Computers and Chemical Engineering 23 (1999) 709–731

Computers & Chemical Engineering

A systematic modeling framework of superstructure optimization in process synthesis

Hector Yeomans, Ignacio E. Grossmann *

Department of Chemical Engineering, Carnegie Mellon University, Pittsburgh PA 15213, USA

Received 22 December 1998; accepted 22 December 1998

Abstract

A systematic framework is presented for the representation of superstructures and derivation of optimization models in process synthesis. The state task network (STN) and state equipment network (SEN) are proposed as the two fundamental representations of superstructures for process systems involving mass, heat and momentum transfer. The mathematical modeling of either of the two representations is performed with generalized disjunctive programming (GDP), and then converted systematically into mixed integer linear programs/mixed integer non-linear programs (MILP/MINLP) problems. The application of this methodology is illustrated with the synthesis of distillation sequences, with and without heat integration, which lead to MILP problems. It is shown that ad hoc models that have been reported in the literature can be systematically derived, and in the case of separation sequences with heat integration, a new improved model is derived. Numerical results for comparing alternative models are also presented.

1. Introduction

In process synthesis, there are two major approaches that a design engineer can take to determine the optimal configuration of a flowsheet and its operating conditions. In one approach the problem can be solved in sequential form, by decomposition, fixing some elements in the flowsheet, and then using heuristic rules to determine changes in the flowsheet that may lead to an improved solution. An example of such an approach is the sequential hierarchical decomposition strategy by Douglas (1988). While this procedure is relatively simple to implement, the sequential nature of the decisions and the heuristic rules that are used can lead to sub-optimal designs.

The second strategy that can be applied to solve a process synthesis problem is based on simultaneous optimization using mathematical programming (Grossmann, 1996). This strategy requires to postulate a superstructure that includes equipment that can be potentially selected in the final flowsheet, as well as their interconnection. The equations of the equipment and their connectivity, and constraints for the operating conditions are then incorporated in an optimization problem where an objective function is specified such as cost minimization or profit maximization. This approach generally requires the use of discrete variables to represent the choices of equipment, with which the model becomes a mixed integer linear or non-linear program (MILP or MINLP). The advantage of mathematical programming strategies for process synthesis is that they perform simultaneous optimization of the configuration and operating conditions. The drawback is that global optimality conditions cannot be guaranteed for nonlinear models unless specific methods for global optimization are used.

Most of the work that has been reported with the mathematical programming approach for process synthesis has concentrated in developing ad hoc models for specific types of problems. For instance, Yee and Grossmann (1990) and Ciric and Floudas (1991) have proposed specialized superstructure representations and MINLP models for heat exchanger networks. Gaminibandara and Sargent (1976), Andrecovich and Westerberg (1985), and Aggrawal and Floudas (1990), have proposed a number of models for selecting distillation sequences. Andrecovich and Westerberg (1985), Flou-

* Corresponding author.
E-mail address: grossman@cmu.edu (I.E. Grossmann)

0098-1354/99/$ - see front matter
PII: S0098-1354(99)00003-4

das and Paules (1988), and Raman and Grossmann (1994), have developed superstructures for heat-integrated distillation sequences with sharp separations. Sargent (1998) has developed a state-task network (STN) representation for the synthesis of non-ideal distillation sequences. Balakrishna and Biegler (1992) and Kokossis and Floudas (1991) have proposed superstructure representations for reactor networks.

