Simultaneous design of separation sequences and whole process energy integration

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Abstract

This paper presents a novel methodology for the optimisation of the preliminary design of heat-integrated multicomponent distillation sequences. This is achieved through use of a reduced process superstructure where the role of splitting each adjacent key component pair is assigned to an individual separation column, greatly reducing the size and complexity of the problem. This methodology uses information about other process streams on site with which heat can be exchanged within the initial design in order to find a plant-wide optimal separation configuration with the aim of reducing the cost of heating provided by utilities including those used for heating and cooling process streams. In order that this model can be formulated as a mixed-integer linear program, this model utilises a discretised temperature grid where stream temperatures are allowed to vary. This methodology was tested on an example of a mixed alkane feed stream, with the sequence changing dependent on the degree of process integration. The method was found to have the potential for significant cost reductions compared to a heuristic design, with the example exhibiting a cost saving of over 50% and a reduction in CO₂ associated with process heating of almost 60%, though the magnitude of these savings is highly dependent on the specific example to which it is applied.

Keywords: Separation process design; energy integration; multicomponent distillation; preliminary design; optimisation

1. Introduction

1.1. Background

The problem of separation of a single multicomponent stream into pure product streams through use of a sequence of distillation columns is one that has received much attention due to the large costs incurred by separation processes. As the most widely used method, Hewitt [1] reported that distillation processes account for an estimated 3% of global energy consumption on account of being used in roughly 95% of industrial liquid separation processes. One major user of distillation processes is the petroleum industry, with the US Department of Energy [2] estimating that distillation accounts for some 40% of process-related energy usage therein. Process heating and cooling utilities, in the form of process steam

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and cooling water, make up a large proportion of this energy requirement, making heat integration within the system to reduce utility usage is very important.

Distillation as a separation method takes advantage of differing boiling points of individual components within a liquid mixture, with simple columns having one feed and two product streams; one vapour distillate stream from the top of the column and one liquid bottoms stream from the bottom. This simple column configuration is the type considered in this work, though other configurations can be used in specific circumstances. The reboiler at the bottom of the column requires the addition of heat, usually from utilities, while the condenser can be used as a source of lower-temperature excess heat. Because of the temperature difference, heat integration between the reboiler and condenser of a single column is not possible, but integration between reboilers and condensers throughout a sequence of integrated columns can be used to reduce the energy penalty. Furthermore, excess heating and cooling duties from other process streams and equipment, such as reactors and furnaces, can be used to further reduce utility demands in separation.

The minimum thermodynamic utility requirement can be calculated through the pinch analysis technique, described by Linnhoff and Hindmarsh [3]. This method, which is based on pre-computed stream temperatures, requires the creation of an energy cascade for each temperature interval, which allows the heat duties and surpluses to be calculated and an overall system utility requirement to be found. In order to provide extra heating and cooling to the system, a number of utilities can be considered; there must be a hot utility able to provide heat to the system at a temperature higher than that of any stream and a cold utility able to cool below the temperature of any stream within the system. Within this work, these are called bracketing utilities, since all stream temperatures must fall between the temperatures at which these utilities are available. There can also be intermediate temperature utilities that can provide heating or cooling within the system, often at a lower cost such as low pressure steam for cheaper heating at lower temperatures.

While pinch analysis is able to find the overall system minimum utility requirement, it is unable to directly identify matches for individual streams and therefore cannot be used for heat exchanger sizing. As a result, any costing data and analysis within this paper will solely consider utility requirement and not heat exchanger or column capital costs. This demonstrates why pinch analysis is best used in the preliminary conceptual design stage, whilst a more detailed model used to assign stream matches and size heat exchangers is required in later design stages to find a more accurate cost estimation for the system.

1.2. Heuristical Process Design

Traditionally, many processes would be designed by using a sequence of design heuristics which give solutions based on previous experience without requiring mathematical analysis. While using these heuristics to design processes will in many cases lead to safe, operable systems, it is likely that optimal solutions for heat and energy management may have been missed. Many historical design heuristics for separation process synthesis exist as two distinct steps; a design for the separation system is created and optimised, after which an optimised energy integration network is designed for the system, corresponding to levels 4 and 5 of the hierarchical process described by Douglas [4]. While this method leads to two

independently optimised layers, it is widely recognised that a simultaneous design of separation and heat integration will give overall utility savings for the system.

Using the heuristical design methodology proposed by Douglas as an example, two independent sets of heuristics are suggested for separation sequence design. The first, which is more general, calls for corrosive components to be removed first, then any remaining reactive components or monomers to be removed after. Next, products should be removed primarily from the distillate streams, and any recycle streams should also be removed as distillates. Based on these simple heuristics, separation sequencing would return the direct sequence in any mixture without corrosive and reactive components.

The second set of heuristics proposed by Douglas are more carefully targeted for simple separation columns (i.e., one distillate stream, one bottoms stream). The following heuristics are to act as a guide to selecting the order of separations.

- Most plentiful first
- Lightest first
- High-recovery separations last
- Difficult separations last
- Favour equimolar splits
- Next separation chosen should be cheapest

Looking at these suggestions demonstrates one of the core weaknesses of a heuristic based design; in many cases, these heuristics directly contradict each other. As an example, if the heaviest component in a mixture is the most plentiful, there will be a direct conflict between the first two heuristics. Because of this, depending on the priority of heuristics used, multiple different solutions may be found, with the selected solution not necessarily optimal.

In a similar vein to heuristical design, Pajula et al. [5] considered case based reasoning (CBR) as a method by which knowledge of existing similar processes is used to guide process design. These analogues are used as initial guesses at separation systems in order to reduce the number of possible solutions to be assessed at an early stage in order to provide quick solutions to these problems which will likely be operable due to their similarities with existing processes. However, in order to use CBR effectively, a large quantity of industrial data is required which is often lacking, leading to an incomplete catalogue of cases and use of a poor basis for making design decisions. Also, CBR will inherently neglect more creative solutions and novel technologies due to the lack of data about them, making it a methodology often used in industries which are more conservative with process design.

1.3. Mathematical Process Design

Detailed mathematical model-based design of separation systems often involves construction of a superstructure system, which is a flowsheet where every possible separation task is considered within the optimisation, with flows progressing in a sequential manner from column to column. As shown by

Thompson and King [6], a superstructure based model will require the analysis of a total of $\frac{(2(N-1))!}{N!(N-1)!}$ possible sequences. By this calculation, a 10 component inlet feed would require evaluation of 4,862 possible sequences to separate the mixture into individual components. As such, using a superstructure leads to a very large model to be solved, meaning that alternative methods are used in order to reduce the problem sizes associated with this.

