

AN INTERNALLY HEAT-INTEGRATED DISTILLATION COLUMN (HIDiC) IN JAPAN

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In Japan, a national research project of an internally heat integrated distillation column (HIDiC) has been conducted since 2002. Several significant outcomes from the project have been obtained. In this paper, the overview of our project is shown, and advances of this project are introduced.

KEYWORDS: energy saving, heat integration

INTRODUCTION

Energy savings of distillation processes are inevitable demands for the sustainable world since the distillation processes are the biggest energy consumers in the chemical industries. To improve energy efficiency of distillation processes, a lot of modifications have been proposed so far, such as thermally-coupled distillation column, which makes a conventional two-column process into a single distillation column. An internally heat-integrated distillation column (HIDiC) is also one of such promising alternatives of the conventional distillation processes to reduce the energy consumptions in the chemical plants. In the HIDiC, the whole or part of the rectifying section is contacted with the stripping section through walls in order to allow heat exchange between these two sections. This structure enables to consume the invested energy more efficiently than the conventional distillation. The early concept of the HIDiC was firstly proposed by Haselden in 1958 for the gas separation [1]. Later the idea was reintroduced by Mah et al. as Secondary Reflux Vaporization (SRV) method [2].

The idea of the HIDiC attracted researchers' interests in Japan, since there were two big worldwide energy crises in 1970's and, at the same time, few energy resources are available in Japan. In 1980's, the early researches were carried out by Takamatsu and Nakaiwa in Kyoto University and a former Agency of Industrial Science and Technology, Japan [3–4]. Their researches were focused on developments of analysis and design methods for HIDiCs. Thus the concept of HIDiCs has been not a mere theory but a practicable technology in Japan. As part of national research projects for energy savings and

environmental protection, the first national HIDiC project has been conducted since 1993. A lot of studies have been made in the project, and finally the bench plant of the HIDiC showed 30% energy reduction for the separation of benzene-toluene binary mixture [5]. In addition to the research progress, Japan became the chair country of COP3 where Kyoto protocol was proposed. In Kyoto protocol, Japan is requested to cut down 6% of emissions of greenhouse gases in 1990 levels for the period 2008–2012. Therefore, the energy savings of industrial processes has become the center of attention in Japan. The present national project for the HIDiCs has been brought into action since 2002. It targets the commercialization of HIDiCs, and now it is in the final stage. Here we report the significant outcomes and the state of the art for the HIDiC technology in Japan.

THE PRINCIPLE OF A HIDiC

A typical structure of a HIDiC is shown in Figure 1. The HIDiC consists of a rectifying section, a stripping section, a reboiler, a condenser, a compressor and a throttling valve. The feed mixture is usually fed into the top of the stripping section. The vapor from the top of the stripping section is compressed by the compressor to raise the pressure of the rectifying section, whereas the pressure of the liquid from the bottom of the rectifying section is decreased by the throttling valve. If the pressure of the rectifying section is

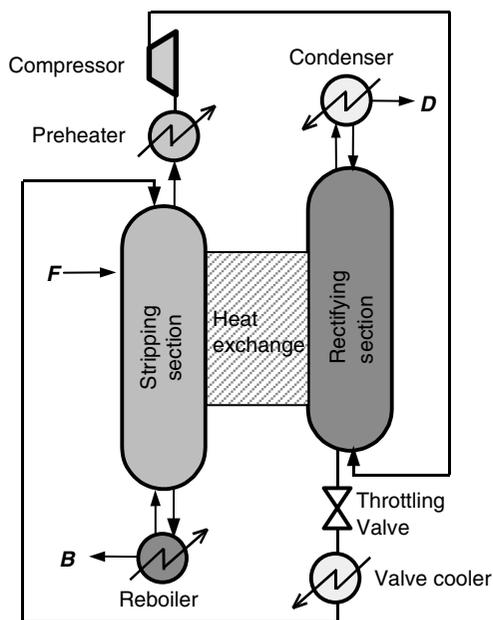


Figure 1. A schematic diagram of the HIDiC

sufficiently higher than that of the stripping section, the temperature of the rectifying section can be also higher than that of the stripping section. Therefore, if the whole or the part of the rectifying section is contacted with the stripping section, heat transfer between these two sections can be expected. The heat transfer will work like side coolers/heaters, leading to smaller reboiler heat duty. Thus the energy saving of the HIDiC is achieved.

