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IDENTIFYING OPTIMAL MIXTURE PROPERTIES FOR HIDiC APPLICATION

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Abstract
Internally heat-integrated distillation columns (HIDiC) are a new design option to provide a sustainable separation system. Although significant energy savings can be achieved using this setup, a considerable increase in investment cost penalizes the application. One major cost driver is the compressor cost, which depends on the relative volatility of the mixture to be separated. While mixtures with large relative volatility are easy to separate in distillation, they require a large pressure increase in the compressor of the HIDiC design, which raises the investment and operating costs of the design. Mixtures with low relative volatility are generally difficult to separate in distillation but favour the application of the HIDiC concept. Although it is clear that close boiling components are favourable from an energetic point of view[10], the influence of the equipment cost can not be neglected. In this contribution, HIDiC column designs for different mixtures are optimized via rigorous MINLP optimization to identify a relative volatility region which provides optimal operating conditions for maximum cost reduction. The mixtures with a low relative volatility favour a highly integrated design, while for a high relative volatility the conventional design is more cost efficient. The optimal design identified for mixtures with low relative volatility is conceptually very similar to a conventional column with heat pump.

Keywords: HIDiC, MINLP, rigorous optimization

1. Introduction

Worries about climate change and depleting resources have drawn attention to more sustainable chemical processes. The consumption of resources in a chemical process is based on the process feedstock and the utility system. Distillation columns are still one of the major consumers for energy from the utility systems. Since the 1950's, different techniques for the reduction of this energy demand have been suggested. These include complex column configurations and energy integration in distillation column networks.

Recently, the concept of a internally heat integrated distillation column (HIDiC) has been suggested[8], which allows for heat integration within a single distillation column. In this design, a compressor is employed to operate a part of the column at elevated pressure. The higher pressure in the rectifying section allows for heat transfer with the trays of the stripping section. The operation of such a column has the potential to be similar to reversible distillation, in which every stage is provided with heating or cooling to allow for an optimal separation[4]. The feasibility of this design has been shown in pilot plant scale[3]. However, no industrial application beyond a pilot plant is known to the authors knowledge.

The major drawback of the HIDiC design is the need for a compressor to connect the vapor streams of the stripping section with the rectifying section. Not only does the compressor require the use of relatively expensive electric utility, it also comes with a significant investment cost. The energy requirement for vapor compression is highly dependent on the relative volatility of the components to be separated; it is estimated in a thermodynamic analysis based on the assumption of isentropic compression. Close boiling mixtures require only a moderate pressure increase and are therefore the favored application for HIDiC and heatpump configurations. In this contribution, the economic performance of HIDiC designs is compared to conventional column designs for six case studies. These binary separation problems were selected based on the relative volatility of the components to be separated; relative volatilities between 1.2 and 5.9 have been selected. The optimal design is determined by means of MINLP optimization, which simultaneously optimizes the column design and the operating point for a minimum total annualized cost (TAC).

2. Modeling

Distillation design is usually based on simulation studies, which require a detailed specification of the distillation column early in the design process. Although design is supported by heuristics, it requires a high manual effort by the design engineer. Another disadvantage is that a good result is not guaranteed. A non-optimal design of the distillation column can lead to significant cost increase and therefore have an impact on the profitability of the process.

Rigorous economic optimization provides an alternative design approach. Here, the distillation column is optimized using rigorous mathematical methods. The optimization modeling allows for the consideration of a high detail on thermodynamics and design and enables the design engineer to identify important design parameters like the number of trays, feed location and column diameter. However, due to the high complexity of the model, good initial guesses are essential in order to robustly converge to a meaningful solution. Good initial guesses can be provided by previous shortcut calculations, as suggested recently by the authors in the 'process synthesis framework'[7]. Due to the ideal nature of the mixtures investigated here, we reduced the model complexity by the application of ideal thermodynamics and Raoults law.