Due to the large number of columns to be assessed in a superstructure, it is important to find a way to reduce problem sizes, such as the MILP method used by Aggarwal and Floudas [7] to construct a heat-integrated separation sequence. With this method, each column is assigned a separation task between two adjacent key components with all possible stream connections allowed in the model to represent the superstructure. As such, the number of columns that must be modelled for an N component mixture is $N - 1$, significantly reducing the number of sequences to be evaluated. An example of this can be seen below in Figure 1 for a three-component mixture.

Figure 1: Example of a reduced superstructure for a three-component problem demonstrating the specified column operations and the network of possible streams

Naka et al. [8] proposed approaching a multicomponent distillation problem by considering each

column as its energy loss within the system. This method uses a system-wide analysis of heating and cooling duties, including heat duties from other process streams. All operating conditions for each item within the sequence must be pre-specified. Since every possible sequence is analysed, the number of possible sequences is as calculated by Thompson and King [6], potentially leading to a large number of sequences for problems with a large number of components.

Novak et al. [9] propose a three-level MINLP model that uses a compact superstructure for designing separation systems, which can then be integrated with a HEN model to find an overall optimal solution to the design problem. In order to reduce the size and complexity of the problem, rigorous models were replaced by simplified approximate models for process unit operations, with logical constraints used to remove infeasible flowsheet options. Since this method uses a MINLP, a phase of initialisation and linearisation must be carried out before the problem can be solved, while a MILP would not require such a step.

Muraki and Hayakawa [10] provided an alternative methodology for synthesising a separation process to produce multicomponent products using simple distillation columns. The proposed method consisted of two stages, with the separation sequence searched for through use of a material allocation diagram, followed by optimisation of the separation sharpness.

Jain et al. [11] considered the use of mathematical optimisation to design a flowsheet simultaneously for separation and heat exchange, using a mixture of simple and complex columns to be determined by constraints on the model. Column sizing and operation are considered in this paper using shortcut methods, with the overall model formulated as a MINLP. Whilst the design of the system includes detail about column sizing and capital cost estimation, there is not enough detail for a fully detailed design and further detailed work would be required.

Lucia and McCallum [12] proposed a methodology by which separation process flowsheets are designed for systems containing multiple columns using energy targeting. A model was designed to provide the optimal minimum energy column sequence from the shortest stripping line approach, found via the minimum boil-up ratio. This is used within a two-level procedure, first proposed by Amale and Lucia [13], to reflect changes in the distillation lines caused by varying product composition. From this iterative process, a global optimisation problem can be formulated, requiring a multi-level MINLP. While this iterative process has many advantages, such as allowing calculations in circumstances where Underwood's equations could not be used (e.g., azeotropic mixtures), the nature of the model makes it less suitable for simple, preliminary process design.

Benz and Cerda [14] propose an MILP model for the synthesis of distillation sequences based on calculation of minimum and maximum heat duties for each column, found via 'rule of thumb' calculations. This is used to find heat-integrated flexible distillation sequences to account for variability in the composition of a feed streams whilst minimising utility costs.

In addition to just using simple columns, more complex columns or configurations can be used to provide alternative separation pathways. Much of the recent work on distillation sequencing has been carried out on thermally coupled columns in an attempt to reduce costs [15],[16], or to use thermodynamically equivalent sequences derived from the thermally coupled sequences in order to further reduce costs [17].

However, this model uses only simple columns and does not consider thermally coupled columns.

Considering mathematical design when considering specific industrial systems, Lee et al. [18] investigated the synthesis of separation systems for olefin molecules with a specified feed and products. Whilst not uniquely considering distillation, this method uses a superstructure containing all possible separators to minimise the total annualised cost of the separation system, with the use of additional disjunctions allowing for heat integration and compression, pumping and pressure reduction to be considered. Lashkajani et al. [19] also considered the olefin separation problem, using a superstructure based optimisation, with the code validated and verified from the literature, in order to find the optimal sequence of distillation columns to separate an olefin feed with heat integration.

When considering separation process synthesis within a crude oil refinery, Liebmann and Dhole [20] proposed a methodology by which a crude oil fractionating column can be designed, optimised for heat recovery. For the initial stage, the fractionating column is considered as equivalent to a system of thermally coupled simple, two product columns through the decomposition method used by Hengstebeck [21]. By visualising the system in this decomposed state, further design and optimisation of the columns and associated heat exchange networks is made easier. Through the decomposition, the method proposed within this paper can also be used for the design of complex columns such as fractionating towers, extending the range of problems that this method can be applied to.

1.4. Objectives

This study will use a novel system of allowing reboiler and condenser temperatures and those of process streams to take values from a discretised grid, whilst allowing a small variation in the process stream and column temperatures from the initial values. This is to represent process flexibility that may be available within process plants, with many process streams varying in practice from their quoted design temperatures. The initial temperature values for the condensers and reboilers are chosen as the normal boiling points of key components. The variation of temperatures is constrained to maintain the temperature difference across the column to be the same as the difference between the boiling points of the two key components. The main advantage of this discretised grid within the reduced superstructure is that it allows the model to be kept linear and to solve quickly compared to other models with similar aims, which being non-linear are computationally more intensive and may have problems with non-convexity.

In this study, we present a mixed-integer linear program (MILP) which can be used to determine the optimum separation sequence from information about the feed and required products with regards to minimising energy input as utilities. This will be achieved through simultaneous design of the separation system with optimisation of the heat exchange network for thermodynamically minimum utility. It is important to note that this heat exchange network design will not include matching streams for heat exchangers, instead focussing on the theoretically optimal network for utility calculations from which a feasible network for energy integration is guaranteed.

2. Mathematical model for construction of optimal heat-integrated multicomponent distillation sequence

2.1. Separation system

Throughout this work, it has been assumed that each column represents the separation between two adjacent key components, which are separated between the distillate and bottoms streams according to an equal distribution factor. It is also assumed that the non-key components are wholly non-distributing and that each column operates with a total condenser and total reboiler. It is assumed that the operating temperatures of the condensers and reboilers are the boiling points of the key components; the boiling point of the light key for the condenser and the heavy key for the reboiler. The heat capacity of the process streams through the condenser and reboiler is determined as being a weighted mean of the heat capacities of the pure components within each stream. These assumptions are sufficient for the conceptual design stage considered here, as the results can be further refined during a subsequent detailed design stage.

