

Recent Advances in Internally Heat-Integrated Distillation Columns (HIDiC) for Sustainable Development

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Internally heat integrated distillation columns (HIDiCs) have attracted industrial attention since HIDiCs are expected to have a strong impact on energy savings in chemical industries. Particularly in Japan, large national research projects for HIDiCs have been conducted, and a HIDiC pilot plant has been constructed. The pilot HIDiC has shown energy savings of more than 60% compared to a conventional distillation column in the separation example of a twelve-component hydrocarbon mixture. In addition, several research projects on HIDiCs have been carried out also in Europe. In this paper, the overview and recent advances in HIDiC technologies of Japan and Europe are introduced.

Introduction

Energy savings of distillation processes are essential for sustainable development since distillation processes are the larger energy consumers in chemical industries. To improve the energy efficiencies of distillation processes, a number of modifications have been proposed so far, such as thermally-coupled distillation columns, which make a conventional two-column process into a single distillation column. Above all, internally heat-integrated distillation columns (HIDiC) are expected to be the most promising alternatives for the conventional distillation processes to reduce the energy consumption in chemical plants. In the HIDiCs, the whole or a part of the rectifying section is in contact with the stripping section through the walls in order to allow heat exchange between these two sections. This structure enables utilize the invested energy more efficiently than conventional distillation. The original concept of the HIDiC was proposed by Haselden in 1958 for gas separation (Haselden, 1958). Later, the idea was reintroduced by Mah *et al.* (1977) as the Secondary Reflux Vaporization (SRV) method.

The idea of HIDiC attracted researchers' interests in Japan, since there were two big worldwide energy crises in the 1970's as well as few energy resources available in Japan. In the 1980's, early researches were carried out by Takamatsu and Nakaiwa in Kyoto University and AIST, Japan (Nakaiwa, 1988; Takamatsu *et al.*, 1988). Their researches were focused on developments in analysis and design methods for HIDiCs, which elevated the conceptual HIDiC theory to be a practical technology. As a part of the national

research projects for energy savings and environmental protection, the first national HIDiC project was launched in 1993. A number of researches have been made in the project, and finally a bench plant of the HIDiC (HIDiC-1) was developed for the first time. The bench HIDiC achieved a 30% energy reduction for the separation of a benzene-toluene binary mixture (Naito *et al.*, 2000, Nakaiwa *et al.*, 2003).

In addition to the research progress, Japan became the chair country of COP3 where the Kyoto Protocol was proposed. In the Kyoto Protocol, Japan committed itself to a 6% reduction in greenhouse gas emissions from 1990 levels in a five-year period from 2008 to 2012. Therefore, the energy savings of industrial processes has become the center of attention in Japan. From 2002, another national project for the HIDiCs was brought into action until 2006. It targeted the commercialization of HIDiCs, and significant advancement towards HIDiC technologies was obtained. In order to make this HIDiC technology put to practical use especially in petrochemical and chemical processes, currently a HIDiC consortium has been conducted by AIST and five companies. Furthermore, Taiyo Nippon Sanso also has a R and D project of HIDiCs for the application of cryogenic air separation.

R and D projects for HIDiCs have also been conducted in Europe. Especially, TU Delft and eight partners had a large R and D project in the Netherlands from 2002 to 2005. They showed the potential energy savings of HIDiC and reduction in CO₂ emission by comprehensive simulations (Olujic *et al.*, 2003). Also, they developed a tray column type HIDiC and the column proved to have sufficient mass and heat transfer efficiencies to achieve the desired energy saving performance by experiments (de Rijke *et al.*, 2006). They also started a new HIDiC project in 2007. On the other hand, a project in the UK was organized by Kaeser and Pritchard, (2006) at the University of Edinburgh. They developed a distillation column with a diabatic section and investigated the

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heat and mass transfer efficiencies by experiments.

In this paper, such important outcomes in HIDiC technologies in Japan are mainly reviewed, and the state of the art of HIDiC technologies including European projects is introduced.

1. HIDiC Principle

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 typically 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 sufficiently higher than that of the stripping section, the temperature of the rectifying section can also be higher than that of the stripping section. Therefore, if the whole or a 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 condenser and reboiler heat duties. Thus, energy saving by HIDiCs is achieved. As its side effect, the volumes of the condenser and the reboiler can be smaller.

Many simulation results show that the HIDiC can be operated at zero external reflux condition, that is, the HIDiC can work without a condenser. If energy is appropriately supplied not by a reboiler, but a feed preheater in the HIDiC, even the reboiler can be eliminated from the process. The ultimate configuration of HIDiCs without a condenser at the top and a reboiler at the bottom is called an ideal HIDiC. On the other hand, a HIDiC with a condenser and a reboiler is called a general HIDiC.

A question might arise from the above explanation regarding why the total energy consumption of the HIDiCs becomes less than those of the conventional distillation columns (CDiCs) even though the HIDiCs require compressor duty in addition to the reboiler heat duty. There are two answers to this question. The first answer is straightforwardly derived by the thermodynamic behavior of a vapor flow; if the temperature rise of a vapor flow after the polytropic compression is equivalent to the one after only heating without the pressure change, the compressor work needed to raise the pressure of the vapor flow is usually less than the heat duty required for the typical heat exchanger. The second answer is that both vapor and liquid flow rates inside the HIDiCs become less than ones in CDiCs due to the internal heat exchange, and it leads to a reduction in the energy consumption of the HIDiCs.

2. Progress in HIDiC Technologies in Japan

The first phase of the HIDiC R and D project was carried out until 1999, and the primary results have been reviewed in our previous paper (Nakaiwa *et al.*, 2003). The second

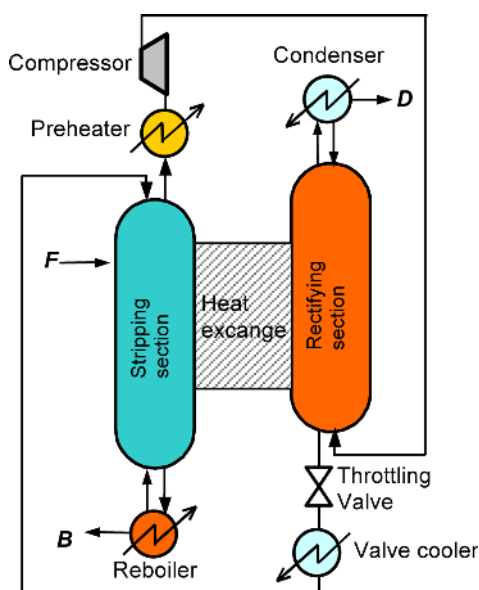


Fig. 1 A schematic diagram of the HIDiC

phase of the HIDiC R and D project was supported by the New Energy and Industrial Technology Development Organization (NEDO), Japan, and organized by the National Institute of Advanced Industrial Science and Technology (AIST), Japan. Five companies (Maruzen Petrochemical Co., Ltd., Kimura Chemical Plants Co., Ltd., Kansai Chemical Engineering Co., Ltd., Kobe Steel Ltd. and Taiyo Nippon Sanso Co., Ltd.) and two universities (Kyoto University and Nagoya Institute of Technology) participated in the project. There were three primary objectives of this second HIDiC R and D project: i) to develop HIDiC simulators for the separation of multicomponent systems and to develop control strategies for HIDiCs, ii) to develop a HIDiC for petrochemical processes (group 1), and iii) to develop HIDiC technology for cryogenic air separation (group 2). The results from these groups are described in the following sections.

