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Heat-Integrated Distillation Columns: Vapor Recompression or Internal Heat Integration?

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Internally, heat-integrated distillation columns (HIDiC) and vapor recompression (VRC) constitute alternative design options to provide sustainable distillation processes. However, the design is often based on heuristic rules or the experience of the designer, as no systematic methodology driven by economics has been established so far. The increased complexity of heat-integrated columns can hardly be dealt with using simulation studies but rather calls for a systematic design procedure. A new design methodology is presented here; it builds on a superstructure, mixed-integer minimization of total annualized cost of operation and rigorous thermodynamic modeling. Optimal VRC and HIDiC designs are identified for the separation of binary, multicomponent, and nonideal mixtures and benchmarked against conventional distillation column designs. A small number of intermediate heat exchangers is optimal for these HIDiC configurations, eventually reducing to a single heat exchanger similar to VRC. Therefore, VRC designs are often more cost efficient due to simpler equipment.
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Introduction

Worries about climate change and depleting resources have drawn attention to more sustainable chemical processes. The energy consumption of a typical (petro-)chemical plant is dominated by the separation section that is often implemented by means of distillation. As distillation columns may simultaneously act as a heat sink (at high-temperatures) and a heat source (at low-temperatures), they are prone for heat integration measures. Since the 1950s, different techniques for the reduction of the energy demand of distillation column sequences have been suggested. It is well-known that significant energy savings can be achieved if complex column configurations[1] and heat integration[2] are used.

Academic research has been focusing on alternative concepts for heat integration in a single distillation column that include vapor recompression (VRC) and internally heat-integrated distillation columns (HIDiC)[3,4] (cf. Figure 1). In these design options, a compressor is used to operate the condenser or an entire part of the column at an elevated pressure. In case of VRC, the condenser is heat integrated with the reboiler at the bottom or at any intermediate tray of the column, where the pressure is chosen to ensure that the boiling temperature of the distillate is above the dew point temperature at the reboiler. In case of HIDiC, an entire section (at the top) is operated at elevated pressure, which allows for direct heat transfer from the rectifying to the stripping section over the entire height of the column section. Such HIDiC designs ideally approach reversible distillation, where every stage is provided with the heating or cooling necessary to allow separation at vanishing driving force.[5] Although heat-integrated designs have been in the focus of academic research after the oil crises in the 1970s, similar ideas have been proposed and are extensively used in industrial air separation for a very long time. A comprehensive review on the history of the development of HIDiC designs has been provided most recently by Shenvi et al.[6] Although the technical feasibility of HIDiC designs has been demonstrated in pilot plant scale,[7] no industrial application exists on plant scale according to the authors' knowledge.

The major economic drawback of VRC and HIDiC designs is the compressor necessary to increase the pressure for the utilization of the heat of condensation. The operation of the compressor not only relies on relatively expensive electrical energy but also comes with significant investment cost. Additionally, a HIDiC design requires the implementation of heat exchange along the height of the column, which is still a challenging problem in terms of equipment design. Recently, several authors have questioned whether heat exchange along the entire height of the column really constitutes a favorable option.[6,8–10] Shenvi et al.,[6] Suphanit[10] as well as the authors of this article[9] have independently proposed to reduce the heat transfer locations to a small number and report negligible impact on the economic performance.

As the number of design degrees of freedoms is significantly increased in heat-integrated column designs compared with conventional ones, the design task can hardly be successfully achieved by simulation studies only. The decisions to be taken during the design of a conventional column include the reboiler duty, the distillate to feed ratio, the feed

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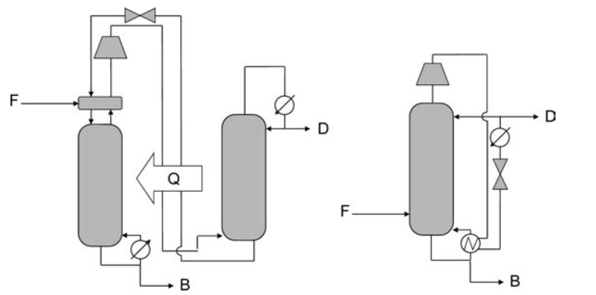

Figure 1. Basic configuration for HIDiC (left) and VRC columns (right).

position, and the number of trays. In case of heat-integrated columns, the number, size, and position of intermediate heat exchangers and the operating pressure of the rectifying section have to be determined in addition. Several authors proposed design methodologies for HIDiC columns in the past. Early design studies were based on heuristic rules shortcut methods[11] and process simulation.[12] For example, Gadalla et al.[13] presented a design approach that combines pinch analysis with a hierarchical design procedure. Hydraulic feasibility and energy consumption were analyzed in a commercial process simulator.[14,15] Ho et al.[16] extended the graphical Ponchon-Savarit shortcut method to tackle heat integration for binary separations. Cabrera-Ruiz et al.[17] used column grand composite curves to determine a suitable column configuration. Shenvi et al.[6] suggested to determine the position of intermediate heat exchangers based on an analysis of the temperature profiles of the column sections. Huang et al.[8] presented a simplified HIDiC design method that relies on heuristic rules and process simulation studies. Most of the design methods presented in the literature require graphical visualization and rely on a commercial process simulator to evaluate the design. They do not consider capital cost for equipment, although any decision on an economically optimal distribution of heat exchangers can only be addressed if capital cost is addressed during the design. Rigorous superstructure optimization can overcome the limitation of existing HIDiC and VRC design methods, if a total annualized cost (TAC) estimate, comprising a weighted sum of operational and capital cost, is chosen as the objective function.[18,19] The optimal design can then be computed by means of mixed-integer nonlinear programming (MINLP), simultaneously optimizing the design and the operating point of the column.

In this contribution, we present a new methodology to design HIDiC and VRC that is based on rigorous optimization. It will be applied to binary, multicomponent, and nonideal mixture separations to illustrate its potential. In case of HIDiC, optimal heat exchanger configurations are determined, which minimize either TAC or energy consumption. Based on the results of these case studies, we confirm that cost-optimal HIDiC designs require only a small number of heat exchange locations. Consequently, conventional shell and tube heat exchangers can be used for implementation, such that there is no need for the development of specialized equipment to realize heat exchange over the entire height of the distillation column. Often, the designs rely on just a single intermediate heat exchanger and are hence very similar to those of VRC. The rigorous design methodology introduced in this contribution is universally applicable and should generally be used for optimal HIDiC and VRC design.

