# Optimal Design of Thermally Coupled Distillation Columns

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This paper considers the optimal design of thermally coupled distillation columns and dividing wall columns using detailed column models and mathematical optimization. The column model used is capable of describing both conventional and thermally coupled columns, which allows comparisons of different structural alternatives to be made. Possible savings in both operating and capital costs of up to 30% are illustrated using two case studies.

## 1. Introduction

Distillation is one of the most energy intensive unit operations, and the optimal design of distillation sequences promises significant savings in both capital and operating costs. The use of complex nonstandard distillation columns, such as that suggested by Petlyuk [*1*], can sometimes lead to substantial savings in capital costs as well as in energy consumption in comparison with conventional one-feed two-product distillation columns. Complex columns are also suitable for retrofit design, since they can often be implemented with only small modifications to existing columns.

Figure 1 shows both the conventional and some nonconventional column arrangements for the separation of a mixture into three different products. Configurations a and b are respectively the so-called direct and indirect sequences. Configurations c and d involve one standard column each and a side stream rectifier and side stream stripper, respectively. It is interesting to note that these configurations are already widely used in the petroleum industry and for air separation using cryogenic distillation (e.g. a side rectifier is used for the recovery of argon), but their use in other sectors is much less frequent.

Configuration e is a fully thermally coupled configuration, commonly known as the *Petlyuk column* [*1*, *2*]. Finally, configuration f is a *dividing wall column* [*3*]. This can be thought of as an attempt to place both columns in a Petlyuk arrangement (configuration e) within a single shell, thereby saving on capital costs. A number of these columns are already in industrial use, with reported savings of up to 30% in both capital and energy costs [*4*].

The rest of this paper is structured as follows. The next section reviews some of the earlier work on studying and optimizing nonstandard distillation columns. Section 3 then presents a column superstructure that can be used for obtaining optimal design and operating conditions for nonstandard columns, including, in particular, Petlyuk and dividing wall columns. Section 4 describes a superstructure within which two or more instances of the above column model can be embedded for the purposes of determining the best column sequence. Section 5 is concerned with the solution of the underlying mathematical optimization methodology. Sections 6 and 7 present two examples of the application of our approach to two difficult separations. Finally, some concluding remarks are made in section 8.

## 2. Earlier Work on Thermally Coupled Column Configurations

The properties of the Petlyuk column and related configurations have been the subject of intensive investigation for more than a decade. The main results and conclusions of these studies are outlined below.

**2.1. Selection of Thermally Coupled Column Configurations.** Some early work on optimal distillation column networks was reported by Tedder and Rudd [*5*]. They compared eight different alternatives, including some thermally coupled ones. The comparison was carried out for six distinct ternary mixtures, by optimizing each alternative for a range of feed compositions. The results were formulated in the form of heuristics for the selection of an appropriate configuration for the separation of a given feed on the basis of its composition and difficulty of separation.

In a series of papers, Glinos and Malone [*6*−*9*] developed a short-cut method for screening different distillation network alternatives based on the Underwood equations. The motivation for their work was the combinatorial explosion of the number of discrete flow sheet alternatives that result from allowing complex column configurations as alternatives. For instance, for a five-component mixture, only 14 distinct designs exist that are based only on simple columns. However, using 8 alternative column designs, this number increases to 110 415!

Due to the assumptions of ideal behavior, the Glinos and Malone method is reliable only for nearly ideal and nonazeotropic systems. The vapor rate and the minimum reflux ratio, both of which are critical for the column design, are calculated for a ternary mixture using equations with a complexity comparable to that of, and derived from, the Underwood equations. The method covers conventional columns, side stream col-

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10.1021/ie9802919 CCC: $18.00 
Published on Web 12/11/1998

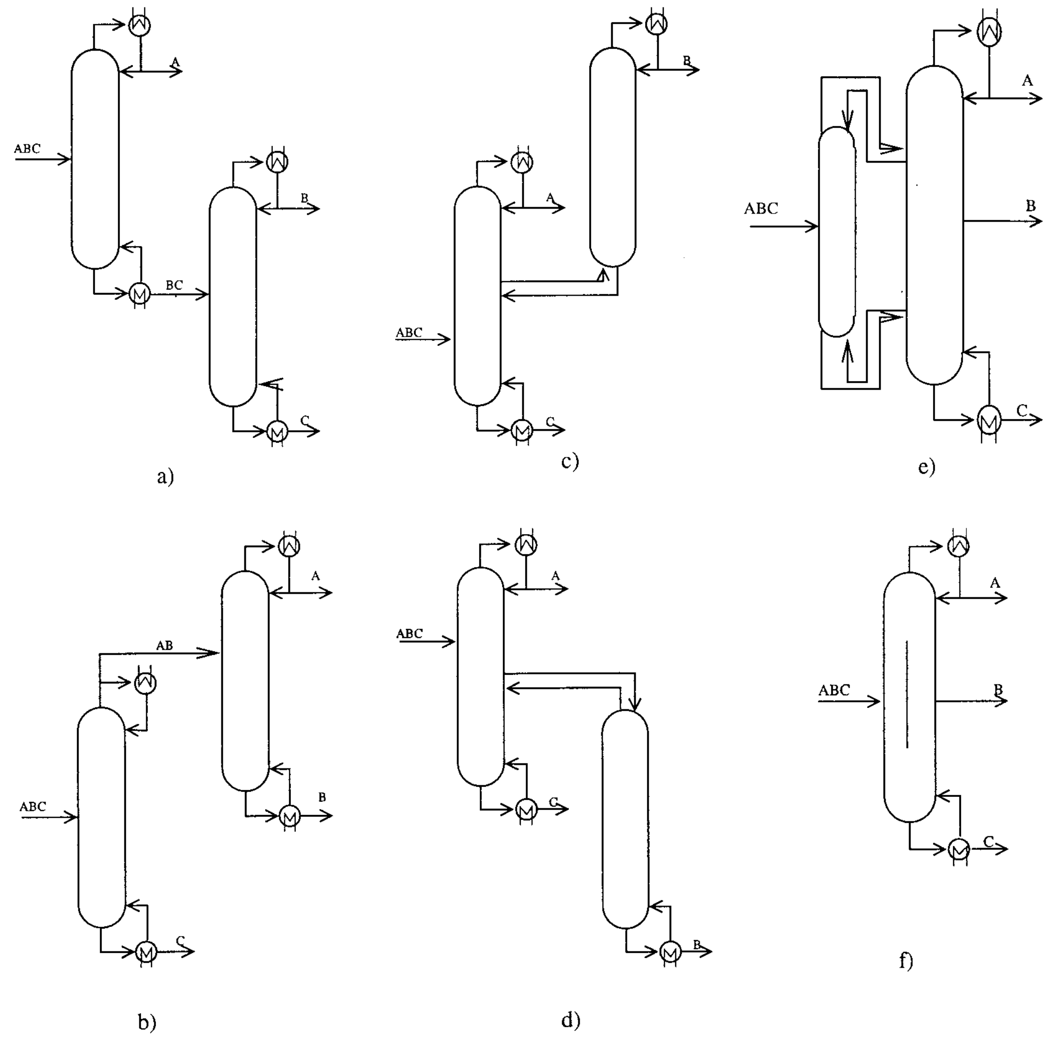



**Figure 1.** Conventional and nonconventional column arrangements for the separation of a mixture into three products.

umns, Petlyuk columns, and others. An extension to a four-component mixture is also possible for all alternatives except the Petlyuk column. In certain cases, Glinos and Malone found that the complex column designs led to savings in both capital and operating costs. In particular, a maximum vapor flow reduction of 50% was found to be possible depending on the feed composition and the relative volatilities of the components in the mixture.

The properties of complex column configurations were also examined by Carlberg and Westerberg [*10, 11*]. On the basis of assumptions of ideal phase equilibrium, constant relative volatilities, and constant molar overflow, these authors extended the Underwood method to configurations with side stream strippers or rectifiers and Petlyuk columns. Temperature-heat diagrams were used to demonstrate some basic thermodynamic properties of these systems. For instance, complex columns are always more energy efficient than conventional columns; this basically means that complex columns are always more favorable with respect to first-law effects. However, they are less favorable with respect to second-law effects due to the larger temperature ranges involved. Therefore, in general, complex columns should be favored provided there is an adequate temperature driving force. This can have an effect on the capital costs for the condenser and the reboiler and also on the operating costs, since utilities at more extreme temperatures tend to be costlier.

Carlberg and Westerberg also showed that energy savings similar to those achieved by complex columns can be obtained using an indirect sequence with a partial condenser in the first column or via heat integration where appropriate.

Annakou and Miszey [*12, 13*] carried out a comparative study of different energy integrated distillation schemes for the separation of a ternary mixture including the Petlyuk column and a heat-integrated two-column scheme. The comparison of the energy savings and the total costs of the investigated schemes show that the two-column heat-integrated scheme is always economically better than the conventional scheme. The Petlyuk column shows considerable energy savings in several cases; however, it can be competitive with the heat-integrated two-column system only in those cases where the concentration of the middle component is high, the split between the first and second components is harder than the split between the second and third components, and the required separation is not too sharp.

It is worth noting that the effect of the required sharpness of the separation could not be quantified in any of the works reviewed in this section, since they were all based on short-cut methods assuming sharp

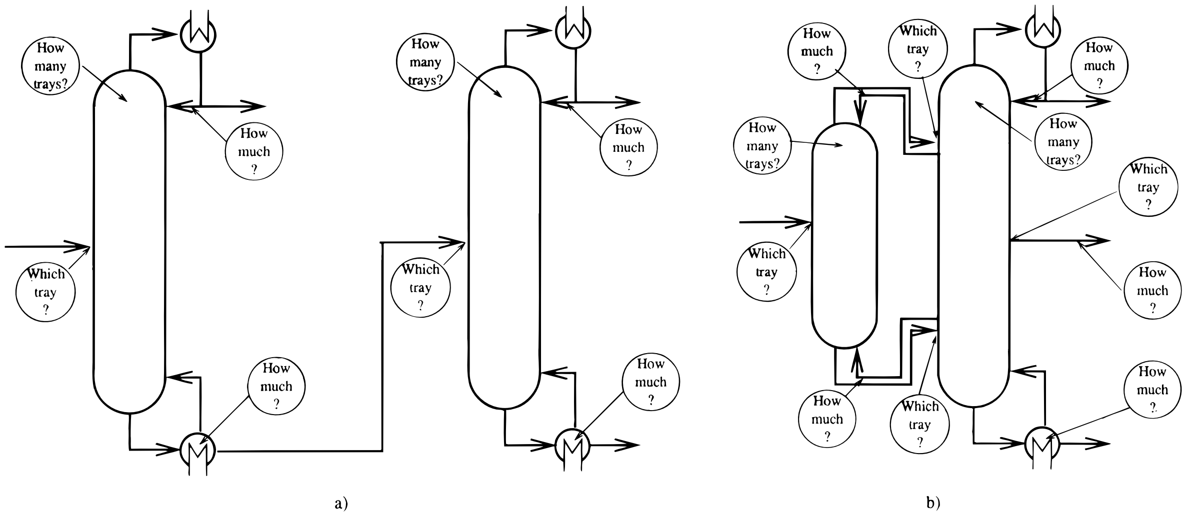



**Figure 2.** Design specifications for (a) direct column sequence and (b) Petlyuk columns.

splits. More detailed studies of Petlyuk columns were carried out by Wayburn and Seader [*14*] using a detailed tray-by-tray model coupled with a differential arc-length continuation technique. Interestingly, these authors observed, for the first time, the existence of multiple steady states in this type of system. Chavez *et al.* [*15*] applied a similar technique to other types of thermally coupled column configuration; a number of different starting points for the continuation paths were used to demonstrate the existence of several solutions, all satisfying the same purity and product flow rate specifications with the same reflux ratio but widely different intercolumn flow rates. Later, Lin *et al.* [*16*] presented a more systematic approach which, by tracing the complete continuation path, managed to produce all solutions from a single starting point.