2.1 Extension of HIDiC technology to the separation of multicomponent mixture

As mentioned above, the energy saving potential of HIDiCs had been studied only by an example of the separation of a benzene+toluene binary mixture in the first HIDiC R and D project. However, practical distillation processes mainly separate multicomponent mixtures. Therefore, the extension of the HIDiC application to the separation of multicomponent mixtures was an inevitable subject in our second R and D project. We had to decide a multicomponent mixture to be separated by the HIDiC, but there had not been any adequate criteria, and there were not even HIDiC simulators for multicomponent separation problems. For this purpose, a Column Ground Composite Curve (CGCC) analysis of multicomponent separation has been devised. In addition, steady-state HIDiC simulation software is also indispensable to analyze the performance of a HIDiC in the separation of multicomponent mixtures as well as to carry out the CGCC analysis. In the following two sections,

Table 1 Design variables and a typical operating condition of HIDiC-2 by a tray column approximation

| Design variables | |
|--------------------------------------|--------|
| N [—] | 84 |
| UA [kW/(K·stage)] | 1.543 |
| N_{HEX} [—] | 41 |
| A typical operating condition | |
| $P_{\text{rec}} \times 10^{-5}$ [Pa] | 1.905 |
| $P_{\text{str}} \times 10^{-5}$ [Pa] | 1.013 |
| R [—] | 0.0 |
| F [mol/s] | 6.8339 |
| D [mol/s] | 0.9139 |
| W [mol/s] | 5.9200 |

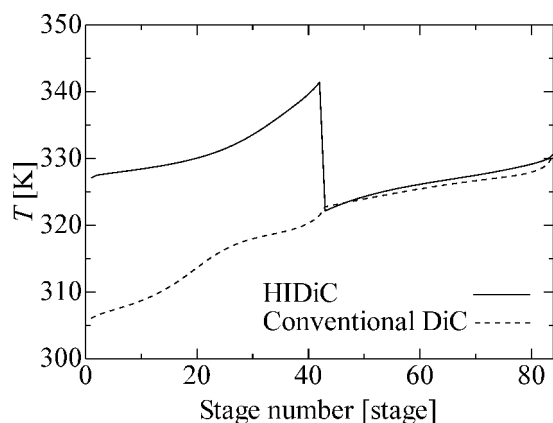


Fig. 2 An example of calculated temperature profiles for the HIDiC and the conventional distillation column by the HIDiC simulator with the equilibrium model

these two significant subjects are described.

2.2 Steady-state simulations of HIDiC for multicomponent separations

A general HIDiC simulator for multicomponent separation with the equilibrium model was developed by the AIST group. The HIDiC is treated as a tray column in the simulator, and the separation problems are solved by a modified Wang and Henke's tridiagonal matrix method. The software can handle design problems for HIDiCs as well as operation ones. With the simulator, the energy savings by HIDiCs in the separation of benzene+toluene, *n*-pentane+cyclopentane+2-methylpentane and even twelve-component hydrocarbon mixtures have been confirmed at various conditions (Iwakabe *et al.*, 2006a, 2006b). A typical example of a simulation condition is shown in **Table 1**, and the calculated temperature and flow rate profiles in the HIDiC and the CDiC for the separation of the twelve-component hydrocarbon mixture are shown in **Figures 2** and **3**. As similar results were also obtained at the other operation conditions, the energy savings by the HIDiC seems to be accomplished by reducing the flow rates inside the column resulting from the internal heat exchange. Then, the HIDiC pilot plant was split into 3 sections (Horiuchi *et al.*, 2008a, 2008b). This allows the specification of different geometries and/or other

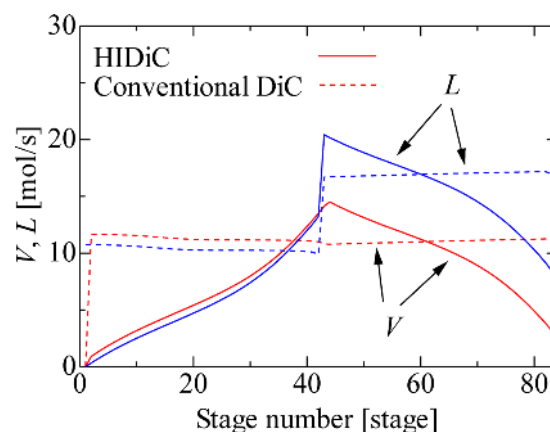


Fig. 3 An example of calculated vapor and liquid flow rate profiles of the HIDiC and the conventional distillation column by the HIDiC simulator with the equilibrium model

Table 2 A comparison of energy consumptions for the HIDiC and the conventional distillation column

| Column type | Conventional DiC | HIDiC |
|--------------------------------------|------------------|--------|
| N [—] | 84 | 84 |
| UA [kW/(K·stage)] | 0 | 1.543 |
| N_{HEX} [—] | 0 | 41 |
| $P_{\text{rec}} \times 10^{-5}$ [Pa] | 1.013 | 1.905 |
| $P_{\text{str}} \times 10^{-5}$ [Pa] | 1.013 | 1.013 |
| R [—] | 11.76 | 0.0 |
| Q_R [kW] | 346.96 | 73.96 |
| Q_{comp} [kW] | 0 | 23.75 |
| $Q_R + 3Q_{\text{comp}}$ [kW] | 346.96 | 141.61 |
| Q_{HEX} [kW] | 0 | 354.23 |
| Energy saving [%] | 0 | 59.2 |

properties (e.g. packing characteristics) in each section. This plant used a structured packing in all sections. Energy consumptions of the HIDiC and the CDiC are compared in **Table 2**. At the given condition, the energy savings by the HIDiC was about 60%. Furthermore, the simulation predicted the experimental data in HIDiC-2 with good accuracy (**Table 3**). The results obtained by the simulator can be utilized in CGCC analysis as mentioned later.