Optimization Model

Distillation design in industrial practice is usually based on simulation studies, which require a detailed specification of the distillation column at an early stage of the design process. Although the design can be effectively supported by heuristic rules, it still requires high engineering effort and bares a high chance to miss the most promising result. Such designs of HIDiC may lead to unnecessarily high cost or energy demand. The full potential of a distillation process is often not exploited.

Deterministic, mathematical optimization using rigorous distillation column models to solve the synthesis problem provides an attractive alternative.[18,19] Rigorous modeling allows to consider a high level of detail with respect to thermodynamic behavior, flow conditions, and equipment design, thus enabling the designer to identify those design parameters which control process performance, which include reboil and reflux ratio, heat exchange area, the number of trays, the feed location, or the column diameter. To this end, the column is modeled by means of an equilibrium tray model. Figure 2 represents a tray of the rectifying section with liquid (L) and vapor (V) streams, and, if applicable, the reflux (R) stream from the condenser and the vapor (V_{BP}) stream from the compressor. A superstructure formulation, in which streams, for example, the reflux or the boil up, can bypass trays, allows for an optimization of the column configuration.

The superstructure model presented in this article is an extension of the one presented by Kraemer et al.[19] for conventional distillation. In case of VRC, the conventional column model has to be extended by a model for the compressor, which is also required to model a HIDiC configuration as presented below. Such an extension of the optimization model results in additional degrees of freedom in case of HIDiC and VRC configurations. For HIDiC, these degrees of freedom include the optimal operating pressure for the rectifying section, and the positions and sizes of the intermediate heat exchangers. In this model, the operating pressure of the stripping section is fixed to deliberately reduce the degrees of freedom by allowing only one variable pressure. The compressor is always located above the feed tray. The superstructure for the design of a HIDiC configuration is displayed in Figure 3.

The superstructure allows heat exchangers to transport heat from the rectifying section, operated at the optimal pressure, to the stripping section at every tray. The combinatorial complexity is pragmatically reduced by permitting heat exchange only between trays of the rectifying and stripping section at the same vertical position. This restriction can be justified by equipment design consideration. To determine

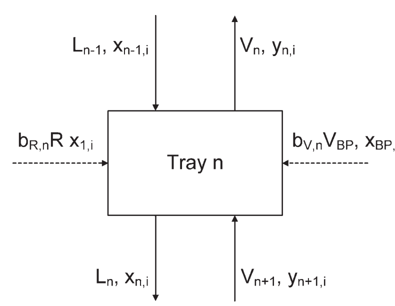

Figure 2. Equilibrium tray with streams in the rectifying section.

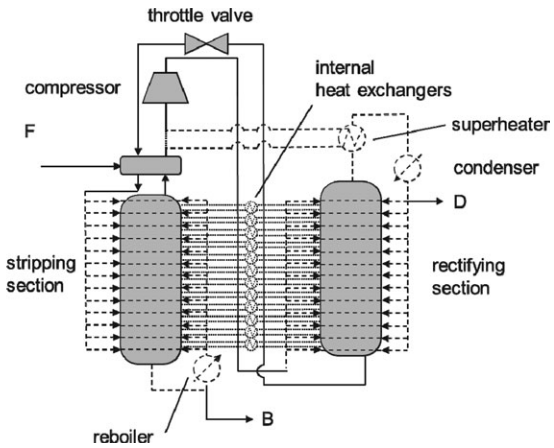

Figure 3. HIDiC superstructure to determine the optimal number of trays and the positions and areas of the internal heat exchangers for both column sections.

optimal heat exchanger connections and tray numbers, column trays can not only be reduced at both ends of the column as it has been shown to be sufficient for simple column design but also at both section ends: the vapor stream from the compressor and the throttled liquid from the valve are decided to bypass trays in the respective section. Thus, column designs are also possible where heat is exchanged at the ends of the column section only (cf. Figure 8). This way, the optimal position of the column feed relative to the column ends is identified by optimizing the number of trays in each column section.

The detailed model of the HIDiC configuration is presented in the following section. The component mole balance for a general tray in the rectifying section including possible reflux and vapor streams from the compressor is given by

$$0 = L_{n-1}x_{n-1,i} + V_{n+1}y_{n+1,i} - L_n x_{n,i} - V_n y_{n,i} + b_{R,n}Rx_{1,i} + b_{V,n}V_{\mathrm{BP}}y_{\mathrm{BP},i}, \quad n \in [2,\mathrm{N_F}[\quad (1)$$

and the corresponding component mole balance for a general tray in the stripping section including possible boil up and liquid streams from the throttle valve is given by

$$0 = L_{n-1}x_{n-1,i} + V_{n+1}y_{n+1,i} - L_n x_{n,i} - V_n y_{n,i} + b_{B,n}By_{N_{\max},i} + b_{L,n}L_{\mathrm{BP}}x_{\mathrm{BP},i}, \quad n \in]\mathrm{N_F}, N_{\max}[\quad (2)$$

The energy balances for every tray in the rectifying and stripping sections are

$$0 = L_{n-1}h_{l,n-1} + V_{n+1}h_{v,n+1} - L_n h_{l,n} - V_n h_{v,n} + b_{R,n}Rh_{l,1} + b_{v,n}V_{\mathrm{BP}}h_{v,\mathrm{BP}} - Q_{D,\mathrm{int},n}, \quad n \in [2,\mathrm{N_F}[\quad (3)$$

$$0 = L_{n-1}h_{l,n-1} + V_{n+1}h_{v,n+1} - L_n h_{l,n} - V_n h_{v,n} + b_{B,n}Bh_{v,N_{\max}} + b_{L,n}L_{\mathrm{BP}}h_{l,\mathrm{BP}} + Q_{B,\mathrm{int},n}, \quad n \in]\mathrm{N_F}, N_{\max}[\quad (4)$$

respectively. Special trays, such as the condenser, the feed tray, and the reboiler, require more specific material and energy balances. The material and energy balances are

$$0 = V_{n+1}y_{n+1,i} - Dx_{n,i} - Rx_{n,i}, \quad n = 1, \quad i \in I \quad (5)$$

$$0 = V_{n+1}h_{v,n+1} - Dh_{l,n} - Rh_{l,n} + Q_D - Q_{\mathrm{pre}}, \quad n = 1 \quad (6)$$