**2.2. Degrees of Freedom in Thermally Coupled Columns.** In view of the numerous advantages of the Petlyuk and divided wall columns listed above, one would expect a wide application of this type of unit operation in industrial practice. However, in reality, relatively few practical applications of such columns have been documented in the literature to date [*4*, *17*]. The process industry's reluctance toward using this type of design can, at least partly, be attributed to concerns regarding potential control problems and the lack of design procedures. Both of these problems are related to the many more degrees of freedom that the Petlyuk column possesses in comparison to conventional columns. This point is illustrated schematically in Figure 2, which identifies the main decision variables associated with the design and operation of (a) a conventional column network (in this case, the direct sequence, cf. Figure 1a) and (b) a Petlyuk column arrangement.

The effect of the degrees of freedom on the operation and control of Petlyuk columns has been the subject of investigation by Lestak and Smith [*18*] and Wolff and Skogestad [*19*]. It was shown that the problem of dealing with the additional degrees of freedom can be solved with a suitable control strategy and that the Petlyuk column is stable and shows a reasonable dynamic behavior. In fact, the dead time involved in its operation tends to be smaller than that for conventional configurations because of the smaller holdup involved. However, purity specifications for the side product are often difficult to fulfill as it is not possible to constrain the levels of both (in the case of a ternary mixture) impurities in it. Moreover, if it is desired to allow for changes in product specifications, the use of multiple side streams is recommended to provide the required flexibility.

**2.3. Design and Optimization of Thermally Coupled Columns.** Due to the relatively large number of degrees of freedom of the Petlyuk column (cf. Figure 2b), the need for optimal design procedures arose relatively early. Of particular interest is the question of how to optimally specify the "recycle" streams from the main column to the prefractionator. The short-cut methods of Glinos and Malone [*9*] give an approximate tool for solving this problem. Also, a heuristic given by Douglas [*20*] recommends that a sharp split be achieved between the light and heavy components in the first column.

Some attempts to combine the tasks of column design and operational optimization in a single step have already been reported in the literature. This avoids the use of potentially misleading heuristics. The first systematic approach was developed by Fidkowski and Królikowski [*21*], who optimized a linear short-cut model for the Petlyuk column with respect to the reboiler vapor flow. The approach was later extended by Fidkowski and Królikowski [*22*] to consider other thermally coupled systems, including those involving side rectifiers and side strippers. The minimization of vapor flow is somewhat similar to a minimum reflux design for a conventional column. However, it is still questionable whether this objective function is appropriate and whether the accuracy of the model chosen is sufficient for design purposes.

Kakhu and Flower [*23*] allow some unconventional column configurations (including the Petlyuk column) in their MILP formulation for the synthesis of heat-integrated distillation sequences. The column model used for their mathematical formulation performs a mass balance and computes the condenser and reboiler duties as linear functions of the feed stream flow rate. The objective function is a linearized version of a cost function taking both operating and capital costs into consideration.

Because of the simple nature of the column model used by Kakhu and Flower, every model within their superstructure is assigned to a specified separation task and column configuration type. Due to this fact and to the effects of the heat integration, the resulting superstructure contains a rather large number of discrete alternatives. For instance, in the case of the separation of a ternary mixture, the superstructure already contains up to 80 columns. This is another manifestation of the combinatorial explosion effect described by Glinos and Malone [*9*].

Triantafyllou and Smith [*24*] present a stepwise design and optimization procedure for Petlyuk columns based on Underwood short-cut models for three linked distillation columns. Their motivation for using Petlyuk columns is the limitation of process integration of conventional distillation columns, often caused by practical constraints. They also give a very practical explanation for the high energy efficiency of the Petlyuk column. In a conventional column, the concentration of the medium boiling component reaches a maximum somewhere in the column. However, this effect is not exploited, but instead, remixing is allowed to take place. On the other hand, in a thermally coupled column, this maximum appears in the main column and defines the optimal side stream stage. Instead of employing minimum reflux or minimum vapor flow as an optimization criterion, Triantafyllou and Smith present a stepwise optimization algorithm which is indirectly concerned with optimizing the cost of the column.

Beyond the optimization of individual conventional and complex column sequences, various approaches have been reported in the literature to formulate superstructures for distillation-based separations including Petlyuk and other unconventional column configurations. Examples include those by Sargent and Gaminibandara [*25*], Kaibel [*3*], and Agrawal [*26*].

It is clear from the above review that a significant body of work has already been devoted to complex distillation columns, and this has led to improved understanding of the operation of these columns and the benefits that may arise from their use. However, a number of deficiencies common to all of the earlier work can also be identified. For instance, only very simple column models have been used for the design and optimization of these columns, and this places a major question mark on the accuracy and validity of any results obtained. Also the assumption of ideal mixture behavior holds only for a small number of examples. Consequently, an accurate quantitative comparison of the different effects is difficult or impossible.

The mathematical optimization approach presented in this paper is based on a detailed column model (section 3). This model is then embedded within a process superstructure that includes thermally coupled column configurations for both three- and four-component separation (section 4).

## 3. Column Model

**3.1. Modeling Requirements.** The first requirement for our mathematical optimization approach is a model for a distillation column that allows the determination of both structural and operational decisions for all types of column that appear in the various configurations of Figure 1. These include a standard multifeed two-product column, the prefractionator, and the main column in the Petlyuk configuration, and the main column and the side stream rectifier and stripper in configurations c and d, respectively.

Structural decisions include the number of trays, the position of any feed streams and side products, and also the existence or otherwise of a condenser and a reboiler. On the other hand, operational decisions include the reflux ratio in the condenser (if the latter exists), the rate of heat input to the reboiler (also if the latter exists), and the flow rates of the various side streams. The latter may include both a side product stream and streams connecting the column being modeled to others in the overall column sequence.

An inspection of Figure 1 also reveals that the phase (liquid or vapor) of some of the streams leaving the column is not entirely determined by their position. Consider, for instance, the stream leaving the side of the first column in configurations c and d. It is clear that, in the former case, this is a vapor while, in the latter one, it is a liquid. However, if we want to formulate a general column model that can be used in a superstructure that allows *both* of these configurations, we need to allow the phase of any such stream to be determined by the optimization itself. Other similar cases include the following:

(i) The top product stream. Albeit usually a liquid, this stream may be a vapor if the column does not incorporate a condenser; this is the case for the second column in configuration d and the first one in configuration e.

(ii) The side product stream. In principle, this can be either a liquid or a vapor stream.

**3.2. Column Superstructure.** We now proceed to describe a column superstructure that embeds all the structural decisions listed above. This is derived from that presented by Viswanathan and Grossmann [*27*, *28*] for standard columns with single and multiple feeds. This type of approach employs detailed tray models with component material balances, energy balances, and phase equilibrium relations. A maximum number of trays that may be used in the column is postulated. The reflux stream is allowed to be returned to any tray in a specified (top) section of the column. Moreover, the feed stream(s) are allowed to be split among all trays at or below the reflux return position. The latter is treated as a discrete decision to be determined by the optimization. The feed split fractions could also be treated as binary variables to ensure that each feed is introduced at a single tray in the column; however, the results reported by Viswanathan and Grossmann [*28*] and later by Smith [*29*] indicate that this condition is fulfilled by the optimal solutions obtained even when using continuous split fractions.

In order to accommodate the requirements described above, our model has the following additional features (see Figure 3):

(i) The condenser model allows partial or no condensation. The top product of the column may be saturated liquid or vapor, or any mixture of these.

(ii) The column has a side product stream (shown as *Side Product* in Figure 3) which may be withdrawn from any tray in the column. This may be saturated liquid or vapor or any mixture of these.

(iii) Additional feed streams may be introduced in either the top tray or the reboiler or both. These are respectively shown as *Top Feed* and *Bottom Feed* in Figure 3.

(iv) A maximum of two feed streams are being considered. If a feed is introduced at a certain tray, a product stream, either liquid or vapor, may also be

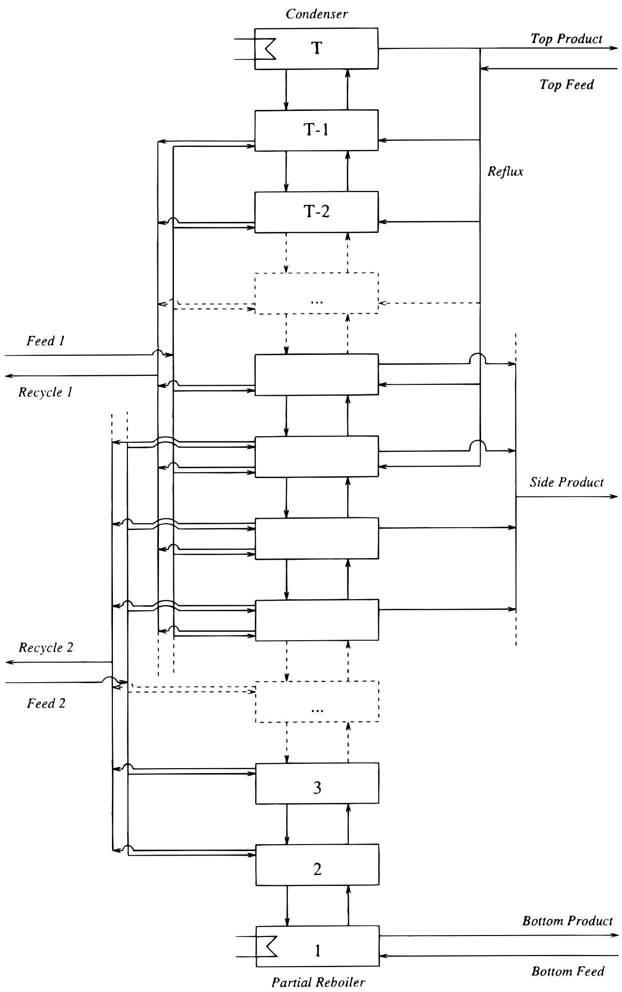



**Figure 3.** Schematic diagram of general column superstructure.

taken from the same tray. These product streams are shown respectively as *Recycle 1* and *Recycle 2* in Figure 3.

