Chemical Engineering Journal 89 (2002) 155–172

Chemical Engineering Journal

www.elsevier.com/locate/cej

A simple method for systematic synthesis of thermally integrated distillation sequences

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Accepted 30 January 2002

Abstract

Thermodynamic approach to heat integration of processes involving separation of multicomponent mixtures is described. An integrability criterion based on geometrical presentation of distillation columns in temperature–enthalpy flow rate diagram was tested by rigorous simulation of distillation columns. The integrability criterion for distillation columns sequence was defined as the minimum sum of products of intracolumn temperatures and their heat flow rates (Min-criterion). Distillation columns sequences were studied according to their profitability in heat integration. Energy-pinch analysis was used for the synthesis of the best heat integrated distillation system. The representation using extended grand composite curve was demonstrated to be a very convenient one for understanding and evaluating the heat integration. The selection of possible distillation columns sequences using rising values of the integrability criterion gave a very good approximation towards the best sequence of heat integrated distillation columns when compared to their total annual cost. Their estimation using rigorous models of distillation columns was shown to be better than using short cut models. The method allows alternative distillation sequences to be evaluated using rigorous calculations, prior to column design. The method is demonstrated with three five- and six-components example problems.

Keywords: Process synthesis; Separation systems; Integrability criterion; Distillation columns sequence; Heat integration; Span of temperatures

1. Introduction

Systematic synthesis of multicomponent separation sequences is an important process design problem in chemical industry. It is concerned with the selection of a separation method and the selection of the best sequence of separators to split a multicomponent mixture into several products of relatively pure species.

For solving separation problems in chemical industry distillation columns are widely employed separators. Their advantage is simple operation and ability to separate various kinds of mixtures into their components. Distillation sequences can be specified by using different methods: heuristic, evolutionary or algorithmic [1,2].

In the past many researchers were engaged with the goal to determine the best sequence of heat integrated distillation columns. Different methods were developed for a simple and quick selection of optimal heat integrated distillation systems. Andrecovich and Westerberg [3,4] based their methods on the assumption that the product of the column condenser or reboiler heat flow rate and the temperature difference between its reboiler and condenser, $\Phi\,\Delta T$, was constant for a single distillation task over a wide range of pressures. Our research [5] showed that in many cases these quantities were not constant in the whole range of operating pressures.

Thermodynamically oriented procedures are commonly applied to a given separation scheme. There are many ways for reducing utility needs which can be considered simultaneously. Umeda et al. [6] used thermodynamic analysis to synthesise energy-integrated distillation systems. They proposed a procedure using a Carnot efficiency vs. heat flow rate diagram ($(T - T_0)/T$ vs. Φ), where the enclosed area between the composite lines for the heat sources and sinks was a measure of the exergy (available energy) losses in the heat exchange subsystem [6,7]. Exergy loses studied by these authors were taken as a good starting point for our approach. Presenting exergy losses in a simplified temperature–enthalpy flow rate (*T*–*I*) diagram it deals with energy savings obtained by heat integration of distillation columns [5].

Many mathematical programming approaches to the synthesis problem are described in the literature. They are usu-

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PII: S1385-8947(02)00012-8

Nomenclature

A	heat exchanger area (m^2)
A_i	utility area (m^2)
C_{ES}	coefficient of ease of separation, defined by Eq. (5) (–)
C_{TA}	total annualised cost (USD/year)
C_U	total annual utility cost (USD/(W year))
D_C	depreciation cost (USD/year)
F_B	amount flow rate of bottoms product (mol/s)
F_D	amount flow rate of overhead (distillate) product (mol/s)
$F_{I,Min}$	integrability criterion (W K)
H	enthalpy (J)
ΔH_m	molar enthalpies difference (J/mol)
I	enthalpy flow rate (W)
ΔI	enthalpy flow rate difference (W)
k	overall heat transfer coefficient (W/(m^2 K))
r	distillate to bottom amount flow rate ratio, F_D/F_B or F_B/F_D (–)
R_{Min}	minimum reflux ratio (mol/mol)
S_T	total savings (USD)
T	absolute temperature of a stream (K)
T_B	boiling point temperature (°C)
T_P	pinch temperature (°C)
T_S	supply temperature (°C)
T_T	target temperature (°C)
T_0	absolute temperature of the atmosphere (K)
ΔT	temperature difference (K)
ΔT_a	intracolumn temperature difference between the condenser and the reboiler (K)
ΔT_e	intercolumn temperature difference between the condenser and the reboiler (K)
ΔT_{Min}	minimum approach temperature (K)
x_i	feed amount fraction (–)

Greek symbols

α	relative volatility of key component to be separated (–)
Δ_i	ΔT or $(\alpha - 1) \times 100$ (K or –)
Φ_a	intracolumn heat flow rate, W
Φ_C	condenser heat flow rate (W)
Φ_{CU}	cold utility heat flow rate (W)
Φ_e	intercolumn heat flow rate, W
Φ_{HU}	hot utility heat flow rate (W)
Φ_R	reboiler heat flow rate (W)

ally supported by rigorous mathematical algorithms. Kattan and Douglas [1] used the simultaneous optimisation of the overall flow sheet combinating it with the method of maximum energy recovery. Some authors [4] solved the problem of distillation sequence synthesis and heat integration by solving a mixed integer linear optimisation problem of the superstructure while others developed another algorithmic approach [6–8]. All these mathematically based approaches give good approximations for promising sequences but demand strong mathematical background, the problem being the superstructure selection.

Heuristic methods use simple empirical rules, which often raise difficulties when the heuristics are in conflict with each other. Having applied the method to find the best non-integrated sequence, the designer would then heat integrate it. The total problem is not solved simultaneously but in two steps. Yet, it does not always give the optimal solution [5]. Evolutionary methods also require strong mathematical background when searching for promising separation systems with evolutionary improvements and evolutionary strategy [1,7].

Drawback of all the methods mentioned is that they either demand strong mathematical background and have no elimination criteria for non-promising candidates or they do not lead to the best heat integrated solutions. A thermodynamic procedure to find the best heat integrated sequence will be represented here, along with examples that illustrate its efficiency in finding the optimal heat integrated distillation systems using rigorous models for distillation columns. The analysis was oriented towards reducing the number of promising schemes to a minimum. The problems studied are too difficult for mathematical analysis to be solved without simplification. The approach was first developed for a system of isolated distillation columns, and then applied to a distillation column train being a part of a chemical process.

2. Heat integration

Distillation columns are substantial energy consumers. Their energy requirements can be considerably reduced when applying heat integration. This can cause significant differences between the best heat integrated and non-integrated sequences [5,9]. Because of lower utilities consumption, heat integrated sequences are cheaper than the non-integrated ones. In order to achieve energy conservation, it is important to establish a systematic method for reducing external energy inputs by an effective exchange of heat flows in distillation trains. Therefore, heat integration is the most decisive factor for finding the best sequence of distillation columns. Effective heat integration needed a better criterion for ranking distillation columns sequences based on thermodynamic properties of the components which had to be separated in the distillation columns sequence and giving a ranking similar to the one established by the total annual costs. According to our previous research [5,9,10], we supposed that the best representation of distillation columns was an extended grand composite curve (EGCC), applied in the temperature–enthalpy flow rate difference (T–ΔI) diagram. For simplicity, it is sometimes assumed that reboiler and condenser heat flow rates are almost the same and the distillation columns are presented with Φ–ΔT rectangles. Actually, distillation columns in the T–ΔI diagrams are usually trapezoids.

The main target in process integration is to make use of every available hot or cold stream match in order to substitute utilities. In the case of multicomponent separation, distillation columns contribute a significant portion of heat flow rates with their condensers and reboilers. The more columns are interconnected into a distillation train heat flow cascade, the higher is the profit of heat integration. Heat is transferred from a condenser of a distillation column with higher temperature to a reboiler of another column or to a process stream with a lower temperature or from a process stream with a higher temperature to a reboiler. The greatest benefit is to make the highest possible extent of condenser–reboiler matches, and process to column heat exchanges.

In process design the distillation column sequence often determines the main part of the process costs with regard to the total annual costs. The most decisive factors for the economics of the whole process are shown in Fig. 1. Here, two important variables should be considered: reboiler/condenser heat flow rate and temperature difference between the reboiler and the condenser in the same column and/or between two columns. Usually, the greatest part of total annual costs are contributed by the hot or cold utilities cost. These are determined first by the type and flow rate of both utilities. The type is fixed by the cold/hot stream temperature or temperature of the available utility, respectively. The second part is the depreciation cost of the equipment. It is determined by the so-called hot or cold utility area (hatched in Fig. 1). Lower heat flow rates result in smaller utility consumption and better flexibility for heat integration. The higher the temperature difference is between the utility and the condenser, and/or the utility and the reboiler, the smaller the condenser and/or the reboiler area. It is recommended to keep the temperature difference between the utilities and the condenser or the reboiler as high as possible. The highest possible temperature differences are desired with the use of the same available utilities. In each heat exchanger network it is therefore recommended to keep the area between the heat integrated columns (intercolumn area) as large as possible. Lower temperature difference inside the column yields better possibilities for heat integration and also larger intercolumn area as shown by the columns D1, D2 and D3 in Fig. 1.

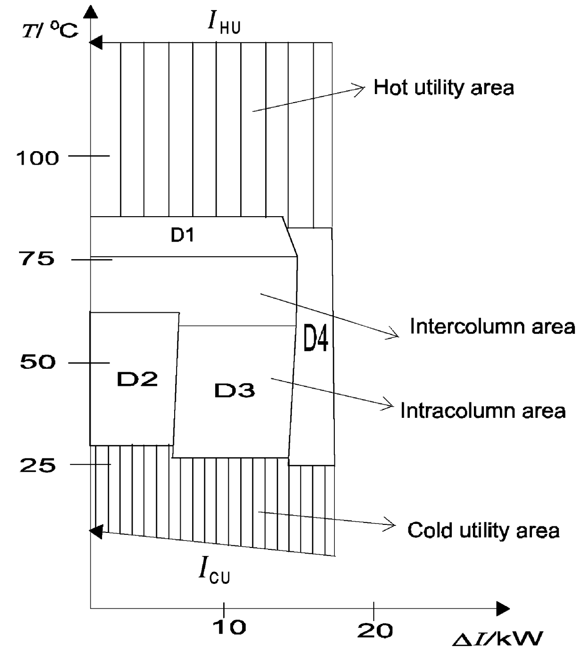

Fig. 1. Integrated columns system and its utility demand.

