Innovation in distillation processes

—Process intensification for energy savings through concept of “detuning” from ideal state —

Masaru Nakaiwa * **and Takao Ohmori**

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A methodology of process intensification was discussed through the concept of “detuning” from the ideal state, especially for the energy-saving continuous distillation processes, which are typical energy consumer in the chemical and petrochemical industries. First, the reversible distillation was shown as the thermodynamically ideal state. Then, it was indicated that several energy efficient modifications of distillation processes can be obtained by “detuning” or simplifying the reversible system. Among these modifications, an internally heat-integrated distillation column (HIDiC) was one of the most promising options. The development of the HIDiC in the national projects was reviewed and the reduction of energy consumption by the HIDiC was estimated to be 60 % of the conventional column from the results of the projects.

Keywords **:** Process intensification, distillation process, energy saving, detuning

1 Introduction

“Process intensification (PI)” is discussed as a new paradigm in the field of chemical engineering where chemical processes and/or their components such as reaction apparatuses and separation devices are investigated. However, the definition of PI at present is not particularly clear. As Hirata mentioned[1], there is only a vague understanding of PI that is shared among the researchers. “Successful implementation of PI results in dramatic improvement in performance.” The goal is to achieve a performance improvement of at least tenfold. It is called a “quantum leap.” Performance improvements that can be achieved through advancement of existing technology by modifying processes and equipment can be limited to a few dozen percent at most. The realization of a “quantum leap” is necessary for a fundamental change, starting from the operating principle and the sizes and shapes of the equipment.

Looking back at the history of PI, it can be understood that the term itself is not new. According to Kuroda and Matsumoto [2], the term PI was used over 30 years ago in the United Kingdom within the context of safe design and size reduction of chemical plants and processes. However, the outset of increasing interest toward PI in the United States of America and Japan was marked by an article by Stankiewicz and Moulijn [3]. The authors’ perspectives on PI have since been changing slowly [4] [5], but a few keywords have remained constant. They are “miniaturization” and “simplification,” represented by efforts to reduce the equipment size and the number of operations in a given process; “increased efficiency,” represented by efforts to boost performance and energy saving; “integration” and “combination,” represented by efforts for simultaneous processing of multiple operations such as distillation and heat transfer, or reaction and separation, using a single unit of equipment; and “enhanced safety,” represented by efforts to minimize waste. Described as such, it is evident that PI has much in common with the concept of Minimal Manufacturing (MM)[6] , which is promoted by National Institute of Advanced Industrial Science and Technology (AIST). The MM concept is to manufacture only as many components and products as required, when required, using only the raw materials and equipment that are truly necessary. Perhaps MM is superior to PI, since PI only deals with chemical processes. However, PI has the same goal as MM. MM attempts to reconcile seemingly conflicting demands, such as energy saving with resource conservation, high efficiency with low cost, and high functionality with new features.

We have developed a heat-integrated distillation column (HIDiC) process where the heat transfer and the distillation operations are integrated, with the goal of energy saving during the distillation process. The process from the basic research to the practical application has not been smooth. Looking back, we can state that it was a R&D called *Full Research*. Also it was not intended from the beginning, but the research can be termed a successful example of PI. What follows is a description of the history, method of conceptualization, and approach to practical application of the HIDiC process in relation to PI. More specifically, we attempt to show that the designing and developing a new process through “detuning” from the thermodynamically ideal state is one method of achieving PI.

2 Approach to energy saving – thermodynamically ideal state and “detuning”

To discuss the innovation and energy saving in distillation

Research Institute for Innovation in Sustainable Chemistry, AIST Tsukuba Central 5, Higashi 1-1-1, Tsukuba 305-8565, Japan
* E-mail : nakaiwa-m@aist.go.jp

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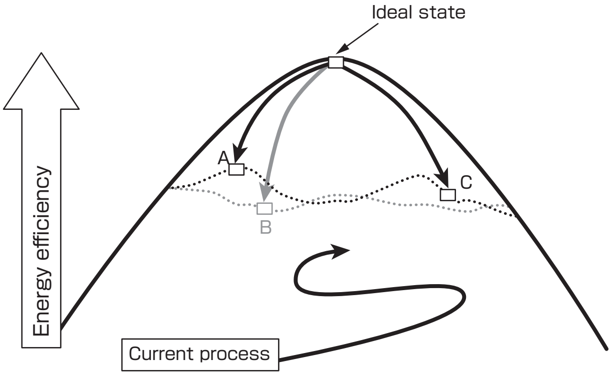
processes in relation to PI, we would first like to explain the concept of "energy saving." Descriptions like "Energy consumption was reduced by 20 % due to energy-saving efforts" are often encountered. Such statements are not so ambiguous and there is no problem as a daily expression. Thermodynamically, however, a closer inspection of them should be carried out, since according to the First Law of Thermodynamics, energy is something that is conserved, not something that can be consumed. If a person gets a 20 % improvement in rate of fuel consumption from purchasing a compact car, what is reduced is the amount of fuel like gasoline that would have been consumed by traveling a given distance, not the energy itself. In an automobile, the chemical energy of fuel is converted through heat into work that is used for moving with the engine (an Otto cycle heat engine for a gasoline vehicle). Except for electric and hybrid vehicles, the term *fuel-efficient engine* refers to one with high energy-conversion efficiency. The upper limit of the efficiency is theoretically determined with thermodynamics under the conditions such as temperature and so on. In other words, there exists a theoretical efficiency limit of converting fuel into moving for the gasoline engine. Improvement of engine efficiency is an important subject for enhancing the rate of fuel consumption in an automobile, but it is basically impossible to achieve the high efficiency that exceeds the limit determined with thermodynamics. As another example, a refrigerator with a compressor is a heat engine called a reverse Carnot cycle, and there is also a theoretical limit of its efficiency, which corresponds to the amount of heat that can be transferred and dissipated from the inside to outside using a given amount of electrical energy. Its limit is theoretically determined with the temperature inside the refrigerator and the outside (room) temperature. It cannot achieve energy saving exceeding the limit of reverse Carnot cycle.

