summarized in Tables 1 and 2. Although the model equations are described for dynamic simulations, only steady-state simulations were carried out in this study. The physical properties needed in the simulation were obtained by Multiflash ver 3.4.3.

3. Results and Discussion

3.1. Effects of the Pressure and the Compression Ratio

The effect of the pressure on the rectifying section, $i.e.$, the compression ratio on the performance of the HIDiC was studied. Figure 4 shows the composition profiles. The relatively volatile components of the feed mixture were not much affected by the compression ratio. However, intermediate volatility components such as cyclopentane and 2-methylpentane were significantly affected by the compression ratio, especially around the feed location ($z = 0.5$). The effects of the compression ratio on the flow rate profiles are shown in Fig. 5. Higher pressure resulted in lower flow rate in the rectifying section, whereas higher compression ratio caused higher flow rate in the stripping section. Figure 6 shows the temperature difference of the bulk liquid between the rectifying and stripping sections. Simulation results with the equilibrium stage model suggested that the temperature difference should increase with the compression ratio for the entire column. However, the opposite phenomena were predicted in the upper part of the column indicating that the temperature difference might not be fully controlled only by the compression ratio. Figure 7 shows the profile of the heat transfer coefficient for the liquid phase. Since the Chilton-Colburn analogy was employed, the shape of the profile is very similar to that of the flow rate.

According to the results, the overall heat transfer coefficient for the internal heat exchange of the HIDiC was calculated based on the temperature difference of the bulk liquid between the rectifying and stripping sections.
The results are shown in Fig. 8. Surprisingly, the value of $U$ was approximately 785 W/(m² K) regardless of the column height, and not very different to 671 W/(m² K) estimated from the operation data of the HIDiC pilot plant\(^8\). These findings suggest that the overall heat transfer coefficient for the internal heat exchange in the HIDiC can be predicted by simulation with the rate based model, if an appropriate analogy between mass and heat transfer is used. From the simulation results based on the present rate-based model assuming the Chilton-Colburn analogy, the difference between the operational and the calculated value of heat transfer coefficient was predicted to be less than 15 %.
3.2 Effect of the Reflux Ratio

Figures 9 and 10 show the effects of the reflux ratio on the composition and the flow rate profiles of the HIDiC, respectively. The composition profile changes with the reflux ratio, especially in the stripping section. External reflux is increased usually by changing the reboiler heat duty. Under such conditions, the flow rates are directly affected by the reboiler heat duty. However, since constant reboiler heat duty was assumed in this study, the reboiler must supply more energy to evaporate intermediate components at lower reflux ratio, leading to lower compositions of intermediate components in the stripping section. In contrast, the energy supplied by the reboiler cannot produce adequate vapor flow that assures mass transfer with increased liquid flow at higher reflux ratio, resulting in more intermediate components.

![Effect of the Reflux Ratio on the Composition Profile](image)

Fig. 9 Effect of the Reflux Ratio on the Composition Profile
components in the stripping section.

The effect of the reflux ratio on the temperature difference between the rectifying and stripping sections is shown in Fig. 11. As mentioned above, the compositions of the intermediate components in the stripping section increased with higher reflux ratio, and the temperature was lower in the stripping section but changed only slightly in the rectifying section at higher reflux ratio. Therefore, the temperature difference is larger at higher reflux ratio. Since the temperature difference is the driving force of internal heat integration, more energy is exchanged between the two sections. As a result, the separation performance of the HIDiC seems to be insensitive to the reflux ratio.

The profile of the liquid phase heat transfer coefficient is shown in Fig. 12. Clearly, the heat transfer coefficient increases with the reflux ratio, which is due to the increase in the liquid flow rate. However, the overall heat transfer coefficient is not affected by the reflux ratio as in Fig. 13, and the average value of $U_{HEX}$ is again around 785 W/(m² K). The simulation results do not indicate the cause of the insensitivity of the overall heat transfer coefficient against the operating variables, but the internal heat transfer of the HIDiC seems to be governed by factors other than the operating variables.