for the condenser

$$0 = Fz_{F,i} + L_{n-1}x_{n-1,i} + V_{n+1}y_{n+1,i} - L_{\mathrm{BP}}x_{\mathrm{BP},i} - V_{\mathrm{BP}}y_{\mathrm{BP},i}, \quad n = N_F \quad (7)$$

$$0 = Fh_F + L_{n-1}h_{n-1} + V_{n+1}h_{l,n+1} - L_{\mathrm{BP}}h_{l,\mathrm{BP}} - V_{\mathrm{BP}}h_{v,\mathrm{BP}} + W_{\mathrm{comp}} + Q_{\mathrm{pre}}, \quad n = N_F \quad (8)$$

for the feed tray, and

$$0 = L_{n-1}x_{n-1,i} - L_n x_{n,i} - V_n y_{n,i} - By_{n,i}, \quad n = N_{\max} \quad (9)$$

$$0 = L_{n-1}h_{l,n-1} - L_n h_{l,n} - V_n h_{v,n} - Bh_{v,n} + Q_B, \quad n = N_{\max} \quad (10)$$

for the reboiler. The energy balance of the condenser (8) also accounts for the energy to superheat the vapor entering the compressor. The thermodynamic behavior is modeled by the closure conditions

$$\sum_i x_{n.i} = 1, \quad \sum_i y_{n,i} = 1, \quad n \in N \quad (11)$$

and the vapor-liquid equilibrium condition for every tray

$$y_{n,i} = K_n(\mathbf{x}_n, \mathbf{y}_n, T_n, p_n)x_{n,i}, n \in N \quad (12)$$

Summation relations for the integer decision variables

$$\sum_n b_{V,n} = 1, \quad \sum_n b_{L,n} = 1, \quad \sum_n b_{R,n} = 1, \quad \sum_n b_{B,n} = 1, \quad V_{N_F} = 0, \quad L_{N_F} = 0 \quad (13)$$

ensure that the reflux and reboil streams as well as the compressed vapor stream and the throttled liquid stream all enter at only one stage, respectively.

Compression is simply modeled be means of the isentropic pressure–temperature correlation

$$T_{\mathrm{out}} = T_{\mathrm{in}}\left(\frac{p_{\mathrm{out}}}{p_{\mathrm{in}}}\right)^{\frac{\kappa-1}{\kappa}} \quad (14)$$

The energy demand of the compressor is determined by the temperature increase. The actual outlet temperature is determined from

$$T_{\mathrm{out}} = T_{\mathrm{in}}\left(1 + \frac{1}{\eta_{\mathrm{comp}}}\left(\left(\frac{p_{\mathrm{out}}}{p_{\mathrm{in}}}\right)^{\frac{\kappa-1}{\kappa}} - 1\right)\right) \quad (15)$$

This equation follows from rearranging Eq. 14 and taking the isentropic efficiency η_{comp} into account. In case of a low isentropic exponent, the vapor feed to the compressor is superheated to prevent partial condensation during compression, which can result in compressor damage. The energy duty of the compressor is determined by the enthalpy change in the compressor, determined from

$$W_{\mathrm{comp}} = V_{\mathrm{BP}}(h_v(T_{\mathrm{out}}) - h_v(T_{\mathrm{in}})) \quad (16)$$

The heat exchange between adjacent trays in the rectifying and the stripping section has to satisfy

$$Q_{D,\text{int},n} = Q_{b,\text{int}n+NF}, \quad n \in [2,\text{N}_\text{F}[\tag{17}$$

while the inequality constraints

$$T_n \geq T_{n+N_F} \quad n \in [2,\text{N}_\text{F}[\tag{18}$$

ensure that heat is only exchanged in the direction of decreasing temperature. The number of trays in the rectifying section is computed as

$$\text{NT}_\text{rect} = \frac{N_\text{max}}{2} - \sum_n \sum_n^{N_\text{max}} b_{R,n} - \sum_{n=1}^{N_F} \sum_{n=1}^{n} b_{V,n} \tag{19}$$

while the number of trays in the stripping section is

$$\text{NT}_\text{strip} = \frac{N_\text{max}}{2} - \sum_n \sum_{n=1}^{n} b_{B,n} - \sum_{N_F}^{N_\text{max}} \sum_n^{N_\text{max}} b_{L,n} \tag{20}$$

Because of the different pressures in the column sections, the shells of a HIDiC configuration are of different diameters. The diameter in the rectifying section is

$$D_\text{col,rect} = \sqrt{\frac{4V_{n-1}}{2\pi} \sqrt{\frac{R_\text{gas} T_{n-1} \sum_i y_{n-1,i} M_i}{p}}}, \quad n = N_F \tag{21}$$

and

$$D_\text{col,strip} = \sqrt{\frac{4V_{n+1}}{2\pi} \sqrt{\frac{R_\text{gas} T_n \sum_i y_{n+1,i} M_i}{p_n}}}, \quad n = N_F \tag{22}$$

in the stripping section. The annual operating costs are determined from the consumption of steam, cooling water, and electricity, that is

$$C_\text{op} = f(Q_B, Q_D, W_\text{comp}) \tag{23}$$

and the capital cost of the column, the heat exchangers, and the compressor are determined by nonlinear cost correlations provided by Guthrie,[20]

$$C_\text{cap} = f(\text{NT}, D_\text{col,rect}, D_\text{col,strip}, A_\text{reb}, A_\text{con}, A_{\text{int},i}, A_\text{pre}, W_\text{comp}) \tag{24}$$

which reflect the economy of scale of the equipment. The TAC is determined as weighted sum of the annual operating cost and annualized capital cost

$$\text{TAC} = C_\text{op} t_a + f_c C_\text{cap} \tag{25}$$

As some of the design variables, like the number of trays, are discrete variables, while others, like heat exchanger areas, flow rates and compositions, are continuous variables, a MINLP problem results. Considering the large scale, the complexity of the HIDiC superstructure and the nonlinearity of the underlying thermodynamic model, it is obvious that this MINLP problem is particularly hard to solve and restricted to local optimization. In this work, a robust and efficient solution of the MINLP problem is achieved by favorable initialization and a clever reformulation of the discrete continuous into a purely continuous problem. This way, the MINLP is converted into nonlinear programming (NLP) problem, which has been shown to solve much more efficiently in case of simple column optimization.[19]

The initialization strategy is based on the process synthesis framework,[21] which comprises of a stepwise design procedure with incremental model refinement. In this framework, design variants are first screened with sophisticated shortcut methods, which also have been extended to HIDiC and VRC configurations.[22] Based on these screening results, the most promising variants are further refined using rigorous optimization. The results from the shortcut step, such as the minimum energy demand and the concentration profile estimates based on pinch points, are used to initialize the optimization. However, such an initialization strategy alone is not sufficient to converge the optimization model presented above. In addition, a simplified model is used first to compute an initialization. It only comprises component mole balances and the vapor-liquid equilibrium relations, but no energy balances. In subsequent solution steps, the energy balances are included again and the optimization model is resolved. In the final step, the model is optimized with respect to the desired objective function, capturing the energy demand or the TAC, for example.

