Recent advances in the internally heat-integrated distillation columns (HIDiC)

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Abstract

Distillation is the most widely used but the most energy-consuming separation process in the chemical industry. The internally **H**eat-**I**ntegrated **Di**stillation **C**olumns (**HIDiC**) are one of the promising alternatives of the conventional distillation processes to reduce the energy consumptions in chemical processes. The HIDiC has a similar structure to a heat exchanger. The rectifying section is contacted with the stripping section through walls in order to allow heat exchange between these two sections. Thus the residual heats in the column can be utilized in itself.

In Japan, a national project on the research and the development of the HIDiC has been conducted, and now it is in an important phase before its commercialization. For the analysis of the energy consumption of the HIDiC, simulation studies have been carried out. The simulation results show that an energy saving of 50 % is expected for the HIDiC when it is applied to the separation of hydrocarbons mixtures. On the other hand, developments of internals and the analysis of their hydrodynamic characteristics by experiments have been also addressed. Here we report such recent advances in the HIDiC.

Introduction

Distillation is said to be the most matured and frequently used technology of separation processes in many literature on chemical engineering. Adding a large amount of thermal energy as a separation agent, products of a chemical process can be highly purified and fractionated in the distillation process. In addition, the production rates of a distillation column are usually higher than the other separation instruments. Therefore, the energy consumption of a distillation process is generally much bigger than the other separation methods. So far, many modifications of distillation columns have been proposed and utilized in the chemical industries. Among such new developments of distillation processes, an internally **H**eat-**I**ntegrated **Di**stillation **C**olumn (**HIDiC**) is one of the promising alternatives. The early concept of the HIDiC was proposed by Haselden with examples of gas separations (Haselden, 1958). In the concept, the compressed feeds or products from a

distillation column can be utilized as an energy source for the distillation columns and it leads to a reduction of the energy consumption of the process. The idea was reintroduced in 1970's as a **S**econdary **R**eflux and **V**aporization (**SRV**) method by Mah et al. (1977), and the general configuration of the today's HIDiC was established. At that time, the Japanese government was paying a lot of efforts to overcome the two energy crisis in 1970's, since almost all energy resources have been imported from other countries. Therefore, there were large demands on the technology for energy savings from all industries in Japan. Since 1980's, researches on distillation columns with the SRV method have been carried out under the name of the HIDiC by Takamatsu and Nakaiwa (Nakaiwa et al., 2003). They further analyzed and modified the original arrangement of the SRV method. From 1990's, a national project on the HIDiC was conducted. In the project, the structure, the performance, the dynamic response and the control strategy for the HIDiC were investigated by both experiments and simulations. With an example of benzene-toluene separation, an energy saving of 30 % in average was achieved by a pilot HIDiC. It was operated even without any condensers, i.e., at zero reflux condition for more than 100 hours. Since then, the commercialization of the HIDiC becomes the next big target. From 2002, a new national project on the HIDiC has been started to industrialize the HIDiC as a key technology for the energy savings of chemical industries.

In this paper, the recent advances of the HIDiC are reported for simulation results of multicomponent separation examples, and developments and evaluations of the internals. Among the advances, the simulation results are mainly shown here. In addition, the HIDiC project in the other countries are also introduced.

