

Assessment of the Implementation of Heat-Integrated Distillation Columns for the Separation of Ternary Mixtures

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ABSTRACT: An ideal HIDiC (internal heat-integrated distillation column) makes the reboiler heat duty equal to zero, with the heat requirements for the separation transferred to a compressor. In real applications, there is a limit on the energy integration that can be achieved depending on the separation problem. In this work, a design method based on the column grand composite curve (CGCC) is presented. The CGCC is used to establish the integration capabilities between the rectifying and stripping sections of the system. The method is used to develop HIDiC separation sequences for ternary mixtures, and the effects of HIDiC implementation on energy, economics and greenhouse-gas emissions savings are addressed. The results of several case studies show that a good deal of energy savings can be achieved with this type of arrangement, but that the work required by the compressor can overcome the energy savings achieved through the process integration.

1. INTRODUCTION

Process intensification has been identified as the need to develop smaller, cleaner, and more efficient technologies.¹ One of the natural candidates for intensification studies is the distillation process, which is characterized by its low thermodynamic efficiency and high energy consumption, aspects that call for the development of more efficient distillation arrangements. By lowering the energy requirements in the reboiler of the columns, such improved schemes would also have a positive effect on factors such as CO₂ emissions and other environmental aspects. One option that has been proposed to improve the energy efficiency of conventional distillation systems is the heat-integrated distillation column (HIDiC). HIDiC schemes are based on the integration of the rectifying and stripping sections of a distillation column, aided by an increase of pressure in the rectifying section in order to provide a proper heat transfer. Studies on heat-integrated distillation columns have typically been carried out using ideal models for binary mixtures, and applications to ternary mixtures have been done with hypothetical ideal models. Among such studies, those by Nakaiwa and co-workers^{2–5} have analyzed several factors for ideal HIDiC systems such as the feed characteristics, the nature of the feed mixture, the temperature gradients, the feed location, and the pressure ratio between the rectifying and the stripping sections of the column. Their results showed that significant energy savings and a higher thermodynamic efficiency can be obtained, although the savings in the total separation costs might not be as significant. Gadalla et al.⁶ analyzed the operating range of HIDiCs as a function of the temperature gradient between the rectifying section (RS) and the stripping section (SS) of the column and showed a possible physical design for these types of systems. Ideal models have also been used to analyze some control aspects of HIDiC systems.^{7–9}

The HIDiC arrangement basically takes the structure of a conventional distillation system (Figure 1) to match the energy

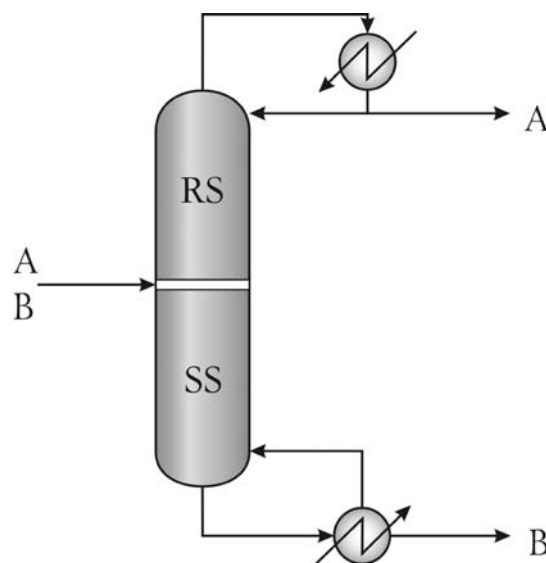


Figure 1. Conventional column with rectifying and stripping sections as defined by the feed stage.

required by the stripping section with the energy available from the rectifying section. In the ideal case, the HIDiC structure (Figure 2) eliminates the need for both heating and cooling requirements; reboilers and condensers would then play a role only for startup procedures. The ideal energy integration between the RS and SS is similar to the conceptual use of heat pumps,¹⁰ for which a higher pressure of the RS is needed to