The goal of heat integration is to minimise total utility consumption and total annual cost of the process. Each heat

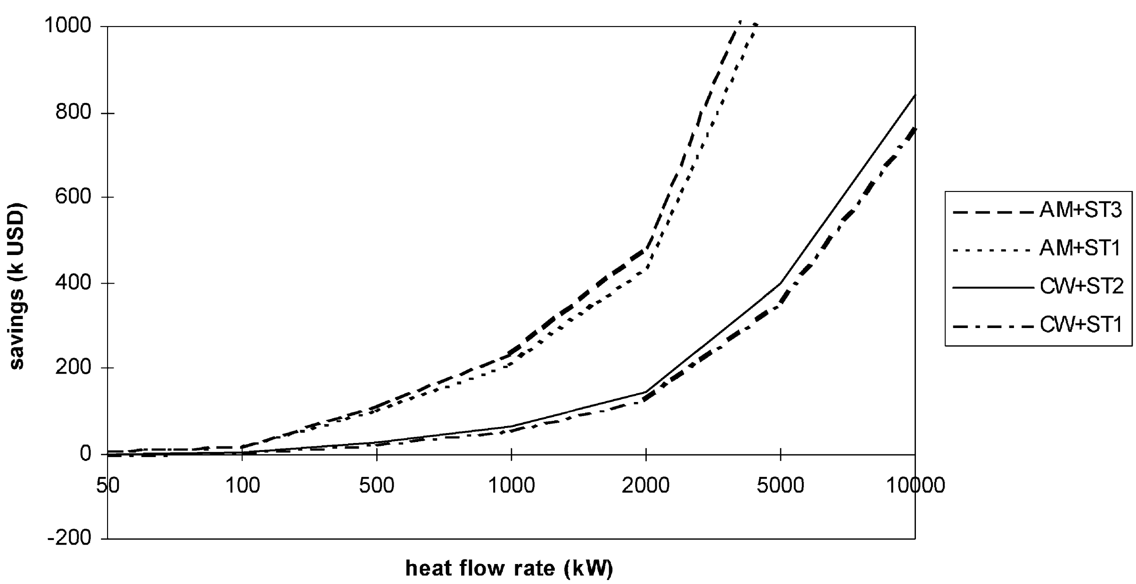

Fig. 2. Influence of different utilities usage on total savings.

integrated (distillation) unit presents a saving which reduces both the hot and the cold utilities. Additional intercolumn heat exchanger area results in savings of both utilities. As the investment in equipment increases, the utilities usage decreases. The difference between the utilities savings and the annualised depreciation cost represents the total savings resulting from the heat integration. Searching for the dependence between the utilities savings and integrated heat flow rates (Fig. 2), a fixed starting temperature difference of 10 K was taken in the heat exchangers. The 20 % of the installed costs were considered to be the annual depreciation costs of equipment. These were calculated according to the literature described later in the paper. Fig. 2 shows the increased savings of operating costs and depreciation with the increased integrated heat flow rate (Φ) from 50 kW to 10 MW in general. Five different utilities were taken for calculations. Two lower curves represent combinations of cheap hot and cold utilities, whereas the other two use more expensive ones. The CW + ST1 curve is a combination of cooling water and low pressure steam (ST1), the CW + ST2 curve of cooling water and medium-pressure steam (ST2). The two upper curves use ammonia (AM) instead of cooling water. AM + ST3 represents the combination of ammonia and the high pressure steam. Therefore, savings are higher in the latter cases and the resulting curves have steeper slopes. Fig. 2 indicates that utilities savings are much higher than the additional depreciation cost of the heat exchangers. The total savings are increasing exponentially with the integrated heat flow rate.

This paper improves a thermodynamic formulation of the heat integrated distillation train synthesis problem, where the separation as well as the heating and cooling task constraints are taken into consideration in a rigorous way. The available utilities for the separation tasks are restricted when their temperatures are not always optimal for the available heating/cooling streams. The aim of the paper is to introduce an improved Min-criterion, which is supposed to supersede the existing ones. The so-called Max-criterion [5] is used for the comparison to test if the new one is actually better.

3. Integrability criterion

The purpose of heat integration is to match hot and cold streams of units in a heat cascade. Our goal was to improve an effective criterion for finding and determining the best sequence of heat integrated distillation columns. In order to handle the minimum possible number of candidates for promising schemes, sequences with less promising possibilities for heat integration should be eliminated. The integrability criterion should measure the ability for heat integration rigorously.

Cost of the intercolumn heat exchanger increases with the increase of heat exchanger area. The larger the area is between the integrated columns, the higher the saving due to heat integration [5]. The situation in the T–ΔI diagram is shown in Fig. 3. Increasing the area between the condenser

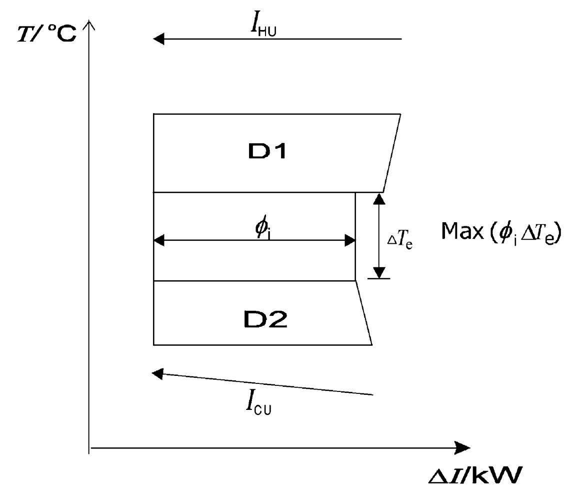

Fig. 3. $\Phi_i\,\Delta T_e$ value criterion in T–I diagram.

of the column D1 and the reboiler of the column D2 increases heat integration effectiveness.

Considering Figs. 1 and 3, it was stated that the maximisation of intercolumn areas, Max $\Phi_i\,\Delta T_e$, was a measure for heat integration [9,10]. On one side effective heat integration requires integration of large heat flow rates with maximum possible intercolumn temperature difference. On the other side this results in shrinking of hot and cold utility areas (A_i) which are mainly a function of reboiler/condenser heat flow rates (Eq. (1)). Here, Φ_i represents a heat flow rate of the appropriate condenser or reboiler and ΔT_e their temperature difference with regard to the appropriate utility. Intercolumn temperature differences are marked by subscript e (ΔT_e). Based on these conclusions the Max-criterion was developed [5,9] (Eq. (2) and Fig. 3):

$$A_i = f\left(\Phi_i, \frac{1}{\Delta T_e}\right) \tag{1}$$

$$F_{Max} = \mathrm{Max}\sum_i(\Phi_i\,\Delta T_{e,i}) \tag{2}$$

Our research showed the importance of thermodynamic properties of a multicomponent feed mixture, that is to be separated into pure components. These properties dictate the geometry of distillation columns in the T–ΔI diagram and the utilities required for the desired separations. In chemical processes other hot and cold streams exist besides those of the distillation columns allowing integration of reboiler or condenser streams with them.

We proceeded our research towards finding an improved criterion which would show the ability of particular distillation columns sequence for heat integration. As an effective aid a minimum integrability criterion ($F_{I,Min}$) was introduced [5] and is now refined. It is defined as a product of temperature difference between the reboiler and the condenser in the same column (intracolumn temperature difference, ΔT_a) and its average heat flow rate (Eqs. (3) and (4) and Fig. 4). In the case of several distillation columns the integrability

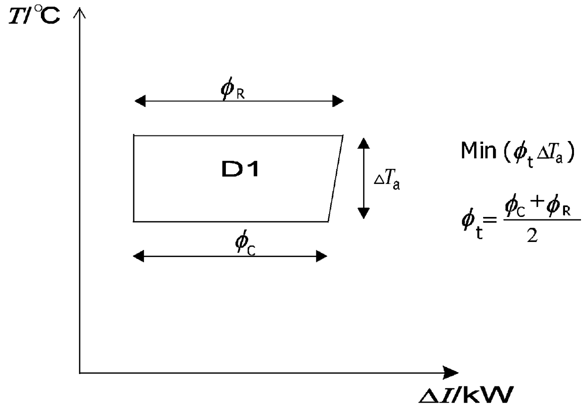

Fig. 4. Integrability criterion in T–I diagram.

criterion was defined as a sum of individual intracolumn $\Phi_t \, \Delta T_a$ products. Actually, both utility areas (hot and cold) are decisive for heat integration. Therefore, in the integrability criterion both contributions are equally represented:

$$F_{I,\mathrm{Min}} = \mathrm{Min} \sum_i (\Phi_t \, \Delta T_{a,i}) \tag{3}$$

$$\Phi_t = \tfrac{1}{2}(\Phi_C + \Phi_R) \tag{4}$$

The smaller the intracolumn temperature difference (ΔT_a) and the average intracolumn heat flow rate (Φ) in the T–ΔI area, the smaller F_I will be. Additionally, distillation columns with small intracolumn areas have better ability for heat integration with other distillation columns or with the process, yielding large intercolumn areas and increasing utilities saving.

Integrability criterion represents the minimum sum of intracolumn areas in the T–ΔI diagram (see Fig. 5). The objective is to reduce the Φ_i and the span of temperatures of the column trapezoids. The latter will increase the possibility of heat recovery between columns and, thus, reduce the overall utility demand. Also, lower heat exchanger area will result in reduction of its depreciation cost.

Ability for heat integration depends on many parameters. Smaller condenser and reboiler heat flow rates result in lower utilities consumption. Distillation columns with a small span of temperatures give large ΔT's between sources and sinks in the match and have good possibilities for heat integration. Therefore, the area between the integrated distillation columns is simultaneously maximised.