Energy saving can be defined as the reduction of amount of energy required to accomplish a given task. There is a theoretical limit in achievable energy saving, which differs for each task. In other words, energy saving is to realize a given function as close to the theoretical limit as possible. In this paper, we use the term "detuning" to refer to the changing the process from the ideal state to one that is feasible. The term "detuning" is generally used when the engine technology developed for the F1-race vehicles is modified for use of commercial car. Modifications usually include reduced cost, increased durability, and improved usability, while the engine performance is decreased. In the field of energy saving, the term "targeting" is often used. It suggests targeting a high-efficiency process with an energy-saving goal, by starting from a current process. In this paper, we take the opposite direction of strategy for the technology development. First, we determine the ideal state, from which no further energy saving is possible (due to requiring unrealistic initial costs and/or equipment setup), and then sacrifice a little energy efficiency to achieve the feasible process. To emphasize this point, we use in this paper the uncommon term "detuning" by intention.

It should be noted that the path of "detuning" from an ideal state to a feasible condition is not one-dimensional. Fig. 1 shows a schematic diagram of the paths taken to enhance energy-saving performance in a certain process. Here, a conventional approach for energy saving with process improvement and refinement corresponds to slowly climbing up from the base of a mountain. On the other hand, the "detuning" process is like arriving first at the summit, then descending while considering energy efficiency as well as the other parameters like cost. In this approach, there are multiple paths that lead down the mountain, and therefore multiple possible destinations. Such an approach has two main characteristics. First, fundamentally unattainable goals are not derived, since the path descends from the theoretical upper limit. Second, when faced with various difficulties to a practical application, it is relatively easy to start again from the peak and seek another "detuning" path. One problem with "detuning" is the risk that a detuned process may become far from the current process. This means, however, that achieving PI that requires "rapid and discontinuous dynamic changes" would become possible.

3 Strategy for energy saving in distillation process through "detuning"

First, we examine the history of energy-saving efforts in the distillation process, while keeping previous discussions in mind. The distillation process is one of the oldest chemical processes: it was actually used to purify perfumes more than two thousand years ago. The principle of distillation is to separate solution components by evaporating them through heating and then condensing them through cooling. Solutions are separated with the difference in the boiling point (BP) of each solution components. The operation involved the heating of the source solution in a container for a given period of time to generate vapor, which was cooled, liquefied, and then collected. This process is known as simple distillation. Subsequently, distillation was used for many applications,



A, B, C: Processes obtained by "detuning" from ideal state

Fig. 1 " Detuning" from ideal state.

including alcohol and acid productions, and as an important technique during the Italian Renaissance for alchemy (not the occult kind, but utilizing the laws of nature). During the late 18th century to early 19th century, a discovery was made that heat is a form of energy, and the concept of evaporative latent heat was introduced, leading to an understanding that the thermal energy contained within a vapor can be used for heating liquids. In the late 19th century, the foundations of the continuous distillation process were established through various experimentations, stimulated by a need of refining crude oil for use as automobile fuel [7][8] . This development of the distillation process from simple to continuous can be regarded as a realization of PI, where “simplification” of the operation as well as “increased efficiency” with high performance and continuous mass processing were achieved. Evaporative latent heat, a major scientific discovery of that time, brought revolutionary changes to the distillation process.