4. Conclusions

The present study developed a HIDiC simulator with the rate-based model and used the correlation of the mass transfer coefficient for structured packings and the Chilton-Colburn analogy to estimate the heat transfer coefficient of the liquid phase. Although the effects of the compression ratio and the reflux ratio on the composition profiles were relatively large, the overall heat transfer coefficient of the internal heat exchange in the HIDiC based on the temperature difference of the bulk liquid between the rectifying and stripping sections was around 785 W/(m² K), regardless of the column height and the operating conditions. This value is not very different from that obtained from the small experimental plant. Therefore, the overall heat transfer coefficient of the HIDiC can be predicted by simulations with
the rate-based model.

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Nomenclatures

\( A \) : heat transfer area \([\text{m}^2]\)
\( A_s \) : interfacial area \([\text{m}^2/\text{m}^3]\)
\( a \) : effective interfacial area \([\text{m}^2/\text{m}^3]\)
\( C \) : heat capacity \([\text{J/mol}\)]
\( D \) : top product flow rate \([\text{kg/s}]\)
\( D_s \) : binary mass diffusion coefficient \([\text{m}^2/\text{s}]\)
\( d \) : diameter \([\text{m}]\)
\( e \) : energy flux \([\text{J/(s m}^2]\)]
\( F \) : feed flow rate \([\text{kg/s}]\)
\( H \) : molar enthalpy \([\text{J/mol}]\)
\( h \) : heat transfer coefficient \([\text{W/(m}^2\text{K})]\)
\( k \) : mass transfer coefficient \([\text{mol/m}^2\text{s}]\)
\( L \) : liquid phase flow rate \([\text{kg/s}]\)
\( N \) : mass flux \([\text{kg/(s m}^2]\)]
\( P \) : pressure \([\text{kPa}]\)
\( q \) : heat duty \([\text{W}]\)
\( R \) : reflux ratio \([-\text{}]\)
\( R_e \) : Reynolds number \([-\text{}]\)
\( S \) : side length of a packing \([\text{m}]\)
\( S_e \) : Schmidt number \([-\text{}]\)
\( T \) : temperature \([\text{K}]\)
\( U \) : overall heat transfer coefficient \([\text{W/(m}^2\text{K})]\)
\( u \) : velocity \([\text{m/s}]\)
\( V \) : vapor phase flow rate \([\text{kg/s}]\)
\( W \) : bottom product flow rate \([\text{kg/s}]\)
\( x \) : liquid phase fraction in mol or mass \([-\text{}]\)
\( y \) : vapor phase fraction in mol or mass \([-\text{}]\)
\( Z \) : height of the section \([\text{m}]\)
\( z \) : normalized height \([-\text{}]\)

\(<\text{Greeks}>\)
\( \Delta d \) : thickness of the wall \([\text{m}]\)
\( \lambda \) : thermal conductivity \([\text{W/(m K)}]\)
\( \rho \) : density \([\text{kg/m}^3]\)

\(<\text{Subscripts}>\)
bottom : bottom of the section
cross : cross sectional area
F : feed
HEX : heat exchange
i : i-th component
j : j-th segment
L : liquid phase
P : constant pressure

\(<\text{Superscripts}>\)
\( L \) : liquid phase
\( V \) : vapor phase

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内部熱交換型蒸留塔の熱・物質移動

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本研究では、速度論モデルを用いた内部熱交換型蒸留塔定常状態操作型シミュレーターを開発した。ここでは熱と物質の同時移動現象を蒸気相移動抵抗支配（液側抵抗は考慮しない）と仮定し、濃縮部-回収部間の伝熱量を蒸留塔内部の状態から推定するために、Cilton-Colburn のアナロジーを用いて伝熱メカニズムをモデル化し、それに対する操作変数の影響について検討した。また、内部熱交換型蒸留塔の移動現象を蒸気相移動抵抗支配（液側抵抗は考慮しない）と仮定し、濃縮部-回収部間の伝熱量を評価した。作成したシミュレーションモデルを用いて計算した結果、熱と物質の同時移動現象を考慮した本モデルを用いることで、実証試験機の結果を比較的良好に予測可能なことが分かった。