2.3 Rate-based simulation model for the HIDiC

As in our previous paper (Naito *et al.*, 2000; Nakaiwa *et al.*, 2003), experimental approaches to zero external reflux operations and operations without a reboiler were made with the HIDiC bench plant (HIDiC-1) in the separation of a benzene-toluene binary mixture. The results showed that HIDiC-1 consumed 40% less energy than the CDiC. Although the equilibrium model mentioned above is simple and concise to predict the performance of HIDiC-1, some phenomena could not be fully predicted since HIDiC-1 was a packed column and the effects of mass and heat transfer on the performance were not taken into account in our equilibrium model. A HIDiC simulator with the rate-based model has already been developed by Taiyo Nippon Sanso Group for their cryogenic air separator with a plate-fin type

Table 3 A comparison of simulation results obtained by the equilibrium model with the observed data in HIDiC-2

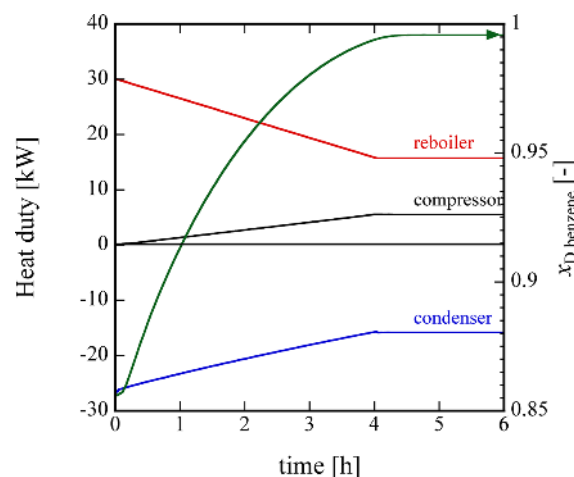
| Compound | HIDiC-2 | | | Simulation | |
|--------------------|----------|----------|----------|------------|----------|
| | z_{Fi} | x_{Di} | x_{Wi} | x_{Di} | x_{Wi} |
| <i>n</i> -Butane | 0.0004 | 0.0034 | 0 | 0.0034 | 0 |
| <i>i</i> -Pentane | 0.0133 | 0.0997 | 0 | 0.0997 | 0 |
| <i>n</i> -Pentane | 0.1167 | 0.8573 | 0.0024 | 0.8573 | 0.0024 |
| 2,2-Dimethylbutane | 0.0036 | 0.0006 | 0.0041 | 0.0002 | 0.0041 |
| Cyclopentane | 0.4300 | 0.0424 | 0.4899 | 0.0395 | 0.4903 |
| 2,3-Dimethylbutane | 0.0184 | 0 | 0.0213 | 0 | 0.0213 |
| 2-Methylpentane | 0.1533 | 0 | 0.1770 | 0 | 0.1770 |
| 3-Methylpentane | 0.0586 | 0 | 0.0677 | 0 | 0.0677 |
| <i>n</i> -Hexane | 0.1040 | 0 | 0.1201 | 0 | 0.1201 |
| Methylcyclopentane | 0.0974 | 0 | 0.1125 | 0 | 0.1125 |
| Benzene | 0.0009 | 0 | 0.0010 | 0 | 0.0010 |
| Cyclohexane | 0.0036 | 0 | 0.0042 | 0 | 0.0037 |

| Heat duty | HIDiC-2 | Simulation |
|-----------------|---------|------------|
| Q_{reb} [kW] | 29.17 | 24.69 |
| Q_{comp} [kW] | 36.09 | 32.12 |
| Q_{cons} [kW] | 131.99 | 121.05 |

vapor–liquid contactor, and it predicted the separation performance. In order to extend the advantage and the capability of the rate-based model, a prototype of the rate-based HIDiC model was developed by our AIST group (Matsuda *et al.*, 2006, 2007, 2008), and simulations of benzene (1)+toluene (2) separation were carried out under similar conditions to the experimental verification of HIDiC-1. In the simulation, a typical non-equilibrium stage model proposed by Krishnamurthy and Taylor with Maxwell–Stefan approach (Krishnamurthy and Taylor, 1985a, 1985b, 1985c) was employed. The differential and algebraic equations in this model were implemented in a commercial process simulator gPROMS® (Process Systems Enterprise Limited, PSE). Although fitting parameters or correlations obtained by experimental results of HIDiC-1 were not used in the simulation, the predicted values are in good agreement with the experimental ones.

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 4**. It verifies that the equipment can be started up as planned. Further runs could then be performed to minimize the startup time until operation. The plots in the figure show the energy input into the system during startup to the steady state operation. The reboiler and condenser duties for both HIDiC configurations were substantially less than those for the conventional column, while the additional compression energy requirement was relatively small. The simulation studies indicate that the HIDiC system can save up to 40% of the energy required for the separation of a benzene+toluene mixture under the conditions.

This rate based model has been further extended to other types of HIDiCs and multicomponent systems. Matsuda *et al.* (2008) compared the separation performance of the compact heat exchanger type HIDiC with the concentric double tube type HIDiC. The results show that the height of the

**Fig. 4** Transient responses of the startup operation

compact heat exchanger type HIDiC can be half that of the concentric double tube type HIDiC. Iwakabe *et al.* (2007) studied the effect of the heat and mass transfer in a general HIDiC by a rate based model and Chilton–Colburn analogy, and predicted the overall heat transfer coefficients in the HIDiC without any operation at data of the HIDiC pilot plant.

2.4 CGCC analysis of multicomponent separations

A classification of practical separation examples by Column Grand Composite Curve (CGCC) was carried out to verify the feasibility of HIDiCs. Basically, CGCC analysis is not very accurate for multicomponent separation problems. However, by choosing the appropriate key components and rearranging the composition profile of multicomponent separation as that of the pseudo-binary separation, CGCC analysis can be performed to predict a tendency of the energy saving potential in the application of HIDiC technology.

Examples of CGCC diagrams for the separation of twelve-component hydrocarbons mixture with a HIDiC are shown in **Figure 5**. The plotted data were calculated from the results of simulations of a HIDiC with the equilibrium model mentioned above. The CGCC diagram for each section shows the minimum reboiler and condenser loads distributed over the temperature ranges of columns, i.e. CGCC describes the energy requirement in each segment or stage in a distillation column. By analyzing the shapes of CGCC diagrams for existing conventional distillation columns, the criteria of mixtures and processes suitable for HIDiCs were proposed (Iuchi *et al.*, 2003). It was also found that the internal heat exchange in the HIDiC covers the majority of heat loads required in each stage at the appropriate design and operating condition as in Figure 5(b).