Fig. 5 shows distillation columns D1 and D2 fully integrated with column D3. The intercolumn area is bigger in case (a) than in case (b). Yet, the comparison shows that the case (a) demands more external utilities than the case (b) ($\Phi_{HU\text{-}1} + \Phi_{CU\text{-}1} > \Phi_{HU\text{-}2} + \Phi_{CU\text{-}2}$). Case (a) has larger heat exchange and the result is larger intercolumn area. On the other side, distillation columns in case (b) have greater ability for heat integration. Their intracolumn areas are smaller than in case (a). This results in lower total annual cost, mainly caused by lower utility consumption.

The size of the column trapezoids varies with the composition, heats of vaporisation and heat flow rates in the condenser/reboiler and with boiling temperature differences between key components which have to be separated. Most of these properties strongly depend on operating pressures.

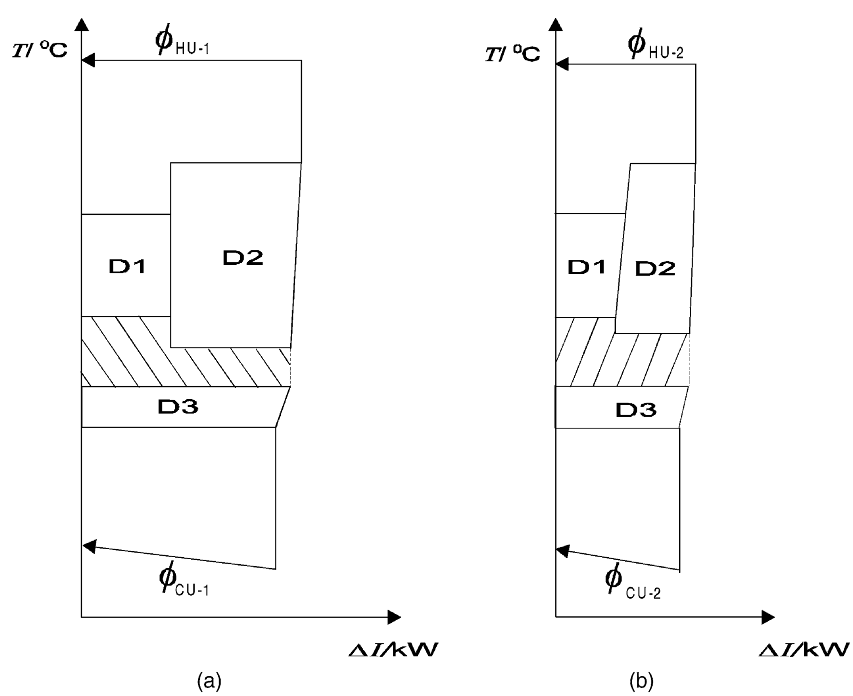

Fig. 5. Comparison of the criteria developed: (a) lower integrability; (b) higher integrability.

In most cases with raising pressure the values of these properties also increase. There are also cases where these increases are not strongly uniform. Therefore, separate splits (separations) should be studied carefully. The selection of pressure mainly depends on the components which need to be separated. In the case of more difficult separations the boiling temperature difference between the light- and the heavy-key component is small, but condenser and reboiler heat flow rates are large.

For further discussion the $\sum \Phi_t \Delta T_a$ criterion was named Min-criterion, whereas the $\sum \Phi_i \Delta T_e$ criterion was named Max-criterion [10]. Min-criterion comprises four fundamental elements which have critical influence on separation costs: external utility consumption, ability for heat integration, utilities saving and HEN + column investment. Min-criterion actually involves the Max-one. Min-criterion has maximal temperature differences between the integrated columns or the columns and utilities as the consequence. The result is smaller heat exchanger area and lower investment. The Max-criterion does not involve the important criterion of minimum external utility consumption, but it was found to be convenient in the case of isolated distillation columns. These columns were studied separately, without any other process unit. Our previous research with short cut models showed that the Min-criterion was better than the Max-one. In this work we wanted to upgrade the proposed short cut method of minimisation of intracolumn areas sum described above to determine the ability for heat integration including other process streams. The improved rigorous simulation method was hoped to be a better one for intensive heat integration.

4. Design method

An improved rigorous synthesis method for finding the best structure of a heat integrated distillation system has been developed. In all case studies only distillation is used as a separation method. The procedure was applied to an isolated distillation columns system first (case study 1) and then together with the rest of the process units, the so-called process system where other process units taking part in heat integration are present (case studies 2 and 3). EGCC was used to find possible integrations between distillation columns of a train and the background process system. The process design method can be described with the following procedure:

1. Analyse thermal structure of the process.
2. Identify all possible splits and determine possible sequences.
3. Analyse each possible split according to its $\Phi_t \Delta T_a$ value (short cut models).
4. Select sequences with the lowest $F_{I,\mathrm{Min}}$ values.
5. Analyse the best sequences economically considering heat integration.
6. Select the most promising schemes and resimulate them with rigorous models.
7. Determine the best scheme of the process.

Three different processes were studied with the procedure proposed. In the first step the thermal structure of the process was analysed (in the case of a system with isolated distillation columns system this step was left out). The thermal structure of the process comprises all process units in which thermal changes occur (heaters, coolers, distillation columns, etc.). The process was separated into a fixed and a variable part. The fixed one refers to the part of the process which is not exposed to thermal changes (heat flow rates and temperatures of its streams do not vary). Heat flow rates and temperatures of heaters and coolers were held constant because of the nature of the process. This part of the process is usually not physically separated from the variable one. In the variable part thermal changes can occur (T_i and Φ_t vary). Further analyses are referred to the variable part only.

In the second step all the possible splits were identified (case studies 2 and 3). Then the splits identified were simulated by using a range of operating pressures, followed by the determination of possible sequences. For each possible split its individual $\Phi_t \Delta T_a$ value was calculated (step 3) using short cut design calculations for each column in a proposed sequence. The $F_{I,\mathrm{Min}}$ values were calculated for the optimum pressure (step 4). The promising sequences were recalculated using the rigorous simulation of distillation columns. Here, possible heat integration of distillation columns, mutual and with the process background was carried out (step 5) obeying rules of pinch method. The whole process flow sheet was simulated for candidate sequences using rigorous models (step 6). With the results obtained, the most promising sequence was selected (step 7).

The pressure is determined by the operating conditions in the process. All the splits should be studied in the range of boundary values of maximum and minimum pressure allowed. Between these ranges the $\Phi_t \Delta T_a$ values are calculated for separate splits.

5. Design procedure

Design procedure described here was used in all the three case studies. Process schemes were taken from the literature and modifications with regard to sequences of distillation columns were carried out. The base flow sheets consisted no heat integrated units.

The ASPEN PLUS process simulator was applied for the simulation of all the case studies. The ASPEN modules DSTWU (short cut) and RADFRAC (rigorous) were used for the simulation of distillation columns sequences. The first one used the short cut model, the latter performed rigorous design calculations for a single-feed, two-product distillation column with a total condenser. It assumed constant amount overflow and constant relative volatilities. Our preliminary

Table 1
Annualised utility cost [9,10]

Utility	Utility cost, C_U (USD/W a)
Chilled water, 5 °C	20.1
Cooling water, 20 °C	5.0
Hot water, 80 °C	60.0
Steam, 138 °C	76.3
Steam, 160 °C	84.4
Steam, 180 °C	94.2
Steam, 200 °C	100.6

investigation [5,9] showed that rigorous models gave very similar results, at most 3–4 % different from the short cut ones but the ranking of the promising sequences differed in case study 1. Therefore, rigorous RADFRAC module was applied for the detailed total costs estimation.

The economics of different sequences was estimated using techniques described in literature [6,7,11,12]. The annual operating time in processes was 8500 h in all the case studies. The cost of equipment was calculated by Guthrie's method [13] using ASPEN PLUS process simulator and 20 % of the installed cost was considered to be the annual depreciation cost of equipment. Prices used for utilities cost estimation are shown in Table 1.

Heat integration was carried out by the use of SuperTarget from Linnhoff March. It was also used for the short cut costs estimation of candidates for promising process schemes in all the three case studies. Obeying the procedure described, hot and cold streams were identified first. Streams of distillation columns were excluded and EGCC was constructed from the GCC of the distillation columns streams and the process ones. Therefore, classical pinch analysis was not applied. Distillation columns represented by trapezoids were graphically adjusted with each other and with the process background EGCC.

Heat flow rate, Φ, can be used when mutually integrating two columns but cannot be used with compressors and chemical reactors—enthalpy flow rate has to be chosen in the latter case. One has to differentiate enthalpy, H (in J), in batch processes from the enthalpy flow rate, I (in W), in continuous ones. Therefore, universal GCC uses $T/\Delta I$ diagrams for the pinch analysis. Enthalpy flow rate can be calculated from difference of molar enthalpies, ΔH_m (in J/mol) and amount flow rate, F (in mol/s): $\Delta I = \Delta H_m F$ [14].

Table 2
Feed compositions and properties for case study 1

	Components	Feed amount fraction, x_i	T_B (°C)	α	C_{ES} [a]
A	Propane	0.05	−42	2.00	5.26
B	*i*-Butane	0.15	−12	1.33	8.25
C	*n*-Butane	0.25	−0.5	2.40	114.55
D	*i*-Pentane	0.20	29	1.25	13.46
E	*n*-Pentane	0.35	36	1.00	–

[a] The coefficients of ease of separation [15]. It is defined as:

$$C_{ES} = r\Delta_i \quad (5)$$

where r is the ratio of the amount of flow rates of products (distillate and bottoms), F_B/F_D or F_D/F_B, depending on which of the two is smaller than or equal to unity; and Δ_i is either a boiling point difference between two components to be separated, $\Delta_i = \Delta T$ or $\Delta_i = (\alpha - 1) \times 100$, with α being the relative volatility or separation factor of the two components to be separated [15]. The second option was taken for calculations shown in Table 1. According to source [15] relative volatilities were calculated at the temperature of 37.8 °C and pressure of 1.72 MPa. Volatilities are relative to the next heaviest component.