The structure of the HIDiC

Figure 1 shows the schematic diagram of a typical configuration of the HIDiCs. As well as the conventional distillation column, the HIDiC has a rectifying and a stripping sections, a reboiler, and a condenser. In addition, a compressor and a throttling valve are equipped for the HIDiC, and either a preheater or a valve cooler, or both may be accompanied with them. By the compressor, the pressure of the vapor flowing up from the top of the stripping section is elevated, and the pressure of the liquid flowing down from the rectifying section is lowered by the throttling valve. If the temperatures for the rectifying and the stripping sections are different from each other and these two sections are contacted through a dividing wall, the temperature difference between the two sections bears a driving force for the heat transfer. Furthermore, when the pressure for the rectifying section is higher enough than the one for the stripping section, the one-way heat transfer from the rectifying section to the other can be expected. This decreases the required reboiler heat duty and the vapor and liquid flow rates inside the column, and enhances the degree of the energy utilization in a distillation column. Thus the HIDiC can realize the reduction of the energy consumption of the distillation process.
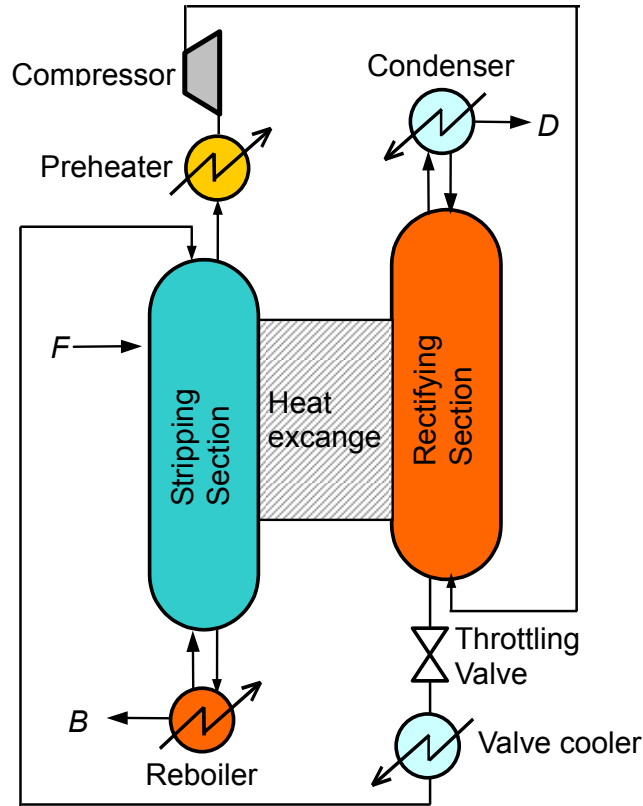
Figure 1. A schematic diagram of the HIDiC.

The model for the HIDiC

The model for the HIDiC is described in the paper by Mah et al. (Mah et al., 1977), and summarized by Nakaiwa with examples of binary separation problems (Nakaiwa, 1988). Since it is based on the tridiagonal matrix method proposed by Wang and Henke (Wang and Henke, 1966), here the difference between the original one and the model for the HIDiC is briefly explained. In order to represent the amount of the heat exchange between the rectifying and the stripping sections, the overall heat transfer coefficient, U, and the heat exchange area, A, are introduced. If the HIDiC is assumed to be a tray column, the amount of the heat exchange between the coupled stages, Q_{ex}, is expressed as follows

$$Q_{ex,j} = U_j A_j (T_j - T_{j+Nhex}) \tag{1}$$

where N_{hex} is the number of the heat exchange stages, and the subscript j denotes the j th stage in the rectifying section. Treating $Q_{ex,j}$ as a duty for a side cooler or a side reboiler, the tridiagonal matrix for the mass and the energy balances is solved. If one takes $A = 0$, $Q_{ex,j}$ becomes also zero and the terms for the heat exchange in the tridiagonal matrix disappear. Thus the Nakaiwa's model for the HIDiC can also handle the conventional column without

any correction. The detail of the model is described in elsewhere (Iwakabe et al., 2004a).

In the following sections, one of separation examples is explained. Since all mixtures studied in the present work consist of hydrocarbons and aromatic hydrocarbons, their vapor-liquid equilibrium (VLE) relationships are nearly ideal. Therefore, the VLE on each stage are calculated by the Soave-Redlich-Kwong (SRK) equation of state (Soave, 1972). The enthalpies and entropies are also estimated by the departure function of SRK equation and heat capacities of components. The compression duty is calculated by assuming the isentropic compression and the ideal behavior of the compressor, that is, the efficiency of the compressor is 100 %. Taking the electricity cost into consideration, the total energy consumption of the HIDiC, Q_{cons}, is calculated by the following equation

$$Q_{cons} = Q_{reb} + 3\ Q_{comp} \tag{2}$$

where Q_{reb} is the reboiler heat duty, Q_{comp} is the compressor duty. The factor of three multiplied to the compressor duty is empirically estimated by the energy cost in Japan.