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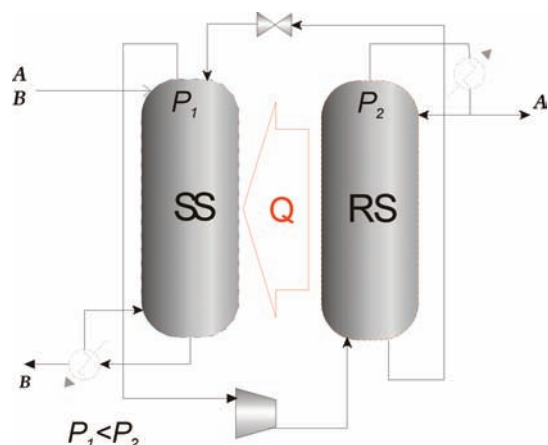


Figure 2. Ideal HIDiC system without reboiler and condenser duties.

provide a higher temperature profile and a suitable temperature difference (ΔT) between the column sections. Regarding the impact or benefits of HIDiC implementation on environmental factors, Gadalla et al.¹¹ reported a study on CO₂ emissions for a HIDiC, changing the heat requirements for the column from 0 to 80 MW; the results from this case study showed that reductions in emissions were possible only for reboiler heat loads lower than 60 MW.

To extend the initial works with the basic design principle of an ideal HIDiC with no need for external heat duty (under the assumption that the heat available in one section would exactly match the heat required by the other section), Gadalla et al.¹¹ showed that there is a range for the effective integration of HIDiC systems when deviations from an ideal model are considered. Recent works have reported design methods for the HIDiC scheme based on models that incorporate thermodynamic aspects, such as the adaptation of the Ponchon–Savarit graphical method for the HIDiC structure reported by Ho et al.¹²

In this work, the implications in terms of energy, economics, and greenhouse-gas emissions of HIDiC implementation are presented. First, the use of the column grand composite curve (CGCC)¹³ is proposed as a basis for the design of HIDiC systems. The advantage of this procedure is that thermodynamic principles are considered to establish the integration feasibility between sections in order to provide the actual energy savings that the HIDiC system can reach. Then, the effects of the work required by the compressor and greenhouse-gas emission levels are addressed. This work takes as a basis separation problems of three-component mixtures and considers sequences of two columns based on HIDiC structures, one that splits the ternary mixture followed by one that performs the remaining binary separation. From these studies, both the individual effects of each column and the overall performance of the HIDiC sequence can be addressed.

2. DESIGN METHODOLOGY

For an ideal distillation system, the heating and cooling requirements are equal. However, the enthalpy deficit profile is not always symmetrical,¹⁰ which means that the heat pump duties (if they were considered) for each section of the column would be different and that the heat distribution along each section would not be uniform and would also change from stage to stage. Therefore, limitations on the energy integration between the sections of the distillation column will arise. This is the basic consideration for the development of this work.

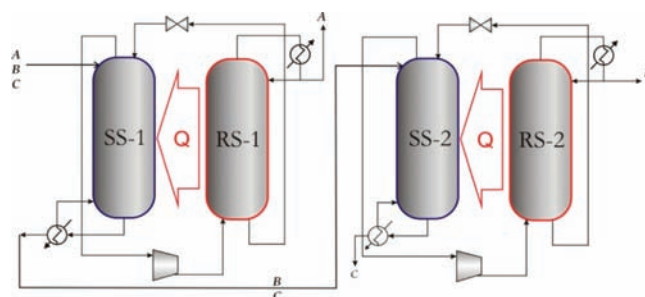


Figure 3. Direct distillation sequence with a HIDiC arrangement.