**3.3. Model Constraints.** The equations and other constraints describing our column superstructure are presented below. The various symbols used are listed and defined in the Notation section at the end of the paper. However, it may be useful to point out here that the column comprises a maximum of $T_{\max}$ stages, with stage 1 being the reboiler and stage $T_{\max}$ the condenser. The reflux is allowed to be returned to any one of trays $T_{\min}$ to $T_{\max} - 1$; this defines the "top section" of the column, the rest (trays 2 to $T_{\min} - 1$) being its "bottom section".

**3.3.1. Component Material Balances**

Reboiler

$$L_2 x_{2,c} + F^{\text{bot}} x_c^{F^{\text{bot}}} = P^{\text{bot}} x_c^{P^{\text{bot}}} + V_1 y_{1,c} \qquad \forall\, c \in \{1...C\} \quad (1)$$

Bottom section

$$L_{t+1} x_{t+1,c} + V_{t-1} y_{t-1,c} + \sum_{i \in N_{\text{feed}}} f_{t,i} F_i x_{i,c}^F = L_t x_{t,c} + V_t y_{t,c} + \sum_{i \in N_{\text{feed}}} f_{t,i} F_i^{\text{outlet}} x_{i,c}^{F^{\text{outlet}}} + s_t S x_c^{\text{S}} \qquad \forall\, c \in \{1...C\},\ t \in \{2...T_{\min} - 1\} \quad (2)$$

Top section

$$L_{t+1} x_{t+1,c} + V_{t-1} y_{t-1,c} + \sum_{i \in N_{\text{feed}}} f_{t,i} F_i x_{i,c}^F + r_t (R x_{T_{\max},c} F^{\text{top}} x_c^{F^{\text{top}}}) = L_t x_{t,c} + V_t y_{t,c} + \sum_{i \in N_{\text{feed}}} f_{t,i} F_i^{\text{outlet}} x_{i,c}^{F^{\text{outlet}}} + s_t S x_c^{\text{S}} \qquad \forall\, c \in \{1...C\},\ t \in \{T_{\min}...T_{\max} - 1\} \quad (3)$$

Condenser

$$V_{T_{\max}-1} y_{T_{\max}-1,c} = P^{\text{top}} x_c^{P^{\text{top}}} + R x_{T_{\max},c} \qquad \forall\, c \in \{1...C\} \quad (4)$$

**3.3.2. Energy Balances**

Reboiler

$$L_2 h_2^{\text{liq}} + F^{\text{bot}} h^{F^{\text{bot}}} + Q_{\text{reb}} = P^{\text{bot}} h^{P\text{bot}} + V_1 h_1^{\text{vap}} \quad (5)$$

Bottom section

$$L_{t+1} h_{t+1}^{\text{liq}} + V_{t-1} h_{t-1}^{\text{vap}} + \sum_{i \in N_{\text{feed}}} f_{t,i} F_i h_i^{\text{F}} = L_t h_t^{\text{liq}} + V_t h_t^{\text{vap}} + \sum_{i \in N_{\text{feed}}} f_{t,i} F_i^{\text{outlet}} h_i^{F^{\text{outlet}}} + s_t S h^{\text{S}} \qquad \forall\, t \in \{2...T_{\min} - 1\} \quad (6)$$

Top section

$$L_{t+1} h_{t+1}^{\text{liq}} + V_{t-1} h_{t-1}^{\text{vap}} + \sum_{i \in N_{\text{feed}}} f_{t,i} F_i h_i^{\text{F}} + r_t (R h_{T_{\max}}^{\text{liq}} + F^{\text{top}} h^{F^{\text{top}}}) = L_t h_t^{\text{liq}} + V_t h_t^{\text{vap}} + \sum_{i \in N_{\text{feed}}} f_{t,i} F_i^{\text{outlet}} h_i^{F^{\text{outlet}}} + s_t S h^{\text{S}} \qquad \forall\, t \in \{T_{\min}...T_{\max} - 1\} \quad (7)$$

Condenser

$$V_{T_{\max}-1} h_{T_{\max}-1}^{\text{vap}}\ P^{\text{top}} h^{P^{\text{top}}} + R h_{T_{\max}}^{\text{liq}} + Q_{\text{con}} \quad (8)$$

**3.3.3. Mole Fraction Normalization Constraints**

$$\sum_{c \in \{1...C\}} x_{c,t} = \sum_{c \in \{1...C\}} y_{c,t} = 1 \qquad \forall\, t \in \{1...T_{\max}\} \quad (9)$$

**3.3.4. Vapor Fraction Definitions**

Top product

$$f_{\text{vap}}^{P^{\text{Top}}} = \frac{h^{P^{\text{top}}} - h_{T_{\max}}^{\text{liq}}}{h_{T_{\max}}^{\text{vap}} - h_{T_{\max}}^{\text{liq}}} \quad (10)$$

Product streams taken from feed trays

$$f_{\text{vap},i}^{\text{F}} = \frac{h_i^{F^{\text{outlet}}} - \sum_{t=1}^{T_{\max}} f_{t,i} h_t^{\text{liq}}}{\sum_{t=1}^{T_{\max}} f_{t,i} (h_t^{\text{vap}} - h_t^{\text{liq}})} \qquad \forall\, i \in \{1...N_{\text{feed}}\} \quad (11)$$

Side product stream

$$f^{\mathrm{S}}_{\mathrm{vap}} = \frac{h^{\mathrm{S}} - \sum_{t=1}^{T_{\max}} s_t h_t^{\mathrm{liq}}}{\sum_{t=1}^{T_{\max}} s_t (h_t^{\mathrm{vap}} - h_t^{\mathrm{liq}})} \tag{12}$$

Vapor fraction bounds

$$0 \le f^{P^{\mathrm{top}}}_{\mathrm{vap}} \le 1 \tag{13}$$

$$0 \le f^{\mathrm{F}}_{\mathrm{vap},i} \le 1 \qquad \forall\ i \in \{1...N_{\mathrm{feed}}\} \tag{14}$$

$$0 \le f^{\mathrm{S}}_{\mathrm{vap}} \le 1 \tag{15}$$

**3.3.5. Definition of Product Stream Compositions**

Top product

$$x_c^{P^{\mathrm{top}}} = f^{P^{\mathrm{top}}}_{\mathrm{vap}} y_{T_{\max},c} + (1 - f^{P^{\mathrm{top}}}_{\mathrm{vap}}) x_{T_{\max},c} \qquad \forall\ c \in \{1...C\} \tag{16}$$

Product streams taken from feed trays

$$x_{i,c}^{F^{\mathrm{outlet}}} = f^{F}_{\mathrm{vap},i} \sum_{t=1}^{T_{\max}} f_{t,i} y_{t,c} + (1 - f^{\mathrm{F}}_{\mathrm{vap},i}) \sum_{t=1}^{T_{\max}} f_{t,i} y_{t,c} \qquad \forall\ c \in \{1...C\},\ i \in \{1...N_{\mathrm{feed}}\} \tag{17}$$

Side product stream

$$x_c^{\mathrm{S}} = f^{\mathrm{S}}_{\mathrm{vap}} \sum_{t=1}^{T_{\max}} s_t y_{t,c} + (1 - f^{\mathrm{S}}_{\mathrm{vap}}) \sum_{t=1}^{T_{\max}} s_t x_{t,c} \qquad \forall\ c \in \{1...C\} \tag{18}$$

**3.3.6. Pressure Profile Constraints.** Here we assume a constant pressure profile in the column:

$$p_t = p_{t-1} \qquad \forall\ t \in \{1...T_{\max}\} \tag{19}$$

An equation relating the pressure drop to the vapor flow could be used instead if desired.

**3.3.7. Physical Property Constraints.** The model includes equations for computing the specific enthalpies of all liquid and vapor streams as functions of their temperature, pressure, and composition. These are generally of the form

$$h_t^{\mathrm{liq}} = h^{l}(T_t, P_t, x_t) \qquad \forall\ t \in \{1...T_{\max}\} \tag{20}$$

$$h_t^{\mathrm{vap}} = h^{v}(T_t, P_t, y_t) \qquad \forall\ t \in \{1...T_{\max}\} \tag{21}$$

It also contains phase equilibrium equations of the form

$$y_{t,c}\phi_c^{\mathrm{v}}(T_t, P_t, y_t) = x_{t,c}\phi_c^{\mathrm{l}}(T_t, P_t, x_t) \qquad \forall\ c \in \{1...C\},\ t \in \{1...T_{\max}\} \tag{22}$$

where $\phi_c^{\mathrm{v}}$ and $\phi_c^{\mathrm{l}}$ are functions computing respectively the vapor and liquid fugacity coefficients of component $c$.

**3.3.8. Logical Constraints**

Normalization of stream split factors

$$\sum_{t \in \{1...T_{\max}\}} r_t = 1 \tag{23}$$

$$\sum_{t \in \{1...T_{\max}\}} s_t = 1 \tag{24}$$

$$\sum_{t \in \{1...T_{\max}\}} f_{t,i} = 1 \qquad \forall\ i \in \{1...N_{\mathrm{feed}}\} \tag{25}$$

Constraint to force all feed locations below the reflux return position

$$f_{t,i} \le \sum_{j=t}^{T_{\max}} r_j \qquad \forall\ i \in \{1...N_{\mathrm{feed}}\},\ t \in \{1...T_{\max}\} \tag{26}$$

Constraint to force the side product location below the reflux return position

$$s_t \le \sum_{j=t}^{T_{\max}} r_j \qquad \forall\ t \in \{1...T_{\max}\} \tag{27}$$

Feed stream ordering constraints. For columns with multiple feed streams ($N_{\mathrm{feed}} > 1$), a degeneracy may occur in the optimal solution as any reordering of the various feed streams is also an equivalent optimal solution. To avoid this, we introduce the following constraints that number the feed streams in the order in which they are introduced in the column, from top to bottom:

$$f_{t,i+1} \le \sum_{j=t}^{T_{\max}} f_{j,i} \qquad \forall\ i \in \{1...N_{\mathrm{feed}} - 1\},\ t \in \{1...T_{\max}\} \tag{28}$$

**3.3.9. Variable Integrality Constraints**

$$r_t \equiv \begin{Bmatrix} 1, \text{ if the reflux is returned to tray } t \\ 0, \text{ otherwise} \end{Bmatrix} \tag{29}$$

$$s_t \equiv \begin{Bmatrix} 1, \text{ if the side product is withdrawn from tray } t \\ 0, \text{ otherwise} \end{Bmatrix} \tag{30}$$

As already pointed out in section 3.2, it is not necessary to enforce explicitly the integrality of the feed stream split fractions $f_{t,i}$.