This model, due to its utilisation of simple distillation columns and the basic preliminary model used, can be used to find separation sequences for basic mixtures of individual components. In particular, it is assumed that the input mixture for the model is non-reacting and that the mixture is zeotropic. These assumptions hold for many separations done within the petrochemical industries, in particular refinery operations where large quantities of energy are used separating very weakly interacting alkane molecules.

For each column considered in this system, a series of material balances must be carried out in order to model the distribution of material within the column. The key components are distributed according to the selected column-specific degree of separation coefficients $S(j)$, with a value of 1 being equivalent to a sharp separation, while non-keys are considered non-distributing.

Since the separation task of each column is known, sensible reflux ratios can be specified for each column. Since this methodology is only for preliminary process design, a more detailed calculation of the true reflux ratios for each column can be made in a subsequent design stage. As such, the method shown here is by design less detailed than some alternatives but is intended to provide an initial estimation of the flowsheet, and is thus simpler and faster to run as an MILP than a comparable MINLP.

Components within the model are labelled within the set i , labelled from the lowest boiling point to the highest, with the number of columns j restricted to one fewer than the number of components in the feed mixture. Columns are labelled according to the separation they carry out, with the first column separating the two components of lowest boiling points, and the second separating the second and third lowest boiling points, and so on.

All terms are defined in the nomenclature section. A material balance is carried out for each component i across each distillation column j ,

$$Z(i, j) = D(i, j) + B(i, j) \quad \forall i, j \quad (1)$$

Component i is distributed across the distillate and the bottoms streams according to Equations 2 to 5, with the light and heavy keys identified through numbering within the sets of columns and components.

The heavy key was identified as being the component numbered one more than the column number and the light key the component numbered the same as the column. These keys are assigned according to a specific column's split fraction $S(j)$, representing the degree of separation achieved. Any component with a higher number within set i than the number of the column $j + 1$ was considered to be a heavy non-distributing non-key component and assigned solely to the bottoms, and where in the case where the number within the set i was lower than j , the species was assigned solely to the distillate as a light non-distributing non-key.

For the case of the heavy key component, the bottoms flowrate from a column j is equal to the inlet flowrate of that component multiplied by the specified split fraction and is given by

$$B(i, j) = S(j) \times Z(i, j) \quad \forall i = j + 1. \quad (2)$$

The distillate flowrate of the light key component is equal to the split fraction multiplied by the inlet flowrate of that component, given by

$$D(i, j) = S(j) \times Z(i, j) \quad \forall i = j. \quad (3)$$

For components below the light key, it is assumed that there is no distribution across the column and as such the flowrate of these components in the bottoms is zero, denoted as

$$B(i, j) = 0 \quad \forall i < j. \quad (4)$$

Similarly, any components with boiling points above the heavy key are assumed to not be present in the distillate streams,

$$D(i, j) = 0 \quad \forall i > j + 1. \quad (5)$$

For the bottoms stream, the total flowrates of each component from each column is split between flows to other columns, denoted $F_{B,tot}$, and flows to product streams denoted $P_{B,tot}$

$$B(i, j) = F_{B,tot}(i, j) + P_{B,tot}(i, j) \quad \forall i, j. \quad (6)$$

The total distillate stream is comprised of flows to other columns $F_{D,tot}$ and flows to products $P_{D,tot}$

$$D(i, j) = F_{D,tot}(i, j) + P_{D,tot}(i, j) \quad \forall i, j. \quad (7)$$

The total flow to products from the bottoms of each column is equal to the sum of all individual flows P_B , of which one goes from the bottoms stream of each column j to each possible product stream p . Similarly, the flow to products from each distillate stream can be found as a sum of all P_D ,

$$P_{B,tot}(i, j) = \sum_p P_B(i, j, p) \quad \forall i, j \quad (8)$$

$$P_{D,tot}(i, j) = \sum_p P_D(i, j, p) \quad \forall i, j. \quad (9)$$

The total flowrate from each bottoms stream allocated to each product is equal to the sum of all individual flows F_B , of which one stream exists from the bottoms of each column j to the inlet of every

column j' . Likewise, the total flowrate from the distillate can be represented in an equivalent manner,

$$F_{B,tot}(i, j) = \sum_{j'} F_B(i, j, j') \quad \forall i, j \quad (10)$$

$$F_{D,tot}(i, j) = \sum_{j'} F_D(i, j, j') \quad \forall i, j. \quad (11)$$

Since the stream from both the bottoms and the distillate is split between product streams and flows to other columns, as can be seen in Figure 2. The mass balance on the flowrate from the bottoms of a column to the products streams is achieved through the use of a discretised split fraction, DSF , which takes one of a finite number of incremental values between 0 and 1. This parameter is used to maintain the linearity of the model instead of using the split fraction of the stream, a continuous variable. This discretised split fraction is multiplied by a binary variable λ to represent this across the set denoted f , with the single value of λ allowing a split fraction to be calculated.

Figure 2: Example flow diagram of splitter for a bottoms stream showing distribution between flows to other columns and to product streams

$$\sum_{f,p} \lambda_{P,B}(j,p,f) \times DSF(f) + \sum_{f,j'} \lambda_B(j,j',f) \times DSF(f) = 1 \quad \forall j \quad (12)$$

$$\sum_{f,p} \lambda_{P,D}(j,p,f) \times DSF(f) + \sum_{f,j'} \lambda_D(j,j',f) \times DSF(f) = 1 \quad \forall j. \quad (13)$$

Each of $\lambda_{P,B}$ and $\lambda_{P,D}$ is only allowed to take one value per stream from j to p , as each stream can only have one split fraction,

$$\sum_f \lambda_{P,B}(j,p,f) = 1 \quad \forall j,p \quad (14)$$

$$\sum_f \lambda_{P,D}(j,p,f) = 1 \quad \forall j,p. \quad (15)$$

The formulation of this discretisation method is shown below in Equations 16 through 19 for the products streams for streams originating both from the bottoms and the distillate, where the discretised split fraction defined above is used to calculate the fraction of the total bottoms stream present in each $\lambda_{P,B}$, and likewise the distillate.

$$P_B(i, j, p) \leq B(i, j) \times DSF(f) + F_{max} \times (1 - \lambda_{P,B}(j, p, f)) \quad \forall i, j, p, f \quad (16)$$

$$P_B(i, j, p) \geq B(i, j) \times DSF(f) - F_{max} \times (1 - \lambda_{P,B}(j, p, f)) \quad \forall i, j, p, f \quad (17)$$

$$P_D(i, j, p) \leq D(i, j) \times DSF(f) + F_{max} \times (1 - \lambda_{P,D}(j, p, f)) \quad \forall i, j, p, f \quad (18)$$

$$P_D(i, j, p) \geq D(i, j) \times DSF(f) - F_{max} \times (1 - \lambda_{P,D}(j, p, f)) \quad \forall i, j, p, f. \quad (19)$$