As for mathematical modeling techniques for superstructures, Kocis and Grossmann (1989) proposed a MINLP modeling decomposition strategy in which the flowsheet is partitioned into process units and interconnection nodes, and where the NLP sub-problems only involve existing units. Non-existing units are sub-optimized with a Lagrangian decomposition to set up the initial MILP master problem. Bagajewicz and Manousiothakis (1992) proposed a 'state space' representation, by decomposing the flowsheet into a block for the distribution network and blocks for different operators, in which design equations for the equipment tasks are included. The operators are typically solved using pinch analysis models for heat and mass exchanger networks. Papalexandri and Pistikopoulos (1996) proposed a modeling strategy where they disaggregate the flowsheet elements into two types of building blocks: a heat and mass exchange block and pure heat exchanger block. Operations such as distillation, reaction, absorption and other unit operations are then represented with these building blocks. Recently, Smith (1996) introduced the 'state operator network', where he considers full connectivity among all the potential equipment in a flowsheet for which no tasks are pre-assigned, since rigorous models are used. The combinatorics are reduced to selecting the equipment. This strategy, however, can have significant convergence difficulties due to the non-convexities that are introduced in the interconnection equations (e.g. bilinear equations). The work of Kocis and Grossmann (1989), Bagajewicz and Manousiothakis (1992), Papalexandri and Pistikopoulos (1996) and Smith (1996) has shown the great importance of representation and modeling in the optimization of flowsheet superstructures.

The goal of this paper is to develop a general representation and modeling framework for systematically deriving process synthesis models. The state task network (STN) and state equipment network (SEN), which are complementary to each other, will be proposed as two basic problem representations for process synthesis. These representations will be modeled through generalized disjunctive programming (GDP) (Raman & Grossmann, 1994) from which specific mixed-integer optimization models can be derived. The representation and modeling of synthesis problems, which will be restricted to linear process models, will be illustrated with sharp distillation synthesis problems with and without heat integration. Numerical examples will be presented to demonstrate the advantages of the proposed representation and modeling strategies.

2. Problem statement

The problem addressed in this paper can be stated as follows: given is a set of equipment, raw materials, products and process alternatives in terms of different choices of tasks and equipment, and the interconnections among them. The objective is to establish a systematic procedure for representing these elements in a superstructure, and for deriving a mathematical programming model with discrete and continuous variables to predict an optimum flowsheet design.

Since this paper is a first step for developing a systematic and comprehensive framework for deriving optimization synthesis models, we will restrict the treatment to problems with linear process models. Furthermore, only flows of material and heat will be considered.

3. General elements of flowsheets

Flowsheets are generally regarded as a network composed of streams and equipment. However, a more general characterization requires three basic elements: states, tasks and equipment.

- States are defined as the set of physical and chemical properties that identify a stream in the process. The definition of a state includes quantitative intensive and extensive properties of a stream, such as composition, temperature, pressure, particle size, heat content, mass flow, etc. However, this quantitative information is limited, and therefore it is necessary to include also qualitative information (Papalexandri & Pistikopoulos 1996). The qualitative properties that can be used to characterize a state are the name of the components that can be present, or will be allowed to be present in the streams, and the phase(s) of the stream. Also, other labels may be required such as a heat exchange label (hot stream, cold stream, stream to be condensed or vaporized), a mass transfer label (rich or lean stream, solvent, solute, absorbent, absorbate) and a momentum transfer label (turbulent or laminar flow, stagnant fluid, fluidized phase). It should be noticed that in general a state corresponds to a process stream, unless the level of detail describing the states is such that several streams can be associated to a single state.
- Tasks can be defined as the physical and chemical transformations that occur between adjacent states. The tasks will correspond to momentum, mass, and energy transfer operations (e.g. distillation, absorp-

tion, reaction, membrane separation, mixing) which are described by conservation, equilibrium and rate equations.

- Equipment are the elements of a flowsheet corresponding to the physical devices that will execute a given task (e.g. reactor, absorber, heat exchanger). The equipment parameters are determined from the corresponding design equations.

4. General framework for process synthesis

The general framework proposed in this paper is composed of three major steps that will be described in detail in this section. These steps are rather general, and can in principle be applied to any synthesis problem to derive a mathematical programming model for predicting an optimal flowsheet configuration.

In the initial step of the proposed framework we will first consider two major superstructure representations: the STN, in which the tasks and states are defined while the equipment assignment is generally unknown, and the SEN in which the tasks and the equipment are defined while the assignment of tasks to equipment must be determined. Based on these network representations, we will model the corresponding synthesis problems with GDP (Raman & Grossmann, 1994; Turkay & Grossmann, 1996a). These logic based methods will then be used as basis for deriving algebraic mixed-integer optimization models.