THE CURRENT HIDiC PROJECT IN JAPAN

The current national project for the development of the HIDiCs has been conducted since 2002 with financially supported by New-Energy and Industrial Technology Development Organization (NEDO), Japan. The project is organized by Energy-Efficient Chemical Systems Group in National Institute of Advanced Industrial Science and Technology, Japan (AIST). The members of the project are AIST, Kyoto University, Nagoya Institute of Technology, Maruzen Petrochemical Co., Ltd., Kimura Chemical Plants Co., Ltd. and Kansai Chemical Engineering Co., Ltd. There are three targets of the project; (1) developments of operation and control methodologies for multicomponent separation by HIDiC (2) R&D of various types of HIDiC for the separation of hydrocarbon mixture (Group 1) (3) development of cryogenic air separator (Group 2). These three subprojects

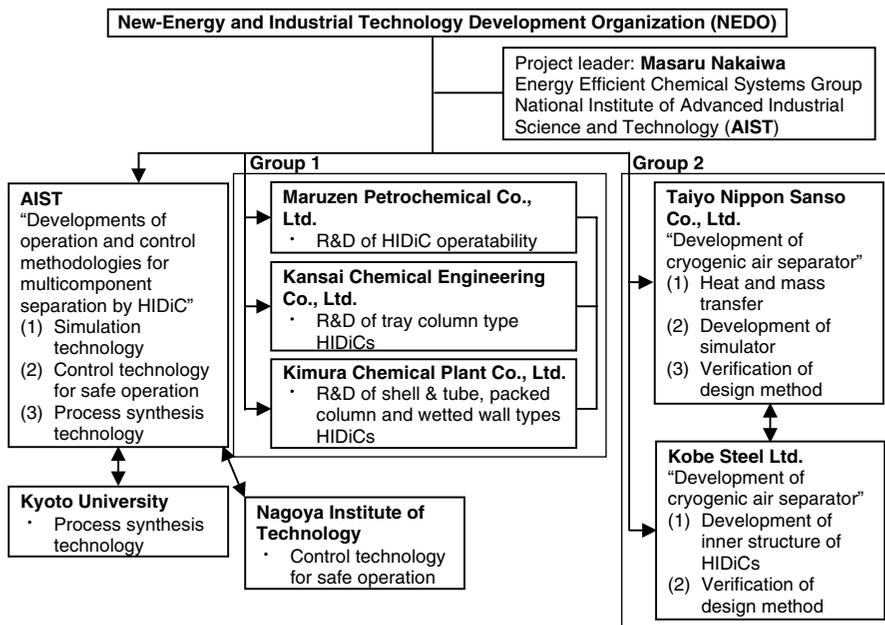


Figure 2. The structure of the national research project for HIDiCs

are individually carried out by each subproject members. The structure of the project is shown in Figure 2. So far, the progress of the project has been presented in several domestic and international conferences [6–7]. In the preceding sections, some of recent advances in each project are shown.

R&D OF VARIOUS TYPES OF HIDiCS

In Group 1, various types of HIDiCs have been designed, and their performances have been studied. Simulation and design technologies for the pilot HIDiC were developed in Group 1. A classification of practical separation examples by Column Grand Composite Curve (CGCC) method was carried out to find the feasibility of HIDiCs. They also developed a dynamic simulator for HIDiCs, and their dynamic characteristics were investigated. Besides the simulation studies, a double tube tray column type HIDiC was developed by Kansai Chemical Engineering Co., Ltd., and its heat transfer characteristics were reported [8]. The experimental overall heat transfer coefficients between the rectifying and the stripping sections, which is indispensable for design and analysis of HIDiCs, were 400–470 W/(m² · K) regardless of the compression ratio. In addition, a shell and tube type HIDiC has been designed by Kimura Chemical Plants Co., Ltd. The cross section is shown in Figure 3. Based on the design, a pilot plant of HIDiC [9–10] was constructed in 2005, and has been continuously operated for 1000 hours. In addition, more than 50% of energy reduction for separation of twelve component hydrocarbons was confirmed by the pilot HIDiC at the various operation conditions.

R&D OF CRYOGENIC AIR SEPARATION WITH THE HIDiC

In Group 2, R&D of cryogenic air separation by the HIDiC has been conducted. A compact heat exchanger-like vapor-liquid contactor was designed and applied to the HIDiC in order to enhance the heat transfer between vapor and liquid flows. Since the compact heat exchanger consists of layered fins, its scale-up is relatively simple. The plate-fin also realizes a counter-current vapor-liquid two-phase flow with only one flow path. Thus the heat exchange devices for the HIDiC can be smaller in size.

