## INTERNALLY HEAT-INTEGRATED DISTILLATION COLUMNS: A REVIEW

## M. Nakaiwa\*, K. Huang\*, T. Endo\*, T. Ohmori\*, T. Akiya\* and T. Takamatsu\*\*

\*National Institute of Advanced Industrial Science and Technology, Tsukuba 305-8565, Japan

\*\*Institute of Industrial Technology, Kansai University, Suita 564-8680, Japan

## ABSTRACT

The heat-integrated distillation column to be addressed in this paper is a special distillation column that involves internal heat integration between the whole rectifying and the whole stripping sections. An overview of the research on this process is presented in this work. It covers from the thermodynamic development and evaluations to the practical design and operation investigations for the process. Comparative studies against conventional distillation columns are introduced and the results obtained show distinctively the drastic advantages in energy efficiency of the process over its conventional counterparts. Some relevant issues of process design and operation are to be stressed and the results of the first of its kind bench-scale plant experimentation are given in great detail.

The application of internal heat integration principle to other distillation-related processes is also discussed in depth. The prospective of the HIDiC and our future research work are then highlighted, finally.

## INTRODUCTION

Distillation columns have been well known for its low energy efficiency. For effecting a separation heat has to be given at a high temperature in the reboiler and simply drawn off at a low temperature in the condenser. For improving its energy efficiency heat pump principle is often adopted, as an effective means, to reuse the rejected low-temperature heat [1], which is generally referred as heat pump assisted distillation column in the literature. Although it is a useful technique for energy savings, it suffers from some strict requirements imposed by mixtures to be separated. Since 1960s, internal heat integration between the rectifying and the stripping sections of a distillation column has gained significant incentives for improving energy efficiency of distillation processes. Freshwater might be the first person to advocate this technique [2]. Flower and Jackson further systematized the

idea and clarified the advantages of this approach through numerical simulations based on the second law of thermodynamics [3]. In terms of the same principle, Mah and his coworkers [4,5] developed and worked with their own process called secondary reflux and vaporization distillation column, which actually included internal heat integration between part of the rectifying and part of the stripping sections. They established the general process configuration to approximate the theoretical model based on the second law of thermodynamics. However, they did not address the problem to which degree the internal heat integration should be adopted between the rectifying and the stripping sections of a distillation column. Takamatsu and Nakaiwa have continued the work on this subject both theoretically and experimentally since 1986 and confirmed firstly by large-scale experimental evaluations the high advantages of these kinds of heat-integrated distillation columns in binary close-boiling mixture separations over conventional distillation columns [6]. In 1995, they noticed that the degree of internal heat integration within a distillation column played a very important role in energy efficiency for a given separation. They proposed, therefore, to further extend the internal heat integration to the whole rectifying and the whole stripping sections and resulted in a sharply different process configuration from conventional distillation columns, which they called heat-integrated distillation column (HIDiC). They found further that the HIDiC was feasible for separations of binary close-boiling mixtures, just as other types of heat pump assisted distillation columns [7,8].

It is worth mentioning here that the HIDiC possesses several very attractive features and it is these features that stimulate us to pursue its realization in practical process engineering. These features include: (i) High energy efficiency. The highest degree of internal heat integration within the HIDiC generally offers itself higher energy efficiency than conventional distillation columns as well as other types of heatintegrated distillation columns, for instance, heat pump assisted distillation columns; (ii) Zero external reflux and reboil operation. Ever since the creation of distillation techniques, it has been the common practice to use condenser and reboiler to generate external reflux and reboil flows for distillation operation. For the HIDiC, the internal heat integration generates these two flows, instead, and thus neither of them is necessary. This may motivate new considerations on distillation process design and operation; and (iii) High potentials and effectiveness of internal heat integration techniques. Internal heat integration is a very efficient means to improve process energy efficiency and can find wide applications within distillation processes. As will be discussed later, it can even facilitate operation of batch distillation columns and pressure-swing distillation processes, which are used for the separation of pressuresensitive binary azeotropic mixtures. Moreover, internal heat integration is not limited only to a single distillation column. It can be considered between two distillation columns that may have no direct connections at all.

Recently, the research on the HIDiC has aroused considerable interests and several research groups have been formed around the world. They have already begun their work on this subject with their emphasis ranging from process design [9,10], and process operations [11], to internal heat and mass transfer mechanism and internal structure arrangement [12,13], respectively. As we have been concentrating on this work for a quite long time, it seems to us that it is necessary to review its current development and predict its prospective. Therefore, the main objective of this paper is to give an in depth summation of our researches on the HIDiC. In the meantime,

considerable emphasis has also been placed on the introduction of applications of internal heat integration principle to other distillation-related processes, for example, batch distillation columns and pressure-swing distillation processes, as it also represents a very important aspect of the HIDiC development.