Fig. 2 shows a schematic diagram of a typical continuous distillation column. The parts of the column above and below the feed position are called the rectifying section and the stripping section, respectively. In the rectifying section, the concentration of low boiling point component is higher than in the feed material, while in the stripping section the concentration of the high BP component is higher than in the feed. The interior of the column is designed such that the gas-phase mixture rising from the lower part and the liquid-phase mixture descending from the upper level come into contact. This is accomplished through the shelf-like constructions known as plates or trays with or without the various shapes of packings. Due to this mutual contact, the phase transition occurs via the transfer of evaporative latent heat between the gas and liquid-phase mixtures. In the vapor phase thus generated, there exists a higher concentration of low BP component while in the liquid phase a higher concentration of high BP component. Therefore, the low and high BP components can be concentrated toward the both ends of the column, respectively. Based on this principle, the separation of liquid mixture in a distillation column inevitably requires that the liquid solution should be converted into vapor with the supply of heat from a reboiler at the bottom of the column. Also, to allow the contact between liquid and vapor for mass transfer throughout the column, the latent heat of condensation must be removed from the vapor to reconvert it back to the liquid in the condenser at the top of the column.

In a continuous distillation column process, cooling at the top of column and heating at the bottom are carried out simultaneously to achieve gas-liquid contact throughout the column. According to thermodynamic analysis, the most efficient operation of distillation can be theoretically done when the continuous heating in the stripping section and the continuous cooling in the rectifying section are conducted along the bottom-to-top direction of column, as shown in Fig. 3[9][10]. This is called an operation of reversible distillation. Specifically, using a distillation column with infinite number of plates, infinitely small amount of heats are used for cooling in each plate of the rectifying section and for heating in each plate of the stripping section, respectively. This is the only thermodynamically ideal and the most efficient operation of continuous distillation, no matter what energy-saving strategies are applied to distillation columns, or whether new energy-saving technologies are developed. As the energy-saving technology becomes more advanced, the operation should approach the reversible distillation. In other words, the effort of energy saving in the distillation process is described as a “detuning” from the reversible distillation process with minimal performance deterioration while achieving economical viability. However, bringing the operation of forced heat transfer such as heating and cooling into mass transfer that is the essence of distillation, would cause significant alterations to the process, and therefore careful consideration of their effects is necessary in developing the desired process. The next step is to consider

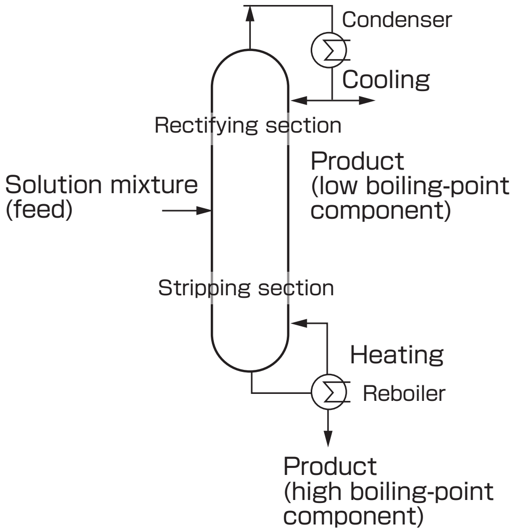


Fig. 2 Conventional continuous distillation column.

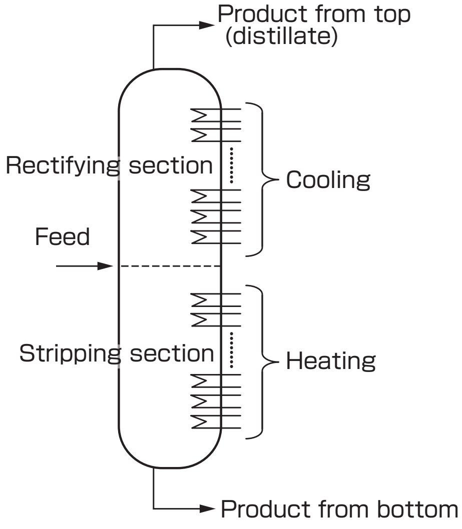


Fig. 3 Reversible distillation operation.

how to configure the equipment for "detuning" while keeping the effects in mind, and this is one process towards realization of PI.

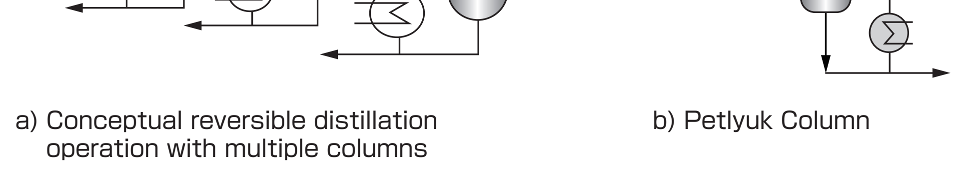
4 Equipment configurations for achieving "detuning"

Interpretation of reversible distillation, an ideal process, is important for performing "detuning." The reversible distillation column shown in Fig. 3 can be interpreted as the equipment involving the infinite number of heaters (reboilers) and coolers (condensers). If one column is divided into multiple (infinite) distillation columns of different heights, the configuration is represented in Fig. 4a). Moreover, by "detuning" to simplify the design to only 2 columns instead of an infinite number of columns, we have the configuration shown in Fig. 4b). Petlyuk et al. studied on this process in the 1960s, and thus this structure is generally called the Petlyuk column[11]. Further simplification along this consideration was carried out, and the resulting distillation process, developed by BASF of Germany, has been commercialized. In Japan, Kyowa Hakko Co., and Sumitomo Heavy Industries, Ltd. have developed this kind of commercial processes[12].