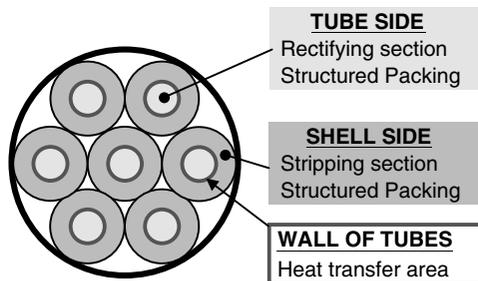


Figure 3. The cross section of the shell and tube type HIDiC

In order to attain sufficient heat transfer, a distribution method for vapor and liquid flows was devised by Kobe steel Ltd. With the plate-fin, a small air separator was developed. Heat and mass transfer correlation was made from the experimental data by Taiyo Nippon Sanso Co., Ltd. According to the correlation, a rate-based simulator was developed and validated by comparing the predicted results with the experimental data. The simulator predicted that an energy saving of 40% can be achieved by the plate-fin type HiDiC in the separation of oxygen from the air. The simulator also showed a possibility of further downsizing of the HiDiC air separator.

STEADY-STATE SIMULATIONS OF THE HiDiC FOR MULTICOMPONENT SEPARATIONS

A steady-state HiDiC simulator was developed in AIST. Energy savings by a HiDiC in the separation of twelve-component hydrocarbons have been confirmed at various conditions [7, 11–12]. Approximately 50% reduction of energy consumption was expected for the HiDiC.

CONTROL STRATEGY FOR HiDiCS

Control strategy for HiDiCs is indispensable for commercialization. A multiloop control system for a HiDiC has been examined by Kyoto University team. Some paring variables sets were determined by using a relative gain array, and controllers were designed by an internal model control for each paring set. As a result, the controllability of HiDiC was almost equivalent to that for the conventional distillation column.

RATE-BASED SIMULATION MODEL FOR THE BENCH PLANT OF THE HiDiC

In our previous paper [5], experimental approaches to zero external reflux and/or no reboiling operation in the HiDiC of benzene-toluene system were made. In the paper, the bench type HiDiC consumed 40% less energy than the conventional distillation column. In order to analyze the HiDiC bench plant more precisely, a rate-based model is needed since the HiDiC bench plant is a packed column. Therefore, a simulator with a rate-based model was developed by AIST for the prediction of the separation performance of HiDiCs with structured packing.

SIMULATION MODEL

In the simulation, a typical non-equilibrium stage model proposed by Krishnamurthy and Taylor's method with Maxwell-Stefan approach [13–15] was employed. The following simple assumptions are made for the present simulation; (1) The condenser and the reboiler are assumed to be at equilibrium (2) Vapor-liquid equilibrium occurs at the vapor-liquid interface (3) Interfacial area for mass and heat transfer is equal to the apparent surface area of the packing (4) Bulk phase is perfect mixing. The liquid hold up for structured packings was calculated using the empirical correlation developed by Xu *et al.* [16], and the binary mass transfer coefficients were estimated using the empirical correlation

Table 1. Basic equations for the rate-based model

Liquid hold up [16]	$h_d = 0.0336u_G^{1/3}u_L^{0.429}$	(1)
Binary mass transfer coefficients [17, 18]	$\frac{k_G d_e}{D_G} = 0.0338 \left\{ \frac{d_e \rho_G (u_{Ge} + u_{Le})}{\mu_G} \right\}^{0.8} \left(\frac{\mu_G}{\rho_G D_G} \right)^{1/3}$	(2)
	$k_L = 2\sqrt{D_L u_{Le}} / (\pi S)$	

developed by Bravo *et al.* [17, 18]. The equations are shown in Table 1. Heat transfer coefficients were computed by using the Chilton-Colburn analogy proposed by King *et al.* [19]. The resulting differential and algebraic equations in this model were implemented in a commercial process simulator gPROMS[®] (Process Systems Enterprise Limited).