In this work a detailed overview of our researches on the HIDiC will be conducted, ranging from thermodynamic development and evaluations to the practical design and operation investigations for the process. Comparative studies against conventional distillation columns will be introduced and show distinctively the big advantages of the HIDiC over its conventional counterparts. Some relevant issues of process design and operation are to be stressed and the results of the first of its kind bench-scale plant experimentation will be given at full length. Some energy-efficient processes that make use of the internal heat integration principle, such as heat-integrated batch distillation columns and heat-integrated pressure-swing distillation columns, are also addressed in a straightforward manner. The prospective of the HIDiC and our future research work will be highlighted, followed by some concluding remarks in the last section of the work.

## THERMODYNAMIC ANALYSIS OF CONVENTIONAL DISTILLATION OPERATION

Figure 1 shows a diagram of a conventional distillation column. In terms of the firstand second- laws of thermodynamics, following equations can be derived.

$$Q_{\text{REB}} - Q_{\text{COND}} + FH_{\text{F}} - DH_{\text{D}} - BH_{\text{B}} = 0$$
(1)  
$$\Delta S = Q_{\text{COND}} / T_{\text{COND}} - Q_{\text{REB}} / T_{\text{REB}} - FS_{\text{F}} + DS_{\text{D}} + BS_{\text{B}} \ge 0$$
(2)

Dissipation energy,  $W_{Loss}$ , is the energy loss due to process irreversibility in the mass and heat transfer, pressure distribution and remixing within a distillation column. It is calculated as follows.

$$\begin{split} W_{\text{Loss}} &= T_0 \Delta S \\ &= Q_{\text{REB}} (1 - T_0 / T_{\text{REB}}) - Q_{\text{COND}} (1 - T_0 / T_{\text{COND}}) + F(H_F - T_0 S_F) \\ &- D(H_D - T_0 S_D) - B(H_B - T_0 S_B) \\ &= Q_{\text{REB}} (1 - T_0 / T_{\text{REB}}) - Q_{\text{COND}} (1 - T_0 / T_{\text{COND}}) - W_{\text{min}} \end{split}$$
(3)

Here,  $W_{min}$  is the minimum energy required by a certain separation and is determined by process operating conditions and product specifications.

$$W_{min} = (DH_D + BH_B - FH_F) - T_0(DS_D + BS_B - FS_F)$$
  
=  $\Delta H - T_0 \Delta S$  (4)



Figure 1. Schematic of a conventional distillation column

For improving the energy efficiency of a distillation column, it is necessary to reduce the dissipation energy,  $W_{Loss}$ , as possible as it can be. For the heat transfer and pressure distribution loss, they can be controlled by effective heat exchanger design and internal flow structure arrangement. For the remixing loss, it can be mitigated by selection of appropriate feed locations. The remained mass transfer loss is, therefore, left as the main reason for the low energy efficiency of a conventional distillation column, as it is closely related to the contacts and distributions of liquid and vapor flows.

The thermodynamic energy efficiency of a conventional distillation column can be defined as

$$\eta_{con} = W_{min} / (W_{Loss} + W_{min})$$
  
=  $W_{min} / (Q_{REB}(1 - T_0/T_{REB}) - Q_{COND}(1 - T_0/T_{COND}))$   
 $\leq W_{min} / (Q_{REB} - Q_{COND})$  (5)

Figure 2 shows a McCabe-Thiele diagram for a conventional distillation column operating at a certain reflux ratio condition. As can be seen, the mass transfer driving force is unevenly distributed along the length of the distillation column, with the smallest value at feed location and increasing gradually away from the feed location to both ends of the distillation column. The uneven distribution of mass transfer force constitutes the main reason of high degree of irreversibility in mass transfer. One can also imagine that even at the minimum reflux ratio operating condition, a great degree of irreversibility in mass transfer still exists within a conventional distillation column.



Figure 2. McCabe-Thiele diagrams for a conventional distillation column and a HIDiC

#### THE CREATION OF THE HIDIC WITH HEAT-PUMP PRINCIPLES

#### A Theoretical Model of the HIDiC

Figure 3 shows a temperature-heat (T-H) diagram of a conventional distillation column separating a benzene and toluene binary mixture. The T-H diagram has been developed based on a thermodynamic equilibrium operation within a distillation column. It can indicate heating and cooling sections of a distillation column and thus guide internal heat integration design. For a binary distillation column, its rectifying section is a cooling section and can provide heat outside. On the other hand, its stripping section is a heating section and needs external heating. These properties provide basis for consideration of internal heat integration within a conventional distillation column.

To reuse the heat available from the rectifying section to the stripping section, one may raise its temperature through heat pumps. With the assumption of a myriad of reversible heat pumps between corresponding stages of the rectifying and the stripping sections, following equations can be obtained.

$$dQ_{\rm R} = T_{\rm R} / T_{\rm S} dQ_{\rm S} \tag{6}$$

$$dW = (T_S - T_R)/T_S dQ_S$$
(7)

Integration of these two equations, one can get

$$\int_{TC}^{TF} \frac{dQ_R}{T_R} = \int_{TF}^{TB} \frac{dQ_S}{T_S}$$
(8)

$$W = \int_{T_F}^{T_B} \frac{T_s - T_R}{T_s} dQ_s = Q_s - Q_R$$
(9)

Eq. 9 has well been illustrated in Figure 3. One can readily understand that at

appropriate operating conditions, distillation processes can become self-content in heat utilization and the separation can be driven by shaft work, only. Neither condenser nor reboiler looks necessary. Furthermore, the pressure elevation from the stripping section to the rectifying section and feed thermal condition must be determined carefully so as to satisfy Eq. 9. This provides us fundamentals and guidelines for the development of a new model of internally heat-integrated distillation column, namely, the HIDiC.