**3.4. Estimation of Capital and Operating Costs.** The capital cost of a column is estimated using correlations and data provided by Douglas [*20*]. These are of the form

$$C_{\mathrm{cap}} = (\mathrm{const}_1)\mathrm{Diam}^{1.06} N_{\mathrm{tray}}^{0.802} + (\mathrm{const}_2)(A_{\mathrm{con}}^{0.65} + A_{\mathrm{reb}}^{0.65}) \tag{31}$$

The appropriate diameter for a certain tray is a given function $f_D$ of the corresponding temperature and vapor flow. We assume that the column has a uniform diameter from top to bottom, which is given by

$$\mathrm{Diam} \ge f_{\mathrm{D}}(T_t, V_t) \qquad \forall\ t \in \{2...T_{\max} - 1\} \tag{32}$$

The actual number of trays in the column is determined by the reflux return position [27, 28]:

$$N_{\text{tray}} = \sum_{t=1}^{T_{\max}} r_t(t-1) \tag{33}$$

Finally, other correlations determine the required heat-transfer areas in the reboiler and condenser in terms of the corresponding temperatures and heat duties:

$$A_{\text{reb}} = A_{\text{reb}}(T_1, Q_{\text{reb}}) \tag{34}$$

$$A_{\text{con}} = A_{\text{con}}(T_{T_{\max}}, Q_{\text{con}}) \tag{35}$$

The column operating cost is assumed to be directly proportional to the heating and cooling loads, $Q_{\text{con}}$ and $Q_{\text{reb}}$, respectively. A *capital charge factor* (CCF) is used to introduce the capital cost into a total annualized cost for the column given by

$$C = (c_{\text{C}} Q_{\text{con}} + c_{\text{H}} Q_{\text{reb}})\Theta + (\text{CCF})C_{\text{cap}} \tag{36}$$

where $c_{\text{C}}$ and $c_{\text{H}}$ are the unit costs of the cooling and heating utilities and $\Theta$ is the annual plant operating time.

**3.5. Degrees of Freedom of the Model.** The model described above contains a large number of degrees of freedom as decision variables for the optimization (cf. Figure 2b). These are listed below:

(i) The number of trays in the column, described by the vector of binary variables $r_t$.

(ii) The location of all feeds, described by the matrix of binary variables $f_{t,i}$.

(iii) The location of the side product stream, described by the vector of binary variables $s_t$.

(iv) The flow rates of all recycle streams $F_i^{\text{outlet}}$; if, at the optimal solution, $F_i^{\text{outlet}} = 0$, then the stream does not exist.

(v) The flow rate of the side product $S$; if, at the optimal solution, $S = 0$, then the stream does not exist.

(vi) The reboiler duty $q_{reb}$, which also determines the existence of the reboiler.

(vii) The flow rate of the reflux stream $R$.

(viii) The vapor fractions of the top product $f_{\text{vap}}^{P^{\text{top}}}$, of the side product $f_{\text{vap}}^{\text{S}}$, and of all recycles $f_{\text{vap},i}^{\text{F}}$.

In principle, the column pressure could also be treated as an optimization decision variable. However, for the purposes of this paper, we have decided *not* to do this because our cost models do not take account of the effects of pressure on the column capital cost, nor do they include the operating costs of any compression that may be necessary. Therefore, in the case studies presented in sections 6 and 7, we treat the column pressures as given.

## 4. Column Sequence Superstructure

The column model described in section 3 has been embedded in a superstructure formulation which describes both conventional and thermally coupled distillation columns. This superstructure is essentially a special case of the State Operator Network formulation by Smith and Pantelides [30], which assumes complete connectivity of all unit operations under consideration. The flow rates of all interconnecting streams are treated as continuous variables; the optimization will then set to zero the flow rates of any nonexistent stream.

As pointed out by Smith and Pantelides, the use of detailed models for the individual unit operations in such a completely connected superstructure has two major implications. First, the only discrete decisions associated with the flow sheet structure are those determining the existence of equipment items; thus, for a plant involving up to two columns, there are only two alternatives involving one or two columns, respectively. In fact, in many cases, it is possible to show *a priori* that the required separation cannot be achieved by a single column, and hence the first of these two alternatives may be discarded. The second implication of the use of detailed models is that the optimal design parameters and operating conditions of each unit (cf. section 3.5) are determined simultaneously with the overall process structure.

**4.1. Superstructures for Production of Three- and Four-Product Streams.** Figure 4 shows the superstructure for a network of two columns, that is, one that is suitable for the separation of a mixture into three distinct products. It is also possible to formulate a comparable structure for the separation of mixtures into more than three distinct product streams. Several configurations of this type have been suggested by Sargent and Gaminibandara [25] and Agrawal [26]. Here, we restrict the possible structures by considering only those that can be implemented in a single shell. This is equivalent to a main column with two prefractionators for the separation of a four-component mixture. A configuration like this can be realized in a single column using two dividing walls or a triangular wall, as suggested by Christiansen *et al.* [31]. For reasons of technical realization and in order to minimize any control problems, communicating streams between the middle sections of the prefractionator and the main column shall not be allowed in this work, since they would correspond to streams passing through the dividing wall.

The superstructure for a four-product stream separation and *some* of the configurations that it encompasses are shown in Figure 5. We note that the superstructure of Figure 4 is just a subset of this one, and all the configurations given in Figure 1 are, therefore, also encompassed by this fairly general structure.

**4.2. Implementability Conditions for Dividing Wall Columns.** It will be noted that neither of the superstructures presented in Figures 4 and 5 includes directly columns with dividing walls. However, as discussed in the Introduction, dividing wall columns can be viewed as Petlyuk arrangements in which both columns are fitted within the same shell.

In the context of the superstructure of Figure 4, we assume that the first column may be included within the shell of the second one if *all* of the following conditions hold:

(i) The reboiler heat load in the first column is zero.

(ii) The condenser cooling load in the first column is zero.

(iii) The number of trays in the first column does not exceed that in the second one. More precisely, the number of trays in the second column between the locations of the two recycle streams returning to the first column must be equal to the number of trays in the latter. This ensures that we have the same number of ideal trays on both sides of the dividing wall.

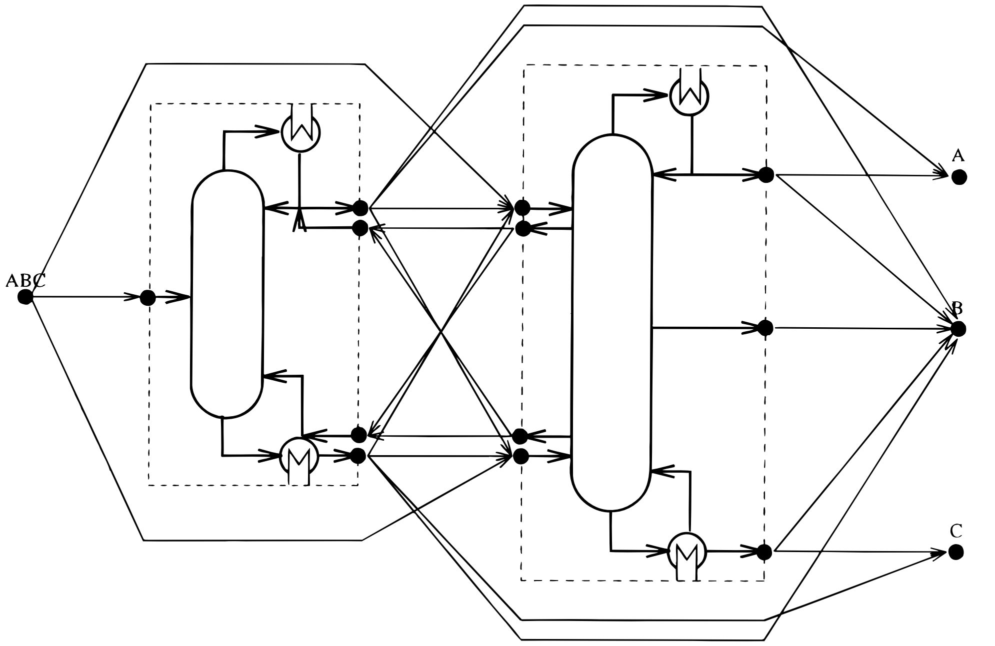



**Figure 4.** Superstructure for separation of the feed stream into three product streams.

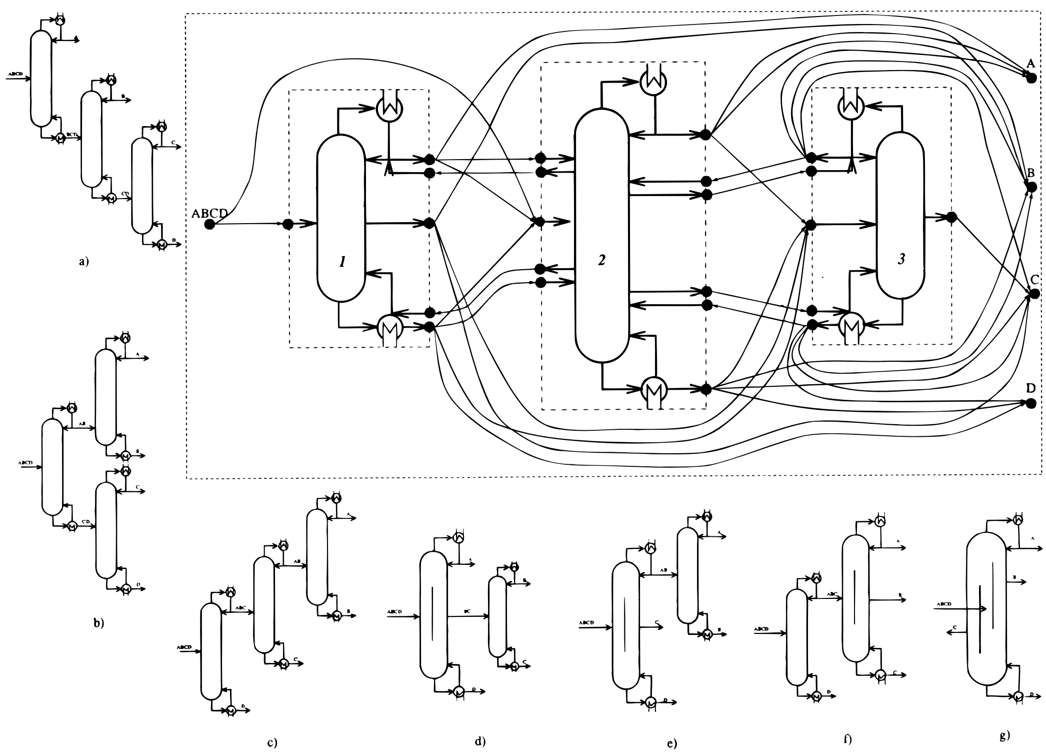



**Figure 5.** Superstructure for separation of the feed stream into four product streams and some of the configurations encompassed by it.

Strictly speaking, the last condition is not always necessary in practice. This is especially the case with packed columns where different types of packing (corresponding to different numbers of equilibrium separation stages) may easily be used on either side of a dividing wall. On the other hand, in the case of tray columns, installing different numbers of trays may somewhat complicate the manufacture of the equipment. This, in turn, will affect the associated capital cost, which may render the simple model used for evaluating the cost of dividing wall columns (cf. section 4.3) invalid. Therefore, for the purposes of this paper, we choose to retain this constraint although it may easily be omitted if so desired.