2.5 Operabilities and controllabilities of HIDiCs

Since the reflux flow rate of a HIDiC is very small, the separation and energy-saving performances are highly dependent on the internal heat exchange and might not be easily controlled by the reboiler heat duty as in the CDiCs. Therefore, an alternative control scheme would be required for HIDiCs. Multiloop control systems for several configurations of HIDiCs and the conventional distillation columns (CDiCs) were studied by Fukushima *et al.* (2006). In their analysis, the purities of the top and the bottom products were defined as controlled variables, and all possible pairings of controlled and manipulated variables for each structure were examined on the basis of a relative gain array to select suitable pairings for each structure. Their analysis showed that HIDiCs have slower dynamics than CDiCs. The controllabilities of HIDiCs were almost equivalent to those of conventional distillation columns so long as suitable control systems are designed. However, the selection of controlled variables for HIDiCs is different from that for conventional distillation columns. For example, not the reflux flow rate, but rather the top product flow rate would be appropriate as a manipulated variable to control the top product purity of HIDiCs against disturbances in feed flow rates and feed compositions, and against changes in set points. The control responses of a CDiC and a general HIDiC with a condenser and a reboiler to a disturbance in the feed flow rate and the setpoint change of the top product flow rate are very similar; however, the ideal HIDiCs showed very different tendencies. They also investigated the effects of the heat transfer area and the compression ratio of the general HIDiC on the control performance, and the controllabilities of the general HIDiC become worse as the internal heat exchange increases.

On the other hand, multiple steady-states were found even in a very simple example of benzene–toluene separation with a HIDiC by Kano *et al.* (2007). They pointed out that the energy balance, particularly concerning the compressor work, of the HIDiC may lead multiple steady-states. Especially when the compression ratio of the HIDiC is maintained at a constant value and the purity of the bottom product is high, multiple steady-states appear in a wide

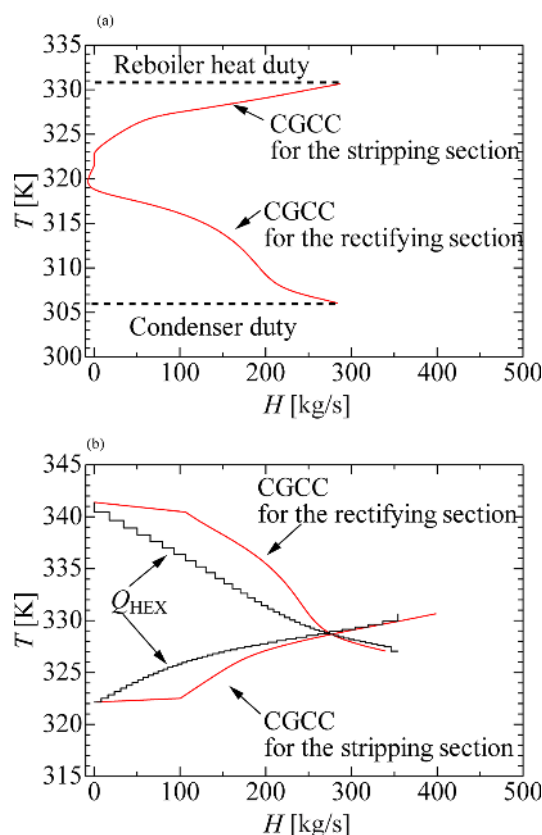


Fig. 5 CGCC diagrams for the separation of a twelve-component hydrocarbon mixture (a) with the conventional distillation column (b) with the HIDiC

range of operating conditions, suggesting that the operation and the control strategies for the HIDiC should be different from those for the CDiCs. Huang *et al.* have been studying the dynamic behavior of HIDiCs (Huang *et al.*, 2006a, 2006b, 2006c, 2006d, 2006e, 2006f, 2006g, 2007a, 2007b). Recently, they reported a new temperature control scheme for HIDiCs (Huang *et al.*, 2008). For distillation processes, a temperature control scheme would be desirable because of the relatively small investment, low maintenance cost, and high reliability. In their analysis, the temperature difference between two stages in the rectifying and the stripping sections, respectively, was designated as a controlled variable to circumvent a side effect of continuous pressure variations in the rectifying section. The temperature control scheme proposed can maintain stable operation around the vicinity of the nominal steady state with improved performance, and tolerable steady state discrepancies in comparison with the direct composition control scheme. They also reported the design and the control of totally heat integrated distillation columns in which the top vapor flow is used as a heat medium of the feed flow of the HIDiC.

2.6 R and D of various types of HIDiCs for the separation of hydrocarbon mixtures

Besides feasibility studies, the internals of HIDiCs were also designed in the group. A double tube tray column type



Fig. 6 The structure of the shell and tube installed in the HIDiC

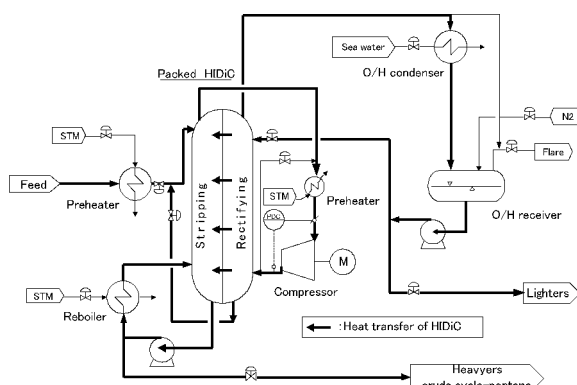


Fig. 7 Process flow diagram of the pilot plant of the HIDiC

HIDiC was developed by Kansai Chemical Engineering Co., Ltd., and its heat transfer characteristics have been reported (Noda *et al.*, 2006). The experimental overall heat transfer coefficients between the rectifying and the stripping sections, which is indispensable for the design and analysis of HIDiCs, were 400–470 W/(m²·K) regardless of the flow rate inside the column and the compression ratio.

The design method of packed column type HIDiCs was developed by Kimura Chemical Plants team through experimental analysis of the heat and mass transfer characteristics (Nakanishi *et al.*, 2008), and a shell-and-tube type HIDiC was designed, as shown in Figure 6. Based on the design, a pilot plant of the HIDiC (HIDiC-2) was constructed at Chiba Factory, Maruzen Petrochemical Co., Ltd. in 2005 (Horiuchi *et al.*, 2006, 2008b). Process flow diagram, structure of HIDiC-2, and photograph of HIDiC-2 are shown in Figures 7–9, respectively. The details of the design and typical operating results are shown in Table 4. HIDiC-2 was continuously operated for 1000h from September 5 to October 16, 2005. HIDiC-2 also successfully achieved the zero external reflux operation that minimizes the energy consumption of the HIDiC. More than a 50% energy reduction for the separation of twelve component hydrocarbons was confirmed by HIDiC-2 at the various operation conditions. Moreover, the design of HIDiC-2 has another advantage in that the capacity of the HIDiC can be easily changed by adjusting the number of shell-and-tubes bundled in the column.