4.1. State task network (STN) representation

The STN representation was introduced by Kondili, Pantelides and Sargent (1993) for process scheduling, but can also be used for process flowsheet superstructures as shown in this section. As mentioned above, this representation will be concerned first with identifying the states and tasks, and then leaving the equipment assignment to a second stage, in similar fashion as in the means-ends analysis strategy (Siirola & Rudd, 1971). Fig. 1 shows the STN representation of a superstructure that involves compression of the feed, reaction, flash and distillation separations with recycle.

In the STN, some of the tasks are conditional and some others must be present in all design alternatives. There is no need to distinguish one from the other at the level of representation, but only at the level of the model. The tasks involve mass transfer, heat transfer, pressure, temperature and phase changes. One or more of these operations may be performed in one task if technically feasible.

Once the states and the tasks are identified, it is necessary to determine what type of equipment can

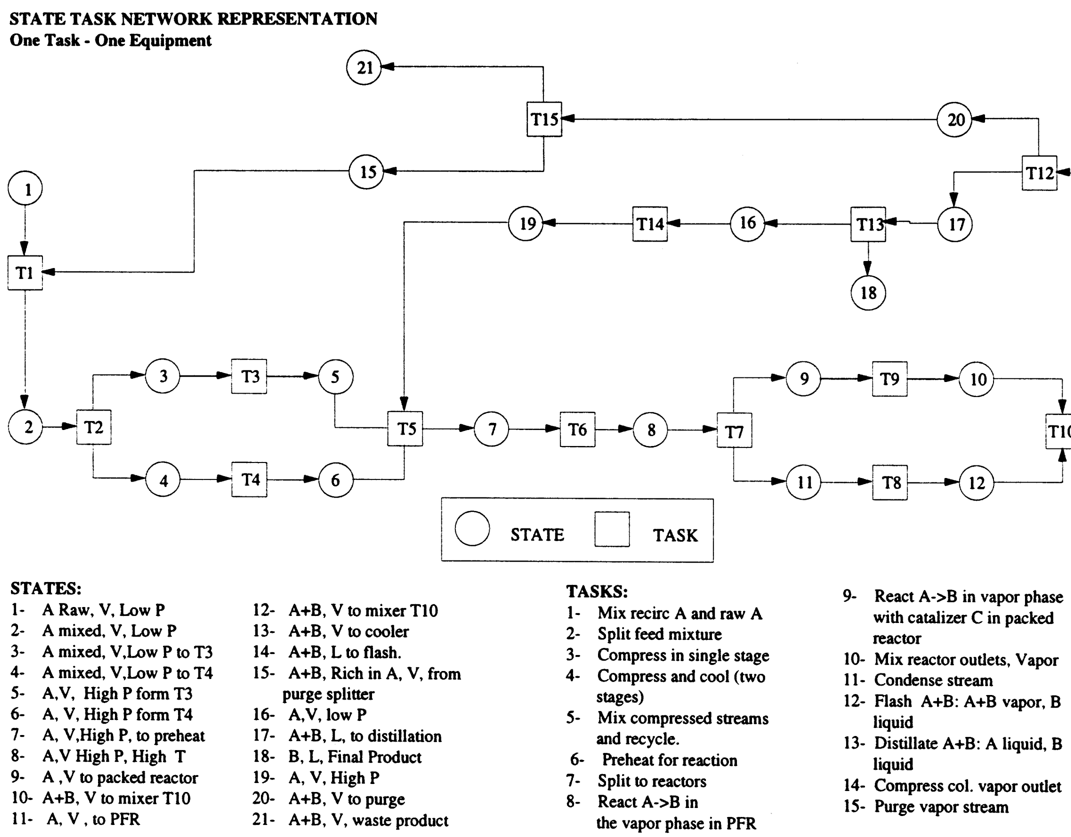

Fig. 1. STN Representation of a general process flowsheet.