To describe the above conditions mathematically, we introduce an extra binary variable $\omega^{(12)}$ defined as follows

$$\omega^{(12)} \equiv \begin{Bmatrix} 1, \text{ if column 1 is fitted within the} \\ \text{shell of column 2} \\ 0, \text{ otherwise} \end{Bmatrix} \qquad (37)$$

and relate it to the variables that we have already introduced (cf. section 33) via the following logical constraints:

$$Q^{(1)}_{\mathrm{reb}} \le M^{(1)}_{\mathrm{reb}}(1 - \omega^{(12)}) \tag{38}$$

$$Q^{(1)}_{\mathrm{con}} \le M^{(1)}_{\mathrm{con}}(1 - \omega^{(12)}) \tag{39}$$

$$N^{(1)}_{\mathrm{tray}} - T^{(1)}_{\max}(1 - \omega^{(12)}) \le \sum_t (f^{(2)}_{t,1} - f^{(2)}_{t,2})t \tag{40}$$

Here the superscripts (1) and (2) denote the two columns, respectively, and $M^{(1)}_{\mathrm{reb}}$ and $M^{(1)}_{\mathrm{con}}$ are upper bounds on $Q^{(1)}_{\mathrm{reb}}$ and $Q^{(1)}_{\mathrm{con}}$, respectively. Note that, because of the cost model adopted for dividing wall columns (see section 4.3), constraint 40 is always active if $\omega^{(12)} = 1$.

A new binary variable $\omega^{(32)}$ and a set of constraints similar to eqs 38–40 can be introduced in the mathematical formulation for the superstructure of Figure 5 to allow for the possibility of incorporating the third column within the shell of the second one.

We note that the column model described in section 3 is valid for the prefractionator even if it is fitted within the same shell as the main column, *provided* heat transfer across the dividing wall is negligible. Lestak and Smith [*18*] provide a detailed discussion of the possible effects of such heat transfer.

**4.3. Capital Cost of Dividing Wall Columns.** The possibility of one column being incorporated within the shell of another introduces a complication regarding the capital cost of the overall arrangement, as this clearly depends on the mechanical engineering details of its manufacture. For the purposes of the present paper, instead of evaluating the cost of the first column separately, we treat its cost as a *surcharge* imposed on the cost of the second column. This surcharge is intended to reflect the additional cost of installing the dividing wall; alternative cost models (e.g. based on a *discount* applied to the combined cost of *both* columns) could also be used within the optimization framework presented here. It should be noted that the increase in shell diameter that is necessary to accommodate both columns within the same shell is *already* taken into account: the diameter of the second column is determined by eq 32, the right-hand side of which normally attains its maximum value for a tray $t$ receiving the *combined* vapor flow rate from both sides of the dividing wall.

To achieve the above effect, we replace eq 31 *for this column* by the following two inequalities:

$$C^{(1)}_{\mathrm{cap}} \ge (\mathrm{const}_1)\mathrm{Diam}^{1.06} N^{0.802}_{\mathrm{tray}} + (\mathrm{const}_2)(A^{0.65}_{\mathrm{con}} + A^{0.65}_{\mathrm{reb}}) - M\omega^{(12)} \tag{41}$$

$$C^{(1)}_{\mathrm{cap}} \ge \mathrm{SF}\, C^{(2)}_{\mathrm{cap}} - M(1 - \omega^{(12)}) \tag{42}$$

where $M$ is a an upper bound on the capital cost of the first column and $SF$ is the surcharge factor. We note that the optimization will ensure that one of these inequalities will be active at the optimal solution, depending on the value of the binary variable $\omega^{(12)}$.

A similar modification is introduced for the capital cost of the third column in the superstructure of Figure 5.

## 5. Optimization Methodology

In practice, the determination of the optimal plant structure using the models detailed in sections 3 and 4 can be guaranteed only if the optimization problem is solved to global optimality. However, despite much recent progress (see, for instance, the review given by Smith [*29*]), global optimization techniques are still far from being able to solve problems of the size and complexity of those considered here. For the purposes of this work, we have relied on the use of a *local* optimization code, CONOPT [*32*] interfaced to the *gPROMS* process-modeling tool [*33*, *34*].

A feasible solution was used as the initial starting point for all the optimizations carried out. Unfortunately, we found that the choice of initial point often influenced the “optimal” solution determined, with the two usually having the same column connectivity. On the other hand, the design (e.g. number of trays, feed and side stream locations, etc.) of the individual columns was found to be much less sensitive to the initial point selected; the same was found to be true for the column operating variables (e.g. reflux ratios, heat duties, etc.).

In view of the above observations, we carried out a number of optimization runs for each of the problems considered. Each run studied a different configuration by setting the flow rates of a subset of the streams in the superstructure to zero. It is worth emphasizing that *all* the decisions listed in section 3.5 for *all* the columns under consideration were treated as variables to be determined by each optimization run. Thus, only the connectivity of the various columns was fixed a priori.

The complications arising from the discrete nature of the decisions associated with the reflux return and side stream locations (cf. constraints 29 and 30) were handled using a branch-and-bound approach. Thus, the integrality requirements for the binary variables $r_t$ and $s_t$ were initially relaxed, allowing them to take *any* value between 0 and 1. If any of these variables was found to have an nonintegral value at the (locally) optimal solution, two more optimizations were performed, with this particular variable being fixed to 0 or 1, respectively (“branching”). This was repeated until all possibilities were considered either explicitly (by fixing variable values as described above) or implicitly (by “bounding”, i.e., by comparing the current value of the objective function with that of the best feasible solution obtained so far). Our experience confirms the observation of Smith [*29*] that this kind of approach terminates within very few (of the order of 5) iterations even when multiple columns with large numbers of trays are considered and that there is no need to enforce the integrality of the feed location variables, as these tend to adopt integral values anyway (cf. section 3.2). Of course, in view of the use of a local optimization code, there is no guarantee that the globally optimal solution is actually obtained. Nevertheless, the approach has been applied successfully to a range of distillation-based separation problems with good results: some of these are described below; others are reported by Dünnebier [*35*].

## 6. Case Study I—The Separation of Close Boiling C4’s

**6.1. Problem Description.** The separation of close boiling C4 hydrocarbons is one of the most difficult separation tasks encountered in refineries. Here we consider a liquid feed mixture of isobutane (4.9 mol %), 1-butene (50.71 mol %), *n*-butane (6.95 mol %), *trans*-2-butene (9.46 mol %), and *cis*-2-butene (27.98 mol %), at its boiling point. The components are listed in the order of decreasing volatility; the primary objective is

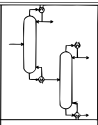
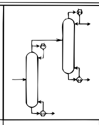
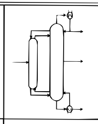
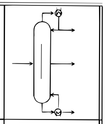
| |  |  |  |  |
|---|---|---|---|---|
| Number of theoretical trays | | | | |
| Column no.1 | 60 | 82 | 58 | 54 |
| Column no.2 | 100 | 67 | 103 | 109 |
| Capital cost[$] | 2,283,720 | 2,121,210 | 2,249,550 | 1,919,060 |
| Reboiler duty [kW] | | | | |
| Column no.1 | 8,147 | 14,412 | 0 | 0 |
| Column no.2 | 13,789 | 6,975 | 15,350 | 14,540 |
| $\Sigma$ | 21,936 | 21,387 | 15,350 | 14,540 |
| Operational cost [$ p.a.] | 1,125,250 | 1,097,040 | 800,870 | 757,370 |
| Relative energy savings [%] | -2.5 | 0 | 28.2 | 32.0 |
| Total annualised cost [$ p.a.] | 2,038,740 | 1,945,520 | 1,700,600 | 1,525,350 |
| Relative cost savings [%] | -4.8 | 0 | **12.6** | **21.6** |

**Figure 6.** Comparison of different structural alternatives for separation of close boiling C4's.

to separate the middle boiling component 1-butene with a purity of 95 mol % and a recovery of 95%. This specification leaves the precise split between the light and heavy components to be determined by the optimization. A similar case study was considered by Triantafyllou and Smith [*24*] as an application of their design and optimization strategy for Petlyuk columns.

The energy requirement of this separation is considerable, and application of heat integration could lead to significant savings. However, to achieve the temperature driving force required for heat integration, one of the columns would have to operate at a higher pressure. Unfortunately, for this separation, such an increase in pressure cannot be achieved without excessive fouling of the reboiler. Consequently, we are led to consider the use of a fully thermally coupled column as an alternative to heat integration. The pressure is fixed at 6 bar throughout the system. The pay back period for the capital is 2.5 years, which corresponds to a capital charge factor (CCF) (cf. eq 36) of 0.4.

**6.2. Problem Solution.** The model has been formulated using the superstructure of Figure 4. The maximum numbers of trays allowed in the two columns were 90 and 115, respectively; these values were found to be sufficiently large for the optimal solution not to lie on the maximum column size constraint.

Vapor and liquid enthalpies for pure components were computed on the basis of cubic polynomials in temperature for the ideal gas-phase specific heat capacity, and the Clausius Clapeyron equation for the enthalpy of vaporization. The vapor pressures $p_c^{\mathrm{vap}}$ of components $c$ at a temperature $T$ were computed using expressions of the form

$$\ln\left(\frac{p_c^{\mathrm{vap}}}{p_c^{\mathrm{crit}}}\right) = \frac{1}{1-z}(\pi_{c,1}z + \pi_{c,2}z^{3/2} + \pi_{c,3}z^3 + \pi_{c,4}z^6) \quad (43)$$

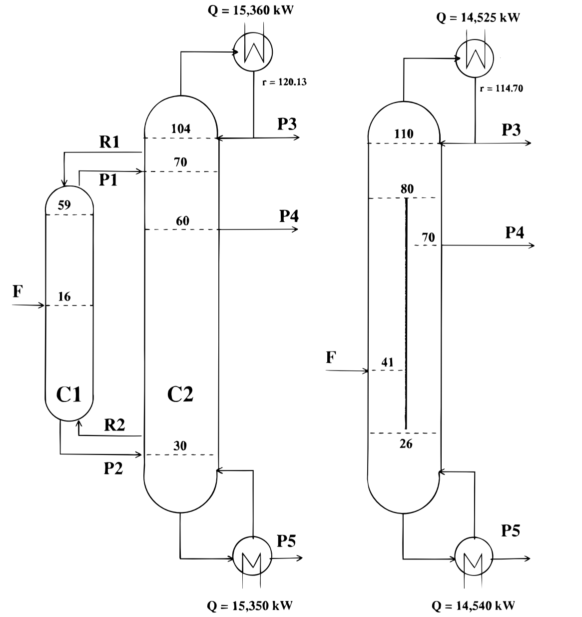
where $z \equiv 1 - T/T_c^{\mathrm{crit}}$, $T_c^{\mathrm{crit}}$ and $p_c^{\mathrm{crit}}$ denote respectively the critical temperature and pressure of component $c$, and $\pi_{c,i}$, $i = 1, ..., 4$ are given coefficients. All necessary data were obtained from Reid *et al.* [*36*]. Ideal mixture behavior was assumed for both the liquid and the vapor phases. The validity of this assumption was checked *a posteriori* by carrying out a simulation of the optimal solutions obtained using the Soave−Redlich−Kwong equation of state. No significant difference in the results was observed.