Light products can be extracted only from the distillate, and heavy products only from the bottoms. Only key components in a given separation can be produced as final products,

$$P_B(i, j, p) = 0 \quad \forall p \neq j + 1 \quad (20)$$

$$P_D(i, j, p) = 0 \quad \forall p \neq j. \quad (21)$$

Flowrates from the bottoms and distillate which are fed to other columns are calculated in the same way as flows to product streams, with use of the discretised split fraction and binary variables λ_B and λ_D ,

$$\sum_{f, j'} \lambda_B(j, j', f) = 1 \quad \forall j \quad (22)$$

$$\sum_{f, j'} \lambda_D(j, j', f) = 1 \quad \forall j \quad (23)$$

$$F_B(i, j, j') \leq B(i, j) \times DSF(f) + F_{max} \times (1 - \lambda_B(j, j', f)) \quad \forall i, j, j', f \quad (24)$$

$$F_B(i, j, j') \geq B(i, j) \times DSF(f) - F_{max} \times (1 - \lambda_B(j, j', f)) \quad \forall i, j, j', f \quad (25)$$

$$F_D(i, j, j') \leq D(i, j) \times DSF(f) + F_{max} \times (1 - \lambda_D(j, j', f)) \quad \forall i, j, j', f \quad (26)$$

$$F_D(i, j, j') \geq D(i, j) \times DSF(f) - F_{max} \times (1 - \lambda_D(j, j', f)) \quad \forall i, j, j', f. \quad (27)$$

Constraints on the minimum acceptable purity, here denoted as $P(i)$, ensure that the purity requirements are met for each individual product, and can be replaced by recovery constraints if necessary. Only if a stream satisfies one of these constraints is it allowed to be assigned to the relevant product stream.

$$P_{tot}(p) = \sum_i F_P(i, p) \quad \forall p \quad (28)$$

$$F_P(i, p) \geq P(i) \times P_{tot}(p) \quad \forall i = p \quad (29)$$

$$F_P(i, p) = \sum_j (P_B(i, j, p) + P_D(i, j, p)) \quad \forall i, p. \quad (30)$$

The inlet to each column is calculated as a sum of some fraction of the overall system inlet feed, here denoted Z_{frac} , and any streams from the distillate or bottoms of other columns. The feed to any individual column is represented by Equation 31. While many other methodologies consider only a sequential separation train, this approach allows the model to recycle streams through columns and to consider complex separation systems with a feed distributed between columns,

$$Z(i, j) = \sum_{j'} (F_B(i, j, j') + F_D(i, j, j')) + Z_{sys}(i) \times Z_{frac}(j) \quad \forall i, j \quad (31)$$

Since Z_{frac} is the fraction of feed fed to each column, the sum of all Z_{frac} must equal unity.

$$\sum_j Z_{frac}(j) = 1. \quad (32)$$

2.2. Heat Integration

One of the largest requirements for energy within process plants is associated with process heating and cooling. By matching high temperature heat sources and low temperature heat sinks, the requirement for external utilities to satisfy heating and cooling requirements is reduced, leading to large cost savings. In particular, it is important to reduce the requirement for hot utilities since process steam is usually more expensive than cooling water (and likely more CO₂ intensive and overall more environmentally damaging).

Heat integration will usually lead to non-linearity of a system due to the expression for heat duty being defined through a bilinear expression, as a product containing a streams flowrate and its temperature difference. Since neither of these variables are bounded, the linearisation methodology used when calculating split fractions cannot be used in this instance. In order to avoid this problem, temperatures have been assigned to values on a discrete grid with 1°C increments. This means that since each interval has a fixed temperature difference of 1°C, the heat duty for each interval can be expressed in a linear manner as an arithmetic sum of all duties present. This is similar to a methodology employed by Andreovich and Westerberg [22] in which column operating conditions were considered as discrete elements of a finite set instead of continuous variables. This was intended to reduce the size of the resulting superstructure in cases where operating conditions would have to be optimised in order to reduce the solution time of the model.

In addition to assigning temperatures to this grid, stream supply and target temperatures have been allowed to vary from their calculated values within bounds of $\pm\Delta T$ to mimic behaviour that can be attained by varying the pressure of the separation columns, reflecting acceptable flexibility in operation windows found in industrial process plants. This is shown for the condensers in Equations 35 and 36. By using this reduced superstructure in which columns are defined instead of stream destinations, the

problem avoids the large solution set associated with the full superstructure even with the temperatures not strictly defined.

2.2.1. Separation Heat Integration

Since each reboiler and condenser operates at a single temperature, the binary variables used to assign these temperatures to the set of temperature intervals T can only have one non-zero value per column,

$$\sum_T T_R(j, T) = 1 \quad \forall j \quad (33)$$

$$\sum_T T_C(j, T) = 1. \quad \forall j. \quad (34)$$

The temperature of the condenser for each column is within ΔT of the pure component boiling point, T_b of the boiling point of the light-key. The temperature assigning binary variable, T_C , has the approach temperature applied to it to ensure that heat exchange would be possible for the column.

$$\sum_T (T \times T_C(j, T)) \leq T_b(i) + \Delta T - T_{app} \quad \forall i = j \quad (35)$$

$$\sum_T (T \times T_C(j, T)) \geq T_b(i) - \Delta T - T_{app} \quad \forall i = j. \quad (36)$$

The reboiler temperatures are found within ΔT of the pure-component boiling point of the heavy key, and the binary variable therefore lies within that range of intervals T ,

$$\sum_T (T \times T_R(j, T)) \leq T_b(i + 1) + \Delta T \quad \forall i = j \quad (37)$$

$$\sum_T (T \times T_R(j, T)) \geq T_b(i + 1) - \Delta T \quad \forall i = j. \quad (38)$$

In order to maintain the temperature difference across the column, whilst still allowing this flexibility, the temperature difference between the reboiler and condenser is fixed as the same between the boiling points of the pure components of the keys, with the added one-degree flexibility built in since the discrete temperature intervals cannot differ by fractions of one degree,

$$\sum_T (T \times T_R(j, T) - T_b(i + 1)) \leq T \times T_C(j, T) - T_b(i) + T_{app} + 1 \quad \forall i = j \quad (39)$$

$$\sum_T (T \times T_R(j, T) - T_b(i + 1)) \geq T \times T_C(j, T) - T_b(i) + T_{app} - 1 \quad \forall i = j. \quad (40)$$