OUTLINE OF BENCH PLANT

Figure 4 shows a schematic diagram of the test plant. The test plant was a double pipe heat exchanger system, which consists of shell and tube column. This plant was 16 m in height and 254 mm in diameter. The shell side was the stripping section, which was operated at nearly atmospheric pressure and packed with Mitsubishi Chemical MC Pack 500. On the other hand, the tube side was the rectifying section. The rectifying section was operated at

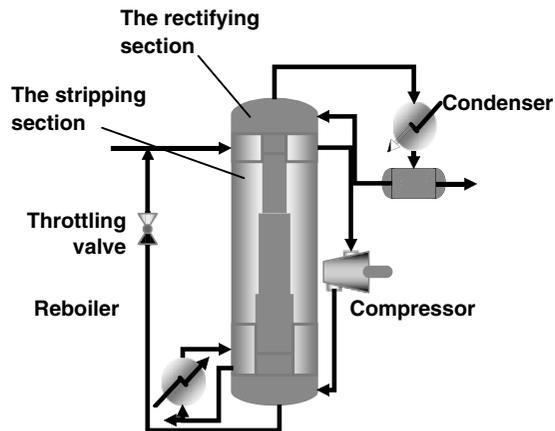


Figure 4. A schematic diagram of a concentric type HIDiC

Table 2. Operating condition for the tube unit of pilot plant

Pressure of the rectifying section	P_r	0.24	[MPa]
Pressure of the stripping section	P_s	0.124	[MPa]
Feed flow rate	F	340	[kl/h]
Reflux ratio	R	0	[-]
Feed composition	x_B	0.5851	[-]
	x_T	0.4148	[-]
Feed thermal condition	q	1	[-]

about twice the pressure of that for the stripping section. The tube side section was packed with MC Pack 350. From the top to the bottom, three different diameters of the section, 140, 165 and 190 mm were used, and their heights were 3, 8, 5 m, respectively.

In order to validate the present simulation model, a comparison of experimental data by a bench-scale HIDiC with the simulated results by the present model was made. Table 2 summarizes the operating condition of the bench plant, which was also used in the simulation. Table 3 shows a comparison of the predicted data, such as temperatures, flow rates and concentrations of top and bottom, by the present model with observed ones. Good agreements are observed between the experimental data and those obtained by the present model.

TYPICAL PHENOMENON OF HIDiC

Figure 5 shows the predicted flow rate profiles in the column. The ordinate is the mass flow rates of vapor and liquid, and the abscissa is the packed height. As can be seen from the figure, the vapor and the liquid flow rates increase from the bottom to the top in the stripping

Table 3. Comparisons of simulation results with the observed data in the pilot plant for the HIDiC

	Temperature				Flow rate		Concentration			
	The rectifying section		The stripping section		Bottom product	Distillate product	The rectifying section			Top
	Bottom Liquid	Top Vapor	Bottom Liquid	Top Vapor			section bottom	Bottom	Top	
$T_{r,b}$	$T_{r,t}$	$T_{s,b}$	$T_{s,t}$	W	D	Benzene $x_{r,b,B}$	Toluene $x_{r,b,T}$	Toluene $x_{w,T}$	Benzene $x_{D,B}$	
	[K]				[kl/h]		[-]			
Experimental	390.3	381.7	388.8	365.1	160	180	0.6226	0.3773	0.9984	0.9932
Calculated	395.9	388.5	385.5	370.6	152	188	0.6326	0.3674	0.9637	0.8835
Deviation [%]	1.43	1.78	-0.85	1.51	-5.00	4.44	1.61	-2.62	-3.48	-11.05

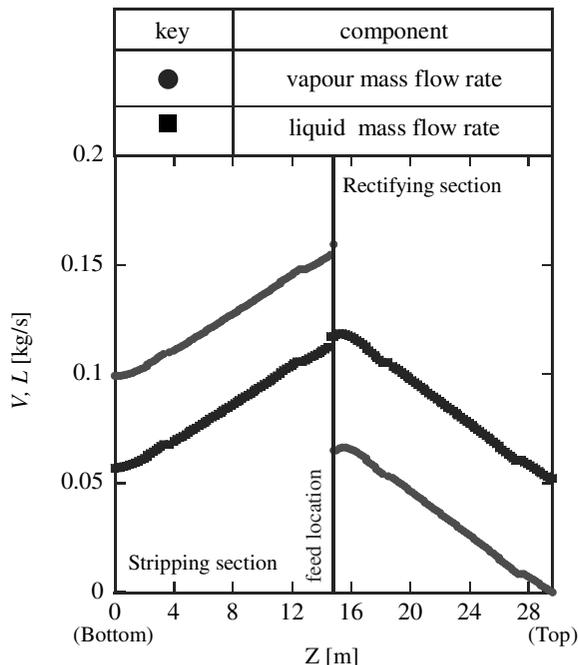


Figure 5. Variation of mass flow rates along the tube unit

section. In the rectifying section, on the other hand, the flow rates decrease from the bottom to the top. It exhibits the evaporation of the liquid in the rectifying section and the condensation of the vapor in the stripping section. The phenomenon occurred because of heat exchange along the tube unit between the rectifying and the stripping sections. Note that the liquid flow rate at the top of the rectifying section is zero, i.e. there is no external reflux.