Figure 3. T-H diagram of a conventional distillation column

Re-examine Figure 2, one can see that internal heat integration leads to the operation curve of the HIDiC exactly the same as the equilibrium one, With the same mass transfer duty, the HIDiC needs apparently less driving force than conventional distillation columns at the minimum reflux ratio operating condition, hence presenting higher energy efficiency.

## The Creation of the HIDIC: A Practical Approximation

It is impossible to develop a HIDiC with its operating curve exactly the same as the equilibrium one, because of the infinite amount of fixed investment, for instance, a infinite number of compressors and stages. We, therefore, have to create an approximate one, making it still possess the majority of the merits of the theoretical HIDiC that is based on the second law of thermodynamics.

As is sketched in Figure 4, a practical configuration for the HIDiC has been created. It possesses such a kind of process configuration that its stripping section and rectifying section are divided into two different columns, while connected through a great number of internal heat exchangers. To accomplish internal heat transfer from the rectifying section to the stripping section, the rectifying section is operated at a higher pressure and a higher temperature than those of the stripping section. For adjusting the pressures a compressor and a throttling valve have to be installed between the two sections. Owing to the heat integration, a certain amount of heat is transferred from the rectifying section and the vapor flow for the stripping section. Thus the condenser or reboiler is, in principle, not needed and zero external reflux operation could be realized. Moreover, as the overhead product is a relative high-pressure vapor flow, it can be seen as a potential hot utility, hence, the process energy efficiency could be considered further improved, if the heat content can be used, effectively.

In Figure 2, one can also examine the operation curve of the approximated practical HIDiC. It is now contracted and shifted away from the equilibrium one, but still takes a quite analogous shape with the latter, thus still assuring the practical HIDiC to have advantages in energy utilizations over conventional distillation columns.



Throttling valve

#### Figure 4. Schematic representation of a HIDiC

## Thermodynamic Analysis of the HIDIC

Takamatsu and Nakaiwa once analyzed the HIDiC with the concept of availability energy [14]. Here we will prove its superiority in energy efficiency by virtue of the thermodynamic energy efficiency mentioned in the preceding section. For the theoretical HIDiC,

$$\eta_{\text{HIDiC}} = W_{\text{min}} / (W_{\text{Loss}} + W_{\text{min}}) = W_{\text{min}} / (W + Q_{\text{F}}(1 - T_0/T_{\text{F}}) - Q_1(1 - T_0/T_1))$$
(10)

By proper process design, one can readily guarantee

$$Q_1 \ge Q_F, \text{ and } T_1 \ge T_F \tag{11a}$$

Yet, the following relationships are held for distillation processes.

$$W = Q_{\rm S} - Q_{\rm R} \approx Q_{\rm REB} - Q_{\rm COND} - Q_{\rm F}$$
(11b)

Thus,

$$\eta_{\text{HIDiC}} \ge W_{\text{min}} / W = W_{\text{min}} / (Q_{\text{REB}} - Q_{\text{COND}}) \ge \eta_{\text{con}}$$
(12)

As per the above thermodynamic analysis, we can see that the theoretical HIDiC is generally more energy-efficient than its conventional counterparts at its minimum reflux ratio operating condition. In practical applications, however, it must be borne in mind that Eq. 12 holds only under proper process design. Moreover, even when Eq. 12 is satisfied, the fact that electricity is generally several times more expensive than heating steams should also be considered in process design. It is, therefore, extremely necessary to check the economical feasibility of the HIDiC before the detailed process development.

## **DESIGN PRINCIPLES OF THE HIDIC**

It should be stressed here that there exist a number of similarities between the HIDiC and conventional distillation columns, though they look sharply different in process configurations. Takamatsu and Nakaiwa once systematically explored these similarities. For the HIDiC there exist similar operating conditions corresponding, respectively, to a maximum reflux ratio and a minimum reflux ratio modes of a conventional distillation column [15]. These two extreme operating conditions are both closely related to the pressure difference between the rectifying and the stripping sections,  $p_r - p_s$ . The minimum reflux ratio operation corresponds to the situation where internal heat integration between the rectifying and the stripping sections causes operating curve tangent everywhere to the equilibrium one, thus requiring an infinitive number of stages, namely, the situation of the theoretical model of the HIDiC. The maximum reflux ratio operation, the situation where internal heat integration is so intensive that operation curve has been made to coincident with the 45° diagonal line, thus requiring a minimum number of stages. One can thus understand that the pressure difference,  $p_r - p_s$ , appears to be a dominating variable for the design of the HIDiC. In terms of the above analysis, a modified McCabe-Thiele design algorithm has already been developed and found effective in process design and analysis [16].