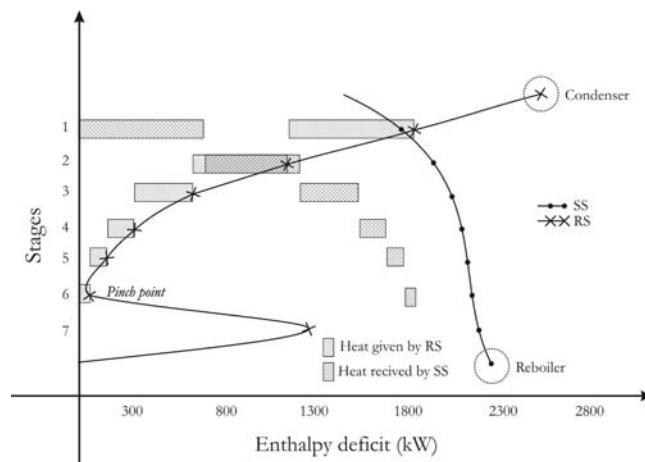


Figure 4. Column grand composite curve as a basis for HIDiC implementation.

2.1. Use of the Column Grand Composite Curve (CGCC). The original design of the HIDiC system is based on a binary separation problem. If one considers the separation of a ternary mixture and the use of only conventional columns, then two columns are required, giving rise to the direct (one by one as overhead products) and indirect (one by one as bottoms products) separation sequences. Each of the individual columns could be integrated to provide a sequence of two HIDiC systems, as shown in Figure 3 for a direct separation sequence.

Aided by the use of the CGCC, one can examine the column profiles to determine the maximum degree of energy integration between sections, which depends on the location of the pinch point. Because the pinch point is not necessarily associated with the feed tray (especially for multicomponent mixtures), limitations for heat integration between the two sections will arise.

A conventional CGCC gives the enthalpy deficit as a function of temperature. However, its representation as a function of the stage number is better suited for distillation applications, because one can visualize the potential for stage-by-stage energy integration. To illustrate this point, Figure 4 shows the results for one separation problem (in this case, it refers to column C1 with mixture M1 and feed F4 of the case studies presented in section 3, for an indirect distillation sequence), in which one can observe that the CGCC of the column sections is not symmetrical and the pinch point does not coincide with the feed stage. There is therefore a limit on the degree of energy integration that can be achieved; in this case, because of limitations on the heat available from the rectifying section, the stripping section cannot meet its energy requirements. This type of analysis is carried out for each

Table 1. Case Studies

mixture	components	ESI
M1	benzene/toluene/ <i>o</i> -xylene	~1 (0.95)
M2	butane/isopentane/pentane	>1 (1.92)
M3	isobutane/butane/pentane	<1 (0.5)

column of the case studies presented in this work. It should be noted that the number of stages required for the separation was calculated first so that the product purities could be achieved, and then the maximum degree of energy integration was detected through the use of the CGCC.

The analysis presented here takes into account the reboiler heat duty (Q_R) for each column of the conventional sequence, the heat duty for the HIDiC system, and the energy required by the compressor (W_s). Comparisons are carried out column by column and on an overall basis, with special attention to the effect of the work load required for the operation of the compressor.

2.2. Thermodynamic Efficiency, CO₂ Emissions, and Cost Analysis. In addition to the energy integration capabilities of HIDiC sequences, the effects of implementing these structures on the thermodynamic efficiency, CO₂ emissions, and economy of the separation systems are also considered. The thermodynamic efficiency can be calculated as

$$\eta = \frac{W_{\min}}{LW + W_{\min}}$$

where LW is the lost work, given by

$$LW = T_0 \Delta S_{\text{irrev}}$$

and W_{\min} is the minimum work, given by an availability balance for the system with lost work equal to zero

$$W_{\min} = \sum_{\text{out}} nb - \sum_{\text{in}} nb$$

where

$$b = h - T_0 s$$

CO₂ emissions were calculated as in Gadalla et al.,¹⁴ assuming the use of natural gas for the reboilers of the conventional sequences and taking into account emissions generated by reboilers and compressors for the HIDiC systems. The use of natural gas was also assumed to calculate the compressor emissions. For the economic analysis, equipment costs were estimated by Guthrie's method^{15,16} using chemical engineering plant cost indexes (CEPCIs) to update costs for 2008. A depreciation time of 10 years and 8600 operating hours per year were assumed.