The model presented in this paper is based on the rigorous MINLP column model presented by Kraemer et al.[6]. Compared to the model of a conventional distillation column, additional degrees of freedom rise from the HIDiC configuration. Most prominent differences are the need to determine the optimal operating pressure for the rectifying section, and the positions of the intermediate heat exchangers. The operating pressure of the stripping section is not variable and set to a value of 1.013 bar, reducing the degrees of freedom by allowing only one variable pressure. In our model, the compressor is located above the feed tray, it is modeled using the isentropic pressure temperature correlation

$$T_{out} = T_{in}\left(\frac{p_{out}}{p_{in}}\right)^{\frac{\kappa-1}{\kappa}} . \tag{1}$$

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

$$T_{out} = T_{in}\left(1 + \frac{1}{\eta_{comp}}\left(\left(\frac{p_{out}}{p_{in}}\right)^{\frac{\kappa-1}{\kappa}} - 1\right)\right). \tag{2}$$

can be determined by rearranging (1) and taking the isentropic efficiency into account[4]. The optimization superstructure for the HIDiC column design is shown in Figure 1. The superstructure allows heat exchangers to transport heat from the pressure optimized rectifying section to the colder stripping section at every tray. The combinatorial complexity is reduced by permitting heat exchange only between trays of the rectifying and stripping section at the same vertical position. In order to enable all meaningful heat exchanger connections and determine the optimal tray number, the column trays can be reduced not only at both ends of the column, which would be sufficient for a simple column design, but at both ends of each section. Thus, column designs are possible where the heat is exchanged at the extreme ends only (cf. Fig. 3). While the feed tray is fixed, the optimal position of the column feed relative to the column ends is identified by optimization of the number of trays in each section. Due to the pressure difference between the column sections, HIDiC columns can have different column diameters for the rectifying and the stripping section. Hence, optimal column diameters are determined for each section separately in this work.

The cost optimal HiDIC design is determined by a minimization of an economic objective function, which contains both operating costs (hot and cold utilities, compressor duty) and capital costs (depreciation costs for column shell, internals, heat exchangers, and compressor). Note that the areas of the heat exchangers contribute with an exponent of 0.65 to the capital costs for the heat exchangers. A minimum number of internal heat exchangers is therefore favored within the economic optimization. Since the design variables (tray numbers) are discrete variables while the heat exchange, flowrates and compositions are continuous variables, a mixed-integer nonlinear optimization problem (MINLP) has to be solved. Considering the large scale and complexity of the HIDiC design and the nonlinearity of the underlying thermodynamics, it is obvious that this MINLP problem is particularly hard to solve. In this work, the robust and efficient solution of the MINLP problem is achieved by a favorable initialization strategy and a reformulation as a purely continuous problem. A brief overview of the HIDiC column model is shown in equations 3 to 21 and Figure 2. A detailed description of the simple column model formulation, the initialization strategy as well as the solution algorithm can be found elsewhere[6].

$$0 = L_{n-1}x_{n-1,i} + V_{n+1}y_{n+1,i} - L_n x_{n,i} - V_n y_{n,i} + b_{R,n}Rx_{1,i} + b_{V,n}V_{BP}y_{BP,i}, \; n \in [2, \mathrm{N_F}[\tag{3}$$

$$0 = L_{n-1}x_{n-1,i} + V_{n+1}y_{n+1,i} - L_n x_{n,i} - V_n y_{n,i} + b_{B,n}By_{N_{\max},i} + b_{L,n}L_{BP}y_{BP,i}, \; n \in]\mathrm{N_F}, N_{\max}[\tag{4}$$

$$0 = L_{n-1}h_{l,n-1} + V_{n+1}h_{v,n+1} - L_n h_{l,n} - V_n h_{v,n} + b_{R,n}Rh_{l,1} + b_{v,n}V_{BP}h_{v,BP} - Q_{D,\mathrm{int},n}, \; n \in [2, \mathrm{N_F}[\tag{5}$$

$$0 = L_{n-1}h_{l,n-1} + V_{n+1}h_{v,n+1} - L_n h_{l,n} - V_n h_{v,n} + b_{B,n}Bh_{v,N_{\max}} + b_{L,n}L_{BP}h_{l,BP} + Q_{B,\mathrm{int},n}, \; n \in]\mathrm{N_F}, N_{\max}[\tag{6}$$

$$0 = V_{n+1}y_{n+1,i} - Dx_{n,i} - Rx_{n,i}, \; n = 1, i \in I \quad (7) \qquad 0 = V_{n+1}h_{v,n+1} - Dh_{l,n} - Rh_{l,n} + Q_D, \; n = 1 \tag{8}$$