The simulation of the HIDiC

Based on the above model, a HIDiC steady-state simulator has been developed by our group and the energy consumptions of both the HIDiC and the conventional distillation column are obtained. In our previous study, three separation problems are studied: (A) benzene-toluene-p-xylene mixture (B) ternary hydrocarbons mixture (C) twelve-component hydrocarbons mixture (Iwakabe et al., 2004a-e). The problem (A) is an extended study of benzene-toluene separation where the energy saving of 30 % by the HIDiC have been confirmed by both experiments and simulations as mentioned above, and an example of the separation of aromatic hydrocarbons that are frequently carried out in the practical chemical processes. The mixture examined in the problem (B) consists of the major three components of the twelve-component hydrocarbons mixture in the problem (C). The problem (C) is the most realistic example since there is a plan to apply the HIDiC to the separation and the separation data for the conventional distillation column is available. The details of the results for these three problems are shown in elsewhere (Iwakabe et al., 2004). In this paper, the new results for the problem (C) are shown in the following discussion. The feed compositions of the mixture are shown in **Figure 2**, and the specification of the separation problem is summarized in **Table 1**. The target component of the top product is n-pentane and its target purity is 84.3 mol%. This purity is equivalent to 99.4 mol% recovery of n-pentane in the feed. For the simulation, the constant pressure drop per stage of 0.3 kPa and Murphree vapor efficiency of 70 % regardless of the components are assumed both for the conventional distillation column and the HIDiC. The pressure of the feed stage of the conventional distillation column and the HIDiC is fixed at 1.013 x 10^5 Pa.

The effects of the total number of stages on the reflux ratio of the conventional distillation column, and the HIDiCs with *UA* of 5.0 kW/(K·stage) and compression ratios 1.6, 1.7, and 1.8 are shown in **Figure 3a**. The reflux ratios required for the HIDiCs are less than

those for the conventional distillation column. Especially for the case with the compression ratio of 1.8, the HIDiC of more than 52 stages can achieve the desired separation even at zero reflux condition. This less value of the reflux ratio for the HIDiC leads to a great reduction of the energy consumption of a distillation column as shown in **Figure 3b**. For example, if the conventional distillation column and the HIDiCs at the compression ratios of 1.6 and 1.7 have 70 stages, the energy consumptions are 302, 238, and 152 kW, respectively, i.e., the energy saving by the HIDiCs at the compression ratio of 1.6 and 1.7 are about 20 and 50 %, respectively. The ratios of the energy consumption for the HIDiC to that for the conventional distillation column at different compression ratios are compared in **Figure 4**. As can be seen from the figure, when the column has more than 40 stages, the HIDiC spends less energy than the conventional column does. In addition, with the increase of the compression ratio, the energy consumption of the HIDiC reduces. However, the lower compression ratio is preferable since the compressor duty occupies a large part of the energy consumption at high compression ratios, even more than the reboiler duty. To decrease the compression ratio, the *UA* value plays very important role. **Figure 5a** shows the inverse-proportional relationship between the compression ratio and *UA* for the HIDiC with 50 stages and at zero reflux condition. For the energy consumption, the compressor duty converted by multiplying a factor of three, $3Q_{comp}$, is about 60 % of the total energy consumption, Q_{cons} at low compression ratios, However, it increases up to about 70 % at high compression ratios, as shown in **Figure 5b**. From these results, the compression ratio around 1.8 and UA about 5 kW/(K·stage) seem to be the most economical and appropriate to satisfy the specification of the separation problem.
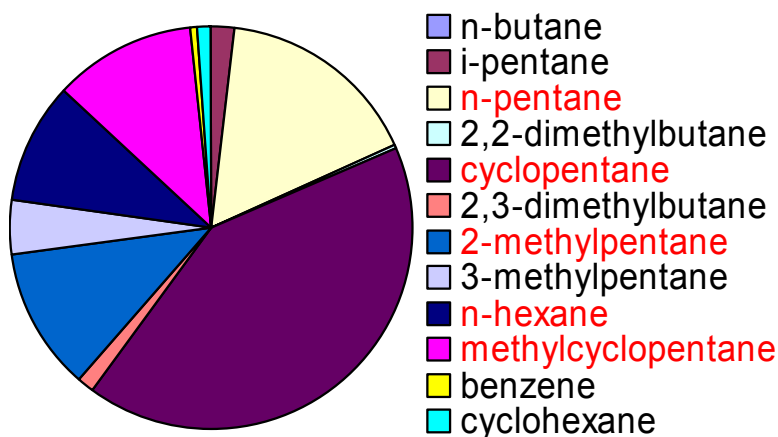