After assigning the condenser and reboiler temperatures to the discrete grid, heat duties must be calculated. For this study, it has been assumed that all columns have a total condenser and total reboiler, so the reflux and boil up ratios must be specified for each column to calculate the overall reboiler and condenser duties. All heat duties and deficits are assigned positive values,

$$Q_C(j) = \frac{R + 1}{R} \times \sum_i (D(i, j) \times \Delta H_{vap}(i)) \quad \forall j \quad (41)$$

$$Q_R(j) = \frac{R}{R+1} \times \sum_i (B(i, j) \times \Delta H_{vap}(i)) \quad \forall j. \quad (42)$$

Once the reboiler and condenser duties have been calculated, they must be assigned to the temperature grid in order to construct heat cascades for pinch analysis. This is achieved by multiplication of the calculated heat duties from Equations 41 and 42 with the binary variables calculated in Equations 33 to 40. The linearisation used is shown below in Equations 43 to 45 for the condenser, and is repeated in Equations 46 to 48 for assignment of the reboiler duties.

$$Q_{C,T}(j, T) \leq Q_C(j) \quad \forall j, T \quad (43)$$

$$Q_{C,T}(j, T) \leq Q_{max} \times T_C(j, T) \quad \forall j, T \quad (44)$$

$$Q_{C,T}(j, T) \geq Q_C(j) - Q_{max} \times (1 - T_C(j, T)) \quad \forall j, T \quad (45)$$

$$Q_{R,T}(j, T) \leq Q_R(j) \quad \forall j, T \quad (46)$$

$$Q_{R,T}(j, T) \leq Q_{max} \times T_R(j, T) \quad \forall j, T \quad (47)$$

$$Q_{R,T}(j, T) \geq Q_R(j) - Q_{max} \times (1 - T_R(j, T)) \quad \forall j, T. \quad (48)$$

2.2.2. General Process Heat Integration

Within a large process plant, there will be other non-separation process streams which will require heating and cooling, usually supplied by utilities. Instead of using utilities to satisfy these requirements, these streams can be integrated with the separation system and each other in order to reduce the overall utility requirements. Since these streams are not connected to the separation process, it is assumed that they are independent and can therefore be treated as standard streams from a generic pinch analysis problem. When analysing the combined problem for heat integration within a whole plant, any streams that are currently supplied by utilities should be included in this analysis, as through integration with this new separation process it is possible that these utility requirements could be reduced or cut altogether. In addition, any streams where atmospheric heating or cooling is used to effect a temperature changed should be included in this analysis as they could provide useful heat sources or sinks for the separation system.

In the same way as the stream temperatures for the separation system, the temperatures of these process streams have been allowed to vary within limits in order to maximise heat exchange matches, representing inherent process flexibility that may be available in the plant.

For hot streams, the target temperature is lower than the supply temperature, so the binary variable for the lower end of the interval Y_l is found in an interval T between $\pm\Delta T$ of the quoted hot stream

supply temperature, less the minimum approach temperature T_{app} to account for the stream being a heat source to ensure matches are feasible,

$$\sum_T Y_l(s, T) = 1 \quad \forall s \in hot(s), T \geq T_T(s) - \Delta T - T_{app} \quad (49)$$

$$\sum_T Y_l(s, T) = 1 \quad \forall s \in hot(s), T \leq T_T(s) + \Delta T - T_{app}. \quad (50)$$

For cold streams, since the supply temperature is the lower of the two, the binary variable for the lower bound of the stream is within an interval T , which is within $\pm\Delta T$ of the given supply temperature instead of the target.,

$$\sum_T Y_l(s, T) = 1 \quad \forall s \in cold(s), T \geq T_S(s) - \Delta T \quad (51)$$

$$\sum_T Y_l(s, T) = 1 \quad \forall s \in cold(s), T \leq T_S(s) + \Delta T. \quad (52)$$

When calculating the upper temperature of the stream, the hot streams will use their supply temperature, again less T_{app} to account for the streams being heat sources, to find a range of acceptable temperature intervals T where the binary variable Y_h can take a non-zero value,

$$\sum_T Y_h(s, T) = 1 \quad \forall s \in hot(s), T \geq T_S(s) - \Delta T - T_{app} \quad (53)$$

$$\sum_T Y_h(s, T) = 1 \quad \forall s \in hot(s), T \leq T_S(s) + \Delta T - T_{app} \quad (54)$$

Similarly, the binary variable Y_h for the cold streams is found for intervals of T within $\pm\Delta T$ of the given target temperature.

$$\sum_T Y_h(s, T) = 1 \quad \forall s \in cold(s), T \geq T_T(s) - \Delta T \quad (55)$$

$$\sum_T Y_h(s, T) = 1 \quad \forall s \in cold(s), T \leq T_T(s) + \Delta T. \quad (56)$$

In order to ensure the model does not allow multiple values for the temperature of each stream,

$$\sum_T Y_l(s, T) = 1 \quad \forall s \quad (57)$$

$$\sum_T Y_h(s, T) = 1 \quad \forall s. \quad (58)$$

The temperature difference between the supply and target temperatures of each stream has been kept constant using Equations 59 and 60 so as to maintain the overall heating or cooling duty,

$$T_S(s) - T_T(s) = \sum_T T \times Y_h(s, T) - \sum_T T \times Y_l(s, T) \quad \forall S \in Shot(s) \quad (59)$$

$$T_T(s) - T_S(s) = \sum_T T \times Y_h(s, T) - \sum_T T \times Y_l(s, T) \quad \forall S \in Scold(s). \quad (60)$$

A binary existence variable, E is used to show the existence of a process stream within any temperature interval T for calculation of the cascades for the entire system,

$$E(s, T) = E(s, T - 1) + Y_l(s, T) - Y_h(s, T) \quad \forall s, T. \quad (61)$$

2.3. Overall System Heat Duties

Since the model determines the temperatures at which the heating and cooling duties for the reboiler must be provided, a heat surplus variable can be created which represents the heat surplus or deficit within each interval. For each reboiler and condenser, the temperature change associated with the phase change in the reboiler or condenser is considered to be 1°C , thus occupying only a single interval. The energy requirements and surpluses from the separation system are also included in the heat surplus as a product of the existence of the stream and its heating duty, meaning that cold streams give positive contributions and hot streams negative to the overall surplus.