REDUCTION OF ENERGY CONSUMPTION

The startup dynamic simulation quantified the pressure and heat exchange during the transition from cold and empty to full operation. The results are shown in Figure 6. It verifies that the equipment could be started up as planned. Further runs could then be performed to minimize the startup time for the full operation. The plots in the figure show the energy input into the system through startup to the steady state operation. The reboiler and condenser duties for both HIDiC configurations are substantially less than those for the conventional column, while the additional compression energy requirement is relatively small. These indicate that the HIDiC system can save to 40% of the energy required for separation.

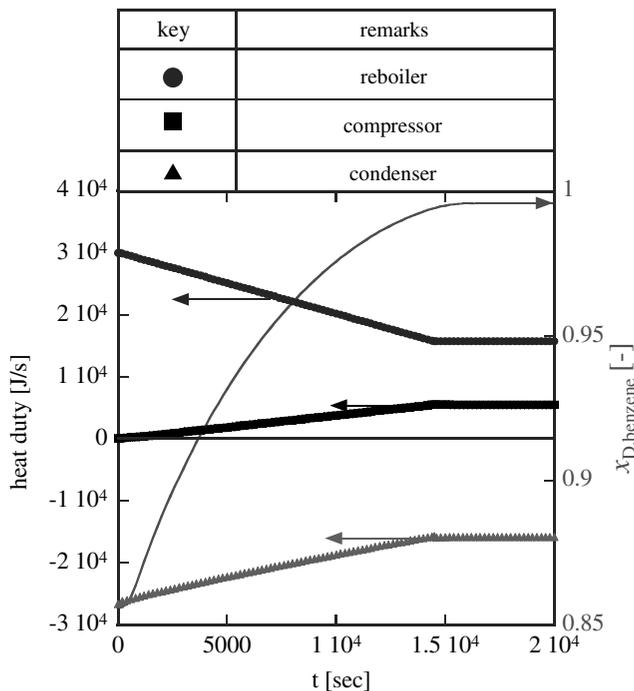


Figure 6. Transient responses of the startup operation

CONCLUSIONS

An overview of a national project on R&D of HIDiCs in Japan was shown in this paper. Several internals for the HIDiCs were developed and their performances were thoroughly evaluated in the project. With the results, a HIDiC pilot plant was developed and operated continuously for 1000 hours. For the separation of the twelve hydrocarbons mixture, the pilot HIDiC showed about 60% reduction of energy consumption than the conventional distillation column.

As well as experimental studies, simulations of separation performance of a bench plant by HIDiC with structured packing were carried out in AIST. A method for prediction of separation performance of a HIDiC was developed by using a rate-based model. A comparison of predicted separation performance of a bench-scale HIDiC by the present model showed excellent agreement with the experimental data.

Now the commercialization of HIDiCs is close at hand. The project members hope that the HIDiC technology will be widely used not only in Japan but also all over the world in near future, and it will also contribute to our sustainable world.

NOMENCLATURE

D	binary diffusion coefficient, m^2/s
d_e	equivalent diameter of channel, m
F	feed flow rate, kl/h
h_d	dynamic liquid holdup, m^3/m^3
k	binary mass transfer coefficient, $\text{kmol}/\text{m}^2\text{s}$
L	liquid phase mass flow rate, kg/s
P	pressure, MPa
q	thermal condition
R	reflux ratio
S	channel side dimension, m
T	temperature, K
u_G	superficial gas velocity, m/s
u_L	superficial liquid velocity, m/s
V	vapor phase mass flow rate, kg/s
x	liquid phase mass fraction
y	vapor phase mass fraction
Z	packed height from top of column

GREEK SYMBOLS

μ	viscosity, $\text{Pa} \cdot \text{s}$
ρ	density, kg/m^3

SUBSCRIPTS

B	benzene
b	bottom
G	gas phase
L	liquid phase
r	rectifying section
s	stripping section
T	toluene
t	top
V	vapor phase

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