Figure 2. The feed compositions of the twelve-component hydrocarbons mixtures.

Table 1. Feed flow rate, top product flow rate, normal boiling points and the feed compositions of the twelve-component hydrocarbons mixture.

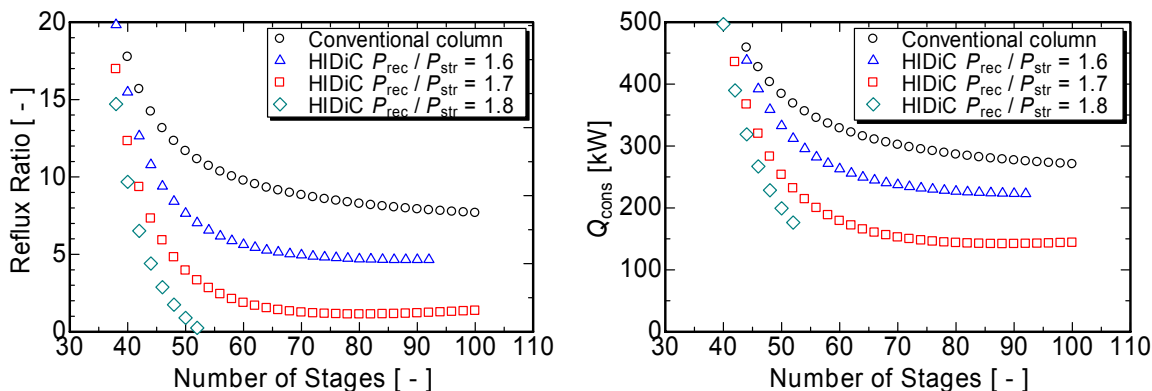| F [mol/s]	D [mol/s]	Component	bp [K]	x_{Fi} [-]
5.778	1.117	n-Butane	272.7	0.0005
		i-Pentane	301.1	0.0197
		n-Pentane	309.3	0.1639
		2,2-Dimethylbutane	322.9	0.0019
		Cyclopentane	322.5	0.4130
		2,3-Dimethylbutane	331.2	0.0139
		2-Methylpentane	333.5	0.1159
		3-Methylpentane	336.5	0.0433
		n-Hexane	341.9	0.0962
		Methylcyclopentane	345.0	0.1154
		Benzene	353.3	0.0043
		Cyclohexane	353.9	0.0120