3. CASE STUDIES

Three ternary hydrocarbon mixtures were considered (see Table 1). For each mixture, the four possible feed compositions listed in Table 2 were assumed. The feed flow rate was taken as 100 kmol/h as saturated liquid. The thermodynamic models were the universal quasichemical (UNIQUAC) model for M1 and the Chao–Seader model for M2 and M3. The mixtures were selected to have different values of the ease of separability index ($ESI = \alpha_{AB}/\alpha_{BC}$, where α_{ij} represents the relative volatility of components i and j and A, B, and C correspond to the light, intermediate, and heavy components, respectively)¹⁷ in order to analyze the effect of the relative difficulty of the split between the light and intermediate components compared to the split

Table 2. Feed Composition for Each Case Study

feed	composition ^a (kmol/h)
F1	33/34/33
F2	40/20/40
F3	30/40/30
F4	15/70/15

^a Values correspond to the order of components listed in Table 1.

between the intermediate and heavy components of the ternary mixture. The analysis based on ESI values presented here provides initial efforts for the establishment of heuristic rules for the implementation of HIDiC structures for the separation of ternary mixtures. Simulations were carried out with the Aspen Plus process simulator. Both direct and indirect separation sequences were considered. In Figure 3, which shows a direct sequence arrangement using HIDiC components, one can notice that, because nonideal (i.e., limited heat-integrated) HIDiC cases are being considered, both a reboiler and a condenser are included for each column of the sequence, in addition to the compressor.

4. RESULTS

Table 3 lists the percentage energy savings provided by the HIDiC sequences with respect to the conventional sequences, without considering the energy consumed by the compressor. Table 4 extends the comparison by including the energy due to compressor operation. The tables report individual savings for each column of the separation sequence, followed by the overall savings for the complete sequence.

It was found that, in most cases, one section limits the integration capabilities of the HIDiC system and the limiting section switches if one moves from a direct to an indirect sequence. Also, the energy savings achieved by one column of the HIDiC structure do not guarantee that the other column will also be energy-efficient. For instance, in Table 3 for the case of mixture M2 with feed F3 and an indirect sequence (IS), the savings provided by column C1 are positive (22%), but C2 shows negative savings (−41%); however, the global savings of 12% for this case reflect the fact that the total energy load for column C1 is higher than that for column C2. This initial analysis takes into account only the reboiler heat loads. When the compressor work is considered, the energy savings drop significantly, mainly for cases M2 and M3 (mixtures with ESI values that are not approximately equal to 1), making HIDiC structures even unsuitable for several of the case studies.

The results show that HIDiC structures are not always convenient choices to carry out a given separation task. Although apparent energy savings are generally observed if one compares the reboiler heat duties, when the energy due to the use of the compressor is taken into account, such a factor can exceed the initial energy savings provided by the HIDiC structure, yielding negative energy savings. The ease of separation index has an effect on the energy efficiency of the HIDiC system, as can be inferred from the results of Table 4. When ESI was approximately equal to 1, the HIDiC system always provided a more efficient energy sequence. This could be interpreted as systems with similar thermodynamic properties that help provide a better coupling between sections, which promotes a higher degree of energy integration. When ESI was higher than 1, the HIDiC structure was more energy-efficient than its direct-sequence counterpart for mixtures with equimolar or nearly equimolar

Table 3. Percentage Heat Duty Savings from the Implementation of HIDiCs

separation sequence	feed	mixture 1			mixture 2			mixture 3		
		ESI \approx 1			ESI > 1			ESI < 1		
		C1	C2	CG	C1	C2	CG	C1	C2	CG
DS	F1	49.29	19.20	35.27	43.08	34.85	38.39	24.76	11.68	21.38
	F2	20.54	42.42	31.69	12.16	25.82	18.18	18.31	-30.55	5.18
	F3	43.11	48.11	45.44	47.23	34.30	39.51	18.56	23.38	19.84
	F4	44.94	46.33	45.52	-24.94	51.21	28.26	57.88	41.66	54.87
IS	F1	86.22	53.48	80.94	-36.91	37.64	-25.40	15.36	38.89	31.30
	F2	32.66	79.14	56.85	71.63	-36.00	67.42	4.39	44.30	29.66
	F3	76.14	47.53	65.58	22.75	-42.22	12.18	22.03	26.08	24.65
	F4	48.51	33.79	43.68	72.20	-2.28	65.34	0.80	5.53	3.80