The optimization procedure outlined here has been implemented in the General Algebraic Modelling System (GAMS).[23] SNOPT[24] has been used to solve the nonlinear programs. All calculations have been performed on a standard desktop computer (single core of Intel Core2 Quad Q9400 @ 2.66 GHz, 4 GB RAM). Vapor-liquid equilibrium calculations for nonideal mixtures are performed in an external user-defined function, which provides function evaluations and derivate information to the solver. This trick reduces the size of the optimization problem in GAMS and enhances the flexibility to choose more complex thermodynamic models.

Optimization Results

In this chapter, the superstructure model is used to determine exemplary optimal designs for HIDiC and VRC configurations. Although any design using VRC will often result in significant energy savings, the TAC of the design is typically of at least as much interest in industrial practice. Consequently, the optimization runs have been based on different objective functions capturing energy demand and TAC as alternative scenarios. Three case studies are selected to illustrate the versatility of the method and to assess the potential of HIDiC and VRC designs. The case studies comprise a set of ideal binary mixtures with increasing relative volatility, an ideal multicomponent separation occurring in styrene production, and a nonideal binary separation motivated by bioethanol production.

Separation of ideal binary mixtures

The optimization is performed first for the separation of different binary zeotropic mixture of equimolar composition into pure component streams with a product purity of 99 mol %. The mixtures, displayed in Table 1, have been selected to cover a range of relative volatility between 1.18 and 4.63, which corresponds to a difference in the boiling points between 5.5 and 51.6 K. The vapor-liquid equilibrium is calculated using Raoult's law and Antoine's equation; the physical property data are taken from the APV72 PURE24

Table 1. Selected Binary Separations

Top Product	Benzene	Benzene	Benzene	Benzene
Bottoms Product	Fluoro-benzene	*n*-Heptane	Toluene	Chloro-benzene
Relative volatility	1.18	1.74	2.49	4.63
Boiling point difference (K)	5.5	18.3	30.5	51.6

database provided by ASPEN Plus.[25] All feed streams are in the state of saturated liquid.

Optimization for minimum energy

To quantify the benefit of a widely distributed heat exchange from a thermodynamic point of view, the HIDiC model is optimized for minimal energy consumption. Consumed electricity and steam are combined in a single energy figure where electricity is weighted by a primary energy factor to take the different exergetic value into account. We assume a primary energy factor of 2, assuming a conversion efficiency of 50% for state-of-the-art power plants. A lower bound on the temperature difference in the heat exchangers of 5 K is defined to constrain the heat exchange area. The nonlinear optimization problem for solving the HIDiC synthesis problem is formulated as

$$\min Q_{\text{total}}$$

$$s.t.\ (1)-(18)$$

Energy optimization is performed twice for all four binary separations. In the first four runs, the number and location of intermediate heat exchange are not constrained while heat exchange is constrained to two or three locations in the second four runs. The results are presented in Table 2 and in Figures 4–7. The figures indicate the relative amount of heat exchanged between the stripping and the rectifying section on adjacent trays. If no constraints are formulated for the heat exchange locations, a widely distributed heat exchange is found to be optimal. In contrast, if the heat exchange is constrained, only a small increase of less than 2% in the energy demand of the investigated HIDiC configuration is observed. If such small energy penalties for constraint heat exchange could be generalized to other separations, it would be highly questionable, whether research and development aiming at new equipment design for distributed heat exchange along the height of the column—the focus of HIDiC research in recent years (cf., e.g., Bruinsma et al.[26])—is promising.

These results correspond to the findings of Shenvi et al.[6] and Suphanit;[10] they both found that HIDiC configurations offer only very little benefit over a design with a small number of heat exchangers in case of high-purity separations. While they derived their conclusions from a qualitative thermodynamic analysis of the temperature profiles, our method results in an optimal design, which quantifies alternative configurations with respect to energy demand using rigorous process models.

Optimization for minimum total analyzed cost

The optimization for minimum energy demand indicates that constraining the number of heat exchange locations to two or three results in a relatively small increase in energy demand of the HIDiC configuration. While an increase in energy demand increases operating cost, a smaller number of heat exchangers most likely lower capital cost. The trade-off between these two cost factors can be systematically addressed by our design method, if they are included in the objective function of the optimization model. Consequently, we formulate and solve the following optimization problem to minimize TAC

$$\min \text{TAC}$$

$$s.t.\ (1)-(25)$$

The operating cost of the column is determined for 8000 h of operation per year with energy cost factors of 3.32 ct/kWh (20 €/t) for steam at 3 bar, 4.15 ct/kWh (25 €/t) for steam at 12 bar and 6.66 ct/kWh for electricity. The cost of cooling water is neglected. Capital cost is based on cost correlations for the equipment involved taken from Guthrie[20] assuming stainless steel equipment and updating the cost according to the 2008 Marshall and Swift index of 1408.8.[27] The capital costs are annualized assuming a depreciation interval of 8 years and an interest rate of 9%. This corresponds to a capital charge factor of 0.18, that is, 18% of the capital costs are charged annually to cover interest on spent capital and depreciation. As equipment cost are accounted for rigorously in this optimization problem, the temperature bounds for the heat exchangers imposed in the optimization problem for minimum energy can be lifted. This optimization problem is solved for all four mixtures given in Table 1 for HIDiC and VRC configurations as well as for a conventional column.