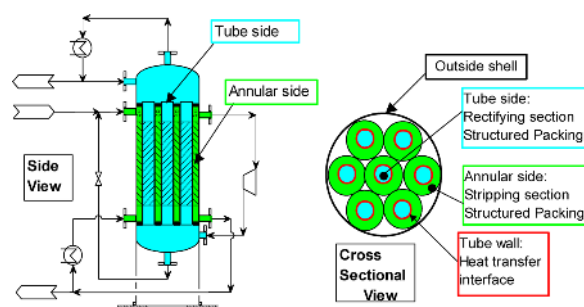


Fig. 8 The cross section of the shell and tube type HIDiC



Fig. 9 The HIDiC-2 pilot plant in Chiba Plant, Maruzen Petrochemical Co., Ltd.

Table 4 Design and operating condition of HIDiC-2

| | |
|--|--------|
| Geometry of HIDiC-2 | |
| Height [m] | 27 |
| Diameter [m] | 1.4 |
| Number of double-tube packed column [—] | 7 |
| Number of theoretical stage for each tube [stages] | 35 |
| Typical operating condition | |
| P_{rec} [kPa] | 190.5 |
| P_{str} [kPa] | 101.3 |
| F [kg/h] | 1900 |
| D [kg/h] | 237.3 |
| R [—] | 0 |
| Q_{reb} [kW] | 29.17 |
| Q_{comp} [kW] | 36.09 |
| Q_{cons} [kW] | 131.99 |

2.7 R and D of cryogenic air separation with the HiDiC

In this group, 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 in the column. Since the compact heat exchanger consists of layered fins, its scale-up is relatively simple. The plate-fin configuration also realized counter-current vapor–liquid two-phase flow with only one flow path. Thus, heat exchange devices for the HiDiC can be smaller in size. In order to attain sufficient heat transfer, a distribution method for vapor and liquid flows was devised by Kobe Steel Ltd. for the plate-fin configuration, a small air separator was developed by Taiyo Nippon Sanso Co., Ltd. and heat and mass transfer correlation was made from the experimental data. According to the correlation, a rigorous simulator with a rate-based model was developed and validated by comparing the predicted results with the experimental data. The simulator predicted that an energy savings of 40% can be achieved by the plate–fin type HiDiC in the separation of oxygen from air. The simulator also showed a possibility to further downsize the HiDiC air separator (Egoshi *et al.*, 2007; Tachibana, 2007). According to the results obtained, a pilot scale cryogenic air separator with the HiDiC technology was built and operated in 2008.

2.8 Implementation cost of HiDiC pilot plant

For the construction cost of the HiDiC pilot plant, the compressor accounted for one third of the total investment. Since the expected feed flow rate for the pilot HiDiC is about 2.0 t/h, the vapor flow rate of the inlet of the compressor is not very large as in practical chemical processes. There were not many choices for the compressor of such a scale, with the price of the compressor being relatively high. Although the compressor has set-point control, the pressure does not make a process variable. That is why attention to the following points is necessary for the operation of the compressor:

1. Operation timing in startup;
2. Compressor surge; and
3. Separation system: decomposition of composition, preparation of oxide material and/or condensation of composition by compression is unsuitable.

On the contrary, the condenser, the reboiler and the preheaters are much smaller than those for the conventional distillation column. Therefore the initial cost for the pilot HiDiC became 1.5 times higher compared to an existing conventional distillation column with the same separation capacity. On the other hand, the operation cost of the HiDiC is significantly affected by the price of electricity. Nowadays, there are various kinds of sources for the power generation such as thermal, nuclear and hydroelectric and, their costs could be very different from region to region. As for Japan, the efficiency of thermal power generation is around 40%. Taking crude oil prices at the end of December 2007 (\$90/bbl) into consideration, the payback time for a typical HiDiC would be 7–8 years (Horiuchi *et al.*, 2008a).

3. HiDiC Projects in Europe

In the Netherlands, the first HiDiC project was conducted from 2002–2005. The project members were Delft University of Technology (TU Delft), MolaTech BV, ABB Lummus, AKZO-Nobel Chemicals BV, BP/Nerefco, Energy Centre of the Netherlands (ECN), DSM, Shell GSI, and Sulzer Chemtech. They carried out systematical research subprojects on the technology, the economy and the ecology of HiDiCs. An annular sieve tray column type HiDiC fully equipped with the heat transfer panels was developed in the project, and the heat and mass transfer in the HiDiC were evaluated by experiments. The results suggest that sufficient internal heat transfer can be expected in their HiDiC. The tray efficiency of the HiDiC with heat transfer panels is almost identical to that of CDiCs under ordinary operating conditions (de Rijke *et al.*, 2006). Individually, ECN designed a thin-plate HiDiC, utilizing alternate parallel narrow channels as rectifying and stripping sections (Hugill and an Dorst, 2005a, 2005b). In addition to these developments of internals, they developed a HiDiC model on the commercial process simulator ASPEN Plus® to achieve model based evaluation of CDiCs, Vapor–Recompression Column (VRCs) and HiDiCs (Olujic *et al.*, 2003). They compared energy losses and utility consumption for these three kinds of columns. As a result, the HiDiC with the lower compression ratio showed the lowest exergy consumption. Surprisingly, the total energy consumption of the HiDiC is almost 10% that of CDiCs. In addition, the implementation cost of a HiDiC for a Propylene–Propane (PP) splitter was highly estimated from a very early stage in the project. In their analysis, the equipment cost of the HiDiC is only 10% higher than a CDiC. Furthermore, the operating cost of the HiDiC is estimated to be 30% less than that of CDiCs. The payback time of the HiDiC for a PP splitter is only 9 months, whereas that of the CDiC is about 20 months. A new conceptual design method of HiDiCs with heat panels was introduced based on the stage temperature profiles and hydraulic calculations (Gadella *et al.*, 2005b 2007; Olujic *et al.*, 2006; Schmal *et al.*, 2006). A variable heat transfer rate design leads to better designs with minimum total cost compared to constant heat transfer rate schemes. Through conceptual design of a HiDiC for a PP splitter (Olujic *et al.*, 2006), they concluded that compressor capital and operating costs are the main factors affecting the economy of a HiDiC. All of these results encourage the commercialization of HiDiCs. As well as these technological approaches, ecological analyses were vigorously carried out by Gadalla *et al.* (2005a, 2005b, 2006). They developed a model for the calculation of CO₂ emissions from distillation columns, and it was applied to several separation examples with HiDiC. The results show that the existing crude oil installations can save up to 83% in CO₂ emissions if a HiDiC configuration is applied to the system.