Another example of "detuning" the reversible distillation from a different viewpoint is discussed next. As a typical characteristic, the concentration of low BP component is higher and the temperature is lower in the upper section of a distillation column. Using the principle of reverse Carnot cycle mentioned above, it becomes possible to take heat from the low-temperature source (rectifying section), to elevate the temperature, and then to supply it to the stripping section where heat is required. The strategy for "detuning" in this line is shown in Fig. 5a)[13]. In this process, the "integration" of distillation column with heat pump is achieved since the condenser plays the role of heat pump. The figure illustrates that the heat taken from the upper section is supplied to the lower section by elevating the temperature with each condenser. This process is close to that of reversible distillation, excluding the supply of work for condensation. Then, further "detuning" gives rise to the process shown in Fig. 5b) where the number of condensers is reduced to only one and the heat exchange operations are carried out at a single point each in the rectifying and stripping sections. The process shown in this figure is generally known as a vapor recompression column (VRC). The VRC is advantageous when the temperature difference between the top and bottom of the column is relatively small, and the enrichment of low-concentration ethanol is an example of commercialized use. In the VRC, the heat exchanges are executed only at the top and bottom of the column, where the temperature difference is the greatest in the apparatus. This makes the "simplification" of equipment much more than for the reversible distillation process, but the divergence from the ideal state is considerable, and thus only limited energy saving is achieved.

5 HIDiC aiming toward more ideal condition

Petlyuk column and VRC process described in the preceding sections are two different ideas for energy-saving distillation, and correspond to the points B and C of Fig. 1. The basic structures of these apparatuses are completely different, but they were both derived from a common methodology of "detuning" from the ideal state. So, are there any other possibilities of "detuning"? The key point of "detuning" for the VRC was exchanging heat only at one location in the rectifying section, and another in the stripping section. The process would be able to approach closer to the ideal if the heat exchange were carried out in the multiple locations instead. However, this would call for the multiple condensers, which would make the strategy impractical. Therefore, to investigate a more realistic equipment configuration that allows for the heat exchange at the multiple locations, we once again turn our attention to the characteristics of a typical continuous distillation column (see Fig. 2). The internal temperature decreases with increasing height, due to the change in concentration of the solution in the continuous distillation column. The same characteristic is exhibited in a reversible distillation process: the cooling temperature


a) Conceptual reversible distillation operation with multiple columns

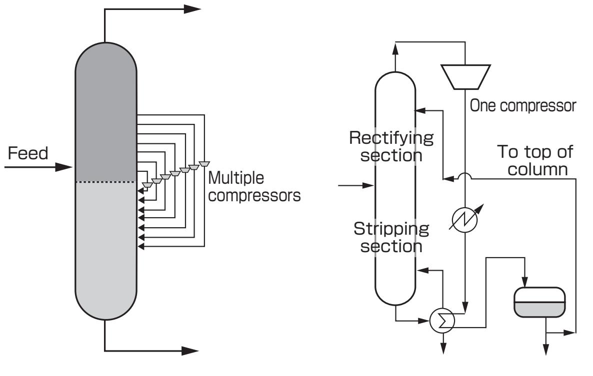
b) Petlyuk Column

Fig. 4 Approach to reversible distillation operation with multiple columns.

in the rectifying section is always lower than the heating temperature in the stripping section. Therefore, the heat taken from the rectifying section cannot be supplied directly to the stripping section. In Fig. 5, the condenser (heat pump) is used to elevate the temperature to solve this problem.

So, how can we achieve the heat transfer in the multiple locations simply? In Fig. 5a), all the cooling points in the rectifying section have a lower temperature than all the heating points in the stripping section. If the temperature of the rectifying section is higher than that of the stripping section, the heat from the cooling points can be supplied directly to the heating points. Would it then be possible to elevate the temperature of the rectifying section? In the separation via distillation, the equilibrium relation of liquid-gas is used. Increasing the pressure results in a higher equilibrium temperature, and we can achieve it by increasing the pressure in the rectifying section to above that in the stripping section. This requires an equipment configuration that the rectifying and the stripping sections are divided, and the pressure of rectifying section are increased until the heat from each cooling point in the rectifying section can be transferred to the corresponding heating point in the stripping section. Increasing the pressure in the rectifying section can be achieved by pressurizing the steam from the stripping section. Only one compressor is required to accomplish this task.