In the following, the results for the separation of benzene and *n*-heptane are presented in more detail. The HIDiC configuration determined by means of optimization has a total number of 46 trays (where the condenser represents the first and the reboiler the last tray), with 23 trays in both, the rectifying and the stripping sections. The rectifying and the stripping sections are operated at pressure of 2.187 bar and 1 bar, respectively. Investigating the optimal solution shown in Figure 8 in more detail, the heat exchange between the column sections is performed in one intermediate heat exchanger between the second and the 45th stage at a level of 504 kW. The reboiler and the compressor duties are 81 and 79 kW, respectively. The vapor is superheated by a temperature increase of 5 K before entering the compressor to avoid condensation. Because of the different pressure levels

Table 2. Energy Demand for Binary Separations; Constrained and Unconstrained Heat Exchange Locations

Top Product		Benzene	Benzene	Benzene	Benzene
Bottom Product		Fluoro-benzene	*n*-Heptane	Toluene	Chloro-benzene
Energy demand	Unconstrained (kW)	255.6	221.0	206.0	223.6
	Constrained (kW)	260.0	221.4	207.4	223.8

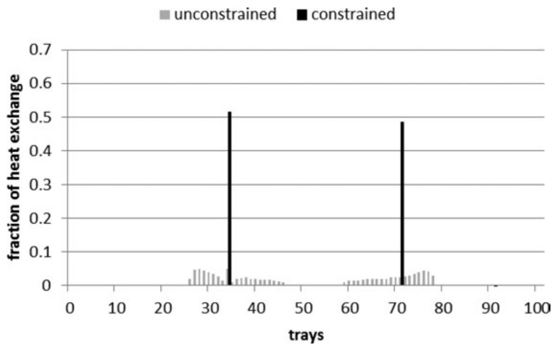

Figure 4. Benzene–fluorobenzene separation: distribution of the fraction of heat exchange for the 100 trays of the rectifying section.

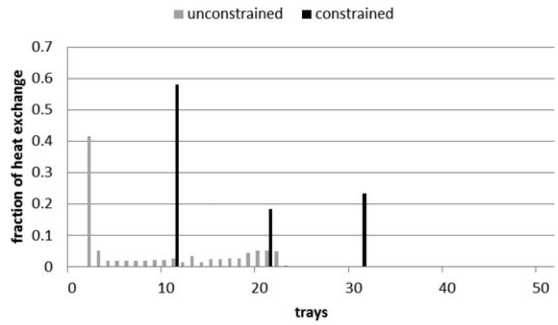

Figure 6. Benzene–toluene separation: distribution of the fraction of heat exchange for the 51 trays of the rectifying section.

and vapor flows, the optimal column diameter differs for the two column sections, 0.647 m for the rectifying section and 0.770 m for the stripping section. This minimum cost design differs significantly from the design minimizing energy demand, where the rectifying section is operated at a pressure of 1.636 bar and the heat exchange is performed in two intermediate heat exchangers.

In case of an optimal VRC design, the column consists of 45 stages. The reboiler energy duty is entirely replaced by condensation of the recompressed vapor amounting to a heat exchange of 600 kW. Before VRC, the vapor is superheated by a temperature increase of 5 K to avoid condensation in the compressor. The vapor is compressed to a pressure of 2.162 bar at a power duty of 66 kW. For the conventional column, 49 stages with a reboiler duty of 586 kW are found to be optimal.

Table 3 displays the major design data for the conventional column as well as for the HIDiC and the VRC configurations. The HIDiC configuration offers a significant cost saving and a tremendous reduction in energy demand compared to the conventional design. However, a difference in the cost structure of the column can be observed. As expected, the compressor and the intermediate heat exchangers almost double the capital cost compared to the conventional column design. The operating cost on the other hand is significantly lower and amount to approximately 65% of the conventional column. The TAC of the HIDiC configuration is 22% lower than the cost of the conventional column. However, even larger cost savings of 42% can be achieved with a VRC design, which shows lower operational and capital cost than the HIDiC configuration.

A brief overview of the results for all four binary separations investigated is displayed in Table 4. The HIDiC configuration does not always show the lowest cost. In case of high relative volatility, the conventional column design shows a cost advantage over the HIDiC configuration based on the specific cost function used in our calculations. While the benzene/*n*-heptane separation still shows cost savings for the HIDiC configuration, the benzene/toluene and the benzene/chlorobenzene separations are more cost efficient with a conventional column. However, the VRC design is always identified to be superior to the HIDiC designs with respect to TAC.

The cost reductions achieved with HIDiC and VRC designs strongly depend on the relative volatility of the mixture to be separated. In particular, higher cost reductions are possible in case of mixtures with low relative volatility. However, a thermodynamic analysis only based on relative volatility does not account for the nonlinear effects of heat exchanger cost correlations, of bounds on temperature difference in heat exchangers and the pressure on capital cost. Hence, an optimization-based design methodology as suggested in this contribution has to be used at least in those cases where the application of HIDiC or VRC configurations appears promising based on a preliminary thermodynamic analysis based on temperature profiles[6] or on relative volatilities.

Trade-off between capital and operating cost

VRC and HIDiC configurations can show significantly lower energy demand compared to a conventional distillation column. However, the increase in capital cost can outweigh

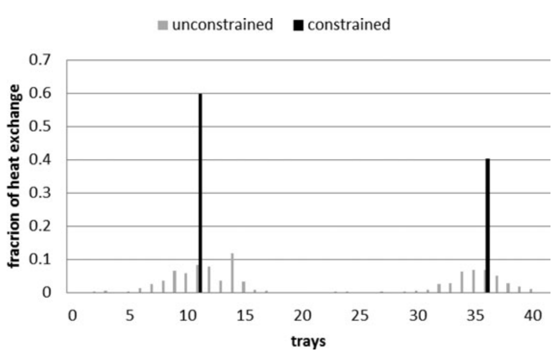

Figure 5. Benzene–*n*-heptane separation: distribution of the fraction of heat exchange for the 40 trays of the rectifying section.

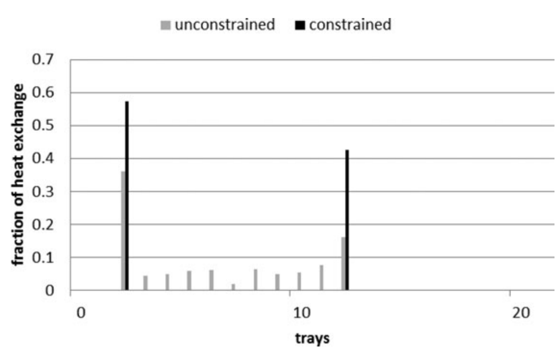

Figure 7. Benzene–chlorobenzene separation: distribution of the fraction of heat exchange for the 21 trays of the rectifying section.