## A WORKING EXAMPLE: SEPARATION OF A BENZENE AND TOLUENE BINARY MIXTURE

Throughout this work, separation of a binary equal-mole mixture of benzene and toluene will be frequently selected as an illustrative example for the design and analysis of the HIDiC. For simplification, an equal latent heat and a constant relative volatility have been assumed. The detailed operating conditions and product specifications are set out in Table 1.

Items	Values
Pressure of stripping section	0.1013 MPa
Feed flow rate	≤300 kmol/h
Feed composition	0.5
Feed thermal condition	0.5
Relative volatility	2.4
Latent heat of vaporization	7000 kcal/kmol
Heat transfer rate	7000 W/K
Overhead product composition	0.995
Bottom product composition	0.005
Operating hours per year (h)	8760
Electricity (\$ kwhr <sup>-1</sup> )	8.43×10 <sup>-2</sup>
Steams (\$ kmol <sup>-1</sup> )	3.03541×10 <sup>-1</sup>
Cooling water (\$ kmol <sup>-1</sup> )	1.06239×10 <sup>-3</sup>
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Table 1. Nominal steady-state operating conditions of the HIDiC

#### **CONCEPTUAL PROCESS DESIGN AND EVALUATIONS**

Conceptual design for a HIDiC and a conventional distillation column has been conducted for the separation example and Figure 5 demonstrates their comparisons [17]. As can be readily seen, the number of stages is drastically increased for the HIDiC over the conventional distillation column. Together with the expenses for the compressor, the total annual cost (TAC) for the HIDiC is more than that for the conventional distillation column by about 50%, demonstrating the very high fixed investment for the HIDiC. Here, TAC represents the fixed investment returned plus operating costs in a year basis.



Figure 5. Comparisons between a HIDiC and its conventional counterpart

Table 2.	Economical	comparisons	of optimal	steady-state	designs
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Items	HIDiC	Conventional DiC	Comparisons
Capital investment (\$US)	2.58205 x 10 <sup>6</sup>	944959	173%
Operating cost (\$US)	302965	713963	57.6%
Payback time (year)	(2.58205×10	<sup>6</sup> - 944959)/(713963- 30	02965) = 3.98

In terms of the above optimum process designs, the payback time for the extra investment can be easily estimated for the HIDiC. As can be seen from Table 2, the HIDiC provides a 57% reduction in operating cost, but at an expense of 1.75 times more fixed investment in comparisons with its conventional counterpart. The payback time is around 4 years and is about the same as that assumed for a conventional distillation column, alone, indicating an economically feasible time period. It should be stressed here that this outcome has been based on an overall heat transfer coefficient, 516.746kcal·m<sup>-2</sup>·K<sup>-1</sup>·h<sup>-1</sup>. As the overall heat transfer coefficient can be as large as, 826.79 kcal·m<sup>-2</sup>·K<sup>-1</sup>·h<sup>-1</sup>, according to our previous pilot-scale experimental measurements, we confidently expect that the payback time could be generally less than 3 years for the HIDiC. The expectation was actually answered by the bench-scale plant experimentation, which will be introduced later in this work. A payback time of 2.78 years was obtained.

## PROCESS SENSITIVITY AND FLEXIBILITY ANALYSIS

#### **Sensitivity Analysis**

As process systems are often subjected to a changeable operating condition due to the consideration of market needs and price variations, it is therefore necessary to ascertain, to a what degree of variations in operating condition, the HIDiC can still hold its higher energy efficiency than conventional distillation columns. Figure 6a illustrates comparisons of the HIDiC with a conventional distillation column, when both the end products have been kept on their specifications, respectively. Here, J<sub>1</sub> represents the operation profit per hour of the HIDiC and J<sub>3</sub>, that for the conventional distillation column. Here, the profit means product values minus operating costs. It is clearly demonstrated that the HIDiC is, only within a certain region, namely, F≤230 kmol/h, more economical than the conventional distillation. As for the influences of feed composition, the HIDiC appears to be always more energy efficient than its conventional counterparts, as is shown in Figure 6b, though the energy efficiency has experienced a considerable magnitude of variations.

As far as the influences of other operating variables have been concerned, they may give strong effects towards the energy efficiency of the HIDiC. Therefore, efforts should be spent to make the HIDiC as insensitive as possible to operating condition variations in process development [18].

#### Influences of Impurities

The binary mixtures to be separated by the HIDiC usually may contain a certain impurity or a third component, though it is in a small amount. The existence of an impurity or a third component, however, influences the energy efficiency of the HIDiC, if no correct measure has been taken in process design. The reason is that process heating and cooling sections have changed and they correspond no longer to the stripping and the rectifying sections, exactly. Instead, the rectifying section can contain a heat section and a cooling section, so can the stripping section. To deal with these situations, configurations of the HIDiC should be modified accordingly based on detailed thermodynamic analysis of the mixture to be separated. In Figure 7 two potential configurations are demonstrated. The left process configuration is for the cases where the impurity or the third component is close to the light component in relative volatility and the right one for the cases where the impurity or the third component.