5.1. Case study 1

The method presented is applied to one simple and two complex case studies. The first one (*case study 1*) is a separation problem of a five components hydrocarbon mixture using isolated distillation columns [1,15,16]. Feed flow rate is 252 mol/s. This is a standard problem, also known as Rathore's problem which is generally used to test separation synthesis algorithms. The mixture with the composition shown in Table 2 is to be separated into pure products. The feed stream contains five light alkane components, available as a saturated liquid at atmospheric pressure. The ASPEN property option for non-polar or mildly polar mixtures (SYSOP5) was chosen.

The most promising heat integrated sequences for case study 1 will be presented in Table 3. The numbering of sequences is taken according to Fig. 6 where all the sequences possible are shown.

The task is to develop a process which would separate the mixture into pure components giving the lowest total annual cost. To simplify the calculations, the following assumptions were used for the case study (and also for case studies 2 and 3, described later) [1,17]:

1. Minimum approach temperature (ΔT_{Min}) is held constant at 10 K.

Table 3
Comparison of total annual cost ranking in case study 1 with different methods

Number	Sequence	C_{TA} (MUSD)	C_{TA} [5] (MUSD)	C_{TA} [1] (MUSD)	C_{TA} [15] (MUSD)	Max $\sum(\Phi_i\,\Delta T_e)$ (MW K)	Min $\sum(\Phi_t\,\Delta T_a)$ (kW K)
1	SEQ-2	2.563	2.289	2.517	0.864	160.1	719
2	SEQ-1	2.678	2.324	2.549	0.858	150.1	721
3	SEQ-3	2.695	2.304	2.539	0.871	127.2	932

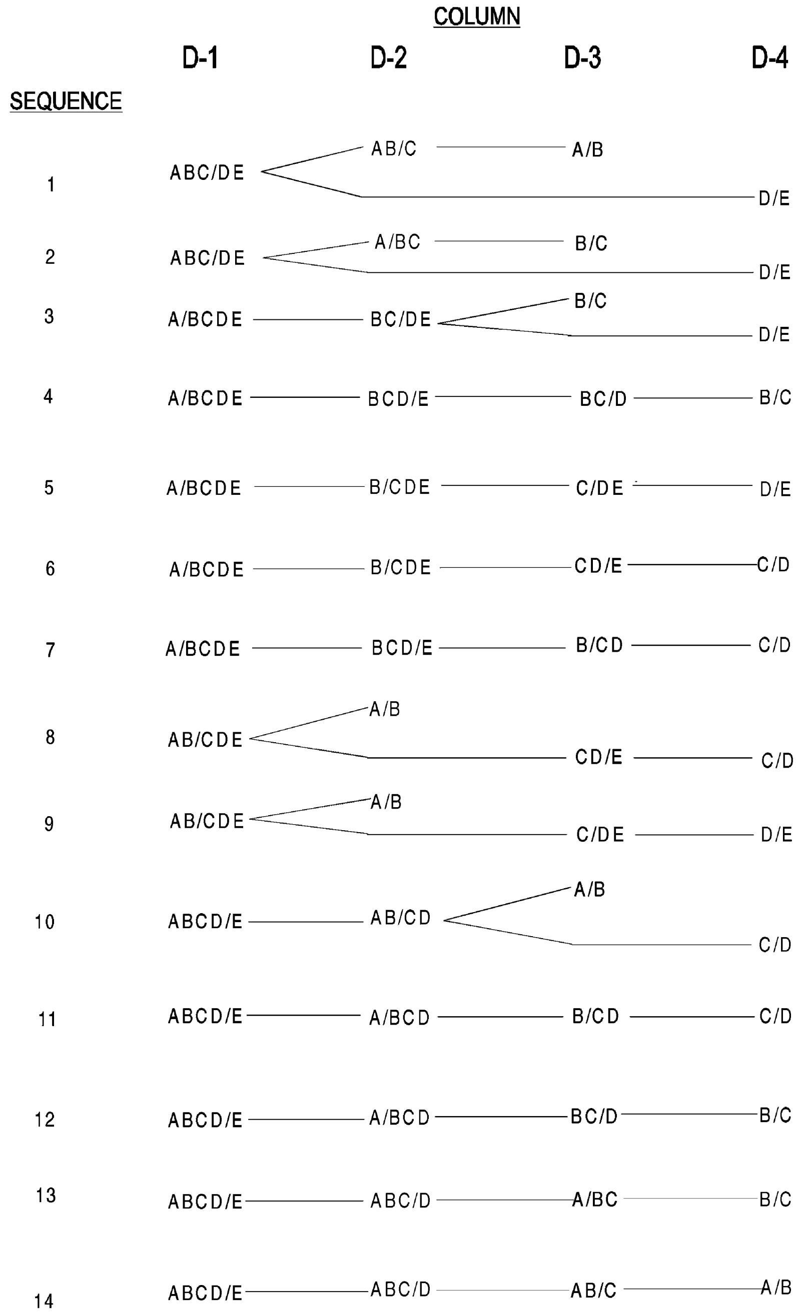

Fig. 6. Sequences studied in case study 1.

2. Column feed and product streams are saturated liquids.
3. Vapour recompression, inter-reboilers and inter-condensers are not allowed.
4. Each distillation column operates at 98 % recovery of the key components.
5. The cost of changing the temperature and pressure of the intermediate streams is negligible compared to the rest of the sequence cost.
6. The reflux ratio (R_B) in distillation columns is held constant at the value of $1.2R_{Min}$.
7. Only total condensers and total reboilers are used.
8. Heat transfer coefficients are held constant at 568 W/(m^2 K) in all heat exchangers.
9. In distillation columns pressure was varied from 1 bar up to 14 bar (case study 1). With these limits very high and very low temperatures were avoided in reboilers and condensers.

5.1.1. Results

In the detailed analysis costs were estimated separately for the fixed and for the variable part of the process using the rigorous RADFRAC model. In the previous research, short cut DSTWU model was used. The cost of the fixed part was the same for all the sequences. Changes occurred in the variable part. In case study 1 there was no fixed part of the process.

For comparison the sequences were classified according to total annual cost. The most promising sequences before and after heat integration were not always the same [5]. We tried to find the best heat integrated sequences by minimising the intracolumns $\sum \Phi_{\mathrm{i}}$ value and also by minimising $\sum(\Phi_{\mathrm{t}}\,\Delta T_{\mathrm{a}})$. The latter was found to be a very good option for estimating the promising heat integrated sequences [5]. The selection of sequences with the best Min $\sum(\Phi_{\mathrm{t}}\,\Delta T_{\mathrm{a}})$ values was based on the minimum areas $(\Phi_{\mathrm{t}}\,\Delta T_{\mathrm{a}})_{\mathrm{i}}$ of trapezoids. Sequences with very large $\sum(\Phi_{\mathrm{t}}\,\Delta T_{\mathrm{a}})$ values were shown as very expensive ones and should not be taken into account for calculations after heat integration. Sequences were classified according to the rising Min $\sum(\Phi_{\mathrm{t}}\,\Delta T_{\mathrm{a}})$ values. The best heat integrated sequence can be selected out of the sequences with the lowest Min $\sum(\Phi_{\mathrm{t}}\,\Delta T_{\mathrm{a}})$ values (see Table 3). The results were compared with the ones obtained by some other authors. According to the literature [1,15] total annual cost estimation made by the other authors is shown for the three best heat integrated sequences only (Table 3). Rigorous ranking is shown to lead to the same sequences. Different ranking was obtained by the heuristic-algorithmic method published in literature [15]. For the heat integration effectiveness the minimum integrability criterion was found to be a very good measure for the ranking of the most promising heat integrated sequences. We can establish that the ranking determined by the integrability criterion of Min $\sum(\Phi_{\mathrm{t}}\,\Delta T_{\mathrm{a}})$ is similar to the total annual cost ranking. For comparison we classified the sequences according to the Max $\sum(\Phi_{\mathrm{i}}\,\Delta T_{\mathrm{e}})$ values, too. The ranking showed as good as the minimum integrability criterion when compared to the total annual cost classification (see Table 3) in the case study 1.

Table 3 compares the three best sequences only because they are the ones available in the literature [1,15]. Another reason is in the fact that the Max $\sum(\Phi_{\mathrm{i}}\,\Delta T_{\mathrm{e}})$ criterion is valid only for the few most favourable sequences in case study 1 (some sequences could have higher Max $\sum(\Phi_{\mathrm{i}}\,\Delta T_{\mathrm{e}})$ values). Here, Min $\sum(\Phi_{\mathrm{t}}\,\Delta T_{\mathrm{a}})$ values are much more promising. In case study 1 both criteria, short cut and rigorous, give the same ranking of the three best sequences, but the sequences following them differ.

Using short cut models only (C^{*}_{TA}) shows different classifications of promising sequences. According to these results [5] the most cost favourable sequence was SEQ-2 again, but followed by sequences SEQ-3 and SEQ-1. Differences between the most promising sequences were very small. In case study 1 the ability for heat integration using the integrability criterion showed its applicability for the selection of the best sequences. Therefore, from this case study we would recommend to use rigorous models for the promising sequences search.

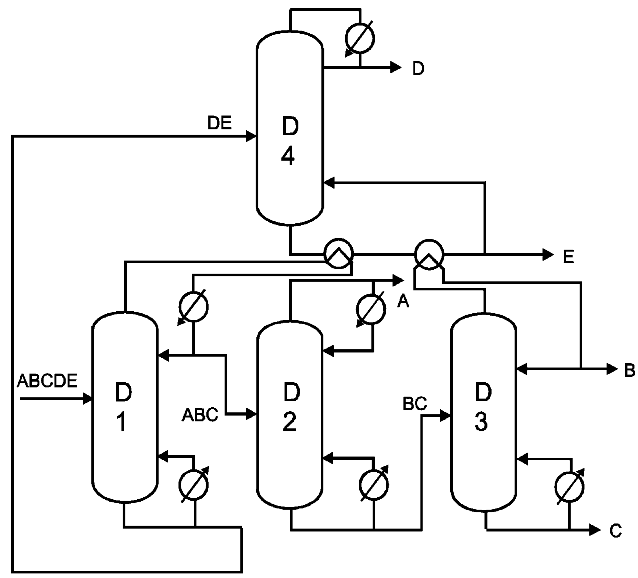

Fig. 7. The optimal process flow sheet for case study 1.