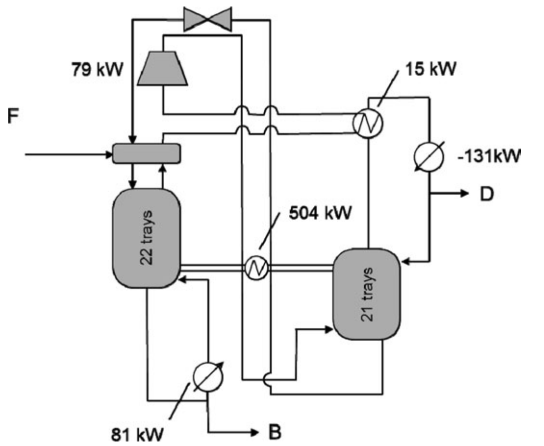

Figure 8. Optimal column configuration and major design data for the separation of an equimolar mixture of benzene and *n*-heptane.

Table 3. Results from Rigorous Column Optimization for the Separation of an Equimolar Mixture of Benzene and *n*-Heptane

	Conventional column	HIDiC Configuration	VRC Configuration
Capital cost (k€/a)	79	141	129
Operating cost (k€/a)	205	71	36
TAC (k€/a)	284	212	165
Pressure (bar)	1.013	2.187/1.013	2.162/1.013
Reboiler duty (kW)	586	81	–
Condenser duty (kW)	593	131	59
Compressor duty (kW)	–	79	66
Number of stages	49	46	45
Feed stage	25	23	23
Diameter (m)	0.810	0.647/0.770	0.819
Computation time (s)	4	76	10

the savings in operating cost. Capital cost enters the TAC calculation by the capital charge factor, which accounts for the fraction of the investment that has to be spent on debt service annually. The choice of the capital charge factor in the design calculations strongly depends on the market environment and the business model of the operating company. Table 5 gives an overview of capital charge factors for different interest rates and depreciation time intervals. In this subsection, the sensitivity of the different designs with respect to the capital charge factor is exemplarily investigated for the benzene/*n*-heptane and the benzene/toluene separations for capital factors in the range of 0.12–0.24, which cover most of the scenarios in Table 5.

Figure 9 displays the results of the design optimizations for the benzene/*n*-heptane separation using different capital charge factors. For the specific cost functions used, the relative order of the cost-optimal designs does not change with the capital charge factor. In all cases, the VRC configuration is the best choice, followed by the HIDiC configuration and the conventional distillation column. The VRC configuration should consequently be selected independently of the economic scenario.

The results are slightly different, if the benzene/toluene separation is optimized using different capital charge factors as shown in Figure 10, where the TAC is displayed for the three design alternatives investigated. Here, the HIDiC configuration is more favorable than the conventional distillation for capital charge factors smaller than 0.14. However, the VRC configuration is still the most competitive choice over the entire range of capital charge factors investigated. Again, the VRC configuration should be selected independently of the economic scenario.

These exemplary calculations show that the economic scenario represented by the capital charge factor can have an impact on the ranking of the separation process alternatives. The thermodynamic properties of the mixture seem to be of major importance. In the two investigated cases, a change in the ranking of the alternatives could only be observed for the separation of the mixture with the higher relative volatility. The trend in the correlation between TAC and capital charge factor is similar for HIDiC and VRC configurations with VRC being always more favorable than HIDiC. The trend of this correlation can change, however, for conventional columns, thus impacting the ranking of the variants.

Ideal multicomponent separation

The optimization-based design methodology suggested in this work is not restricted to binary mixtures. Rather, it is (in principle) applicable to separations involving any number of components. In this section, the design methodology is applied to a multicomponent separation, for example, the separation of the product from the reactor effluent in styrene production. Styrene is separated from a mixture of benzene, toluene, etyhlbenzene, and styrene at a purity of 99 mol %. The exemplarily chosen separation task assumes a bottom product to feed ratio of 0.645, a saturated liquid feed at a flow rate of 10 mol/s and a composition of $x_{\text{benzene}} = 0.006$, $x_{\text{toluene}} = 0.013$, $x_{\text{ethylbenzene}} = 0.336$, and $x_{\text{styrene}} = 0.645$. The separation task is known to be difficult, because the boiling point difference between the distillate and the bottoms product at an operating pressure of 0.3 bar is only 11.6 K. HIDiC designs have been investigated in simulation studies for this mixture before.[28]

All cost data are the same as in the case studies involving binary mixtures. The vapor-liquid equilibrium is calculated from Raoult's law and Antoine's equation; the physical property data are again taken from the APV72 PURE24 database provided by ASPEN Plus.[25] Because of the relatively low isentropic exponent of the compressed vapor mixture, the vapor has to be superheated before entering the compressor to prevent condensation during the compression for the HIDiC configuration by 5 K and for the VRC

Table 4. Summary of Results for the Investigated Binary Separations

Top Product	Benzene	Benzene	Benzene	Benzene
Bottom Product	Fluoro-benzene	*n*-Heptane	Toluene	Chloro-benzene
Relative volatility	1.18	1.74	2.49	4.63
TAC conv. column (€/a)	836	284	160	126
TAC HIDIC (€/a)	472	212	177	147
TAC VRC (€/a)	384	165	133	126

Table 5. Capital Charge Factors for Different Interest Rates and Depreciation Time Intervals

	Depreciation Time (a)			
Interest Rate (%)	6	8	10	12
8	0.216	0.174	0.149	0.133
10	0.230	0.187	0.163	0.147
12	0.243	0.201	0.177	0.161
14	0.257	0.216	0.192	0.177

configuration by 12 K. The vapor is superheated using part of the condenser energy.

The design is optimized for TAC. Because of the increased number of components and the large number of trays required for the separation of the close-boiling mixture, the number of equations in the NLP problem is increased significantly. Therefore, convergence is more difficult to achieve than in case of the binary separations investigated before. However, the utilization of the stepwise initialization and the tight bounds on the variables allow for a good convergence at moderate computational effort. The design optimization of the ideal-multicomponent separation was performed on a desktop computer (single core of Intel Core2 Quad 2.66 GHz, 4 GB RAM) in less than 100 s.