As well as the bracketing utilities, intermediate utilities such as lower pressure steam or warm water are considered in order to reduce the overall cost of the utilities within the system. These utilities are incorporated within the calculation of the heat surplus for each interval, with an interval-assigned hot and cold utility term. Each temperature will be associated with a cost depending on the type of utility to be used. Both the hot and cold utilities here must be given positive values.

$$SH(T) = \sum_j (Q_{R,T}(j, T) - Q_{C,T}(j, T)) + \sum_s (E(s, T) \times FCp(s)) - HU_tDuty(T) + CU_tDuty(T) \quad \forall T. \quad (62)$$

In order to enforce the use of bracketing utilities, the maximum and minimum values of the cascade are set at zero. This has the effect that any heat duties and surpluses within the system have to be satisfied with utilities, allowing the total utility requirements to be found,

$$CH(T_{max}) = 0 \quad (63)$$

$$CH(T_{min}) = 0. \quad (64)$$

This surplus heat vector is turned into a cascade by summation to find a cumulative heat available or required at the associated temperature. By making the cascade heat a positive variable, the adjusted cascade heat is calculated in a single step, with the pinch point where the cascade heat value is zero [23],

$$CH(T + 1) = CH(T) + SH(T + 1) \quad \forall T. \quad (65)$$

Utilities have different values depending on their costs and availability at the plant considered. The objective function consists of an expression for calculation of the cost of operation in \$ per hour, based on the cost of different utilities and the requirement of each to be used. If costs are given per tonne of utility, the duty of each stream in MJ per tonne must be calculated; for steam, this is equivalent to the enthalpy

of vaporisation, while for cooling water this is the heat capacity across a 5°C increase. In general, cooling water costs are provided per unit of energy so the energy density of the utility is not required,

$$H = 8000 \times \sum_T (HUtCost(T) \times HUtDuty(T) + CUtCost(T) \times CUtDuty(T)). \quad (66)$$

It is important to note that capital costs of the columns or heat exchangers are not included in this model, with the cost, and therefore the objective function of the model, based purely on the operating cost of utilities. It is not anticipated that capital costs will change the sequences selected, as column costings are expected to be broadly similar across sequences, but this has not yet been assessed. This work will be extended in the future to assess the cost implications associated with the capital costs of the columns, which will be assessed simultaneously to see if this has any impact on the sequence.

3. Case Study - Alkane Separation

3.1. Data and Problem construction

The light end products of the crude oil distillation process are separated together, comprising predominantly of a mixture of light alkanes. The composition of this light ends feed will vary depending on the operation of the fractional distillation unit and the type of crude used as feed to the refinery, and can be highly variable. This example has a feed of five components ranging from propane to pentane, with the separation system pressurised to 6 bar to account for the sub-zero boiling points of some components under standard conditions. Input data for this problem is displayed below. It is assumed that the degree of separation within each column is 99% between key components, with non-distribution of non-keys, and all products are required to have a purity of at least 95%. The minimum approach temperature for heat exchange is 10°C and steam costs and temperatures, shown in Table 2, have been taken from the example considered by Mascia et al. [24]. When calculating annualised costs, it is assumed that the system is to operate for 8,000 hours over the course of the year.

In addition to the cost savings associated with reduced utility requirements, optimising the system in order to reduce the cost through utilities also has the benefit of reduced fuel requirements for heating, and hence reductions in CO₂ emissions associated with the separation sequence. For the purpose of this paper, simplified calculations associated with process CO₂ emissions have been carried out, simply to illustrate that reducing the costs associated with separation sequences often has the secondary effect of reducing the process CO₂ emissions from utilities. Process CO₂ emissions were calculated by assuming that steam was raised by burning natural gas (methane) in a boiler with an 85% efficiency in order to satisfy hot utilities, whilst assuming that process CO₂ emissions associated with cooling water as a cold utility are negligible. Whilst this oversimplistic calculation will not be an entirely accurate representation of the CO₂ savings associated with the process, these figures suggest an additional benefit of this optimisation with the reduced emissions.

In order to let the model integrate the separation system with other process streams, sample streams have been devised with their temperature data shown in Table 3. The models will be solved using the GAMS software with the IBM ILOG CPLEX optimizer version 24.7.4 [25].

Component	Inlet Flowrate (kmol/hr)	Boiling Point (°C)	Enthalpy of Vaporisation (MJ/kmol)
Propane	25.4	8.0	19.0
i-Butane	136.1	44.7	21.4
n-Butane	226.8	57.5	22.6
i-Pentane	281.4	91.6	25.2
n-Pentane	157.5	100.5	26.1

Table 1: Inlet feed composition and pure component properties

Stream	Supply Temperature (°C)	Cost per GJ (\$/GJ)
Cooling water	20	0.190
LP Steam	144	6.094
MP Steam	184	8.004
HP Steam	245	11.154

Table 2: Cost and temperature data of available utilities

Stream Identifier	Supply Temperature (°C)	Target Temperature (°C)	Stream heat capacity (FCp) MJ/hr
H1	120	70	40
H2	140	50	50
H3	170	60	40
H4	90	35	20
C1	35	100	40
C2	120	180	50
C3	50	90	60
C4	80	120	30

Table 3: Sample temperature and heat capacity data of external streams from other processes

3.2. Baseline Scenario

The baseline scenario represents current practice in many plants, with external heat sources satisfied through heat integration elsewhere in the plant, and a new direct sequence distillation system self-integrated where possible. A representation of this problem is shown below in Figure 3. This will be used as the base-case scenario to illustrate the advantages that using this model to propose alternative configurations can offer. Temperatures of the reboilers and condensers for each column are allowed to vary from the pure component boiling points at 6 bar (the system pressure used in the initial problem) by up to 10 °C per column, and the auxiliary streams from other parts of the system have a maximum allowable temperature difference of 5°C. Costs have been estimated with data from Table 2 used in the calculations for both the auxiliary streams and the separation system.

Figure 3: Direct Sequence of separation for olefin separation example

Stream	Supply Temperature (°C)	Heating duty for system (GJ/hr)	Cost of utility (\$/yr)	CO ₂ emissions (tons/yr)
Cooling water	20	2.349	3,600	0
LP Steam	144	10.859	529,500	5,960
MP Steam	184	0	0	0
HP Steam	245	0	0	0

Table 4: Utility requirements and costs for the Direct Separation Sequence

Stream	Supply Temperature (°C)	Heating duty for system (GJ/hr)	Cost of utility (\$/yr)	CO ₂ emissions (tons/yr)
Cooling water	20	4.370	6,600	0
LP Steam	144	0.190	9,300	75
MP Steam	184	1.110	71,000	610
HP Steam	245	0	0	0

Table 5: Duty and associated cost of each utility for the auxiliary streams

This gives a combined total annual operating cost of \$620,000, with the utility cost of the auxiliary streams costing \$87,000 per year and the separation system \$533,000. The auxiliary streams utility requirements are satisfied through a combination of cooling water, low- and medium-pressure steam, with the medium pressure steam responsible for the majority of the cost. By contrast, the utility costs for the separation are entirely due to the requirement for low-pressure steam to provide heat to the system, contributing to 85% of the total costs. In order to reduce this, it is likely to be beneficial to change configuration to attempt to replace the requirement for hot utilities with lower-cost cold utilities. Assuming that the steam is generated in a natural gas boiler with 85% efficiency, and that cooling water does not have inherent associated CO₂ emissions, the total emissions from this process are 6,650 tons per year from the combustion of natural gas.