Figure 6. Comparisons between HIDiCs and their Conventional Counterpart



Figure 7. Potential configurations for dealing with impurities

#### **Flexibility Consideration**

Although the HIDiC is self-content in heat utilization, adding a trim-condenser and trim-reboiler can, however, enhance process operation flexibility, substantially [19, 20]. Figure 8 shows a HIDiC with a trim-condenser and a trim-reboiler and its performances in energy utilizations are also illustrated in Figure 6, where J<sub>2</sub> stands for its operating profit per hour. It is readily to understand that the HIDiC with a trim-condenser and a trim-reboiler can stand larger variations in operating condition than before and still keep its higher energy efficiency than conventional distillation columns. This characteristic helps to increase the applicability of the HIDiC to various chemical and petrochemical process systems. However, it should be reminded here that a certain degree of loss in energy efficiency of the HIDiC has to be experienced. As has been demonstrated in both the figures, J<sub>2</sub>-J<sub>3</sub> is less than J<sub>1</sub>-J<sub>3</sub> in most of the preferred operating regions.



Figure 8. Trim-condenser and trim-reboiler enhance process flexibility

## PROCESS DYNAMICS AND OPERATION

## **Process Dynamics**

One concern from applications of internal heat integration is the complicated process dynamics that may be produced in the resultant processes. It is, therefore, imperative to investigate if it really happens. Figures 9 and 10 illustrate step responses of the HIDiC, when the pressure difference between the rectifying and the striping sections,  $p_r - p_s$ , and the feed thermal condition, q, have been disturbed in magnitudes by 1%, respectively, in both positive and negative directions. Here, the transient responses of the light component, benzene, are shown. It is readily to see that the influences of the feed thermal condition, q, to the HIDiC appear to be much stronger than those of the pressure difference,  $p_r - p_s$ .

An interesting phenomenon observed from the process step responses is the two different time constants associated with the pressure difference,  $p_r - p_s$ , and the feed thermal condition, q [21]. The one of the feed thermal condition, q, is much larger than the one of the pressure difference,  $p_r - p_s$ . The existence of two time constants is apparently due to the special configuration of the HIDiC. The higher the product specifications become, the more distinctive the difference between the two time constants will be. When the pressure difference,  $p_r - p_s$ , becomes zero, it will be expected that only one time constant exists within the HIDiC. It should be stressed here that designing the HIDiC with sharply different time constants will be, to a certain degree, beneficial to the process operation, because it can lead to a less degree of interaction in the dynamic state between the overhead and the bottom control loops. In other words, the HIDiC with a high-pressure difference would be more operation resilient than the one with a low-pressure difference. However, the former must afford more energy consumption than the latter. Therefore, a careful tradeoff must be exercised between process design economics and process operation.

According to the operation characteristics of the HIDiC, it is reasonable to call the feed thermal condition, q, a variable for material balance control and the pressure difference,  $p_r - p_s$ , a variable for energy balance control. As the HIDiC is very sensitive to changes in the feed thermal condition, q, it is imperative to tightly tune the control system with the feed thermal condition, q, as its manipulative variable. For example, a cascade control system is a better choice than a single-loop control system.



Figure 9. Transient responses of the HIDiC after ±1% perturbations in pressure difference. (a) overhead product, (b) bottom product



Figure 10. Transient responses of the HIDiC after ±1% perturbations in feed thermal condition. (a) overhead product, (b) bottom product

#### **Process Startup**

Due to the no-reboiler and no-condenser structure, it is impossible to startup the HIDiC by itself and it is therefore necessary to carry out the operation by means of an external trim-condenser and an external trim-reboiler. An effective procedure for process startup was already proposed recently [22]. A very important step towards smooth process startup is that inverse heat transfer from the stripping section to the rectifying sections must be avoided, otherwise, not only consumption of extra energy but also risks of potential operation problems might happen.

#### **Process Operation**

It is impossible to run the HIDiC in the same way as its conventional counterparts because of the no-reboiler and no-condenser structure. It is therefore necessary to synthesize different control systems for the HIDiC. Reconsider Eq. 9, one may find that the left side is the energy consumption of the compressor, which is closely related to the pressure difference between the rectifying and the stripping sections,  $p_r$  $-p_{s}$ . The right side is the unbalance of heat loads between the rectifying and the stripping sections. Feed thermal condition, q, is the dominating variable to change this unbalance. As, for any separations, Eq. 9 must be satisfied, it is not difficult to understand that the pressure difference between the rectifying and the stripping sections,  $p_r - p_s$ , and feed thermal condition, q, can be potential control variables for the process operation. A schematic diagram for a control configuration of the HIDiC is shown in Figure 11 and a typical response to a +5% step change in feed composition is illustrated in Figure 12. It is readily to see that both the overhead and bottom products can be maintained accurately to their desired steady state values. Extensive simulation studies have confirmed the feasibility of this control configuration as well as other alternatives [23,24,25,26].