Optimization for minimizing TAC is performed for HIDiC and VRC configurations and for a conventional column. The HIDiC configuration consists of 75 stages, 37 in the rectifying and 38 in the stripping section. No reboiler is required, as the heat is entirely provided by two intermediate heat exchangers, which transfer 571 kW between the second and the 53rd stage and 1065 kW between the 23rd and the 74th stage. The compressor duty is 115 kW to increase the pressure from 0.3 bar in the stripping section to 0.517 bar in the rectifying section. The TAC of the HIDiC configuration is 384 k€/a. The VRC design consists of 122 stages, 41 in the rectifying and 81 in the stripping section. The reboiler heat duty of 1350 kW is entirely provided by the recompressed vapor. The pressure is increased from 0.3 to 0.576 bar at a compressor duty of 95 kW. The TAC of the VRC design is 372 k€/a. The conventional column consists of 120 stages, 49 in the rectifying and 71 in the stripping section. The reboiler duty is 1343 kW. The TAC of the conventional column is 611 k€/a. Surprisingly, the HIDiC design has a significantly lower number of stages compared with the VRC and the conventional column designs. However, this comes at the price of an increased reflux rate to achieve the desired purity. The detailed results of the design optimization are displayed in Table 6. Figure 11 shows the structure of the HIDiC configuration.

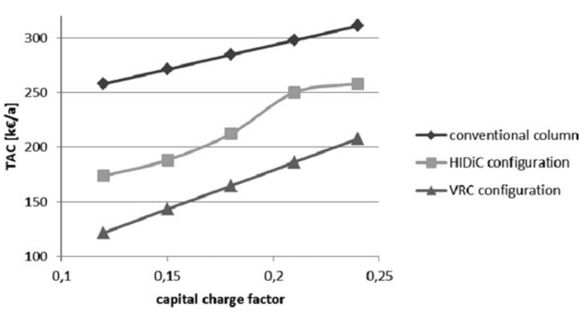

Figure 9. TAC for the benzene/*n*-heptane separation using different capital charge factors.

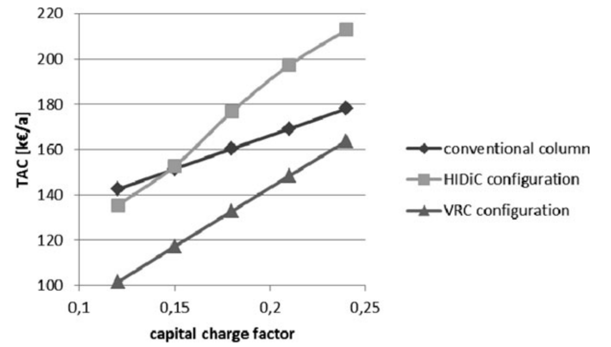

Figure 10. TAC for the benzene/toluene separation for different capital charge factors.

This result confirms the findings in the case studies involving binary mixtures: a small number of intermediate heat exchangers is cost optimal. In particular, the HIDiC design offers significant cost savings of 37% compared to the conventional column design. However, similar to the binary cases, a VRC design can even further reduce the cost by 39% compared to the conventional column design. In contrast to the binary separations, the cost difference between VRC and HIDiC designs is small.

Nonideal binary separation

The last case study addresses nonideal separations. In particular, we choose a binary water/ethanol separation, because of its relevance to biorenewables processing, for example, in the production of bioethanol or in solvent recovery in the Organosolv process[29] for biomass pretreatment and fractionation. The separation task investigated is the removal of water from an equimolar mixture of ethanol and water to increase the ethanol mole fraction in the mixture to 80%. The nonideal vapor-liquid equilibrium is modeled by the non-random two-liquid (NRTL) activity coefficient model. All the physical property data are taken from the APV72 PURE24 database provided by ASPEN Plus.[25] Vapor-liquid equilibrium calculations are performed using an external user-defined function, which is called by the solver via GAMS. The relatively high isentropic exponent of the vapor mixture does not require superheated vapor at the compressor inlet.

A HIDiC and a VRC design are investigated and benchmarked against a conventional column design. The three designs are optimized for TAC for a feed flow rate of 10 mol/s. All the economic data are the same as before. The

Table 6. Results of the Rigorous Optimization of the Styrene Separation

	Conventional Column	HIDiC Configuration	VRC Configuration
Capital cost (k€/a)	235	321	320
Operating cost (k€/a)	376	63	52
TAC (k€/a)	611	384	372
Pressure (bar)	0.3	0.517/0.3	0.576/0.3
Reboiler duty (kW)	1343	–	–
Condenser duty (kW)	1342	102	95
Compressor duty (kW)	–	115	95
Number of stages	120	75	122
Feed stage	49	37	41
Diameter (m)	1.509	1.476/1.673	1.533
Computation time (s)	36	97	74

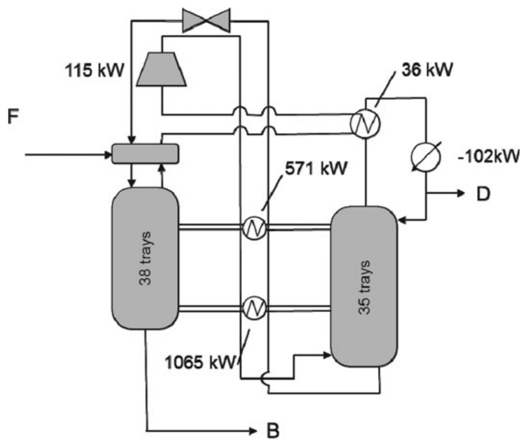

Figure 11. Optimal configuration of the styrene separation process.