Table 4. Percentage Energy Savings Including the Compressor Work for HIDiCs

separation sequence	feed	mixture 1			mixture 2			mixture 3		
		ESI \approx 1			ESI > 1			ESI < 1		
		C1	C2	CG	C1	C2	CG	C1	C2	CG
DS	F1	22.09	-22.07	1.51	2.11	13.62	8.67	9.96	-50.89	-5.73
	F2	-9.02	16.89	4.19	-47.62	3.31	-25.17	-54.31	-93.70	-76.35
	F3	14.80	19.26	16.88	14.02	11.91	12.76	-24.04	-37.64	-27.65
	F4	12.28	23.43	16.91	-107.28	32.28	-9.78	23.68	4.60	20.14
IS	F1	67.31	22.64	60.10	-96.98	-4.74	-82.73	-63.73	13.60	-11.33
	F2	-7.04	56.63	26.80	48.05	-99.78	42.26	-93.70	20.31	-21.50
	F3	47.87	11.71	34.52	-26.05	-103.8	-38.71	-53.63	-3.06	-20.84
	F4	15.57	-3.26	9.39	50.05	-60.42	39.88	-116.9	-28.83	-61.03

Table 5. Comparison of Thermodynamic Efficiencies (%) for Conventional and HIDiC Systems

feed	sequence	M1		M2		M3	
		conventional	HIDiC	conventional	HIDiC	conventional	HIDiC
F1	DS	10.65	17.03	4.98	6.20	11.99	11.96
	IS	4.19	11.03	5.95	3.62	12.61	11.04
F2	DS	13.52	20.28	4.36	4.13	12.23	9.03
	IS	11.61	16.94	1.62	3.86	13.01	9.55
F3	DS	10.05	18.27	4.88	6.38	11.03	11.42
	IS	8.61	15.04	5.20	4.56	12.33	8.81
F4	DS	5.40	14.41	3.59	4.18	5.59	11.49
	IS	5.57	6.99	2.27	6.43	7.06	4.02

feed composition; for other types of feed mixtures, the indirect HIDiC structure was more efficient when compared to the conventional indirect sequence. When ESI was lower than 1, HIDiC structures were less energy-efficient than the conventional sequences for almost all of the cases. As a consequence of these results, one can conclude that systems with high energy demands to carry out the separation would be good for HIDiC implementation, because the savings in reboiler duties in those cases are more likely to be higher than the work load required for the operation of the compressor.

Because each individual split of the ternary mixture can be done through a conventional or integrated arrangement, one can detect possible combinations to yield hybrid structures that would be energy-efficient. For instance, from the results of Table 4, if one

considers mixture M1 with feed F1 and use of a direct sequence structure, then column C1 should be integrated in the form of a HIDiC structure and column C2 should be a conventional column, whereas for mixture M1 but with feed F2, the first column should be a conventional column and the second one should be a HIDiC system. In each case, the most energy-efficient individual choice is selected to produce the best possible hybrid arrangement of HIDiCs and conventional distillation columns.

Additional aspects of the energy efficiency of HIDiC sequences were also addressed. Table 5 shows a comparison of thermodynamic efficiencies between HIDiC and conventional distillation sequences. For practically all cases, HIDiC structures improve this factor, which is expected because a HIDiC system can be viewed as a distillation tower with a heat pump.