Figure 11. A control strategy for the HIDiC



Figure 12. Responses of the HIDiC to a +5% feed composition upset

## **Process Design and Operation**

As discussed earlier, it is sometimes necessary to consider the interaction between process design economics and process operation in order to guarantee enough resilience for process operation. For the HIDiC, the symmetrical structure between the rectifying and the stripping sections intensifies the interaction between the overhead and bottom product control loops, significantly. It is sometimes beneficial to modify the process configuration and Figure 13 demonstrates two alternative process designs for the HIDiC. As can be seen, the symmetry between the rectifying and the stripping sections has been broken in these two processes. As a result, the performances of process operations could be improved substantially compared with the general process configuration as shown in Figure 4 [27,28], however, with the expense of a certain degree of loss in energy efficiency.



Figure 13. Other potential configurations of the HIDiC

#### **BENCH-SCALE EXPERIMENTAL EVALUATIONS OF THE HIDIC**

#### **Practical Realizations of the HIDiC**

Although the HIDiC appears to be very attractive in energy efficiency, it poses great difficulties in finding it a very effective configuration. Difficulties comes from not only

the arrangement of enough heat transfer area between the rectifying section and the stripping section, but also the possible degradation in mass transfer between vapor and liquid phases, due to the influences of internal heat integration. To seek an appropriate solution, we have already developed and evaluated experimentally several configurations for the HIDiC and obtained very deep insights into the associated problems [29, 30, 31]. As a typical example, a concentric configuration will be introduced here, which is very similar, in structure, to a single-tube and single-shell heat exchanger (see Figure 14). The tube side and shell side work as the rectifying and the stripping sections, respectively, and they are both furnished with packed distillation columns, in this situation. Special design has been undertaken so as to guarantee enough contact between vapor and liquid phases in both sections, making them actually very different from those conventional packed distillation columns. Further detailed discussions can be found, elsewhere [32, 33].

It should be mentioned here that the example shown here is, by no means, the best configuration for the HIDiC. It is just a suitable one for fitting the purposes of the bench-scale experimental evaluations, as will be discussed in the next subsection, for example, the very low processing capacity, F = 3.28 kmol/h.

## Layout of the Bench-Scale Plant

The simplified layout of the bench-scale HIDiC is shown in Figure 14. The plant is about 27 m in height and 0.254 m in diameter. Feed is introduced to the process at a constant flow rate and several temperature and pressure sensors are installed along the length of the HIDiC. A trim-condenser and a trim-reboiler have been affiliated to the process, due to the necessity of process startup and flexibility, as discussed earlier. The levels of reflux drum and trim-reboiler are maintained by the overhead and bottom product flows, respectively and the overhead and bottom products are mixed together and recycled back to the feed tank [34].



Figure 14. Layout for the bench-scale plant

## **Process Startup**

During process startup the inverse heat transfer from the stripping to the rectifying sections must be avoided, as has been discussed in the preceding section. To

enhance the pressure difference between the rectifying and the stripping sections as soon as possible, one needs to start the overhead trim-condenser at a proper time later than the bottom trim-reboiler. Based on a startup operation procedure devised, it was found that no special difficulties were encountered during startup operation. Generally speaking, around 10 hrs were needed for the process to reach its normal steady state although further reduction of this time period seems to be possible.

#### Steady State Operation with External Reflux

Steady state operations with external reflux were obtained directly after startup operation. More than 100 hrs of continuous operation have been performed and no special difficulties were encountered during the experimental tests. The obtained results suffix to manifest that the process can be operated very smoothly, just as its conventional counterparts. It was these valuable results that gave us confidence to farther perform external reflux-free operations, as will be introduced in the next subsection.

#### Steady-State Operation with no External Reflux

We achieved external reflux-free operation by reducing external reflux rate and, meanwhile, increasing the pressure difference,  $p_r - p_s$ , between the rectifying and the stripping sections, gradually. Figure 15 shows three typical operation results. As can be seen the operation style of the bench-scale plant could be easily shifted to the reflux-free mode from the startup period (Figure 15a). The internal heat integration between the rectifying and the stripping sections could function as an efficient means to generate internal liquid and vapor flows (Figure 15b). Figure 15c illustrates the time history of the overhead and bottom temperatures of the rectifying and the stripping sections, respectively, demonstrating stable operation of the bench-scale plant. Figure 16 shows the steady state heat and mass balances for the bench-scale plant at the same conditions as in Figure 15. It can be readily seen that the internal heat integration between the rectifying and the stripping sections plays a very important role in the process operation.



Figure 15. A typical reflux-free operation result of the bench-scale plant



Figure 16. Heat and mass balances for the bench-scale plant

## Energy Efficiency of the HIDiC

Table 3 compares the operating costs between the HIDiC and a conventional distillation column, which is designed for the same separation task. The comparisons clearly demonstrate the advantages of the HIDiC. As can be seen, the HIDiC without external reflux is about 27.2 % more energy efficient than the conventional distillation column [35]. This outcome is, however, much lower than that calculated in the conceptual process design. It is mainly due to the reasons that the bottom trim-reboiler is still in operation and the overhead product is not used to preheat the feed in this case. Furthermore, very different processing capacities also sharpen the gap between these two calculation results.