A retrofitting approach that introduces heat transfer plates on sieve trays in CDiCs has been studied by Pritchard's group at the University of Edinburgh. They reported the effects of the flow rate of the heat exchange

medium and the vapor load on the heat transfer coefficients and the Murphree efficiency (Kaeser and Pritchard, 2006). The experimental distillation column was operated in heating and cooling modes by changing the temperature and the flow rate of the heat exchange medium. In the heating mode, the experimental data of heat transfer coefficients were a 0.9 power relationship with the temperature difference between the plate and the bulk fluid on the tray, suggesting a nucleate boiling mechanism. In addition, the Murphree efficiency tends to increase with the heat transfer in the heating mode. In the cooling mode, the quenching leads to a bubble regime with low froth height, and consequently very low Murphree efficiencies. In both heating and cooling modes, the vapor velocity has little or no effect on the heat transfer coefficient from the tray to the fluid on the tray, but significantly affects the Murphree efficiency.

4. Perspective of HIDiC Technology

As many outcomes have been obtained in the national project, the next step in the R and D of HIDiCs becomes very important to spread HIDiC technology into practical applications. The applicability of HIDiCs is easily evaluated by CGCC analysis, even only with the data obtained by the existing conventional distillation processes. The equilibrium model can predict the energy consumption of HIDiCs with only simple assumptions such as the feed and product flow rates and compositions. The rate-based model approach would be a powerful tool to solve design and operating problems of HIDiCs since it has a strong ability to estimate the heat and mass transfer phenomenon inside the column in detail. Combining these three approaches, feasibility studies on HIDiCs can be carried out much easier than before.

Possible applications of HIDiC technology are not limited to ordinary conventional distillation processes. As mentioned in the example of bioethanol purification, a part of azeotropic distillation processes can also be replaced by HIDiCs. Reactive distillation processes are suggested to be a good application of HIDiCs by Huang *et al.* (2006f, 2006g). Furthermore, heat integration among two or more individual distillation columns as proposed by Kataoka *et al.* (2009) might bear a new separation sequence of distillation columns. A bioethanol purification process by a HIDiC with a membrane separator was proposed by the Nagoya Institute of Technology (NIT) team (Hashimoto *et al.*, 2009; Abe *et al.*, 2010). The ethanol concentration in the crude product of bioethanol obtained via fermentation process is usually low. Since the primary purpose of the production of bioethanol is to provide an eco-friendly fuel in place of fossil fuels, its purification process should also be eco-conscious. Therefore, a configuration to apply HIDiC technology to bioethanol purification was investigated. In the conventional separation method for the dehydration of ethanol, the azeotrope formed by ethanol and water is usually broken by adding solvents (entrainers such as benzene or cyclohexane). However, it requires another distillation tower to separate solvents from the aqueous ethanol mixture. To avoid

azeotropic distillation, membrane separation is thought to be one of the potential alternatives, and there have been many proposals for hybrid distillation systems. The concept of a hybrid separation system was extended to the HIDiC (Jana, 2010). The proposed configuration of the HIDiC with a membrane separator showed a possibility for energy savings in bioethanol purification.

According to a report from the Statistics Bureau and Statistical Research and Training Institute, Japan (2006), the annual CO₂ emission in Japan was 1178×10^6 t-CO₂ in the year 2002, a 12.2% increase from the 1990 level. Above all, the amount of CO₂ emission from chemical processes was about 70×10^6 t-CO₂. In these chemical processes, there are currently around 3,000 distillation columns in operation. If 30% of these columns are replaced by HIDiCs, the energy savings per year is estimated to be 1.557×10^9 L-crude oil, which is equivalent to a reduction in CO₂ emissions of 4.163×10^6 t-CO₂ (Horiuchi *et al.*, 2008a). About a 6% reduction in energy consumption in chemical plants can be achieved by only the substitution of 30% of the conventional distillation columns in Japan with HIDiCs.

Conclusion

An overview of a national project for the R and D of HIDiCs in Japan and Europe has been presented in this paper. Simulations of the separation performances of prototype and pilot plant HIDiCs with structured packing have been carried out in AIST, Kyoto University and Nagoya Institute of Technology. As well as simulation studies, several internals and new configurations and sequences of HIDiCs have been developed, and their performances have been thoroughly evaluated in the projects. Owing to these efforts, a HIDiC pilot plant was constructed and operated continuously for 1000 h. For the separation of a twelve hydrocarbon mixture, the pilot HIDiC showed an about 60% reduction in energy consumption over existing conventional distillation columns under the same separation condition. Moreover, technologies for HIDiCs have greatly progressed in Europe as well. HIDiC projects conducted in the Netherlands have been very successful, not only in terms of simulation works such as propylene+propane splitter examples and the development of internals, but also the evaluation of energy savings, cost and environmental effects. On the other hand, fundamental studies by Kaeser and Pritchard (2006) have clarified the relationship between the separation performance and the internal heat exchange of the HIDiC, and suggested that retrofit implementation of HIDiC technology to the CDiCs would be a good option.

At present the commercialization of HIDiCs is close. All of the project members hope that the HIDiC technology will be widely used not only in Europe and Japan, but also all over the world in the near future, and it will contribute to sustainable developments.

Nomenclature

| | | |
|---|-------------------------------------|-------------------------|
| A | = heat transfer area per stage | [m ² /stage] |
| D | = top product flow rate | [mol/s, kL/h or kg/h] |
| F | = feed flow rate | [mol/s, kL/h or kg/h] |
| H | = heat duty in CGCC analysis | [kg/s] |
| L | = liquid phase flow rate | [kPa] |
| N | = number of stages | [—] |
| P | = pressure | [kPa or MPa] |
| Q | = energy flow rate | [kW] |
| R | = reflux ratio | [—] |
| T | = temperature | [K] |
| U | = overall heat transfer coefficient | [kW/m ² ·K] |
| V | = vapor phase flow rate | [mol/s, kL/h or kg/h] |
| W | = bottom product flow rate | [mol/s, kL/h or kg/h] |
| x | = mole or mass fraction of liquid | [—] |
| y | = mole or mass fraction of vapor | [—] |
| z | = feed mole fraction | [—] |

Subscript

| | |
|--------|-------------------------|
| bottom | = bottom of the section |
| D | = top product |
| comp | = compressor |
| cons | = consumption |
| F | = feed |
| HEX | = heat exchanger |
| i | = i-th composition |
| L | = liquid phase |
| reb | = reboiler |
| rec | = rectifying section |
| str | = stripping section |
| top | = top of the section |
| V | = vapor phase |
| W | = bottom product |