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

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

$$0 = L_{n-1}x_{n-1,i} + V_{n+1}y_{n+1,i} - L_{BP}x_{BP,i} - V_{BP}y_{BP,i}, \; n = N_F \tag{11}$$

$$0 = L_{n-1}h_{n-1} + V_{n+1}h_{l,n+1} - L_{BP}h_{l,BP} - V_{BP}h_{v,BP} + W_{comp}, \; n = N_F \tag{12}$$

$$\sum_i x_{n.i} = 1, \sum_i y_{n,i} = 1, \; n \in N \quad (13) \qquad y_{n,i} = K_n(\mathbf{x}_n, \mathbf{y}_n, T_n, p_n)x_{n,i}, \; n \in N \tag{14}$$

$$\sum_n b_{V,n} = 1, \; \sum_n b_{L,n} = 1, \; \sum_n b_{R,n} = 1, \; \sum_n b_{B,n} = 1, \; V_{N_F} = 0, \; L_{N_F} = 0 \tag{15}$$

$$W_{comp} = V_{BP}cp(T)(T_{out} - T_{in}) \tag{16}$$

$$NT_{rect} = N_{\max} - \sum_n \sum_n^{N_{\max}} b_{R,n} - \sum_n \sum_{n=1}^{n} b_{B,n} - \sum_{n=1}^{N_F} \sum_{n=1}^{n} b_{V,n} - \sum_{N_F}^{N_{\max}} \sum_n^{N_{\max}} b_{L,n} \tag{17}$$

$$Q_{D,\mathrm{int},n} = Q_{b,\mathrm{int}\,n+NF}, \; T_n \geq T_{n+N_F} \quad n \in [2, \mathrm{N_F}[\tag{18}$$

$$D_{col,strip} = \sqrt{\frac{4V_n}{2\pi}\sqrt{\frac{RT_n \sum_i y_{n,i}M_i}{p_n}}}, \; n = N_{\max}, \; D_{col,rect} = \sqrt{\frac{4V_{n+1}}{2\pi}\sqrt{\frac{RT_{n-1} \sum_i y_{n-1,i}M_i}{p}}}, \; n = N_F \tag{19}$$

$$C_{op} = f(Q_B, Q_D, W_{comp}), \; C_{cap} = f(NT, D_{col}, A_{reb}, A_{con}, A_{\mathrm{int},i}, W_{comp}) \tag{20}$$

$$\min TAC = C_{op}t_a + f_c C_{cap} \tag{21}$$

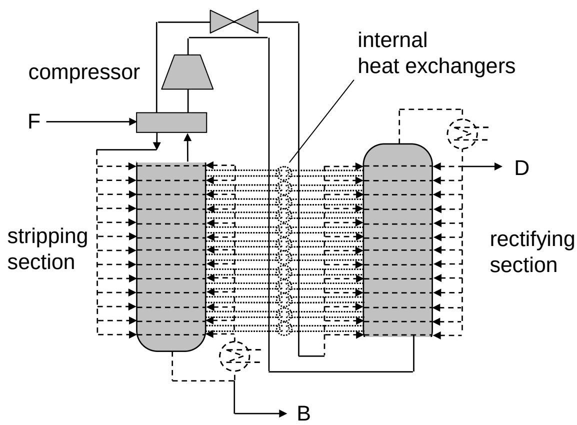

Figure 1. Superstructure to determine the optimal Number of trays and the position of internal heat exchangers for both column sections