As explained in section 5, separate optimization runs were carried out for different column connectivities. The optimal solutions obtained are outlined in Figure 6. The solution for the dividing wall column was obtained using the modifications described in sections 4.2 and 4.3 with a surcharge factor of 15% ($SF = 0.15$, cf. constraint 42). The initial guess for this optimization was the optimal Petlyuk design obtained previously. We note that this guess violates the dividing wall implementability constraint (eq 40), since there are only 40 trays in the main Petlyuk column between the upper and lower recycle streams while the prefractionator has 58 trays. However, the final dividing wall design obtained does satisfy the constraint.

Compared to the best conventional design (in this case, the indirect sequence), the Petlyuk column saves 28.2% of the total energy and 12.6% of the total cost, and the dividing wall column 32.0% and 21.6%, respectively. We note that the savings achieved by two columns sharing the same shell allows the dividing wall arrangement to be implemented at a significantly lower capital cost while simultaneously reducing the reboiler heat duty.

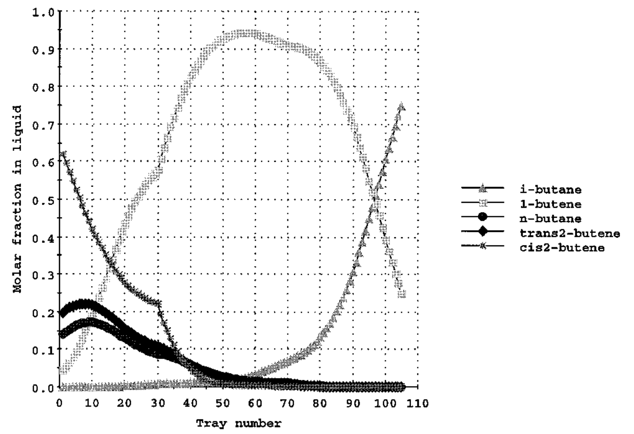
The detailed design and operating parameters of the Petlyuk and dividing wall columns may be seen in Figure 7. It can be seen that the imposed purity and recovery specifications are satisfied. However, stream P3 still contains a relatively high amount of the valuable product 1-butene. This indicates that a modified objective function taking account of the revenue realized by the sale of 1-butene might be more appropriate than an arbitrary recovery specification. Fortunately, this type of modification can easily be incorporated within the objective function discussed in section 3.4 (cf. eq 36).

Some insight into the high thermodynamic efficiency of the Petlyuk column in comparison with more conventional designs can be obtained from the composition profiles shown in Figure 8. The upper part of the figure

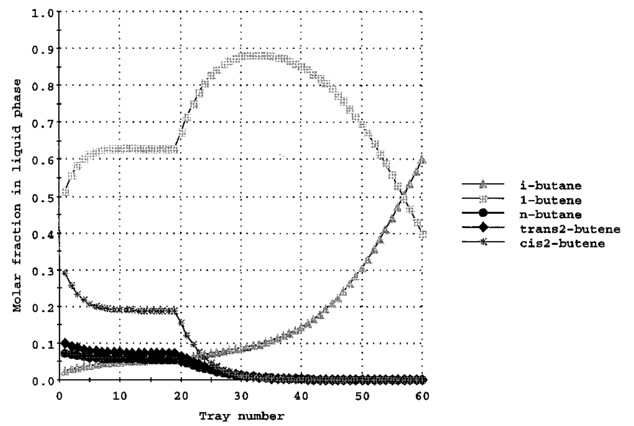



| | Petlyuk Column | | | | Dividing Wall Column | | | |
|---|---|---|---|---|---|---|---|---|
| | F | P1 | P2 | R1 | R2 | P3 | P4 | P5 |
| Flow [kmol/hr] | 500.0 | 1296.6 | 1614.5 | 1139.6 | 1271.5 | 21.7 | 253.6 | 224.8 |
| i-butane [%] | 4.90 | 7.79 | 1.29 | 7.09 | 1.30 | 75.53 | 3.21 | 0.00 |
| 1-butene [%] | 50.71 | 91.30 | 54.76 | 92.05 | 60.20 | 24.47 | 95.00 | 3.28 |
| n-butane [%] | 6.95 | 0.49 | 9.18 | 0.46 | 9.03 | 0.00 | 1.00 | 14.34 |
| trans2-butene [%] | 9.46 | 0.35 | 11.25 | 0.32 | 10.63 | 0.00 | 0.71 | 20.25 |
| cis2-butene [%] | 27.98 | 0.06 | 23.52 | 0.08 | 18.84 | 0.00 | 0.08 | 62.13 |

**Figure 7.** Optimal design and operating parameters for Petlyuk and dividing wall columns for separation of close boiling C4's.




(a)




(b)

**Figure 8.** Composition profiles for (a) the main Petlyuk column and (b) the first column of the direct sequence.

shows the composition profiles in the main column of the optimal Petlyuk column. The two kinks at trays 30 and 70 correspond to the location of the two pairs of feed and recycle streams. The side product is taken precisely at the location of the maximum concentration of the middle component at tray 60. The prefractionator gives a top product which is nearly free of the heavy components *n*-butane, *trans*-2-butene, and *cis*-2-butene, and a bottom product nearly free of the light component isobutane.

For comparison purposes, the lower part of Figure 8 shows the composition profiles in the first column of the optimal direct sequence. A maximum concentration of the middle component 1-butene in the middle region of the column can clearly be seen. However, as already pointed out by Triantafyllou and Smith [*24*], this effect is not exploited by sequences of simple distillation columns. Instead, the remixing that occurs results in thermodynamic inefficiency. To a certain extent, this can be avoided by using a column with a side stream, but the achievable purity of the side stream may be limited.

**6.3. Effects of Product Purity Specifications.** As described in section 2, earlier work reported in the literature has generally made use of short-cut methods based on the assumption of sharp splits. Therefore, it is difficult to apply the rules and heuristics derived to predict the performance of Petlyuk or dividing wall columns in cases of nonsharp separations. Our optimization approach, being based on detailed column models, does not suffer from this limitation.

To examine further the influence of the product purity requirements on the designs obtained, we consider two new cases with desired purities of 1-butene of 97 and 98 mol %, respectively. The designs obtained in section 6.2 were used as the initial guesses for the corresponding runs in the first case; the solutions obtained were then used as the initial guesses for the corresponding second-case runs.

The results for the different structural alternatives for the two cases are summarized in Tables 1 and 2, respectively. When these results are compared with each other and with the results of Figure 6, it is clear that the energy costs increase significantly as the purity specification is tightened. Also, the *relative* advantage of the Petlyuk and dividing wall columns in comparison with the more conventional configurations generally decreases with increasing purity. However, the *absolute* differences in performance remain high.

It is worth mentioning that Annakou and Miszey [*13*] suggested that thermally coupled columns should be considered only for lower product purities, with heat-integrated column sequences being preferable for higher purities. Unfortunately, for the reasons explained in section 6.1, such sequences are not suitable for this particular separation.

As a further investigation, the desired product purity was relaxed to a lower level of 90% while still requiring a recovery of 95% of the 1-butene. A simple mass balance shows that, in this case, it may be possible to satisfy these constraints using only a single column without producing a separate light fraction. Alternatively, a single column with a side stream may also provide a feasible solution to this problem. The problem has been solved using the following three initial guesses, taken from the results reported in Figure 6: (i) the optimal solution of the original Petlyuk column design, (ii) the optimal design of the direct sequence, and (iii) the optimal design of the indirect sequence.

All optimizations result in a single column being identified as the locally optimal solution; in the first case, this is a column with a side product stream, and in the other two cases, this is a standard one-feed two-product column. The key design and operational parameters for these columns are presented in Figure 9. The annualized costs of the two columns are \$1,000,-130 p.a. and \$1,202,560 p.a.; thus, the side stream column is, in fact, 16.8% cheaper than the standard one.

## 7. Case Study II—Separation of Alkane Mixtures

**7.1. Problem Description.** Here we consider the separation of a stream of 50 kmol/h of a mixture of 10 mol % 2-methylbutane, 40 mol % pentane, 40 mol % hexane, and 10 mol % heptane. The feed is boiling liquid, and the pressure in the columns is fixed at 3 bar. The mixture is to be separated into streams of hexane and heptane with a purity of 99 mol % each, and a stream of pentane with a purity of 95 mol % recovering 93% of the pentane in the feed stream. This problem involves both simple separation tasks (like the hexane/heptane one) and a more difficult one (2-methylbutane/pentane).

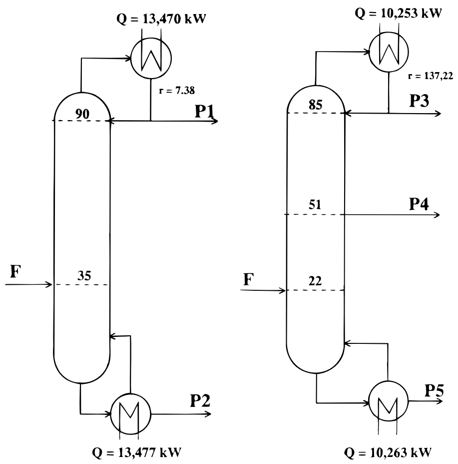
This separator network is designed using the three-column superstructure displayed in Figure 5. This formulation provides both an approach for the optimal design of the extension of the Petlyuk idea to a four-

**Table 1. Comparison of Different Structural Alternatives for a Required Product Purity of 97%**

| | direct sequence | indirect sequence | Petlyuk configuration | dividing wall column |
|---|---|---|---|---|
| no. of theoretical trays | | | | |
| column no. 1 | 77 | 105 | 63 | 61 |
| column no. 2 | 108 | 83 | 113 | 113 |
| capital cost ($) | 2,680,240 | 2,583,850 | 2,564,920 | 2,136,410 |
| reboiler duty (kW) | | | | |
| column no. 1 | 11 424 | 13 874 | 0 | 0 |
| column no. 2 | 13 614 | 9 081 | 17 378 | 17 682 |
| Σ | 25 068 | 22 955 | 17 378 | 17 682 |
| operational cost ($ p.a.) | 1,284,370 | 1,177,500 | 905,220 | 921,050 |
| relative energy savings (%) | −9.2 | 0 | 24.3 | 23.0 |
| total annualized cost ($ p.a.) | 2,356,460 | 2,211,040 | 1,931,210 | 1,775,630 |
| relative cost savings (%) | −6.6 | 0 | 12.7 | 19.7 |

**Table 2. Comparison of Different Structural Alternatives for a Required Product Purity of 98%**

| | direct sequence | indirect sequence | Petlyuk configuration | dividing wall column |
|---|---|---|---|---|
| no. of theoretical trays | | | | |
| column no. 1 | 88 | 90 | 63 | 63 |
| column no. 2 | 108 | 77 | 113 | 113 |
| capital cost ($) | 2,900,230 | 2,588,940 | 2,824,770 | 2,344,660 |
| reboiler duty (kW) | | | | |
| column no. 1 | 13 218 | 14 955 | 0 | 0 |
| column no. 2 | 13 690 | 11 876 | 21 010 | 20 951 |
| Σ | 26 908 | 26 781 | 21 010 | 20 951 |
| operational cost ($ p.a.) | 1,380,290 | 1,359,500 | 1,094,400 | 1,091,310 |
| relative energy savings (%) | −0.5 | 0 | 21.5 | 21.8 |
| total annualized cost ($ p.a.) | 2,540,380 | 2,395,070 | 2,224,320 | 2,029,200 |
| relative cost savings (%) | −6.1 | 0 | 7.2 | 15.3 |




Standard Column / Sidestream Column

| | F | P1 | P2 | P3 | P4 | P5 |
|---|---|---|---|---|---|---|
| Flow [kmol/hr] | 500.0 | 267.7 | 232.3 | 12.7 | 267.64 | 219.7 |
| i-butane [%] | 4.90 | 9.15 | 0.00 | 71.47 | 5.71 | 0.07 |
| 1-butene [%] | 50.71 | 90.00 | 5.45 | 28.53 | 90.00 | 4.13 |
| n-butane [%] | 6.95 | 0.57 | 14.30 | 0.0 | 2.22 | 13.11 |
| trans2-butene [%] | 9.46 | 0.28 | 20.03 | 0.0 | 2.07 | 19.01 |
| cis2-butene [%] | 27.98 | 0.00 | 60.22 | 0.0 | 0.0 | 63.68 |

**Figure 9.** Comparison of different structural alternatives for a required product purity of 90%.

component separation and a rich superstructure containing all structural alternatives already discussed in the previous sections. In the case of a three-column flow sheet, the number of alternative structures increases significantly. There are already five different conventional arrangements, another four containing a dividing wall column with one wall, one with a column with two dividing walls, and several others with a side stripper and/or rectifier.