Literature Cited

- Abe, H., H. Mori, Y. Hashimoto and S. Iwata; "Energy-Saving Performance of the Compressor Free HiDiC," Society of Chemical Engineers, Japan (SCEJ) 42th Autumn Meeting, **R308**, Kyoto, Japan (2010)
- de Rijke, A., W. Tesselaar, M. A. Gadalla, Z. Olujic and P. J. Jansens; "Heat and Mass Transfer Characteristics of an Annular Sieve Tray," Proceedings of the Distillation and Absorption 2006, IChemE Symposium Series No. 152, pp. 181–189, London, U.K. (2006)
- Egoshi, N., K. Ishikawa, S. Sakaue and H. Kawakami; "Air Separation Experiments by Plate-Fin Type HiDiC," Society of Chemical Engineers, Japan (SCEJ) 39th Autumn Meeting, **B117**, Sapporo, Japan (2007)
- Fukushima, T., M. Kano and S. Hasebe; "Dynamics and Control of Heat Integrated Distillation Column (HiDiC)," *J. Chem. Eng. Japan*, **39**, 1096–1103 (2006)
- Gadalla, M. A., Z. Olujic, L. Sun, A. de Rijke and P. J. Jansens; "Pinch Analysis-Based Approach to Conceptual Design of Internally Heat-Integrated Distillation Columns," *Chem. Eng. Res. Des.*, **83**, 987–993 (2005a)
- Gadalla, M. A., Z. Olujic, P. J. Jansens, M. Jobson and R. Smith; "Reducing CO₂ Emissions and Energy Consumption of Heat-Integrated Distillation Systems," *Environ. Sci. Technol.*, **39**, 6860–6870 (2005b)
- Gadalla, M. A., Z. Olujic, A. de Rijke, P. J. Jansens, M. Jobson and R. Smith; "Reducing CO₂ Emissions of Internally Heat-Integrated Distillation Columns for Separation of Close Boiling Mixtures," *Energy*, **31**, 2409–2417 (2006)
- Gadalla, M. A., L. Jiménez, Z. Olujic and P. J. Jansens; "A Thermo-Hydraulic Approach to Conceptual Design of an Internally Heat-Integrated Distillation Column (i-HiDiC)," *Comput. Chem. Eng.*, **31**, 1346–1354 (2007)
- Haselden, G. G.; "An Approach to Minimum Power Consumption in Low Temperature Gas Separation," *Chem. Eng. Res. Des.*, **36**, 123–132 (1958)
- Hashimoto, Y., T. Watanabe, K. Song, Y. Ota, T. Hamaguchi and H. Mori; "Heat Transfer Analysis of Tray-Type HiDiC," 8th World Congress of Chemical Engineering, 61, Montréal, Canada (2009)
- Horiuchi, K., K. Yanagimoto, K. Kataoka and M. Nakaiwa; "Energy-Saving Characteristics of Heat Integrated Distillation Column Technology Applied to Multi-Component Petroleum Distillation," Proceedings of the Distillation and Absorption 2006 (IChemE Symposium Series No. 152), pp. 172–180, London, U.K. (2006)
- Horiuchi, K., K. Keigo, K. Iwakabe and M. Nakaiwa; "Evaluation of Economical and Environmental Performance of an Internally Heat-Integrated Distillation Column (HiDiC)," *Kagaku Kogaku Ronbunshu*, **34**, 444–447 (2008a)
- Horiuchi, K., M. Nakaiwa, K. Iwakabe, K. Matsuda and M. Toda; "Intensification of the Process Flow in the Pilot Plant of the Internally Heat Integrated Distillation Column (HiDiC)," *Kagaku Kogaku Ronbunshu*, **34**, 70–75 (2008b)
- Huang, K., K. Matsuda, T. Takamatsu and M. Nakaiwa; "A Simple Method for Modeling Process Asymmetry," *J. Chem. Eng. Japan*, **39**, 448–452 (2006a)
- Huang, K., K. Matsuda, T. Takamatsu and M. Nakaiwa; "The Influences of Pressure Distribution on an Ideal Heat-Integrated Distillation Column (HiDiC)," *J. Chem. Eng. Japan*, **39**, 652–660 (2006b)
- Huang, K., K. Matsuda, K. Iwakabe, T. Takamatsu and M. Nakaiwa; "Graphical Synthesis of an Internally Heat-Integrated Distillation Column," *J. Chem. Eng. Japan*, **39**, 703–708 (2006c)
- Huang, K., K. Matsuda, K. Iwakabe, T. Takamatsu and M. Nakaiwa; "Choosing More Controllable Configuration for an Internally Heat-Integrated Distillation Column," *J. Chem. Eng. Japan*, **39**, 818–825 (2006d)
- Huang, K., K. Matsuda, K. Iwakabe, T. Takamatsu and M. Nakaiwa; "Interpreting Design of an Ideal Heat-Integrated Distillation Column through Exergy Analysis," *J. Chem. Eng. Japan*, **39**, 963–970 (2006e)
- Huang, K., K. Iwakabe, M. Nakaiwa and A. Tsutumi; "Towards Further Internal Heat Integration in Design of Reactive Distillation Columns—Part I. The Design Principle," *Chem. Eng. Sci.*, **60**, 4901–4914 (2006f)
- Huang, K., M. Nakaiwa and A. Tsutumi; "Towards Further Internal Heat Integration in Design of Reactive Distillation Columns—Part II. The Process Dynamics and Operation," *Chem. Eng. Sci.*, **61**, 5377–5392 (2006g)
- Huang, K. J., S.-J. Wang, K. Iwakabe, L. Shan and Q. Zhu; "Temperature Control of an Ideal Heat-Integrated Distillation Column (HiDiC)," *Chem. Eng. Sci.*, **62**, 6486–6491 (2007a)
- Huang, K., L. Shan, Q. Zhu, J. Qian, L. Shan, Q. Zhu and J. Qian; "Design and Control of an Ideal Heat-Integrated Distillation Column (Ideal HiDiC) System Separating a Close-Boiling Ternary Mixture," *Energy*, **32**, 2148–2156 (2007b)
- Huang, K., L. Shan, Q. Zhu, J. Qian, L. Shan, Q. Zhu and J. Qian; "A Totally Heat-Integrated Distillation Column (THiDiC)—The Effect of Feed Pre-Heating by Distillate," *Appl. Therm. Eng.*, **28**, 856–864 (2008)
- Hugill, J. A. and E. M. an Dorst; "Design of a Heat-Integrated