Optimal process flow sheet of the case study 1 is shown in Fig. 7 (abbreviations D1–D4 indicate distillation columns). The T–ΔI diagram of the best heat integrated sequence in the case study 1 is shown in Fig. 8. It should be pointed out that temperatures of condensation and evaporation are considered to be constant during our research. Therefore, condenser and reboiler heat flow rates are shown by horizontal lines in Fig. 8.

For comparison, we are representing the analyses of the sequences obtained when using heuristic rules [15]. This method was taken for the same reason in case studies 2 and 3, too. According to them the most promising heat integrated sequence was the sequence SEQ-1. Comparison with the literature shows that in the case study 1 all the best three heat integrated sequences (SEQ-2, SEQ-3, SEQ-1) have the most favourable $\sum(\Phi_{\mathrm{t}}\,\Delta T_{\mathrm{a}})$ values (compare the results in Table 3).

5.2. Case study 2

To confirm the conclusions obtained from case study 1 a more complex system of distillation columns was taken into consideration. It consists a variable part which could influence heat integration of distillation columns and the method developed here.

The second case study represents a separation problem of a six-products mixture for recovery of ethylene and light products from an oil-pyrolysis gas stream. The objective of the case study is to design a separation train for the recovery of nine components from steam-cracked oil involving about two dozen compounds. Total flow rate of the feed is 53.4 kg/s, its temperature is 333 K and the pressure is 136.5 kPa [19]. Process flow sheet of the case study 2 is shown in Fig. 9. It actually represents the sequence SEQ-1,

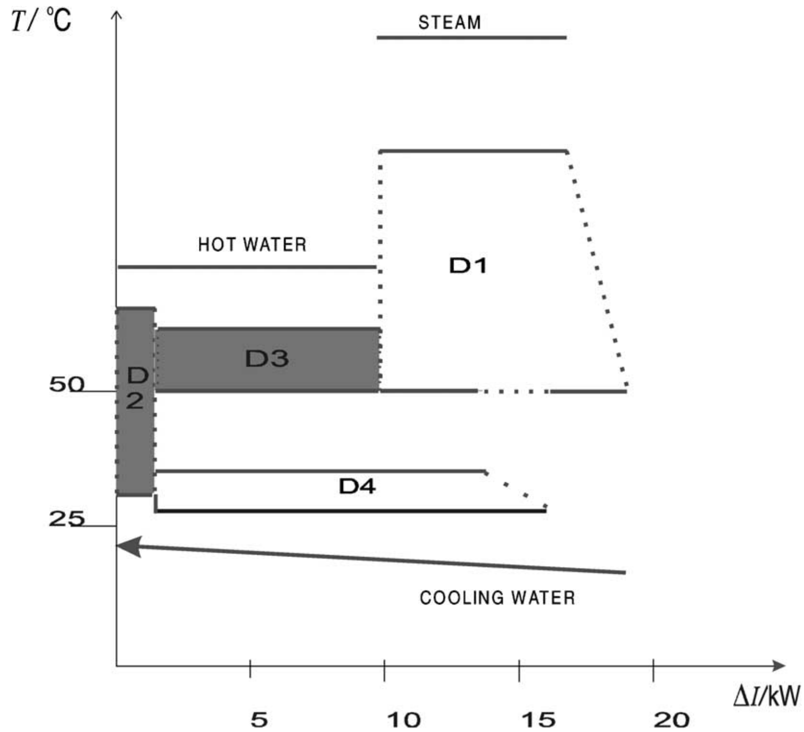

Fig. 8. T–ΔI diagram of the best heat integrated sequence in case study 1 (see Fig. 5).

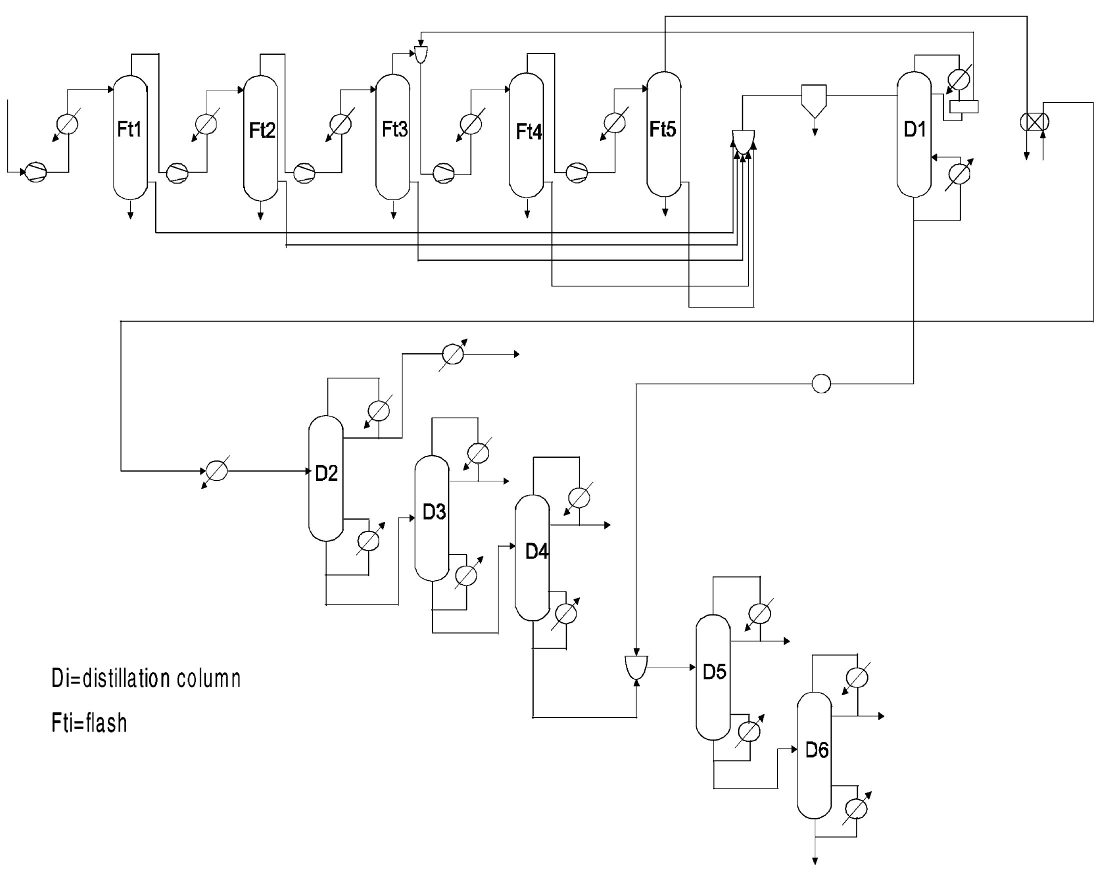

Fig. 9. Process flow sheet for case study 2.

Table 4
Feed compositions and properties for case study 2

	Components	Feed amount fraction, x_i	T_B (°C)
A	Hydrogen + methane	0.48	−161.5
B	Ethylene	0.28	−103.75
C	Ethane	0.06	−88.6
D	Propylene + propane	0.08	−47/−42.1
E	1,3-Butadiene + *trans*-2-butene	0.04	−4.45/4
F	*n*-Pentane	0.06	68.73

Table 5
Stream data for the fixed part of the process

Stream	T_S (°C)	T_T (°C)	Φ (kW)
Hot-1	100.00	39.00	19306.5
Hot-2	90.00	39.00	5987.4
Hot-3	92.00	39.00	5618.0
Hot-4	91.00	39.00	5278.0
Hot-5	95.00	17.00	8595.6
Hot-6	15.00	−63.00	14843.4
Cold-1	−119.00	38.00	5024.0
Cold-2	−32.00	−31.00	1228.0

one of the possible sequences shown later in Fig. 11. All process schemes have the same flow sheet of the flash drums including the first column D1 (the fixed part), different sequences occur in the second part (columns D2–D6). This part should be optimised. The composition of the feed mixture is shown in Table 4. It presents the feed stream to column D2. Bottom product from the column D1 contains negligible fractions of heavier components (components with four or more C-atoms) and is taken to the column D5. Its impact on the process can almost be neglected. Operating pressures are taken from the literature [18], the maximum being 3.2 MPa with deviations down to atmospheric pressure.

For simplicity and for better comparison of different sequences the same assumptions as in case study 1 were made.

Because of the light components, cooling temperatures are often below 0 °C. Refrigeration at these temperatures is the reason why the cost of cold utilities is very high and dominant in the process. Therefore, it is economically more favourable to separate products one-by-one in overhead than in the bottoms. For this purpose low temperature cooling methane, ethane and propylene were taken from the process.

For better understanding of the process some additional data are given for the fixed part of the process in Table 5. From these the GCC could be generated.