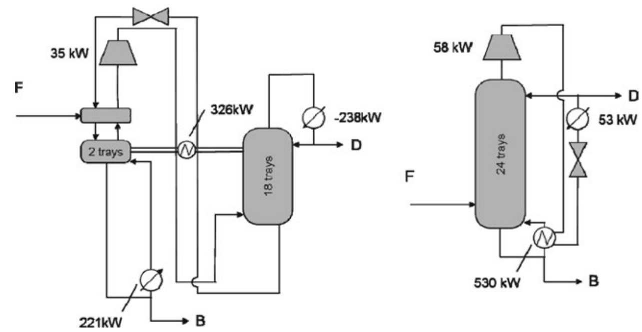

Figure 12. Optimal configurations for HIDiC (left) and VRC (right) designs for the ethanol/water separation.

resulting HIDiC configuration consists of 24 stages, 19 in the rectifying and 5 in the stripping section. A single intermediate heat exchanger transfers 326 kW between the second and the 22nd stage of the column. The rectifying and the stripping sections are operated at pressures of 1.799 and 1013 bar, respectively. The reboiler and the compressor duties are 221 and 35 kW. The TAC of the HIDiC process is 192 k€/a. The VRC design consists of a column with 26 stages, 22 in the rectifying and 4 in the stripping section. The vapor is compressed to a pressure of 2.811 bar and condensed. This way, the external utility of 530 kW provided to the reboiler is completely replaced. The compressor duty is 58 kW. The TAC of the VRC design is 132 k€/a. The optimized conventional column consists of 26 stages, 22 in the rectifying and 4 in the stripping section. The reboiler heat duty is 522 kW. The TAC is 205 k€/a. The design results are summarized in Table 7, and a sketch of the HIDiC and the VRC configurations is provided in Figure 12. All design optimizations were performed on a desktop computer (single core of Intel Core2 Quad 2.66 GHz, 4 GB RAM) in reasonable computation times of less than 1 min.

For this separation, the VRC design is found to be more cost effective than the HIDiC and the conventional column designs. A cost savings of 36% compared to a conventional distillation column can be achieved. In contrast, the HIDiC configuration is not attractive because predicted savings amount to 6% compared to the conventional column and are, therefore, quite low. Similar to the benzene/*n*-heptane separation, a single column shell and lower pressure with higher relative volatilities in the rectifying section gives the VRC design a competitive advantage over the HIDiC design.

Table 7. Results of the Rigorous Optimization for the Ethanol/Water Separation

	Conventional Column	HIDiC Configuration	VRC Configuration
Capital cost (k€/a)	59	111	101
Operating cost (k€/a)	146	81	31
TAC (k€/a)	205	192	132
Pressure (bar)	1.013	1.799/1.013	2.811/1.013
Reboiler duty (kW)	522	221	–
Condenser duty (kW)	517	238	53
Compressor duty (kW)	–	35	58
Number of stages	26	24	26
Feed stage	22	20	23
Diameter (m)	0.449	0.463/0.523	0.538
Computation time (s)	8	43	28

Conclusions

HIDiC have the potential of significant cost and energy savings compared with conventional distillation columns. So far, high equipment cost, not yet fully satisfactory designs of internal heat exchangers, and the lack of a systematic design methodology has hindered the application of these attractive design variants in industrial practice. In this contribution, we investigate under which circumstances HIDiC or VRC configurations offer favorable design options for cost-optimal heat-integrated distillation. A new methodology relying on rigorous optimization for minimizing TAC is introduced to identify cost-optimal conceptual designs of heat-integrated distillation systems. The rigorous optimization significantly reduced the engineering effort compared with tedious sensitivity studies in process simulations. In contrast to conventional column or VRC designs, the HIDiC problem results in a more complex formulation including additional degrees of freedom, such as the operating pressure of the rectifying section, the feed tray position, the number of trays, and the location and size of the internal heat exchangers. Different objectives can be used in design optimization; we have used energy demand and TAC in the case studies of this work. A continuous reformulation of the mixed-integer optimization problem together with a favorable, stepwise initialization strategy allow for fast and robust convergence of the solution algorithm. The performance of the new design methodology and the achievable economical benefit are demonstrated on several separation case studies, including a quaternary and several binary ideal mixtures as well as a nonideal binary mixture. Compared to a conventional column design, significant cost savings can be achieved by heat-integrated designs. However, HIDiC and VRC designs are shown to be only more favorable than conventional columns for close boiling mixtures, since the required pressure increase for the rectifying section is relatively small.

The locations of the intermediate heat exchangers to transfer heat from the rectifying to the stripping section are important design decisions for HIDiC configurations. In literature, internal heat exchange uniformly distributed over the entire height of the column has often been claimed to be optimal. However, the optimal intermediate heat exchangers configuration found in all cases investigated in this work consists of a small number of relatively large heat exchangers. Configurations designed for minimal energy demand with a limited number of possible heat exchangers show only small increase in energy demand compared to the case where heat exchangers are allowed to connect any of the trays. In economic optimization, the trade-off between operation and capital cost clearly favors a small number of heat exchangers. This result, though confirming less rigorous findings in previous studies, is in conflict with the predominant understanding of HIDiC design, which assumes uniformly distributed heat exchange over the entire height of the column. Such a heat exchange configuration poses serious problems with respect to equipment design, which are yet largely unsolved. However, the optimal designs identified in our case studies, all showing a small number of intermediate heat exchangers can be easily built using conventional equipment, for example, column shells and heat exchangers.

VRC designs have resulted in lower TAC compared to HIDiC designs in all the investigated case studies. Given the similarity between VRC and HIDiC with a single intermediate heat exchanger, the simpler design of VRC with a single column shell, and the generally higher relative volatilities at lower operating pressure in the rectifying section, this is result is finally not surprising.

Based on our findings as well as other researchers in the literature,[6,10] we postulate the heuristic that—maybe with rare exceptions—VRC designs are typically superior over HIDiC designs with respect to energy and cost savings, but both designs often outperform conventional distillation, in particular if mixtures with relatively small driving forces are to be separated.

Acknowledgments

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Notation

A = heat exchanger area, m^2
b = binary variable
B = boil up flow rate, mol/s
C = cost, €
D = diameter of column shell, m
F = feed flow rate, mol/s
f_c = capital charge factor
h = specific enthalpy, kJ/mol
I = set of components
K = equilibrium constant
L = liquid flow rate, mol/s
N = set of stages
NT = number of trays in section
p = pressure, bar
Q = heat flow, kW
R_{gas} = gas constant
R = reflux flow rate, mol/s
T = temperature, K
t_a = annual operating time
TAC = total annualized cost, €/a
V = vapor flow rate, mol/s
W = work, kW
x = mole fraction of liquid phase
y = mole fraction of vapor phase
z = mole fraction of feed

Greek letters

η = isentropic efficiency
κ = isentropic exponent

Subscripts

cap = capital
comp = compressor
con = condenser
in = inlet
int = intermediate heat exchanger
max = maximum
n = stage number
op = operational
out = outlet
pre = preheater
reb = reboiler
rect = rectifying section
strip = stripping section

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