Items	Energy consumption (kW)	Comparison	
Conventional (R =1.5)	36.5	100.0 %	
HIDiC (R = 0.0)	26.5	72.8 %	

Table 3. Comparisons between the bench scale plant and a conventional distillation column

## FURTHER INTENSIFICATION OF THE HIDIC

As mentioned in the preceding section, the overhead product of the HIDiC is a highpressure vapor flow and it can be reused as a potential hot utility for the feed preheating, giving rise to a further intensified process configuration, as is shown in Figure 17.



Figure 17. Schematic representation of the intensified HIDiC

It has been proven that this process has a pole at the origin and hence becomes an open-loop integrating process [36]. It is apparently caused by the heat integration between the overhead product and feed flows. It is, therefore, impossible to design the HIDiC as an open-loop stable process and this is the drawback introduced by the heat integration between the overhead product and feed flows.

It is interesting to examine the effect of mass transfer delay from the overhead of the HIDiC to the feed preheater,  $\tau_d$ , to the intensified HIDiC (Figure 18). Even reinforced with a large value of  $\tau_d$ , it cannot change the process into an open-loop stable process. When  $\tau_d$  is small, the interaction between the two heat integration designs, namely, that between the rectifying and the stripping section and that between the overhead product and feed flows, becomes strong and consequently causes the process to have a large process gain. The extreme case comes when  $\tau_d$ =0, the dynamics for q becomes a pure integrator and possesses the largest process gain, therefore, benefiting process operation. On the contrary, when  $\tau_d$  becomes large, the interaction turns to be weak and causes the process to have a small process gain, therefore, worsening process operation.



Figure 18. Dynamic behavior of the intensified HIDiC

To examine the process operation feasibility, we investigated system performances with the same control configuration as before, namely, the control system shown in Figure 11. A typical response to a disturbance of +5% in feed composition is also shown in Figure 12. It elucidates that the intensified HIDiC can still be operated quite smoothly with the pressure difference between the rectifying and the stripping sections,  $p_{r}p_s$ , and feed thermal condition, q, although a certain degree of deteriorated performances have been observed.

#### EXTENSIONS OF INTERNAL HEAT INTEGRATION PRINCIPLE TO OTHER DISTILLATION-RELATED PROCESSES

#### A Heat-Integrated Batch Distillation Column

The principle of internal heat integration can also be applied quite analogously to batch distillation columns. Figure 19 shows a schematic of such an application, where internal heat integration between its rectifying section and reboiler has been considered. Takamatsu and his co-workers recently presented a systematic analysis and comparisons of this process with conventional batch distillation processes [37]. It was demonstrated that the energy efficiency could be improved sharply compared with its conventional counterparts. The reason could be attributed mainly to the lower reflux rate than the minimum one and, as a result, higher distillate rate of its product.



Figure 19. Schematic of a heat-integrated batch distillation column

# Facilitating Pressure-Swing Distillation Processes for the Separation of Binary Pressure-Sensitive Azeotropic Mixtures

Separation of binary pressure-sensitive azeotropic mixtures with pressure-swing distillation (PSD) process can be facilitated with internal heat integration. A PSD process is one of the simplest and yet most economical techniques for separating binary azeotropes, provided that the azeotropic composition is sensitive enough to the changes in operation pressure. It consists of two distillation columns. One is operated at a relative low pressure and temperature and the other is at a high pressure and temperature. By this pressure elevation the azeotropic point can be broken. Furthermore, it provides also convenience for considerations of internal heat integration between the high-pressure (HP) and low-pressure (LP) distillation columns.

Figure 20 demonstrates an application to the separation of a binary minimum azeotropic mixture: acetonitrile-water system [38]. Table 4 compares the internally heat-integrated PSD process with its conventional counterpart at three different scenarios. The three scenarios differ only in feed composition with their values being, 0.05, 0.2, 0.5, respectively. The comparison illustrates the economic advantages brought about by internal heat integration within the PSD process. Although the acetonitrile-water binary mixture is not an appropriate system for studying internal heat integration within PSD processes, it still results in a 4% reduction in fixed investment for the scenarios 2 and 3. With regard to scenario 1, extra investment should be expended, nevertheless, resulting in a 9% reduction in operating cost. From Table 4, it is reasonable to reach a conclusion that when the internal circulation rate is large (feed composition is large), the fixed investment could be reduced with internal heat integration, and when the internal circulation rate is small (feed composition is small), the operating cost could be reduced.

It is interesting to note that internal heat integration has reduced the fixed investment for the scenarios 2 and 3. It is, however, in great contrast to conventional belief that internal heat integration generally requires extra fixed investment. For the scenarios 2 and 3, the internal circulation rates are relative large, requiring, therefore, large heating and cooling duties for both distillation columns. Internal heat integration reduces these duties and hence the heat exchange area for all the condensers and reboilers. In contrast, the internal heat exchange area incorporated between the HP and the LP distillation columns is much smaller than those reductions at all reboilers and condensers, because of the large pressure difference, and thus high temperature driving force from the HP to the LP distillation columns. As heat exchangers usually take a great portion of fixed investment for distillation columns, it is not difficult to understand why the total fixed investment has decreased after internal heat integration.