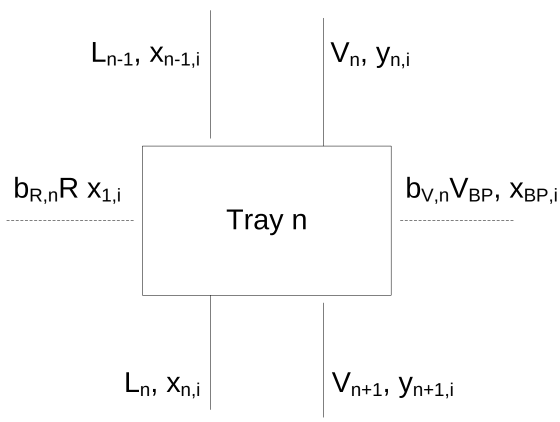

Figure 2. Equilibrium tray with streams for the rectifying section

3. Results

The model described in the previous section is applied to six different binary zeotropic mixtures with a feed flowrate of 10 mol/s. The separation task was the separation of the pure components from a mixture of equimolar composition. These binary mixtures are benzene/fluorobenzene, benzene/heptane, benzene/toluene, benzene/3-methyl-heptane, benzene/chlorobenzene and benzene/xylene. The desired product purity is 99%. The isentropic compressor efficiency is 72%. The operating cost of the column was determined for 8000 hours of operation per year with a energy price of 3,32 ct/kWh (20 €/t) for the 3bar steam, 4.15 ct/kWh (25 €/t) for 12bar steam and 6.66 ct/kWh for electricity. Cost functions for the equipment are taken from Guthrie[2]. Annualized capital cost are determined for a five year depreciation.

The optimization results for the benzene/fluorobenzene (relative volatility 1.18) separation will be discussed in more detail. Due to the close boiling nature of the mixture, 101 trays are optimal to achieve the desired product purity. The optimal solution identified for the HIDiC setup does not have a traditional reboiler and therefore does not require steam utility. The heat requirement of the stripping section is entirely fulfilled by the heat transferred from the rectifying section. The close boiling mixture requires only a moderate pressure increase of 0.37 bar. Since the energy balance of the column has to be fulfilled, there is still a need for a condenser to remove heat from the separation system.

Looking at the selected structure in more detail (cf. Figure 3), the heat exchange between the columns is optimally performed in one intermediate heat exchanger. Compared to a conventional column design, the total number of heat exchangers did not change. The position of the intermediate heat exchanger is at the top of the rectifying section and at the bottom of the stripping section, replacing the reboiler and partially the condenser. This design corresponds to a conventional column with heat pump, the only difference is that the entire rectifying section is operated at elevated pressure. Due to the elevated pressure, the optimal column diameter differs for the two column sections, 1.35 m for the rectifying section and 1.64 m for the stripping section.

Table 1. Results from rigorous column optimization for the separation of an equimolar mixture of benzene and fluorobenzene

	conventional column	HIDiC
capital cost	509580 €/a	546670 €/a
operating cost	584070 €/a	95920 €/a
TAC	1093650 €/a	642590 €/a
operating pressure	1.013 bar	1.383/1.013 bar
reboiler duty	2090 kW	-
condenser duty	2090 kW	160 kW
compressor duty	-	180 kW
number of trays	127	101
feed tray	52	50
diameter	1.504 m	1.513 / 1.625 m
computation time	6.7 s	135.9 s

A conventional column design was also optimized to have a base of comparison. Table 1 displays the major design specifications for both the HIDiC setup and the conventional column. The HIDiC configuration provides a significant cost saving compared to the conventional design. However, a difference in the cost structure of the column can be observed. As expected, the compressor and intermediate heat exchangers increase the capital cost of the design by 7%. The operating cost of the column is significantly lowered by approximately 83%. The total annualized cost of the HIDiC column is 41% lower than the cost of the conventional column.

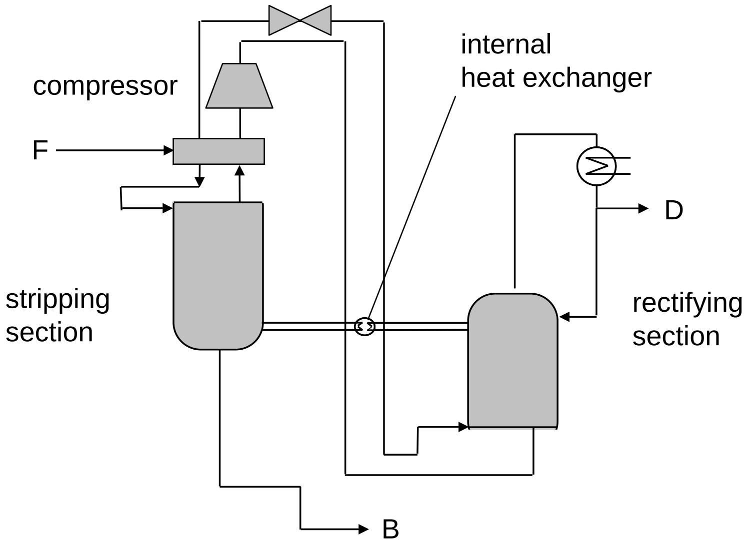

Figure 3. Optimal column configuration for the fluorobenzene / benzene separation