Table 6. Percentage Reductions in CO₂ Emissions from the Use of HIDiC

feed	sequence	M1		M2		M3	
		with comp	without comp	with comp	without comp	with comp	without comp
F1	DS	-2.57	34.51	-10.41	28.60	-49.50	21.30
	IS	51.33	74.09	-101.3	-22.48	-33.57	24.37
F2	DS	3.39	35.83	-43.97	12.74	-109.2	5.18
	IS	9.15	45.96	39.66	72.49	-51.02	18.94
F3	DS	7.28	39.99	-5.58	29.25	-44.62	-9.50
	IS	31.06	63.33	-47.21	19.37	-52.46	9.81
F4	DS	12.51	45.52	-36.74	12.94	7.42	54.87
	IS	5.17	43.68	24.14	57.37	-89.33	0.49

Table 7. Percentage Reductions in Total Annual Cost from the Implementation of HIDiCs

feed	sequence	column	mixture 1	mixture 2	mixture 3
F1	DS	C1	4.11	-36.79	-191.4
		C2	-13.64	-47.53	-13.55
	IS	C1	57.67	-162.2	-107.8
		C2	-19.96	-1.03	-144.7
F2	DS	C1	-17.63	-96.04	-315.2
		C2	-10.79	-49.86	-147.2
	IS	C1	-43.35	-492.5	-126.6
		C2	-0.38	-54.60	-152.4
F3	DS	C1	-9.00	-18.60	-193.1
		C2	-12.08	-52.38	-92.18
	IS	C1	-16.70	-706.7	-96.16
		C2	10.17	-106.9	-155.4
F4	DS	C1	-39.87	-224.4	-54.56
		C2	0.15	-30.14	5.36
	IS	C1	-38.05	-41.65	-280.5
		C2	10.22	-58.40	-176.3

Greenhouse-gas emissions are expected to be lowered through the use of integrated distillation sequences such as the HIDiC system. The effects of HIDiC implementation with respect to conventional direct and indirect distillation sequences on CO₂ emissions are reported in Table 6. It can be seen that savings in CO₂ emissions are not always provided by the use of HIDiC structures and that the results are clearly affected by the ESI value. When ESI was approximately equal to 1, the HIDiC structure turned out to reduce CO₂ emissions for almost all of the cases. However, for mixtures with ESI values far from 1, the use of the compressor adversely affects the potential advantages of the HIDiC system, making this structure less environmentally friendly than the conventional distillation sequences. Therefore, ternary mixtures with equal difficulties in their A/B and B/C splits provide the best incentive for HIDiC implementation, from both energy and environmental perspectives.

To complement the study, an economic analysis in terms of total annual cost estimations was conducted, with the results given in Table 7. In most cases, the compressor cost exceeded the savings in reboiler duties, to yield higher TAC values for HIDiC structures than for the conventional sequences. Hybrid structures based on minimum TAC values can again be identified. For instance, for mixture 1 and feed F1 with either direct or indirect sequence, the first column should be implemented as a HIDiC

system, whereas the second one should be implemented as a conventional column, with the indirect structure giving higher economic potential. Overall, the economic incentive for HIDiC implementation seems to be even more restrictive than that based on energy or environmental aspects.

5. CONCLUDING REMARKS

A method for the design of HIDiC systems has been presented. The method uses the column grand composite curve as a basis. The CGCC directly provides the thermodynamic capabilities for energy integration for HIDiC systems. As a consequence, one can check the deviation from the zero theoretical heat load for reboilers of an ideal HIDiC for a given application. This methodology was applied to a series of problems for the separation of ternary mixtures. A relevant result from our analysis is that savings that are observed between the reboiler loads of HIDiC and conventional sequences can be overcome by the work required by the compressor. The addition of the compressor also affects CO₂ emissions, to make some of the HIDiC sequences unfavorable from an environmental perspective. Based on the characterization of mixtures through the ease of separability index, it was observed that the best ternary mixtures for HIDiC implementation were those in which the separation between the light and intermediate components is equally as difficult as that between the intermediate and heavy components. The use of HIDiC structures therefore seems to be restricted to special cases. Another line of potential application has been shown here, considering the implementation of hybrid systems of HIDiCs and conventional distillation columns: we have shown that for several ternary separation problems, there exist such combinations that provide energy-efficient and environmentally friendly separation sequences.

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■ NOMENCLATURE

b = thermodynamic availability

h = enthalpy

LW = lost work

s = entropy

T_0 = temperature of the surroundings

W = work

Greek Letters

η = thermodynamic efficiency

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