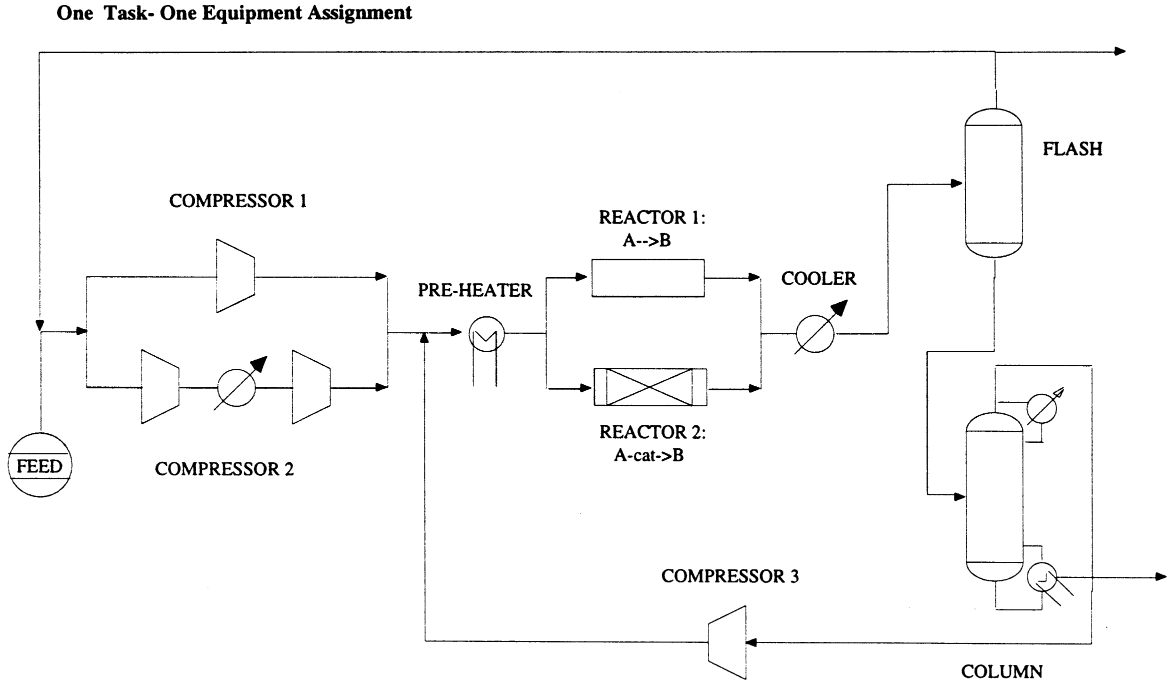

Fig. 2. One task–one equipment (OTOE) assignment for the STN representation.

perform each task, and then assign it to the corresponding tasks. There are two cases for this purpose:

- One task–one equipment (OTOE) assignment: in this case each task is assigned to a single equipment unit. If a task can be executed by two different equipment, the tasks will have to be redefined to distinguish one from the other.
- Variable task-equipment (VTE) assignment: in this case, a set of equipment that can perform all the tasks needed in the flowsheet is identified first. The assignment of the equipment to the tasks is then considered as part of the optimization model. In this way, a single equipment unit can be assigned to different tasks, and a single task can be assigned to different equipment.

It is important to notice that for the OTOE case the equipment assignment is explicitly performed a-priori, while for the VTE case it is an unknown to be determined. In the case of the OTOE, the representation by tasks and equipment is identical.

If in Fig. 1 we consider the OTOE case, we simply replace the blocks for the tasks with equipment, and the network representation reduces to the one in Fig. 2. For the VTE case the equipment is not pre-assigned to the task, as shown in Fig. 3. Note for instance that for tasks T3 and T4 the same compressor can be assigned.

4.2. State equipment network (SEN) representation

As discussed above, it is possible to develop a process synthesis representation that includes the different states of the process, and the equipment that are likely to be used. Once these elements are specified, the different tasks (one or more) that each equipment can perform must be determined. We define this representation as the SEN representation.

This state-equipment representation was developed initially by Smith (1996) who considered full connectivity among the states and equipment. The tasks that can take place in a specific equipment are not pre-specified, which is equivalent to a VTE assignment.

The construction of the SEN generally leads to a smaller combinatorial problem for the selection of equipment. However, the disadvantage is the implicit combinatorial complexity that is present in the possible equipment interconnections. An example of the SEN representation for a similar process as in Fig. 1 can be seen in Fig. 4.