The desired pay back time is 5 years, which corresponds to a capital charge factor (CCF) (cf. eq 36) of 0.2. Configurations involving columns with one or two dividing walls were handled using the modifications described in sections 4.2 and 4.3 applied to the first and/or the third columns. The surcharge factor for *one* column (either the first or the third) sharing the second column's shell was set at 15% (SF = 0.15, cf. constraint 42). For the case of *both* the first and the third columns sharing the second column's shell, the surcharge factor was set at 10% for *each* one of them.

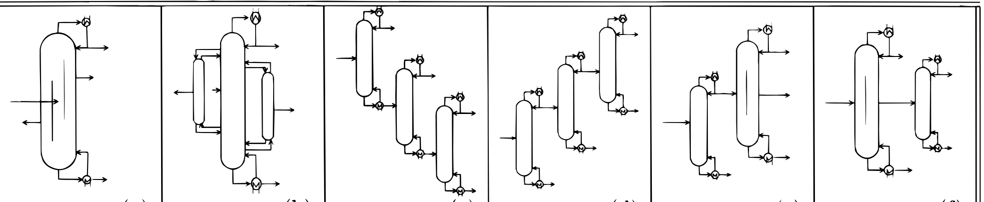
The necessary physical properties were computed in a manner similar to that used for Case Study I (see section 6.2).

**7.2. Problem Solution.** The model was formulated allowing a maximum number of 45 trays in the first and third columns and 55 trays in the second column. To minimize problems with locally optimal solutions in the column connectivity, the model was optimized for several fixed connectivities corresponding to six different structural alternatives using a strategy similar to that described in the previous section. For all cases involving dividing wall columns, the corresponding Petlyuk optimal designs were used as initial guesses for the optimizations.

A comparative summary of the key results is given in Figure 10. We note that the two sequences of conventional columns (alternatives c and d) result in both the highest annualized cost and the highest energy consumption among all those considered. The relative savings of the other structures are expressed with respect to the cheaper of these two conventional configurations.

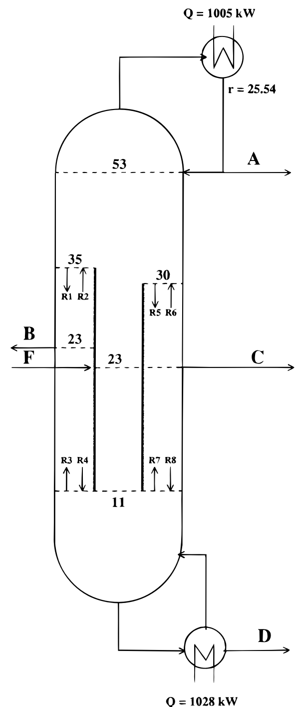
The best design, with a total annualized cost of $112,560 p.a., is a column with two dividing walls (alternative a). The optimal design and operational parameters for this solution are displayed in Figure 11.

It is also interesting to note that the most energy efficient alternative is configuration (b). This structure, first proposed by Agrawal [*26*], involves a main column surrounded by two satellite columns. The results indicate that this type of arrangement can achieve up to one third energy savings in comparison to conventional



| Structural Alternative | (a) | (b) | (c) | (d) | (e) | (f) |
|---|---|---|---|---|---|---|
| Number of theoretical trays | | | | | | |
| Column no.1 | 24 | 28 | 38 | 21 | 20 | 20 |
| Column no.2 | 53 | 52 | 30 | 29 | 50 | 49 |
| Column no.3 | 19 | 21 | 23 | 39 | 27 | 29 |
| Capital cost[$] | 295,240 | 359,970 | 327,030 | 338,270 | 306,410 | 314,280 |
| Reboiler duty [kW] | | | | | | |
| Column no.1 | 0 | 0 | 359 | 583 | 578 | 0 |
| Column no.2 | 1,028 | 983 | 402 | 431 | 754 | 786 |
| Column no.3 | 0 | 0 | 718 | 613 | 0 | 389 |
| $\sum$ | 1,028 | 983 | 1,480 | 1,626 | 1,331 | 1,175 |
| Operational cost [$ p.a.] | 53,510 | 51,170 | 79,990 | 84,630 | 69,590 | 61,130 |
| Relative energy savings [%] | 30.5 | 33.5 | 0 | -9.9 | 10.0 | 20.6 |
| Total annualised cost [$ p.a.] | 112,560 | 123,180 | 142,400 | 152,310 | 130,870 | 124,400 |
| Relative cost savings [%] | **21.0** | **13.5** | 0 | -7.0 | 8.1 | **12.6** |

**Figure 10.** Comparison of different structural alternatives for alkane separation.




| | F | A | B | C | D | R1 | R2 |
|---|---|---|---|---|---|---|---|
| Flow [kmol/hr] | 50.0 | 5.40 | 19.58 | 20.13 | 4.88 | 83.87 | 67.47 |
| 2-methyl-butane [%] | 10.00 | 74.82 | 4.89 | 0.01 | 0.00 | 13.68 | 16.07 |
| Pentane [%] | 40.00 | 25.18 | 95.00 | 0.19 | 0.00 | 86.32 | 83.93 |
| Hexane [%] | 40.00 | 0.00 | 0.11 | 99.00 | 1.00 | 0.00 | 0.00 |
| Heptane [%] | 10.00 | 0.00 | 0.00 | 0.80 | 99.00 | 0.00 | 0.00 |

| | R3 | R4 | R5 | R6 | R7 | R8 |
|---|---|---|---|---|---|---|
| Flow [kmol/hr] | 56.61 | 53.43 | 36.54 | 32.00 | 30.07 | 14.47 |
| 2-methyl-butane [%] | 0.86 | 0.30 | 10.98 | 12.60 | 0.00 | 0.00 |
| Pentane [%] | 9.88 | 5.16 | 57.61 | 65.65 | 0.00 | 0.00 |
| Hexane [%] | 62.10 | 65.76 | 31.15 | 21.75 | 95.24 | 88.64 |
| Heptane [%] | 27.16 | 28.78 | 0.27 | 0.00 | 4.75 | 11.35 |

**Figure 11.** Optimal design of dividing wall column for alkane separation.

sequences. The use of a divided wall column (configuration a) does not lead to lower energy consumption in this case; this is because the upper bounds on the number of trays in the prefractionator (eq 40) proves to be somewhat restrictive. However, alternative a is cheaper overall because of the capital savings achieved by having all columns situated within the same shell.

Finally, it is worth noting that configuration f is the one suggested by the heuristics presented in the work of Glinos and Malone [*9*] and Nicolaides and Malone [*37*]. Albeit not the best one in this case, this solution does achieve substantial savings of 20.6% in energy and 12.6% in total cost over the conventional sequences.

## 8. Conclusions

This paper has considered the design of column sequences involving both conventional and fully thermally coupled columns. The latter include both Petlyuk-type columns and columns with one or two internal dividing walls. Our approach is based on the use of detailed column superstructures coupled with mathematical optimization, and it allows the simultaneous determination of both the design and the operational characteristics of the individual columns.

Two higher level superstructures for the separation of mixtures into three and four distinct product streams, respectively, were also proposed. However, our experience has been that these are currently of limited use because the underlying mathematical problems cannot be solved to global optimality, and local optimization codes tend to get trapped within the flow sheet connectivity corresponding to the initial guess used. For this reason, our approach has been to use these superstructures only as a means for systematically generating a number of candidate column connectivities and then to optimize the columns and the streams connecting them within each such fixed connectivity. Future advances in global optimization techniques may render this kind of approach unnecessary, allowing the direct optimization of the superstructures presented in this paper.

Another important issue is that of thermophysical properties. In principle, the formulation presented here is not limited to any particular method of computing physical properties. However, the examples studied were concerned with fairly ideal mixtures, and it is likely that nonidealities (e.g. such as those encountered in azeotropic mixtures) will both increase the computational complexity and introduce further difficulties regarding the global optimality of the solutions obtained. On the other hand, the physical property correlations used in this paper have removed some of the restrictive assumptions of much of the earlier work on thermally coupled distillation systems, namely those of constant relative volatility and constant molar overflow. As pointed out in a recent paper [*38*], "*for real mixtures, relative volatilities are generally not constant along the length of the distillation column*", from which it is concluded that analyses based on constant relative volatility assumptions "*...may need to be done for more than one set of values of relative volatilities*".

Notwithstanding the mathematical difficulties mentioned above, the approach presented has been applied successfully to several examples [*35*], two of which have been described in this paper. Our results confirm the conclusions of earlier work in this area that substantial benefits in both operating and capital costs may be achieved by the use of nonstandard distillation columns. Moreover, because of the nature of the mathematical models used in our work, our solutions contain the detailed information that is necessary in order to design and operate these columns, thus helping to overcome some of the obstacles associated with the practical use of such equipment in the past. Of course, the operability and control of such columns remain challenging problems that are currently receiving attention in the literature (see, for instance, the recent work by Abdul Mutalib and Smith [*39*] and Abdul Mutalib *et al.* [*40*]).

## Acknowledgment

G.D. would like to thank Professor Sebastian Engell of the University of Dortmund for his guidance and support during this project. The financial support of the Ernest-Solvay-Stiftung is gratefully acknowledged.