Figure 3. The effects of the total number of stages (a) on the reflux ratio required to obtain the specified purity of the top product, and (b) on the energy consumptions of the conventional distillation column and the HIDiC with UA = 5.0 kW/(K·stage).
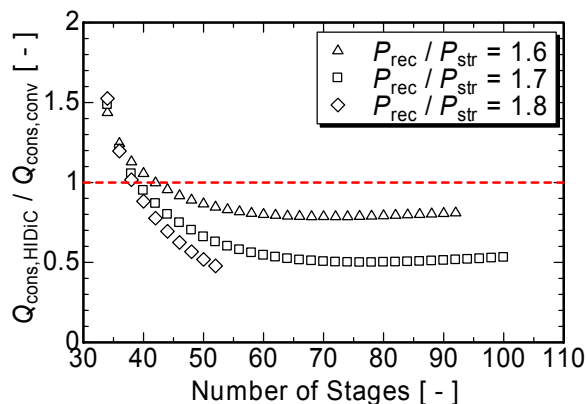

Figure 4. The comparison of the ratio of the energy consumption for the HIDiC to that for the conventional distillation column at various compression ratios.
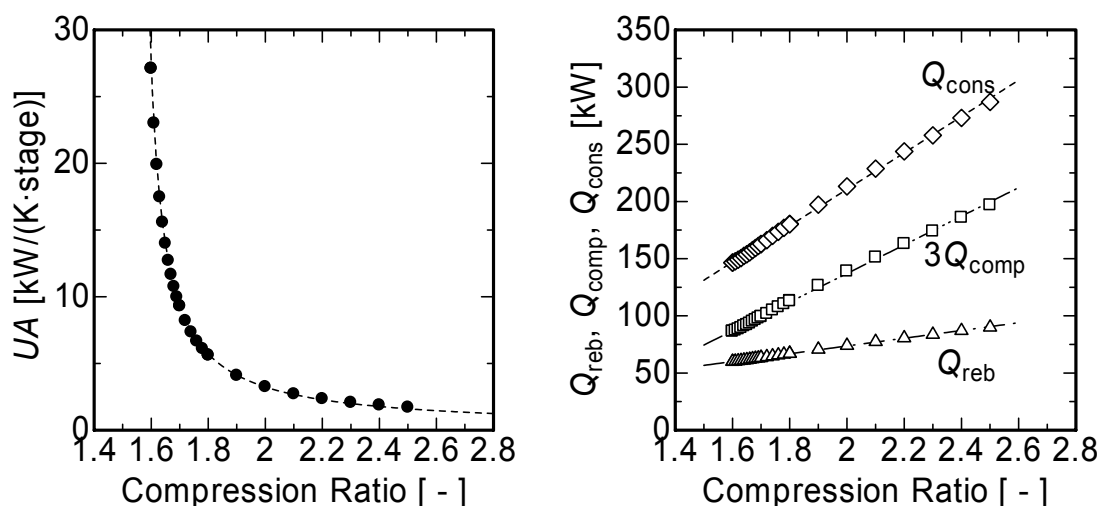
Figure 5. (a) The relationship between the compression ratio and *UA* for the HIDiC with 50 stages and at zero reflux condition. (b) The effects of the compression ratio on the energy consumptions for the HIDiC.

Developments and evaluations of internals for the HIDiC

Developments and evaluations of internals for the HIDiC are one of the important branches of the HIDiC project in Japan. In this section, several studies presented in the sessions related to separation technology of the Japanese domestic meetings on chemical engineering are summarized.

Kataoka et al. developed a twin plate-columns system thermally linked together with heat transfer tubes, and the plate efficiency and the heat transfer characteristics were evaluated. They reported that the rate of the heat transfer from the high temperature column to the low temperature column was not influenced directly by either of the vapor phase flow rates of the two columns. They also showed that the experimental overall heat transfer coefficient between the lower- and higher-pressure columns lied in the order between 400 and 1000 $W/(m^2 \cdot K)$, and within the operating condition of internal heat integration the average plate efficiency were in the range between 40 and 90 % (Kataoka et al., 2003; Kataoka et al., 2004). Nakanishi et al. developed a wetted wall type HIDiC that consists of a shell and tube heat exchanger in which the shell side is a wetted wall and the tube is packed with regular packings (Nakanishi et al., 2003). The overall heat transfer coefficient for the column was also about 1000 $W/(m^2 \cdot K)$.