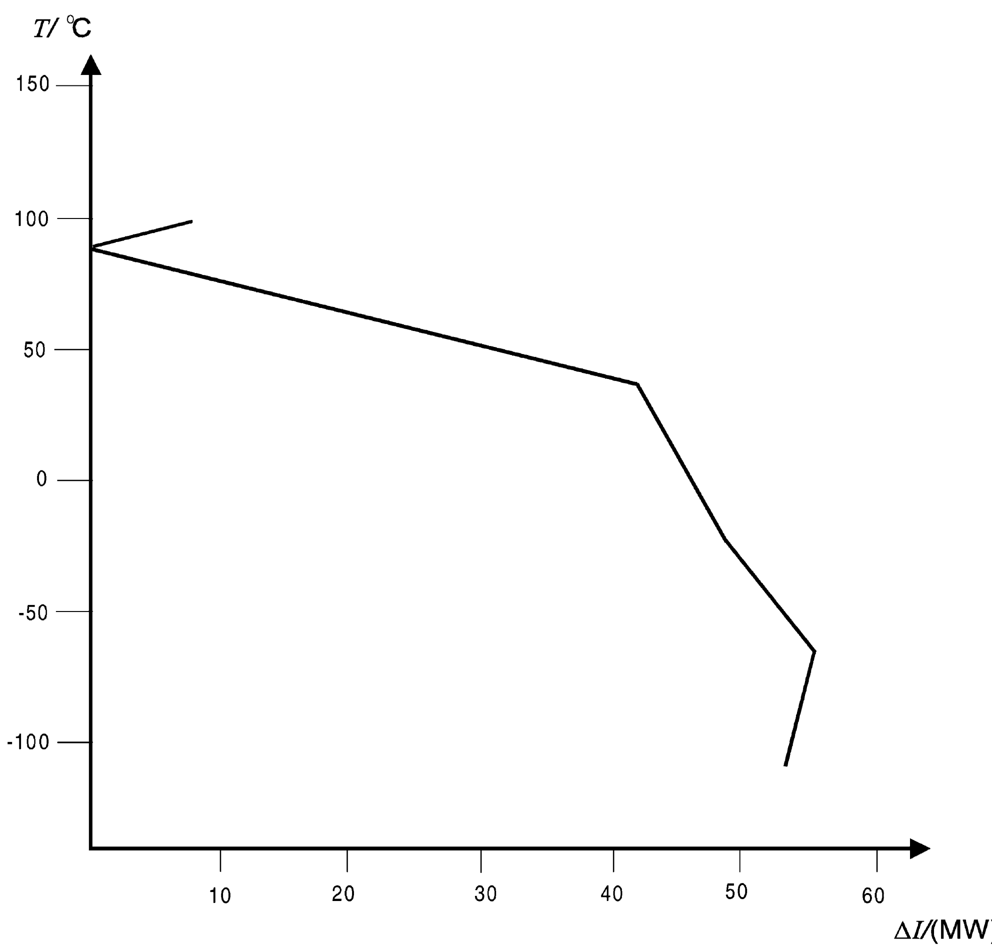

Fig. 10. Grand composite curve for the fixed part of the separation process.

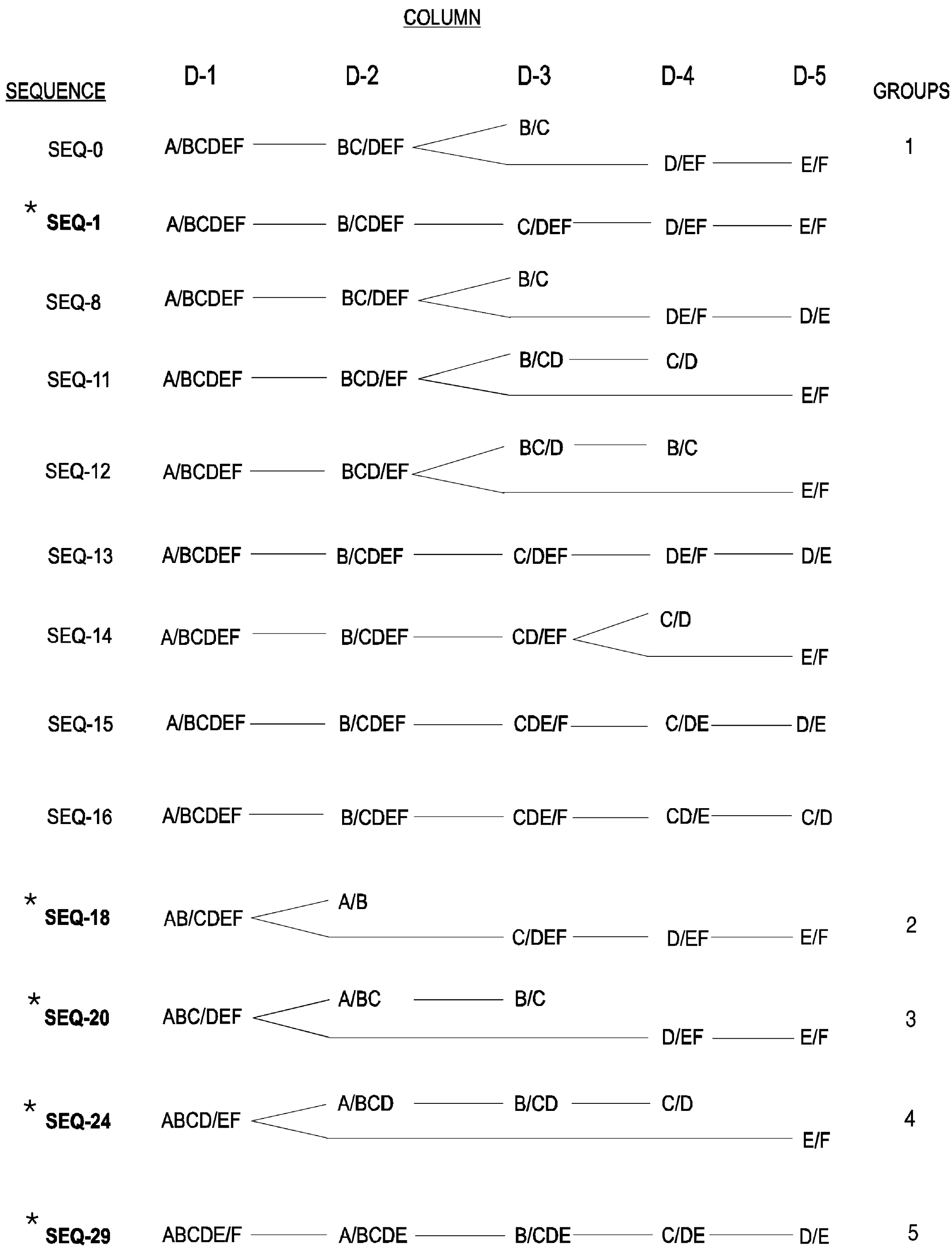

Fig. 11. Sequences studied by the short cut method.

5.2.1. *Results*

As in case study 1 the process is separated into a fixed and a variable part. Altogether, there are 12 hot and eight cold streams. The fixed part consists of seven hot and three cold streams, whereas the variable part contains five hot and five cold streams. There is a great surplus for cold utilities demand (Fig. 10). The reason is in the fixed part of the process which needs about 52 MW of cold utilities (coolers downstream the compressors).

In this case study 42 different potential schemes exist. These were classified into five groups using the sequence in the first column as the criterion (Fig. 11). This separation has a decisive influence on the whole process economics. It is the main reason to use the criterion of first column split for formation of groups of sequences.

One sequence was selected from each group only. For a better comparison, we had chosen sequences where products were separated one-by-one as overhead products. In the next

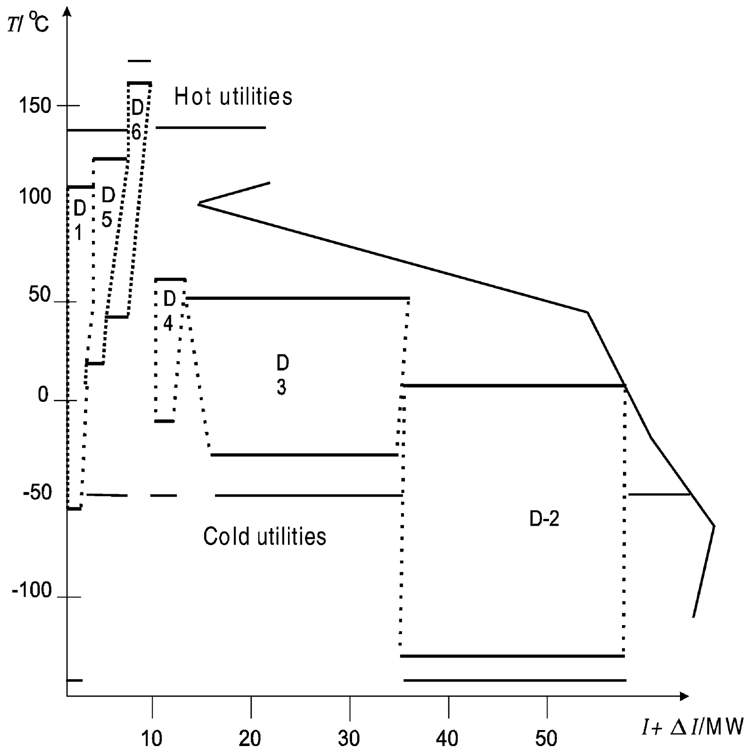

Fig. 12. T–$I+\Delta I$ diagram of the best process scheme.

step economics of the five selected sequences from Fig. 11 were studied. Typical sequences with successive numbers 1, 18, 20, 24 and 29 are marked with an asterisk. Thirteen different sequences were chosen for comparison from 42 sequences existing altogether (all of which have been exposed to simple short cut cost estimation).

In the variable part of the process the components were separated in the five distillation columns. The best distillation columns sequences were integrated with the process heat source. Thus, the fixed and the variable part of the process were integrated (see Fig. 12). The best heat integrated sequences were ranked according to the integrability criterion. Five of the best sequences were ranked similar to the C_{TA} evaluation with only one exception. Sequence SEQ-0 has a smaller Min $\sum(\Phi_t\,\Delta T_a)$ value then the sequence SEQ-12, whereas all the other sequences follow the same ranking as those using total annual cost (C_{TA}). Sequence SEQ-0 has better integrability properties but in this specific case the sequence SEQ-12 has lower C_{TA} (see Table 6).

The economic comparison of the sequences showed that our ranking was very good. Results in Table 6 were calculated for the complete process and they confirmed our conclusions. Here, distillation columns are heat integrated mutually and with the other process streams. The best heat integrated sequences, SEQ-1 and SEQ-13, are those with the lowest total utilities cost. In all the other good sequences (also SEQ-12, SEQ-0, SEQ-14) components are removed mostly as distillates. The two best sequences obey this rule in all the columns (SEQ-1) or in all but one column (SEQ-13).