Once the temperature of the rectifying section exceeds that of the stripping section, various methods can be considered for the heat transfer. The simplest one is to arrange the direct contact between the rectifying and the stripping sections. The HIDiC aims to achieve the high efficiency by closely approximating the reversible distillation process, with the addition of compression [14] (see Fig. 6). We consider that this is also a type of “detuning,” and that it corresponds to point A in Fig. 1. The HIDiC also can be regarded as an “integration” process of distillation, heat transfer, and heat pump. A better energy-saving performance can be achieved in the HIDiC for a system of liquid mixture that is difficult to separate in a conventional distillation column because the temperature difference between the top and bottom of the column is small. In the propylene/propane mixture that the difference of BP is small, our evaluation shows that the HIDiC can separate the mixture using only 1/10 of the energy consumption in a conventional distillation column.



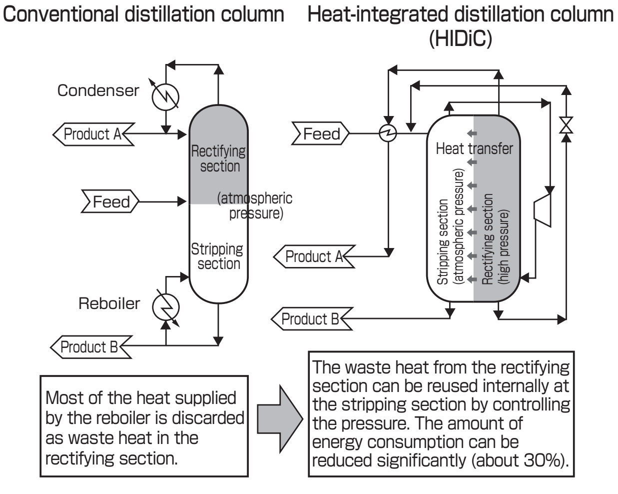
a) Conceptual process with multiple compressors

b) Vapor recompression process

Fig. 5 Approach to reversible distillation operation with multiple compressors.

Table 1 briefly summarizes the characteristics of the HIDiC, VRC, and Petlyuk column. The “Initial cost” in the table is the construction cost of a distillation column. The difference in total construction cost of distillation process is not as significant as that in construction cost of the column, because additional costs of plumbing and measuring instruments are required. “Applicability” corresponds to the number of industrially relevant solution systems to be processed. The HIDiC is particularly suitable for the solution systems where the temperature gradient in the bottom-to-top direction of column is relatively uniform. The key products for the petrochemical industry such as the mixture solutions of benzene/toluene/xylene and purification of crude cyclopentane are classified in this category. On the other hand, Petlyuk column is suitable for removing impurities of low concentration while VRC is for separating solutions with small BP difference. A conventional distillation column can be applicable in a wider range of solution systems, and the initial cost is generally favourable. The HIDiC realizes the concept of reversible distillation operation more faithfully, and has a wider range of application with achieving higher energy efficiency compared against Petlyuk column. It has also more advantage of energy efficiency since there is great improvement on the problem that the large temperature increase via compression is required in the VRC. In the HIDiC, the increase in energy efficiency of more than 20 % is expected over the Petlyuk column and the VRC, though it would depend on the separation specifications.

Professor Richard Mah of Northwestern University in the USA published the basic concept of the HIDiC in the 1970s for the air separation by cryogenic distillation [15].



Most of the heat supplied by the reboiler is discarded as waste heat in the rectifying section.

The waste heat from the rectifying section can be reused internally at the stripping section by controlling the pressure. The amount of energy consumption can be reduced significantly (about 30%).

Fig. 6 Internally heat-integrated distillation column (HIDiC).

He called this system Secondary Reflux and Vaporization (SRV) distillation (see Fig. 7). However, only its theoretical possibility was noted in the paper. It was shown that the energy saving can be achieved for the low-temperature systems that have a small difference in BP, such as air separation; but its potential for practical application was not considered in the paper. Professor Mah himself seemed to think that its practical potential was low. In fact, he submitted no patent application for this idea, and therefore what might be called a 'master patent' for the HIDiC does not exist. Professor Mah was not an expert in separation via distillation. Rather, he studied on an optimization of process flow using graph theory. After the first oil crisis of the mid-1970s, it seemed that he started investigating on energy saving of the distillation process, the most energy-consumption process in chemical plants. At the time, it was a common practice to wrap the distillation column with insulation materials to try to minimize any heat transfer from/to the outside except for the top and bottom of the column. (This is still a common practice today.) Thus, it was completely beyond the common sense to deliberately allow heat to enter and exit from the column body. According to Professor Kazuyuki Shimizu of Kyushu Institute of Technology, who worked on the SRV under Professor Mah at the beginning of the 1980s, the response to their investigation presented at the meeting of the American Institute of Chemical Engineers (AIChE) was very cold, and it was labelled as "a research just for publishing a paper" and almost ignored. Thereafter, several proposals on the HIDiC like concept were made in the form of research papers and patents in the U.S.A., but all of them suggested ideas or concepts only, and no realization research has been carried out.