The separation model took only 0.167 seconds to solve with the CPLEX solver before convergence to an integer optimal solution within the tolerances set in the model. The additional process streams' heat exchange model is solved separately and takes 0.122 seconds with the same software and solver. More detailed information and data from the simulation can be found in the electronic supplementary information.

3.3. Separation Sequence Integration

Using the model described in Section 3, the separation sequence has been optimised to reduce the overall utility requirements. This involves removing constraints on the separation sequence and allowing the model to find the optimal configuration, in this instance one shown below in Figure 4. In this configuration, the majority of the feed is again fed into the first column for the separation between propane and i-butane, with the bottoms fed to a column which separates the butane from the isopentane. This column does not form any final products, but instead two streams containing two components each, which are separated in subsequent columns to form the remaining four product streams.

Figure 4: Optimised separation sequence for olefin separation without further process integration

Stream	Supply Temperature (°C)	Heating duty for system (GJ/hr)	Cost of utility (\$/yr)	CO ₂ emissions (tons/yr)
Cooling water	20	13.391	20,400	0
LP Steam	144	5.775	281,600	3,170
MP Steam	184	0	0	0
HP Steam	245	0	0	0

Table 6: Duty and associated cost of each utility for the separation system

The total annual utility cost of this separation system is \$302,000, giving a total combined cost of \$389,000. Compared to the previous case with the direct sequence, this combined operating cost is 37% lower. By using this non-conventional sequence, the system balances the requirements between hot and cold utilities more evenly, with a greater proportion of the total utility made up of the cheaper cooling water than the steam. CO₂ emissions associated with the burning of natural gas for steam production

are reduced by 42% compared to the base case, with total emissions of 3,860 tons per year. This reflects the change in the process configuration and shows the importance of mathematical optimisation, since heuristic methods would be unlikely to suggest this solution.

This model takes considerably longer than the baseline scenario, with the separation system optimisation taking 1033.3 seconds with the CPLEX solver. With the other process streams not included at this stage, the problem is solved independently as above and takes 0.122 seconds. Detailed solution data, including temperatures and flow destinations, can be found in the electronic supplementary information.

3.4. Plant-wide Integration

While typical mathematical design would concern itself only with integration of the separation system, it is important also to consider the other process streams on plant that are currently satisfied through use of utilities. This system incorporates these process streams into the optimisation, allowing the model to select the optimal configuration for the separation system including integration with the wider process. This results in a different sequence from that proposed above, with the new sequence shown in Figure 5, due to the additional heat exchange between the process streams and the columns' reboilers and condensers.

Stream	Supply Temperature (°C)	Heating duty for system (GJ/hr)	Cost of utility (\$/yr)	CO ₂ emissions (tons/yr)
Cooling water	20	15.712	24,000	0
LP Steam	144	3.915	191,000	2,150
MP Steam	184	1.110	71,000	610
HP Steam	245	0	0	0

Table 7: Duty and associated cost of each utility for the combined separation and auxiliary heating systems

Figure 5: Optimised separation sequence for olefin separation with external process integration

The total annual operating cost of this combined system is \$286,000 for both the separation system and the process streams to have their heating and cooling requirements satisfied, 53% lower than the direct sequence and 26% lower than the optimised sequence. CO₂ emissions of 2,760 tons per year from the natural gas-fired boiler are 58% lower than in the base case, and 28% lower than if just the separation sequence is optimised. This clearly demonstrates the advantages of integrating the heat exchange network of the separation system with other process streams, instead of optimising both networks independently. It is important to note that by considering the process streams with the separation system, the optimal separation sequence actually changes to make better use of the available heat duties from the other process streams, demonstrating that attempting to retrofit the separation system with other process streams will not provide the overall optimal network.

In contrast to the previous model, which considered only the separation system and not the additional process streams, when these are combined for this model the total solution time is 105.07 seconds, significantly faster than the previous calculation. More detailed information and data pertaining to this result can be found in the electronic supplementary information, including temperatures, flowrates and

heat duties.

4. Conclusion

Due to the high cost associated with the use of utilities within chemical plants, heat integration has been used to reduce the overall utility requirements of these processes, thus reducing cost, CO₂ emissions and environmental damage. In particular, in the petroleum industry distillation is responsible for 40% of process energy usage. It is therefore important to consider how energy usage can be reduced in these distillation sequences at an early stage of process design. Heat integration is typically considered after the initial process design stage (including column sequencing) has been completed and is often limited to considering only one process at a time. By using mathematical optimisation when designing a process with utility requirements as one of the major considerations, in many cases significant cost savings can be achieved.

This article proposes using a mixed integer linear program (MILP) for the preliminary design phase of a separation sequence synthesis which incorporates other streams from within the overall plant in order to reduce overall utility requirements. This optimisation framework considers the system as an integrated process which has the potential to interact with other plant areas instead of as a self-contained operation within the plant, allowing improvements to the overall use of heat within a facility.

This model considers temperatures being allowed to vary to mimic behaviour from a process plant, due to process flexibility or column pressurisation but in discrete intervals. This allows subtle changes in the default column temperatures that may allow for a greater degree of heat integration, with some streams able to exchange heat which would not be able to previously. By assessing this in the preliminary design, opportunities for optimal heat management will not be missed when considering a more detailed design.

An alkane separation case study has been used to demonstrate the optimisation methodology and its associated benefits. By using the mathematical optimisation system to ascertain the optimal separation configuration, in certain cases where the direct sequence is non-optimal, significant utility cost savings can be found. The magnitude of these cost savings is highly dependent on the input data to the problem but the example here demonstrates that simply changing the configuration can lead to operating cost savings of over 50%.

In addition to cost savings, using this method to reduce utility use within the plant can lead to significant reductions in CO₂ emissions associated with natural gas combustion for the purpose of raising steam. Using the example above, total emissions associated with utilities can be reduced by almost 60% when compared to the base case, and almost 30% when compared to a system where the separation process is optimised in isolation. This once again demonstrates the benefits of carrying out a plant-wide optimisation when considering heat integration and management.