## 9. Notation

*Parameters*

$C$ = number of components within the column
$N_{\mathrm{feed}}$ = number of different feed streams
$T_{\min}$ = minimum number of stages below the reflux return position
$T_{\max}$ = maximum number of all stages within the column including the condenser (stage $T_{\max}$) and the reboiler (stage 1)
CCF = capital charge factor
$c_{\mathrm{C}}$ = unit cost of cooling utility
$c_{\mathrm{H}}$ = unit cost of heating utility
SF = surcharge factor for dividing wall columns
$\Theta$ = plant operating time per annum

*General Variables*

$F_i$ = molar flow of feed $i$
$h_i^F$ = specific enthalpy of feed $i$
$x_{i,c}^F$ = mole fraction of component $c$ in feed $i$
$P^{\mathrm{top}}$ = molar flow of the top product
$h^{P^{\mathrm{top}}}$ = specific enthalpy of the top product
$x_c^{P^{\mathrm{top}}}$ = mole fraction of component $c$ in the top product
$P^{\mathrm{bot}}$ = molar flow of the bottom product
$h^{P^{\mathrm{bot}}}$ = specific enthalpy of the bottom product
$x_c^{P^{\mathrm{bot}}}$ = mole fraction of component $c$ in the bottom product
$S$ = side product molar flow rate
$x_c^{\mathrm{S}}$ = mole fraction of component $c$ in the side product
$h^{\mathrm{S}}$ = specific enthalpy of the side product
$s_t$ = fraction of $S$ taken from tray $t$
$R$ = reflux molar flow rate
$F_i^{\mathrm{outlet}}$ = molar flow rate of the outlet stream at the location of feed $i$
$x_{i,c}^{F^{\mathrm{outlet}}}$ = mole fraction of component $c$ in the outlet stream at the location of feed $i$
$h_i^{F^{\mathrm{outlet}}}$ = specific enthalpy of the outlet stream at the location of feed $i$
$F^{\mathrm{bot}}$ = molar flow rate of feed to reboiler
$x_c^{F^{\mathrm{bot}}}$ = mole fraction of component $c$ in the reboiler feed
$h^{F^{\mathrm{bot}}}$ = specific enthalpy of reboiler feed
$F^{\mathrm{top}}$ = molar flow rate of feed to top tray
$x_c^{F^{\mathrm{top}}}$ = mole fraction of component $c$ in the top tray feed
$h^{F^{\mathrm{top}}}$ = specific enthalpy of top tray feed
$A_{\mathrm{con}}$ = heat exchange area in condenser
$A_{\mathrm{reb}}$ = heat exchange area in reboiler
$f_{\mathrm{vap}}^{P^{\mathrm{top}}}$ = vapor fraction in top product
$f_{\mathrm{vap},i}^{F}$ = vapor fraction in outlet stream $i$
$f_{\mathrm{vap}}^{\mathrm{S}}$ = vapor fraction in side product
$Q_{\mathrm{reb}}$ = reboiler heat duty
$Q_{\mathrm{con}}$ = condenser heat duty
$C_{\mathrm{cap}}$ = capital cost of the column
Diam = column diameter
$N_{\mathrm{tray}}$ = number of trays in the column
$\omega^{(kl)}$ = binary variable denoting whether column $k$ is fitted within the shell of column $k$

*Variables on Each Tray*

$x_{t,c}$ = liquid-phase mole fraction of component $c$ on tray $t$
$y_{t,c}$ = vapor phase mole fraction of component $c$ on tray $t$
$h_t^{\mathrm{liq}}$ = specific liquid enthalpy on tray $t$
$h_t^{\mathrm{vap}}$ = specific vapor enthalpy on tray $t$
$L_t$ = liquid molar flow rate leaving tray $t$
$V_t$ = vapor molar flow rate leaving tray $t$
$T_t$ = temperature on tray $t$
$p_t$ = pressure on tray $t$
$f_{t,i}$ = fraction of feed $i$ to tray $t$
$r_t$ = fraction of reflux returned to tray $t$

## Literature Cited

(1) Petlyuk, F. B.; Platanov, V. M.; Slavinskii, D. M. Thermodynamically optimal method for separating multicomponent mixtures. *Int. Chem. Eng.* **1965**, *5*, 555–561.

(2) Wright, R. O. Fractionation apparatus. U.S. Patent 2,471,-134, 1949.

(3) Kaibel, G. Distillation columns with vertical partitions. *Chem.-Ing.-Techn.* **1987**, *10*, 92–98.

(4) BASF AG. Innovation award 1995: Split wall for the environment. Press Release available on the World Wide Web at URL http://www.basf.de/basf/html/e/entwick/innov/trenn95.htm, 1997.

(5) Tedder, D. W.; Rudd, D. F. Parametric studies in industrial distillation. *AIChE J.* **1978**, *24*, 303–315.

(6) Glinos, K.; Malone, M. Minimum reflux, product distribution, and lumping rules for multicomponent distillation. *Ind. Eng. Chem. Process Des. Dev.* **1984**, *23*, 764–788.

(7) Glinos, K.; Malone, M. Minimum vapor flows in a distillation column with a sidestream stripper. *Ind. Eng. Chem. Process Des. Dev.* **1985**, *24*, 1087–1090.

(8) Glinos, K.; Nikolaides, I. P.; Malone, M. New complex column arrangements for ideal distillation. *Ind. Eng. Chem. Process Des. Dev.*, **1986**, *25*, 694–699.

(9) Glinos, K.; Malone, M. Optimality regions for complex column alternatives in distillation systems. *Chem. Eng. Res. Des.* **1988**, *66*, 229–240.

(10) Carlberg, N. A.; Westerberg, A. W. Temperature-heat diagrams for complex columns. 2. Underwood's method for side strippers and enrichers. *Ind. Eng. Chem. Res.* **1989**, *28*, 1379−1386.

(11) Carlberg, N. A.; Westerberg, A. W. Temperature-heat diagrams for complex columns. 3. Underwood's method for the Petlyuk configuration. *Ind. Eng. Chem. Res.* **1989**, *28*, 1386−1397.

(12) Annakou, O.; Mizsey, P. Operability investigation of energy integrated distillation schemes. *Hung. J. Ind. Chem.* **1996**, *24*, 155−160.

(13) Annakou, O.; Mizsey, P. Rigorous comparative-study of energy-integrated distillation schemes. *Ind. Eng. Chem. Res.* **1996**, *35*, 1877−1885.

(14) Wayburn, T. L.; Seader, J. D. Solutions of systems of interlinked distillation columns by differential homotopy-continuation methods. In *Proceedings of the 2nd Internatinal Conference on Found. Computer-Aided Process Design*; Westerberg, A. W.; Chien, H. H., Eds.; CACHE Publications: Ann Arbor, Michigan, 1984; pp 765−862.

(15) Chavez, R.; Seader, J. D.; Wayburn, T. L. Multiple steady-state solutions for interlinked separation systems. *Ind. Eng. Chem. Fundam.* **1986**, *25*, 566−576.

(16) Lin, W−J.; Seader, J. D.; Wayburn, T. L. Computing multiple solutions to systems of interlinked separation columns. *AIChE J.* **1987**, *33*, 886−897.

(17) Rudd, H. Thermal coupling for energy efficiency. *The Chemical Engineer* **1992**, *27*, Distillation Supplement.

(18) Lestak, F.; Smith, R, The control of a dividing wall column. *Chem. Eng. Res. Des. 71*, **1993**, 307.

(19) Wolff, E. A.; Skogestad, S. Operation of integrated three-product (Petlyuk) distillation columns. *Ind. Eng. Chem. Res.* **1995**, *34*: 2094−2103.

(20) Douglas, J. M. *Conceptual Design of Chemical Processes*; McGraw-Hill Chemical Engineering Series; McGraw-Hill: New York, 1988.

(21) Fidkowski, Z. T.; Królikowski, L. Thermally coupled system of distillation columns: Optimization procedure. *AIChE J.* **1986**, *32*, 537−546.

(22) Fidkowski, Z. T.; Królikowski, L. Minimum energy requirements of thermally coupled distillation systems. *AIChE J.* **1987**, *33*, 643−653.

(23) Kakhu, A. I.; Flower, J. R. Synthesising heat-integrated distillation sequences using mixed integer programming. *Chem. Eng. Res. Des.* **1988**, *66*, 241−254.

(24) Triantafyllou, C.; Smith, R. The design and optimisation of fully thermally coupled distillation columns. *Trans. 1nst. Chem. Eng.* **1992**, *70*, 118−132.

(25) Sargent, R. W. H.; Gaminibandara, K. Optimum design of plate distillation columns. In *Optimization in Action*; Dixon, L. W. C. Ed.; Academic Press: London, 1976; pp 267−314.

(26) Agrawal, R. Synthesis of distillation column configurations for a multicomponent distillation. *Ind. Eng. Chem. Res.* **1996**, *35*, 1059−1071.

(27) Viswanathan, J.; Grossmann, I. E. An alternative model for finding the number of trays required for a specified separation objective. *Comput. Chem. Eng.* **1993**, *17*, 949−955.

(28) Viswanathan, J.; Grossmann, I. E. Optimal feed locations and number of trays for distillation columns with multiple feeds. *Ind. Eng. Chem. Res.* **1993**, *32*, 2942−2949.

(29) Smith, E. M. B. On the Optimal Design of Continuous Processes. Ph.D. Thesis, University of London, 1996.

(30) Smith, E. M.; Pantelides, C. C. Design of reaction/separation networks using detailed models. *Comput. Chem. Eng.* **1995**, *19*, S83−S88.

(31) Christiansen, A.; Skogestad, S.; Lien, K. Complex distillation arrangements: Extending the Petlyuk ideas. *Comput. Chem. Eng.* **1997**, *21*, S237−S242.

(32) Drud, A. S. *CONOPT: A system for large scale nonlinear optimisation − Reference manual*; ARKI Consulting and Development A/S: Bagsværd, Denmark, 1995.

(33) Barton, P. I.; Pantelides, C. C. Modeling of combined discrete/continuous processes. *AIChE J.* **1994**, *40*, 966−979.

(34) Oh, M.; Pantelides, C. C. A modelling and simulation language for combined lumped and distributed parameter systems. *Comput. Chem. Eng.* **1996**, *20*, 611−633.

(35) Dünnebier, G. Case Studies in Optimal Process Design − Separator Networks. Diploma thesis, Lehrstuhl für Anlagensteuerungstechnik, Fachbereich Chemietechnik, Universität Dortmund, Dortmund, Germany, 1997.

(36) Reid, R. C.; Prausnitz, J. M.; Poling, B. E. *The Properties of Gases and Liquids*; McGraw-Hill: New York, 1986.

(37) Nikolaides, I. P.; Malone, M. Approximate design and optimization of a thermally coupled distillation with prefractionation. *Ind. Eng. Chem. Process Des. Dev.* **1988**, *27*, 811−818.

(38) Agrawal, R.; Fidkowski, Z. T. Are thermally coupled distillation columns always thermodynamically more efficient for ternary distillations? *Ind. Eng. Chem. Res.* **1998**, *37*, 3444−3454.

(39) Abdul Mutalib, M. I.; Smith, R. Operation and control of dividing wall distillation columns. part 1: Degrees of freedom and dynamic simulation. *Trans. Inst. Chem. Eng.* **1998**, *76*, 308−318.

(40) Abdul Mutalib, M. I.; Zeglam, A. O.; Smith, R. Operation and control of dividing wall distillation columns. part 2: Simulation and pilot plant studies using temperature control. *Trans. Inst. Chem. Eng.* **1998**, *76*, 319−334.

*Received for review* May 13, 1998
*Revised manuscript received* October 27, 1998
*Accepted* October 28, 1998

IE9802919