Another important subject of our HIDiC project is a cryogenic air separation by the HIDiC equipped with a plate-fin type flow path. Kinoshita et al. constructed an aluminum plate fin type heat exchanger, and its H.E.T.P. was ranged from 0.2 to 0.4, which is almost equivalent to that for the ordinary regular packings (Kinoshita et al., 2003). Since the plate-fin is made by aluminum, it is very light and small. Tachibana et al. experimentally

confirmed that the pressure of the oxygen product in the packed column type HIDiC was about 259 kPaG, that is lower than the pressure of the entrance of the high pressure tower in the typical air separation system of 420 kPaG (Tachibana et al., 2003). Therefore, the cryogenic air separation process can be operated at less pressure by the HIDiC, that is, the reduction of the compressor duty is expected.

The HIDiC project in Europe

A HIDiC project has been conducted by Olujic et al. in the Delft University of Technology in the Netherlands, and some companies in Europe also belong to the project. They have shown that the energy saving of 90 % will be achieved if the HIDiC is applied to the separation of propylene and propane (C3 splitter) (Olujic et al., 2003). In the project, a conceptual design procedure of the HIDiC utilizing the pinch analysis has been proposed (Olujic et al., 2004). The reduction of CO_2 emission by the HIDiC with no reboiler is estimated to be 83 % compared to the conventional column, and even 36 % compared to the pump assisted distillation column (Gadalla et al., 2004). They have also experimental setups for testing the hydraulics of the HIDiC. As well as the Japanese HIDiC project, the commercialization of the HIDiC is in sight.

Concluding Remarks

The recent advances in the HIDiC project in Japan were introduced, especially, the simulation results for the separation problem of the twelve-component hydrocarbons mixture were explained in detail. For the separation problem, the energy consumptions of the HIDiC were about 50 % of those for the conventional distillation column. Among the design and the operating variables, the compression ratio and *UA* significantly affected the performance of the HIDiC. The other important subjects for the HIDiC are developments and evaluations of the internals suitable for the HIDiC. Although these internals are designed for the HIDiC, their overall heat transfer coefficients were equivalent to those for the conventional columns. On the other hand, it has been confirmed by experiments that the pressure of the oxygen product in the cryogenic air separation can be lowered by the HIDiC, which will lead the reduction of the total energy consumption of the process. The HIDiC is also studied in the European project conducted by Olujic et al., and interesting results for C3 splitter have been presented, especially the energy saving of 90 % is predicted for the separation. They will also industrialize the HIDiC in very near future.

In the Japanese project, there is a plan to separate the twelve-component hydrocarbons, which is one of the model mixtures in our simulation studies, by the pilot HIDiC next year. The authors hope that the HIDiC is utilized in many chemical processes, and give a great contribution to the sustainable world.

Acknowledgment

Nomenclature

A	=	heat transfer area per stage	$[m^2 \cdot stage^{-1}]$
B	=	bottom product flow rate	$[mol \cdot s^{-1}]$
bp	=	boiling point at 1.013×10^5 Pa	[K]
D	=	distillate flow rate	$[mol \cdot s^{-1}]$
F	=	feed flow rate	$[mol \cdot s^{-1}]$
N	=	number of stages	[-]
P	=	pressure	[Pa]
Q	=	energy flow rate	[kW]
R	=	reflux ratio	[-]
T	=	temperature	[K]
U	=	overall heat transfer coefficient	$[kW \cdot m^{-2} \cdot K^{-1}]$
x	=	liquid composition	[-]

<Subscript>

comp	=	compressor
cons	=	consumption
B	=	bottom product
D	=	distillate
F	=	feed
hex	=	heat exchange
i	=	i th component
j	=	j th stage
reb	=	reboiler
rec	=	rectifying section
str	=	stripping section

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