By integrating the separation process with other process streams within the plant, additional cost savings can be made. Even where changing sequence reduces utility costs from the base case, there is the potential for further cost reductions, even if the overall transferred heat from these auxiliary streams is small relative to the heat duties of the separation system. In a case where the auxiliary streams contribute

roughly one sixth of the utility cost to a system, integrating them within the separation sequence can lead to the cost reduction of changing sequence increasing from 38% to 53%.

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Nomenclature

Dimensions/Indices

f - Fraction of stream, used to keep splitter equations linear

i/p - Used to represent components, p used generally to denote a flow of component i to the product stream representing component p

j/j' - Denotes the required column. j' used in equations as a target column where there is a flow from column j to column j'

s - Denotes process streams not involved in separation that are considered in determining minimum utility requirements

$SHot(s)$ - Subset of process streams which have a surplus heat that they can supply to the system

$SCold(s)$ - Subset of process streams which have a heat deficit that must be supplied by the system

T - Temperature intervals, starting from 1 and rising in increments of 1°C

Parameters

$CUtCost$ - Cost associated with cooling water in $\$/MJ$

$DSF(f)$ - Discretised grid for assignment of stream fractions for exiting splitters

F_{max} - Maximum flowrate possible in system, must be larger than total flowrate into first column in $kmol/hr$

$FCp(s)$ - Product of flowrate and heat capacity of stream s with units $MJ/(hr.K)$

$HUtCost(T)$ - Relative cost associated with heating utility available at temperature T in $\$/MJ$

$\Delta H_{vap}(i)$ - Standard enthalpy of vaporisation of pure component i in kJ/mol

$P(i)$ - Specification of required product purity for each component i

$R(j)$ - Reflux ratio for column j

$R_{min}(j)$ - Minimum reflux ratio for each column j

$S(j)$ - Degree of separation between key components associated with column j

T_{app} - Minimum approach temperature for effective heat transfer in $^{\circ}C$

$T_b(i)$ - Standard boiling point of pure component i in $^{\circ}C$

$T_S(s)$ - Supply temperature of process stream s in $^{\circ}C$

$T_T(s)$ - Target temperature of process stream s in $^{\circ}C$

$Z_{sys}(i)$ - Flowrate of component i in overall system inlet in $kmol/hr$

Variables

- $B(i, j)$ – Flowrate of component i in bottoms from column j in $kmol/hr$
 $B_{tot}(j)$ – Total flowrate of bottoms stream from column j in $kmol/hr$
 $CH(T)$ – Calculated cascade heat available at temperature T in MJ/hr
 $CUtDuty$ - Required duty of cooling water in system in MJ/hr
 $D(i, j)$ – Flow of species i in distillate of column j in $kmol/hr$
 $D_{tot}(j)$ – Total flowrate of distillate stream from column j in $kmol/hr$
 $F_B(i, j, j')$ – Flow of species i in bottoms of column j fed to column j' in $kmol/hr$
 $F_{B,split}(i, j, j', f)$ – Determination of split fraction of bottoms from column j to each column j'
 $F_{B,tot}(i, j)$ – Flow of species i in bottoms of column j fed to all other columns combined in $kmol/hr$
 $F_D(i, j, j')$ – Flow of species i in distillate of column j fed to column j' in $kmol/hr$
 $F_{D,split}(i, j, j', f)$ – Determination of split fraction of distillate from column j to each column j'
 $F_{D,tot}(i, j)$ – Flow of species i in distillate of column j fed to all other columns combined in $kmol/hr$
 $F_P(i, p)$ – Total flow of component i in product stream p in $kmol/hr$
 H – Weighted objective variable to determine system utility requirements in $\$/yr$
 $HUtDuty(T)$ - Heating duty required at temperature T in MJ/hr
 $P_B(i, j, p)$ - Flowrate of i from bottoms of j fed to products p in $kmol/hr$
 $P_{B,split}(i, j, p, f)$ – Determination of split fraction of bottoms from column j to product p
 $P_{B,tot}(i, j)$ – Flow of species i in bottoms of column j fed to all products combined in $kmol/hr$
 $P_D(i, j, p)$ - Flowrate of i from distillate of j fed to products p in $kmol/hr$
 $P_{D,split}(i, j, p, f)$ – Determination of split fraction of distillate from column j to product p
 $P_{D,tot}(i, j)$ – Flow of species i in distillate of column j fed to all products combined in $kmol/hr$
 $P_{tot}(p)$ – Total flow of all components in product stream p in $kmol/hr$
 $Q_C(j)$ – Total heat duty across condenser of column j in MJ/hr
 $Q_{C,T}(j, T)$ – Heat duty of condenser associated with column j available at temperature T in MJ/hr
 $Q_R(j)$ – Total heat duty across reboiler of column j in MJ/hr
 $Q_{R,T}(j, T)$ – Heat duty of reboiler associated with column j available at temperature T in MJ/hr
 $SH(T)$ – Calculated surplus heat available at temperature T in MJ/hr
 $T_C(j, T)$ – Temperature of the condenser of column j allocated to discrete temperature T in $^{\circ}C$
 $T_R(j, T)$ – Temperature of the reboiler of column j allocated to discrete temperature T in $^{\circ}C$
 $Z(i, j)$ – Flow of species i in inlet of column j in $kmol/hr$
 $Z_{frac}(j)$ – Proportion of feed fed to the inlet of column j

Binary Variables

- $E(s, T)$ - 1 if stream s present within temperature interval T , 0 otherwise
 $T_{C,A}(j, T)$ – 1 if adjusted temperature of condenser of column j is equal to T , 0 otherwise
 $T_{R,A}(j, T)$ – 1 if adjusted temperature of reboiler of column j is equal to T , 0 otherwise
 $Y_l(s, T)$ - 1 if lower temperature boundary of process stream s is located at temperature T , 0 otherwise

$Y_h(s, T)$ - 1 if higher temperature boundary of process stream s is located at temperature T , 0 otherwise

$\lambda_D(j, j', f)$ - 1 if split fraction from distillate of column j to column j' is $DSF(f)$, 0 otherwise

$\lambda_B(j, j', f)$ - 1 if split fraction from bottoms of column j to column j' is $DSF(f)$, 0 otherwise

$\lambda_{PD}(j, p, f)$ - 1 if split fraction from distillate of column j to column p is $DSF(f)$, 0 otherwise

$\lambda_{PB}(j, p, f)$ - 1 if split fraction from bottoms of column j to products p is $DSF(f)$, 0 otherwise

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