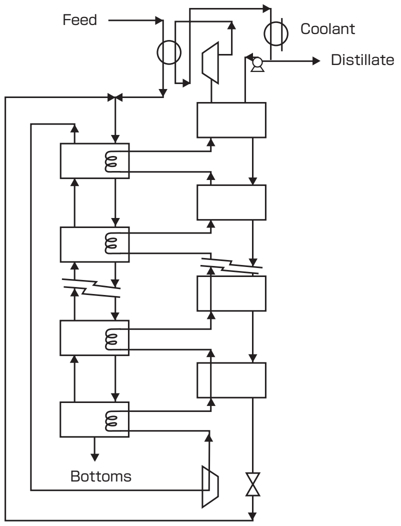
6 Road to Practical Applications of HIDiC

Several years after Professor Mah first published the paper mentioned above, the idea of HIDiC had attracted the attention of Professor Takeichiro Takamatsu of Kyoto University. He began to study on the fundamental characteristics of the process. Professor Takamatsu investigated on exergy analysis and thermodynamic analysis of chemical processes at the time, and he seemed to take notice of the high energy efficiency of HIDiC process. In the mid-1980s, the authors uncovered the characteristics of this process theoretically and experimentally at the National Chemical Laboratory for Industry through the collaborative research with Professor Takamatsu. First, we demonstrated theoretically that the column height and number of plates required as well as the heat transfer area, which allows us to prepare the overall column design, can be obtained by specifying the separation conditions and operation pressure. Also we showed that the operation pressure in the high-pressure side should be as low as possible for energy saving; but there is a thermodynamical limit of the value, which can be determined by providing the separation condition. Experimentally, it was proved using the small apparatus that the amount of heat required at the bottom of the column can be reduced via the internal heat-exchange. However, in this experimental apparatus, the size of compressor was larger than the column and its efficiency was low, we were unable to show the overall performance of energy savings considering the electrical power input.

Table 1 Comparison between HIDiC and other energy-saving distillation technologies.

	Energy efficiency	Applicability	Operation range	Initial cost
HIDiC	○	○	○	△
VRC	△	△	△	△
Petlyuk	△	×	△	○

The "Super Heat Pump Energy Accumulation System" project was in progress at the Agency of Industrial Science and Technology in those times, and we anticipated that the HIDiC technology would be positioned as an energy-saving industrial process where the heat pump in the wide sense was used. In the 1990s, a few groups of university researchers in U.K., France, and Hungary started investigating experimentally, but no prospect for realization was obtained. In Japan, on the other hand, the "Broad Area Energy Utilization Network System Technology (Eco-Energy City)" project was carried out from 1993 to 2000 in the New Energy and Industrial Technology Development Organization (NEDO). The research and development of HIDiC was conducted within the project by AIST and three companies (Kimura Chemical Plants Co., Ltd.; Maruzen Petrochemical Co., Ltd.; and Kansai Chemical Engineering Co., Ltd.), based on the findings from the collaborative research with Professor Takamatsu and AIST. In December of 1999, a



Mah R.S.H., et.al., AIChE Journal, Vol. 23(5), 651-658 (1977)

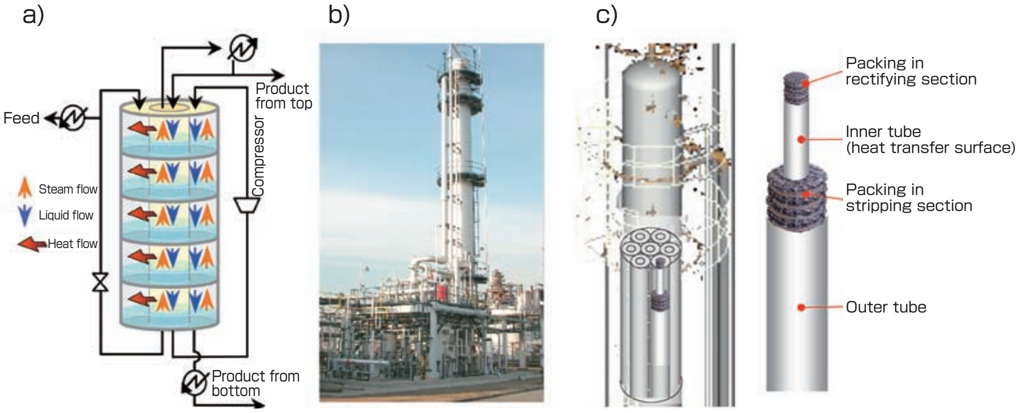
Fig. 7 Concept of Secondary Reflux and Vaporization distillation.

prototype column with diameter of about 300 mm and height of about 25 m was constructed in the Chiba Plant of Maruzen Petrochemical, and the continuous operation of over 100 hours was successfully achieved first in the world although it processed a small throughput of 300 kg/h with benzene/toluene system.