Figure 20. Separation of binary azeotropes with a heat-integrated PSD

 Table 4. Reductions in heat loads and costs of internally heat-integrated against conventional PSD processes

Items	Values		
	Scenario I	Scenario II	Scenario III
Cooling duty load	21.5%	9.50%	9.10%
Heat duty load	9.8%	8.60%	8.10%
Fixed cost	-9.6%	4.70%	4.10%
Operating costs	7.8%	0.53%	0.41%

#### Internal Heat Integration between different Distillation Columns

Similar to the case of the PSD process discussed in the last subsection, internal heat integration principle can also be applied to two distillation columns that may have no connections at all. Provided feasible to consider internal heat integration both principally and economically between two different distillation columns concerned, it can generally lead to more benefits in energy utilization than those just based on condenser-reboiler heat integration structure, only. If one examines a distillation train system, either the direct or the indirect sequences, he may readily find that there exist many opportunities to consider this type of internal heat integration. It is considered to hold even higher potentials of applications than the HIDiC.

## PERSPECTIVES OF THE HIDIC

Separation of multi-component mixtures is usually much more energy-consuming than that of binary mixtures and higher potential of energy savings can thus be expected, if an effective multi-component HIDiC can be developed. Thermodynamic analysis of multi-component mixture separation indicates that a very complicated configuration is necessary if equilibrium operation has been required, making its industrial approximation an extremely challengeable problem. In our new project on internal heat integration approved this year by our government, great efforts will be paid on this problem. Commercialization of the HIDiC is now underway. It is anticipated that the first practical application will appear in the forthcoming several years. In order to extend the applications of the HIDiC to the separation of binary mixtures having a little bit large relative volatilities, schemes with partial internal heat integration between the rectifying section and the stripping section have also been under development.

#### CONCLUSIONS

A HIDiC has been developed through effectively applying heat pump principles to conventional distillation columns. In contrast to other heat pump assisted distillation columns, this process involves internal heat integration between the whole rectifying and the whole stripping sections and thus possesses high potential of energy savings. It has been proved from both the thermodynamics and bench-scale experimental evaluations that it really holds much higher energy efficiency than conventional distillation columns for those close-boiling binary mixture separations. Simulation studies and experimental operation results have also confirmed that the process can be operated very smoothly with no special difficulties found, yet.

A number of problems must be considered simultaneously during the HIDiC design, for example, flexibilities for operating condition changes, influences of an impurity or a third component, and operation performances, etc. They impose constraints on the energy efficiency that can be achieved. Therefore, trade-off between process design economics and process operation appears to be very important and has to be carried out very carefully.

Practical development of the HIDIC has gained great progress, in the meantime. Several configurations for the HIDiC have been developed and investigated up to now. It is anticipated that the first commercial application of the HIDiC will appear in the next few years.

By applying the same principle of internal heat integration, other energy-efficient processes, for example, heat-integrated PSDs and heat-integrated batch distillation columns, can be created. Internal heat integration can also be applied to two distillation columns that may have no direct connections. These represent another very important research and application areas for internal heat integration applications within distillation processes.

So far, development of the HIDiC has primarily been confined to binary mixture separations. As multi-component mixture separations represent major applications of distillation columns, development of corresponding HIDiC techniques is necessary and stands for an extremely challenging topic for our future work.

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## NOMENCLATURE

В	bottom product, kmol/s
$(C_1)$ $(C_2)$	composition controller
D	distillate product, kmol/s
F	feed flow rate, kmol/s
FC	flow control
Н	enthalpy, kJ
$J_1$	operation profit per hour of a HIDiC, \$US/h
$J_2$	operation profit per hour of a HIDiC with a trim-condenser and a trim-
	reboiler, \$/h
$J_3$	operation profit per hour of a conventional distillation column, \$US/h
L	liquid flow rate, kmol/s
LC	level control
n	number of total stages
p <sub>r</sub>	pressure of rectifying section, kPa
p <sub>s</sub>	pressure of stripping sections, kPa
q	thermal condition of feed
Q	heat load, kJ
R	reflux ratio
S	entropy, kJ/K
Т	temperature, K
t	time, h
V	vapor flow rate, kmol/s
W	shaft work, kJ
$W_{Loss}$	dissipation energy, kJ
W <sub>min</sub>	minimum energy, kJ
X	mole fraction of liquid
y	mole fraction of vapor
Z <sub>f</sub>	feed composition
$\tau_d$	mass flow delay from overhead to feed preheating, h
η	efficiency
Dic	distillation column
HIDiC	heat-integrated distillation column
HP	high pressure
LP	low pressure
PSD	pressure-swing distillation
TAC	total annual cost
Subscripts	
В	bottom
con	conventional
COND	condenser
D	distillate
F	feed
R	rectifying section
REB	reboiler
S	stripping section
0	environment
1	top stage of HIDiC

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