A brief overview of the results for the other binary separation case studies is displayed in Table 2. The HIDiC setup does not always come with cost savings. In case of high relative volatility, the conventional column design has a cost advantage over the HIDiC setup based on the given cost functions for capital and operating cost. However, these ratio is highly sensitive to the energy prices for steam and electricity.

A highly integrated design is only found to be optimal for cases with low relative volatility. In the cases of high relative volatility, the level of heat integration is lower for the cost optimal solution. A good indicator for this is the presence of a reboiler. While the reboiler is totally replaced for the cases with low relative volatility, this is not the case for the case studies with higher relative volatility. These generally do not show cost savings compared to the conventional column design. Although the HIDiC model itself should converge to a conventional column in these cases, the bounds required to achieve convergence enforce a HIDiC design.

Table 2: Summary of results for the binary case studies

top product	benzene	benzene	benzene	benzene	benzene	benzene
bottom product	fluoror-benzene	n-heptane	toluene	3-methyl-benzene	chloro-benzene	xylene
Relative volatiltity	1.18	1.74	2.49	3.19	4.63	5.85
reboiler	no	no	yes	yes	yes	yes
operating pressure HIDiC [bar]	1.383	2.269	3.013	1.895	2.468	3.084
TAC HIDIC [€/a]	643	262	178	194	147	148
TAC conv. column [€/a]	1094	325	178	170	135	133
TAC ratio	0.59	0.81	1.00	1.14	1.09	1.11

The results from the rigorous column optimization correspond qualitatively to the thermodynamic analysis. The HIDiC design is favourable in the case of mixtures with low relative volatility of the components. However, thermodynamic analysis does not take the nonlinear influence of heat exchanger cost, temperature bounds of utilities and the influence of pressure on the capital cost in to account. Since the conventional design is also covered by the model formulation, the extended rigorous optimization model should be used in cases where the application of HIDiC seems possible from a thermodynamic point of view.

4. Conclusions

In this contribution, the optimal HIDiC column configuration for six different binary mixtures is identified using MINLP optimization. The extension of an existing MINLP model to a HIDiC setup requires more complex formulations, especially for the optimization of the feed tray position, the number of trays and the location of internal heat exchangers. In order to optimize the pressure level of the stripping section, ideal thermodynamics based on Antoine's equation and Raoult's law were applied.

Compared to a conventional column design, cost savings can be achieved. However, this is highly dependent on the relative volatility of the components to be separated. The results confirm the thermodynamic analysis that HIDiC is favourable for close boiling mixtures, since the required pressure increase for the rectifying section is relatively small. The optimal solutions identified based on our cost function indicate that a relatively small number of intermediate heat exchangers is cost optimal. This is probably due to the fact that the nonlinear cost functions favour a small number of relatively large heat exchangers. It is a very interesting result, that heat exchange over the entire length of the column is not cost optimal.

In the case of very close boiling mixtures, the reboiler is completely replaced by the intermediate heat exchanger, reducing the HIDiC structure basically to a conventional column with heat pump. Hence, the operating window for HIDiC application is bounded on both sides of a relative volatility region. At low relative volatility, heat pump designs are cost effective and at higher relative volatility, conventional column designs are more cost effective. Unfortunately, the HIDiC design with 'true' intermediate heat exchangers (which do not entirely replace the reboiler) were all more expensive than the conventional column design.

In future work, additional modelling effort is required to improve the HIDiC MINLP model. Especially the implementation of nonideal thermodynamics is required to make the model widely applicable. The need for good initial guesses could be satisfied by an initialization based on sophisticated shortcut methods[1,9], as it has been shown for others cases of rigorous column optimization.

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