In the sequences with A/BCDEF split, $x = 48\,\%$ of all the components are removed in the first column (component A is in great surplus). Here, the importance of the heuristics “most abundant first” shows its great value. It is decisive for the selection of the best sequence. Our short cut comparison of all the 42 sequences showed that sequences with the early separation of most abundant components are more cost favourable than those which do the same split later. The results of the latter are not shown in this paper.

Table 6
Min- and Max-criteria ranking compared with total annual cost

Rank	Sequence	T_P (°C)	Min $\sum(\Phi_t\,\Delta T_a)$ (MW K)	Max $\sum(\Phi_i\,\Delta T_e)$ (GW K)	C_{TA} (MUSD/year)
1	*SEQ-1	8.7	2.000	349.1	25.002
2	*SEQ-13	100.1	2.590	273.9	25.925
3	*SEQ-12	99.1	3.247	218.2	28.492
4	*SEQ-0	93.6	2.860	269.7	28.584
5	SEQ-14	100.0	3.480	195.9	28.712

For comparison, the best heat integrated sequences were also determined using the Max $\sum(\Phi_\mathrm{i}\,\Delta T_\mathrm{e})$ criterion. The most cost favourable ones were assumed to be those with the largest $\sum(\Phi_\mathrm{i}\,\Delta T_\mathrm{e})$ values. All the best heat integrated sequences have large $\sum(\Phi_\mathrm{i}\,\Delta T_\mathrm{e})$ values. Comparison of the best five sequences is shown in Table 6. Results of the other sequences are not presented, but the short cut economical comparison showed that the studied ones (Fig. 11) were the most favourable. Especially, favourable are sequences with Max-values over 200 GW K. They all belong to the first group of sequences. Sequences SEQ-20 and SEQ-24 have large $\sum(\Phi_\mathrm{i}\,\Delta T_\mathrm{e})$ values, too, but their reboilers and condensers have large heat duties, high span of temperatures in the columns, and pinch temperatures around 0 °C. As heat flow should not be transferred across the pinch, with the exception of heat integration between the columns, heat integration was limited and these sequences had low possibilities for heat integration.

Comparison of the most promising sequences, obtained by the Min- and Max-criteria gives in this case the same results. It has to be pointed out that results obtained by the Max-criterion are based on the short cut simulation, whereas the promising sequences obtained by the Min-criterion were based on the rigorous simulation. Therefore, in this case study short cut and rigorous approach led to the same final solutions. As presented in Table 6 all the five best sequences were ranked in the same way according to short cut and rigorous approach. In spite of that the rigorous method presented here gives more trust in result.

The pinch temperature is very important for heat integration capability. All heat integrated sequences in case studies 1 and 3 have pinch temperatures around 100 °C. The sequence SEQ-1 of this case study has much lower pinch temperature (8.7 °C) but it has the lowest Min $\sum(\Phi_\mathrm{t}\,\Delta T_\mathrm{a})$ value and, therefore, excellent possibilities for columns to be integrated with the process. Sequences of the other four groups have pinch temperatures between −10 and +5 °C.

According to the results obtained from the EGCC (Fig. 12), it is desirable to integrate as many streams under 95 °C as possible. Thus, process streams of the fixed part are used as heat source for the reboilers, and utilities needed otherwise for the reboilers and process streams are saved. Low pinch temperature increases low temperature cold utility requirements because of the large surplus of hot streams.

Total utilities cost plays the most important role in the total annual cost estimation of separation processes with more than four or five components, where expensive utilities are needed for operation of distillation columns. Although the investment in the process equipment can be very high, one should take into account the fact that in companies the equipment depreciation per year could be 10 %. We studied how the ranking of different sequences was changing with regard to the depreciation per year from 10 to 50 % including interest on capital (see Table 7).

Sequence SEQ-1 is the best from the 25 % depreciation per year on and only for the higher depreciation factor the best sequence is SEQ-13. The lower the depreciation per year, the more cost favourable is the sequence SEQ-0. By the 10 % depreciation per year, SEQ-0 becomes the third best one, immediately following sequences SEQ-1 and SEQ-13. T–ΔI diagram of the best sequence is shown in Fig. 12.

Table 7
Fractional depreciation per year influences the total annual cost ranking of the best sequences

Rank	Depreciation per year (%)				
	50	25	16.7	12.5	10
1	SEQ-13	SEQ-1	SEQ-1	SEQ-1	SEQ-1
2	SEQ-1	SEQ-13	SEQ-13	SEQ-13	SEQ-13
3	SEQ-12	SEQ-12	SEQ-12	SEQ-12	SEQ-0
4	SEQ-14	SEQ-14	SEQ-0	SEQ-0	SEQ-12
5	SEQ-0	SEQ-0	SEQ-14	SEQ-14	SEQ-14

In literature [19] the optimal heat integrated sequence was obtained by using four heuristic rules. The resulting sequence at 20 % depreciation was the SEQ-0 one. Our method showed that this sequence was only the fourth or fifth best one in the total annual cost classification (see Table 7). Those sequences which separate more components as distillates (SEQ-1, SEQ-13 and SEQ-12) were identified as better ones in most cases. Additional annual savings of up to 14 % were found as compared with the sequence SEQ-0. It is obvious that the rigorous (Min) method presented here gives better estimation of promising sequences than heuristic rules.

5.3. Case study 3

The third case study is a separation problem of a six-product mixture resulting from acetaldehyde production by ethanol dehydrogenation. It is a process system with reactor, coolers, heaters, a flash, an absorber, pumps, valves and distillation columns. The composition of the mixture is shown in Table 8. Fig. 14 actually represents the sequence SEQ-7. All process schemes have the same flow sheet of the fixed part including column D1, different sequences occur in the second, variable part (columns D2–D6). The mixture given in Table 8 presents the feed stream to column D2. The total feed flow rate is 4 kg/s, its temperature is 315 K and its pressure is 101.32 kPa [20]. Actually, there is a seventh component—hydrogen, but it is separated in the absorber. In this case study all the splits of the components

Table 8
Feed compositions and properties for case study 3

	Key components	Feed amount fraction, x_i	T_B (°C)	C^*_ES [15]
A	Acetaldehyde	0.43	21	42.25
B	Ethyl acetate	0.01	77	0.79
C	Ethanol	0.34	78	6.21
D	Water	0.20	100	4.79
E	*n*-Butanol	0.01	117	0.01
F	Acetic acid	0.01	118	–

Table 9
Stream data for the fixed part of the process

Stream	T_S (°C)	T_T (°C)	Φ (kW)
Cold-1	64.80	329.85	9310.0
Hot-1	329.85	152.85	2446.0
Hot-2	65.40	64.80	10.4
Hot-3	242.10	46.80	6879.0

were not as sharp as in case studies 1 and 2. The reason is in the formation of the ethanol–water azeotrope and the two-phase liquid mixture butanol–water. As in case study 1 the DSTWU module was used for the short cut estimation and RADFRAC module for the detailed total costs estimation. The last one includes the model for simulating the azeotropic distillation columns, too.

Therefore, the resulting separation gives no sharp splits in columns D2–D6. In our research this has not been overseen. The lower purities of final products did not affect the economic estimation of particular sequences.

Stream data for the fixed part of the process are given in Table 9. From these the GCC could be generated. Other hot/cold streams result from the six distillation columns.

5.3.1. Results

In case study 3 the same rigorous method as in case studies 1 and 2 was applied. The same procedure as given in Section 5 (steps 1–7) was applied. Data used in the rigorous simulation were based on the same number of stages, operating pressures and reflux ratios as with the short cut DSTWU model. The promising heat integrated sequences are shown in Fig. 13. Results of the best heat integrated sequences are shown in Table 10. The situation in case study 3 is more complex than in case studies 1 and 2.

Comparison with the results obtained by use of short cut models only (C^*_{TA}) [5] shows somewhat different classification of the best three sequences. The promising sequence was SEQ-7 followed by sequences SEQ-6 and SEQ-3. In this case study the use of rigorous models has shown to be more promising. Short cut models gave promising candidates, but their ranking was not exactly optimal.

For comparison promising sequences ranked according to Max $\sum(\Phi_i \, \Delta T_e)$ values are also shown. The Min-criterion has been proved to be better and more effective than the Max $\sum(\Phi_i \, \Delta T_e)$ one. Actually, the Max-criterion was found

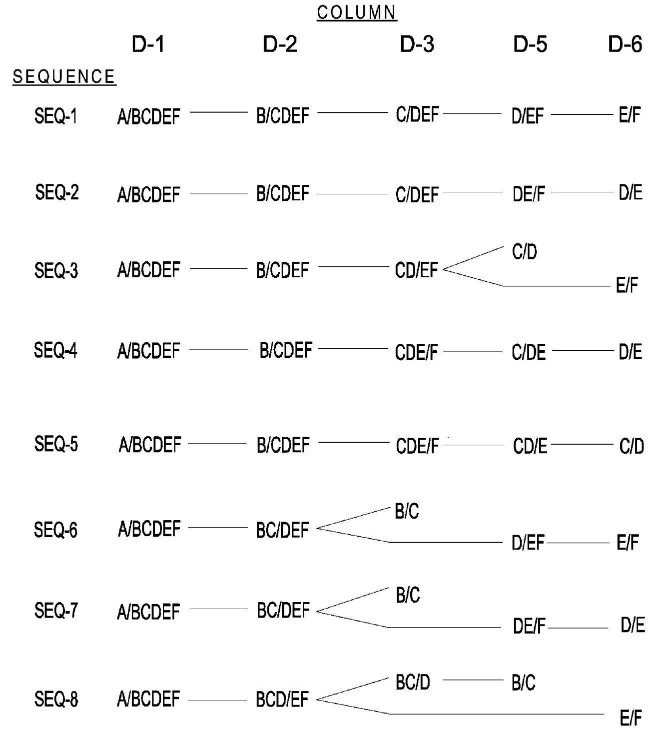

Fig. 13. Sequences studied by using integrability criterion in case study 3.

not to be appropriate in the case study 3 at all. This is confirmed by the results shown in Table 10, too. Other process streams influence heat integration in this case and, therefore, intercolumns area is not the only important criterion for the total annual cost classification.