In an effort to further develop this achievement, the Ministry of the Economy, Trade and Industry (METI) and the NEDO launched on September of 2002 the “Development of Energy Saving Distillation Technology through Internal Heat Exchange” project in the New Global Warming Prevention Technology Program[16]. One of the authors acted as a leader of this project. The companies participated are Kimura Chemical Plant Co., Ltd., Maruzen Petrochemical Co., Ltd., and Kansai Chemical Engineering Co., Ltd., the target of which is for the petrochemical industries; and two companies that aimed for the air separation market, namely Nippon Sanso Corporation (currently Taiyo Nippon Sanso Corporation) and Kobe Steel Ltd.. The project focused on a double-tube design, in which the stripping section was the outer tube; and the rectifying section was the inner tube with structured packings, as shown in Fig. 8a). At the end of FY 2003, the construction of a pilot plant (see Fig. 8b)) for 12-component mixture system was decided at the Chiba Plant of Maruzen Petrochemical. As illustrated in Fig. 8c), this plant consisted of 7 double-tubes bundled together. Several considerations to realize PI were carried out in the design of plant, such as the inner tubes with 3 settings for diameters according to the gas-liquid load.

Construction of the pilot plant proceeded smoothly, and the first trial operation was conducted in FY 2005. Eventually, the continuous operation of 1000 h was achieved, and the operations under varied conditions and without external heat source were also carried out. In all the conditions, the stable operations demonstrated the energy reduction performance evaluated in the design stage. The amount of energy saving obtained during the experimental operations was 62 % in primary energy compared with the present distillation column. The reduction in required reboiler heat was 290 to 320 Mcal/h with an input electrical energy of about 30 Mcal/h into the condenser, and thus the ratio between the input electrical energy and the thermal energy gained (Coefficient of performance in heat pump) was approximately 10, which is quite high. After the completion of NEDO project, AIST and three companies in the petrochemical industries mentioned above with two new companies, namely Mitsubishi Chemical Corporation and Toyo Engineering Corporation, established a consortium to investigate on the commercialization of the process and prevailing the technology. The results were taken back to each company and Kimura Chemical Plant opened up the commercial activities on FY 2008. In foreign countries, the research and development of HIDiC has been started from the viewpoint of global environmental issues. Specifically, the research with the aim of commercializing the HIDiC process was begun on January of 2002 in the Netherlands, and currently it is in the second phase of research and development. Many prominent companies in Europe are participating in this project, with Delft University of Technology and Energy Research Centre of the Netherlands.

Why was Japan able to launch commercialization first? The answer to this question is not clear, but we believe it is due to the difference in strategy of realizing the characteristics of HIDiC where the distillation and heat transfer are integrated. The goals of researches in the U.S.A. in the 1980s and Europe in the 1990s were mainly to increase the amount of heat transferred from the rectifying section to the stripping section. Therefore, the consideration of material transfer and distillation performance was presumably insufficient. For example, multiple heat pipes were proposed to use for the heat transfer between the rectifying and stripping sections in a patent submitted in the U.S.A. in 1980[17] . This would achieve a sufficient heat transfer, but the realization of equipment would not be possible due to difficulty in



Basic flows of HIDiC (inner tube corresponds to the upper part of conventional column, and the outer annulus the lower part)

View of the pilot plant (in the Chiba Plant of Maruzen Petrochemical Co., Ltd.)

Internal structure of pilot plant (7 double-tube unit)

Fig. 8 Internal structure of HIDiC and pilot plant.

manufacturing such a design of equipment as well as the obstruction of liquid flow within the column caused by the heat pipes. On the other hand, we studied via simulation technology to prevent decreased distillation performance, and adopted a simple double-tube structure that led more straightforwardly to practical applications. If the amount of heat transfer was not enough, we made the heat transfer area larger by extending the height of column. While the HIDiC process is a realization of PI by "integration" of the distillation and heat transfer, we believe our success was due to the strategy of maintaining distillation performance even in pursuing energy saving through the "integration" of heat transfer into the distillation.

7 Conclusion

In this paper, an approach for the development of energy-saving technology for the present process was shown with the concept of "detuning" from the ideal state. We also discussed the development of energy-saving distillation processes, including heat-integrated distillation column (HIDiC). In the case described here, the procedures were: a) to determine the thermodynamically ideal state for the process to be examined; b) to perform "detuning" from the ideal state down to more attainable conditions; c) to evaluate the energy-saving performance, cost, equipment configuration, and other considerations for realistic feasibility after "detuning"; and d) if the evaluation result is not satisfactory, return to b) and explore other "detuning" paths. The course of b) → c) → d) forms a loop, which we consider to be one of the paths to the realization of PI. Actually, we believe that the development of HIDiC followed this path to reach the practical application stage. In summary, we described our thoughts and approaches to practical application of the HIDiC process, which is an energy-saving distillation process, from the perspectives of PI and "detuning," We hope this paper will help the progress of *Synthesiology*.