- Distillation Column based on a Plate-Fin Heat Exchanger," ECN Publication, **ECN-RX-05-157**, 1-19 (2005a)
- Hugill, J. A. and E. M. an Dorst; "The Use of Compact Heat Exchangers in Heat-Integrated Distillation Columns," ECN Publication, **ECN-RX-05-156**, 1-20 (2005b)
- Iuchi, K., M. Nakaiwa and H. Tatsumi; "Applicability of Internally Heat Integrated Type Energy Saving Distillation Column (HIDiC)," Society of Chemical Engineers, Japan (SCE) 36th Autumn Meeting, **B1P03**, Sendai, Japan (2003)
- Iwakabe, K., M. Nakaiwa, K. Huang, T. Nakanishi, T. Ohmori, A. Endo and T. Yamamoto; "Performances of an Internally Heat-Integrated Distillation Column (HIDiC) in Separation of Ternary Mixtures," *J. Chem. Eng. Japan*, **39**, 417-425 (2006a)
- Iwakabe, K., M. Nakaiwa, K. Huang, T. Nakanishi, A. Røsjorde, T. Ohmori, A. Endo and T. Yamamoto; "Energy Savings in Multicomponent Separation Using an Internally Heat-Integrated Distillation Column (HIDiC)," *Appl. Therm. Eng.*, **26**, 1362-1368 (2006b)
- Iwakabe, K., K. Matsuda, M. Nakaiwa, K. Horiuchi, T. Nakanishi, A. Horiguchi, K. Kubo, T. Ohmori, T. Yamamoto and S. Kataoka; "A Rate-Based Model Approach To Internally Heat Integrated Distillation Columns (HIDiC)," AIChE 2007 Annual Meeting, **168d**, Salt Lake City, U.S.A. (2007)
- Jana, A. K.; "Heat Integrated Distillation Operation," *Appl. Energy*, **87**, 1477-1494 (2010)
- Kaesler, M. and C. L. Pritchard; "The Impact of Heat Transfer on Murphree Tray Efficiency," *Energy*, **31**, 3108-3116 (2006)
- Kano, M., T. Fukushima, H. Makita and S. Hasebe; "Multiple Steady-States in a Heat Integrated Distillation Column (HIDiC)," *J. Chem. Eng. Japan*, **40**, 824-831 (2007)
- Kataoka, K., H. Noda, H. Yamaji, M. Mukaida and M. Kaneda; "A Compressor-Free HIDiC System for Recovery of Waste Solvent Mixtures," 8th World Congress of Chemical Engineering, 512a, Montreal, Canada (2009)
- Krishnamurthy, R. and R. Taylor; "A Nonequilibrium Stage Model of Multicomponent Separation Processes, Part I: Model Description a Method of Solution," *AIChE J.*, **31**, 449-456 (1985a)
- Krishnamurthy, R. and R. Taylor; "A Nonequilibrium Stage Model of Multicomponent Separation Processes, Part II: Comparison with Experiment," *AIChE J.*, **31**, 456-465 (1985b)
- Krishnamurthy, R. and R. Taylor; "A Nonequilibrium Stage Model of Multicomponent Separation Processes, Part III: The Influence of Unequal Component-Efficiencies in Process Design Problems," *AIChE J.*, **31**, 1973-1985 (1985c)
- Mah, R. S. H. Jr. J. J. Nicholas and R. B. Wodnik; "Distillation with Secondary Reflux and Vaporization: A Comparative Evaluation," *AIChE J.*, **23**, 651-658 (1977)
- Matsuda, K., K. Huang, M. Nakaiwa, T. Ohmori, A. Endo, T. Yamamoto, S. Kataoka K. Iwakabe T. Nakanishi, K. Kataoka and T. Takamatsu; "Simulation of Ternary Distillation in a Heat Integrated Distillation Column (HIDiC) with a Rate-Based Model," *J. Chin. Inst. Chem. Eng.*, **37**, 467-474 (2006)
- Matsuda, K., K. Iwakabe, K. Kubo, A. Horiguchi, Y. Weifang, H. Kosuge, S. Kataoka, T. Yamamoto, T. Ohmori and M. Nakaiwa; "Rate-Based Modeling for Internally Heat-Integrated Distillation Column (HIDiC) - Binary System," *J. Jpn. Pet. Inst.*, **50**, 162-168 (2007)
- Matsuda, K., K. Iwakabe, M. Nakaiwa, H. Kosuge, S. Kataoka, T. Yamamoto and T. Ohmori; "Simulation of Multicomponent Separation in Internally Heat Integrated Distillation Column using the Compact Heat Exchanger System," *Kagaku Kogaku Ronbunshu*, **34**, 64-69 (2008)
- Naito, K., M. Nakaiwa, K. Huang, A. Endo, K. Aso, T. Nakanishi, H. Noda and T. Takamatsu; "Operation of a Bench-Scale Ideal Heat Integrated Distillation Column (HIDiC): An Experimental Study," *Comput. Chem. Eng.*, **24**, 495-499 (2000)
- Nakaiwa, M.; "Fundamental Research on Energy Savings of Distillation Processes," Ph. D. Thesis, Kyoto University, Japan (1988)
- Nakaiwa, M., K. Huang, A. Endo, T. Ohmori, T. Akiya and T. Takamatsu; "Internally Heat-Integrated Distillation Columns: A Review," *Chem. Eng. Res. Des.*, **81**, 162-177 (2003)
- Nakanishi, T., N. Adachi, K. Aso, K. Iwakabe, K. Matsuda, K. Horiuchi, M. Nakaiwa and T. Takamatsu; "Development of a High Performance Distributor for an Internally Heat-Integrated Distillation Column," *Kagaku Kogaku Ronbunshu*, **34**, 224-229 (2008)
- Noda, H., T. Mukaida, M. Kaneda, K. Kataoka and M. Nakaiwa; "Internal Column-to-column Heat Transfer Characteristics for Energy-Saving Distillation System," Proceedings of the Distillation and Absorption 2006 (IChemE Symposium Series No. 152), pp. 737-744, London, U.K. (2006)
- Olujić, Z., F. Fakhri, A. de Rijke, J. Graauw and P. J. Jansens; "Internal Heat Integration —The Key to an Energy-Conserving Distillation Column," *J. Chem. Technol. Biotechnol.*, **78**, 241-248 (2003)
- Olujić, Z., L. Sun, A. de Rijke and P. J. Jansens; "Conceptual Design of an Internally Heat Integrated Propylene-Propane Splitter," *Energy*, **31**, 3083-3096 (2006)
- Schmal, J. P., H. J. Van der Kooi, A. de Rijke, Z. Olujić and P. J. Jansens; "Internal versus External Heat Integration—Operational and Economic Analysis," *Chem. Eng. Res. Des.*, **84**, 374-380 (2006)
- Statistics Bureau & Statistical Research and Training Institute, Ministry of Internal Affairs and Communications; CO₂ Emissions from Energy Use in Japan, <http://www.stat.go.jp/english/> (2006)
- Tachibana, H.; "Application Technology of HIDiC for Low Purity Oxygen Production Process," *Taiyo Nippon Sanso Giho*, **26**, 29-31 (2007)
- Takamatsu, T., V. Lueprasitsakul and M. Nakaiwa; "Modeling and Design Method for Internal Heat-Integrated Packed Distillation Column," *J. Chem. Eng. Japan*, **21**, 595-601 (1988)