In case study 3 operating pressures were selected as in case study 2. It was desired to keep pressure in the columns sequences cascading downwards as the separation proceeded, approaching the ambient one. This was provided in order to keep the cost of changing stream pressures at the minimum and to allow safer operation (if the columns were near ambient conditions). Pressure selection was varied from the atmospheric value up to 500 kPa. Thus, the purchase of new compressors and pumps for increasing pressure in distillation columns was avoided. Optimal process flow sheet of case study 3 is shown in Fig. 14.

$T/(I + \Delta I)$ diagram of the best heat integrated sequence in the case study 3 is shown in Fig. 15. Distillation columns are presented as *T–I* trapezoids.

Table 10
Comparison of integrability criterion and total annual cost ranking in case study 3[a]

Rank	Sequence	C_{TA} (MUSD)	C_{TA} [5] (MUSD)	Min $\sum(\Phi_t \, \Delta T_a)$ (kW K)	Max $\sum(\Phi_i \, \Delta T_e)$ (GW K)
1	SEQ-6	1.464	1.487	246	99.168
2	SEQ-3	1.500	1.732	256	128.854
3	SEQ-7	1.539	1.464	257	55.914
4	SEQ-1	1.562	1.777	260	34.949
5	SEQ-4	1.658	1.829	317	67.298

[a] Results with the short cut model.

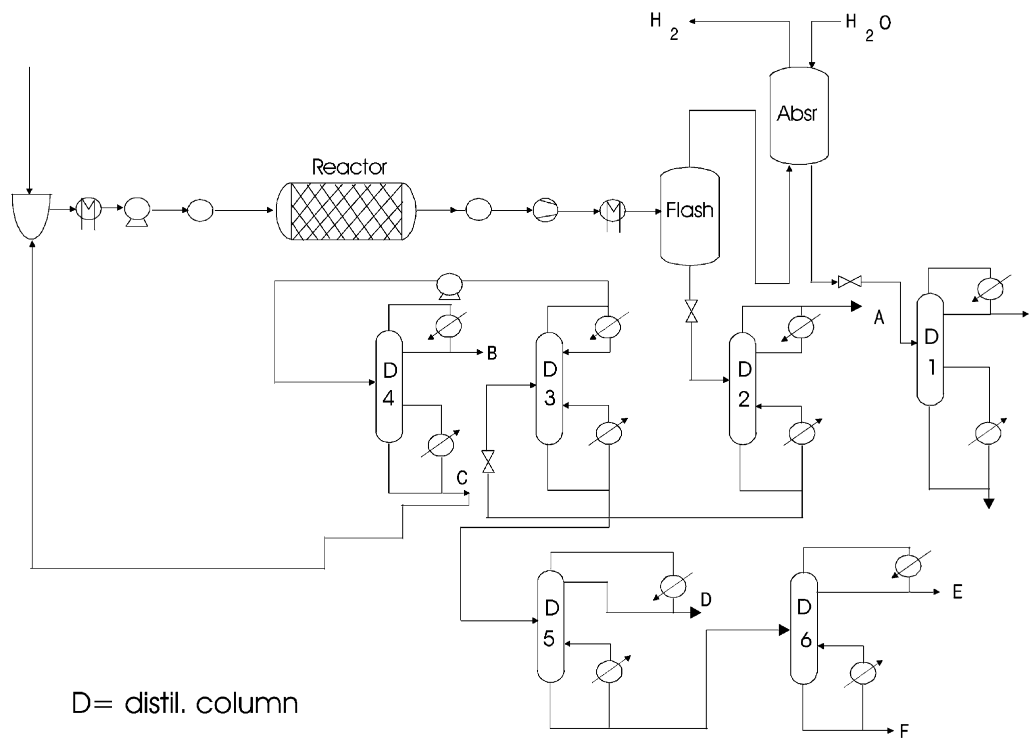

Fig. 14. Process flow sheet for case study 3.

In case study 3, the C_{ES} coefficient for the A/BCDEF separation is by far the highest (Table 8). Therefore, all the best heat integrated sequences are expected to have the A/BCDEF separation as the first one. The optimal sequence obtained by using a simple heuristic method [15] was the integrated sequence 6 (SEQ-6), too. The evaluation showed this sequence to be the best in the total annual cost ranking after heat integration, having the lowest sum of the F_I values. Using the heuristic rules for non-integrated sequence the second best sequence is SEQ-7, the third one being SEQ-8, which is only the eighth best using total annual cost ranking after heat integration. It is obvious that the rigorous method based on the Min-criterion gives better estimation of promising sequences than heuristic rules and Max-criterion.

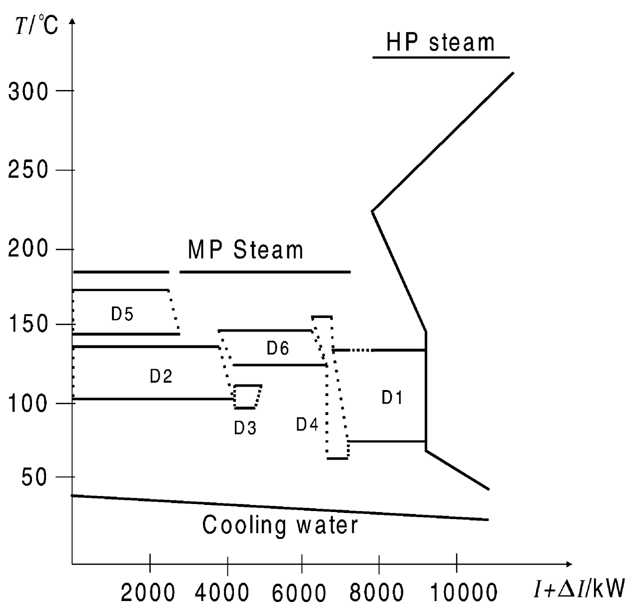

Fig. 15. T–$I + \Delta I$ diagram of the best heat integrated sequence in case study 3.

6. Discussion

Rules for ranking of integrated distillation columns have been revisited using a rigorous method for distillation columns when determining the optimal sequence. The Min-criterion with rigorous column models has been proven to be more effective and appropriate in this respect, more than with short cut models. It is suitable for isolated distillation trains and also in combination with process systems. It is based on the minimisation of intracolumn Φ and ΔT of distillation columns trapezoids (integrability criterion). The intra- and intercolumn areas have been explained in Fig. 1. When minimising the utility consumption minimum external utility consumption (Φ_{HU} or Φ_{CU}, respectively) and maximum heat exchange between the integrated columns is reached. Actually, the Min-criterion includes the maximisation of $\sum(\Phi_i \, \Delta T_e)$ areas between the integrated columns. A column with a low span of intracolumn temperatures has better possibilities for heat integration and lower heat exchanger areas. The situation is similar when more columns are connected in series or even if they form a cascade. These statements are valid for use of short cut or rigorous models of distillation columns.

The criterion of minimising intracolumn $\sum(\Phi_t\, \Delta T_a)$ using rigorous plate to plate simulations was confirmed as an upgraded option to the short cut model, developed in our previous work, in the case of complex processes, where besides the distillation columns other process units also apply and contribute their heat flow rates to the heat integration. Obviously, process parameters which can be represented by trapezoids in the extended GCC, i.e. the heat flow rate (Φ) and the span of intracolumn temperatures (ΔT_a) are decisive for heat integration. The most cost favourable columns have wide and low trapezoids in T–I or T–ΔI diagram. They enable integration of large heat flow rates and high flexibility in stacking, they have high degree of integrability.

Comparison of short cut and rigorous approaches gives similar results in all the case studies. It shows that short cut models are quite good for estimation of promising sequences. They also give enough information needed for most economic calculations. The proposed method is a good combination of short cut and rigorous models. The former are simple and fast for use, give basic results needed, whereas rigorous ones serve for more precise cost estimation.

The main advantage of the method presented here when compared to the algorithmic and evolutionary ones is its screening value. It eliminates inappropriate sequences and reduces the time needed for finding the promising ones. It is not completely automated yet and can be regarded as a semi-automated procedure. Therefore, we cannot compare computational times of different approaches but comparing calculations by hand it is faster than the other approaches except the heuristic one. The method is simple to apply and use, does not need any special computational knowledge besides the process simulation. It gives identical optimum sequences as the more rigorous and time-consuming computer methods and it is more sound than the heuristic ones.

7. Conclusions

The Integrability criterion is applicable for fast screening of process alternatives giving the best sequences of heat integrated distillation columns. The comparison of the best heat integrated sequences obtained by the rigorous method and detailed total cost analysis showed almost the same ranking. Isolated distillation column sequences with the optimum heat integration were the best also when treated with the rest of the process background GCC.

The method described is applicable to the analysis and synthesis of the best heat integrated processing systems. It has been successfully applied to five- and six-component mixture separation problems and is also convenient for process systems with distillation columns trains. The rigorous method upgrades the short cut Min- and Max-criteria from our earlier work and is more convenient for processes where streams other than the column ones appear. According to the results obtained from all the three case studies it could be concluded that the integrability criterion based on minimisation of intracolumn areas alone is efficient enough for short cut selection of candidates for promising sequences. For the detailed cost estimation rigorous simulation is recommended.

The method proposed has short-comings and limitations as all methods do. The integrability criterion is unable to select operating pressures in the columns of a distillation sequence itself. It does not include absolute temperatures, which are heavily important for heat integration, but these temperatures are implicitly determined by the pressure selection. The approach does not include the dependence of utility costs on utility temperature and it does not include the stream temperatures explicitly. Temperatures are included in the GCC. Possible constraints are very important and should be considered with special attention.

We shall try to apply this knowledge in the optimisation of other thermally integrated multicolumn processes to minimise limitations of the method proposed.

Acknowledgements

The authors gratefully acknowledge the Ministry of Education, Science and Sports of Slovenia for enabling the research. We would like to thank the reviewers for their helpful suggestions.

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