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Author

Masaru Nakaiwa
Graduated from the Department of Chemical Engineering, Kyoto University in 1980. Joined the National Chemical Laboratory for Industry, Agency for Industrial Science and Technology in 1980. Participated in overseas research at the Kansas State University, U.S.A. in 1990 -1991. Leader of Energy-Efficient Chemical Systems Group, Research Institute for Green Technology, AIST from 2001. Currently, the Director of the Research Institute for Innovation in Sustainable Chemistry. Engaged in the researches mainly on energy saving in chemical processes and application of nonlinear chemical systems. For this paper, took the charge of the part related to the HIDiC process with the "detuning."

Takao Ohmori
Graduated from the Department of Chemical Engineering, Graduate School of Engineering, Kyoto University in 1984. Joined the National Chemical Laboratory for Industry, Agency for Industrial Science and Technology in 1985. Participated in overseas research at the West Virginia University, U.S.A. in 1991 -1992. Leader of Energy-Efficient Chemical Systems Group, Research Institute for Innovation in Sustainable Chemistry, AIST from 2006. Currently, the Principal Research Scientist of the Research Institute for Innovation in Sustainable Chemistry. Engaged in the researches mainly on nonlinear phenomena, complex system, and process intensification. For this paper, took the charge of the part related to the process intensification.

Discussion with Reviewers

1 Comparison with current processes

Question and comment (Koichi Mizuno)

I understand that HIDiC is superior to VRC and Petlyuk in terms of energy saving and operability. However, there are some problems in terms of manufacturing cost, performance, and so forth, as referred in the paper. For example, the cost of HIDiC manufacturing might be higher due to its complex structure. What do you think of this point? Also, what are the differences in distillation performance when this process is compared with the other processes?

Answer (Masaru Nakaiwa)

In general, a distillation column is entirely custom-made since the specifications of the apparatus are different according to the property of the solution to be separated, the level of product purity required, and the processing volume. Thus, the manufacturing cost depends on the specifications. For the pilot plant of HIDiC (at the Chiba plant of Maruzen Petrochemical Co., LTD.; fractional distillation of gasoline with 12 components at the commercial scale feed of 12,000 ton/year), the cost was 230 million yen. The amount of reduction in annual running cost via the energy saving of HIDiC is about 20 million yen when calculated at the crude oil price of 60 US dollar/barrel. While the manufacturing cost of an ordinary column at the same scale is uncertain, the response of Kimura Chemical Plants Co., Ltd. to user inquiries is: “It is about twice as much as a conventional type.” Applying this to the pilot plant, when the reduced annual running cost is subtracted from the cost difference between HIDiC and an ordinary column, the simple payback time is 5.75 years. I think the actual cost would be slightly different since the pilot plant has somewhat special specifications such as the use of temperature measurement system for investigations. The detailed descriptions are summarized in the *Kagaku Kogaku Ronbunshu*, 34(4), 444 (2008). For the distillation performance of HIDiC without the heat transfer, it is about the same as the ordinary process.

2 Energy consumption in ideal state and “detuning” process

Question and comment (Koichi Mizuno)

Energy saving was achieved considerably in this study. Is it possible to quantitatively describe the difference in energy consumption between the ideal state and HIDiC?

Answer (Masaru Nakaiwa)

We have not done such a calculation since the amount of energy consumption depends on the several conditions. HIDiC is a “detuning” process from the reversible distillation operation that is ideal state, but an additional factor of the heat pump effect is involved. The total energy consumption in HIDiC is significantly influenced by the energy efficiency in the compression process. Here, the thermodynamically ideal state of this process is a reverse Carnot cycle, as described in the paper. The energy efficiency of a compression heat pump that is currently used in industries is at most 50 % of the reverse Carnot cycle. Therefore, there is a possibility of reducing the energy use by half at least. If we discuss the minimum energy consumption for the separation of mixture thermodynamically apart from HIDiC, it is possible to achieve a reduction of energy consumption of at least tenfold. Exergy analysis is useful to estimate the value of such a reduction quantitatively.

3 Detuning from ideal state

Question and comment (Hiroshi Tateishi)

I think Fig. 1 is easy to understand as a concept, yet the meanings of A, B, and C, respectively, are unclear. As I understand that an approximate operation is carried out under some conditions with “detuning” from the ideal state, can you include any explanations from such a viewpoint in the figure?

Answer (Masaru Nakaiwa)

We explain a general concept of “detuning” in Fig. 1. Petlyuk column and VRC process do not quantitatively correspond to B and C of Fig. 1. To clarify that Fig.1 illustrates a general concept, we added the explanation to the figure.

4 Explaining key points of research on HIDiC

Question and comment (Hiroshi Tateishi)

I think the overall structure of paper where the R&D of HIDiC is overviewed from a higher stage of the innovation in distillation process is good. However, I think it is necessary to emphasize the significance of research on HIDiC at AIST using some specific examples. Can you give us more detailed explanation for how the theoretical analysis, numerical simulation and technological developments on HiDiC have been integrated at AIST?

Answer (Masaru Nakaiwa)

The description in chapter 6 was revised from the viewpoint of the contributions of AIST.