An important feature of the SEN is that the state definition is not unique, since the properties of the stream coming out of a certain equipment will be defined by the particular task that the equipment performs. Therefore, the state definition will have to consider all the possible realizations of the streams that will originate from a certain task, which can complicate the modeling stage.

If every equipment is restricted to perform one single task in the SEN, it is possible to obtain the same representation as with the STN/OTOE, provided that the same tasks that appear in the SEN appear in the STN representation. The SEN representation is also useful for retrofit design problems, as it shows explicitly the existing equipment of the given problem. Finally, one can also often determine ahead of time the number of equipment that is needed, in which case the optimization reduces to assigning the tasks and determining the interconnections of the equipment.

4.3. Generalized disjunctive programming (GDP) modeling

The second step of the proposed framework for process synthesis corresponds to the modeling of the chosen representation, STN or SEN, as a mathematical programming problem. Since there will be conditional tasks or equipment that might be selected or not in the final flowsheet, it is necessary to use a discrete mathematical programming model. The use of disjunctive programming (Balas, 1979) is of particular interest, since process synthesis problems naturally lead to models where the solution space is disjoint, and there is a strong logic on the connectivity among the different tasks (Raman & Grossmann 1993, 1994).

In order to use GDP (Raman & Grossmann, 1994) to model the STN or SEN representations, it is necessary to identify the conditional constraints from among those that must hold for all synthesis alternatives. The conditional constraints will be represented with disjunctions and assigned a Boolean variable that represents its existence (if the Boolean variable takes a value of 'true'). In general mixers and splitters can be considered conditional tasks. However, if the equations that are applied to the mixer and splitter are only mass and energy balances, these constraints do not involve any type of discrete decision or discrete variable assignment for them to be valid. For this reason they are considered permanent in this paper.

4.4. GDP models for STN representation

In order to formulate the GDP model, the following sets and variables must be defined. Let $t \in \mathrm{T}$ define the set of tasks in the superstructure, where $\mathrm{T} = \mathrm{T_P} \cup \mathrm{T_C}$ and $\mathrm{T_P}$ is the set of permanent tasks (valid for all design alternatives) and $\mathrm{T_C}$ is the set of conditional tasks that may be selected. Let $s \in \mathrm{S}$ define the set of states, and $j \in \mathrm{E}$ define the set of equipment units. Let $\mathrm{I}_t = \{s \mid s$ is an input state of task $t\}$, and $\mathrm{O}_t = \{s' \mid s'$ is an output state of task $t\}$. The variables z_t, x_s and d_j are used to represent the operating variables in the tasks, the flow and state variables interconnecting the states, and the design variables for the equipment, respectively. The function $h_t(z_t, x_s, x_{s'})$ represents the equations (mass balances, energy balances, etc.) and constraints corresponding to task t, and $r_j(d_j, x_s, x_{s'}, z_t)$ represents the equations and constraints corresponding to a particular equipment design. Finally, $f(d_j, z_t)$ represents the cost function in terms of the design and control variables, d_j and z_t.

If the OTOE case is considered for the STN superstructure, the equations and constraints from equipment and tasks can be integrated in the vector $g_t = [h_t(z_t, x_s, x_{s'}), r_{j'}(d_{j'}, x_s, x_{s'}, z_t)]^T$ where $j' \in \mathrm{Q}_t = \{j' \in \mathrm{E} \mid j'$ is associated with task $t\}$, and $\bigcap_{t \in T} Q_t = \emptyset$. The GDP model for STN/OTOE representation is then as follows,

$$\text{(P-STN1):} \quad \min \sum_{t \in \mathrm{T}} c_t + \sum_{s \in S} \alpha_s x_s \tag{1}$$

$$\left.\begin{array}{l} s.t.\ \ g_t(d_{j'}, z_t, x_s, x_{s'}) \le 0 \\ \quad c_t = f(d_{j'}, z_t) \end{array}\right\} \quad \begin{array}{ll} j' \in Q_{t'} & t \in T_P \\ s \in I_t, & s' \in O_t \end{array} \tag{2}$$

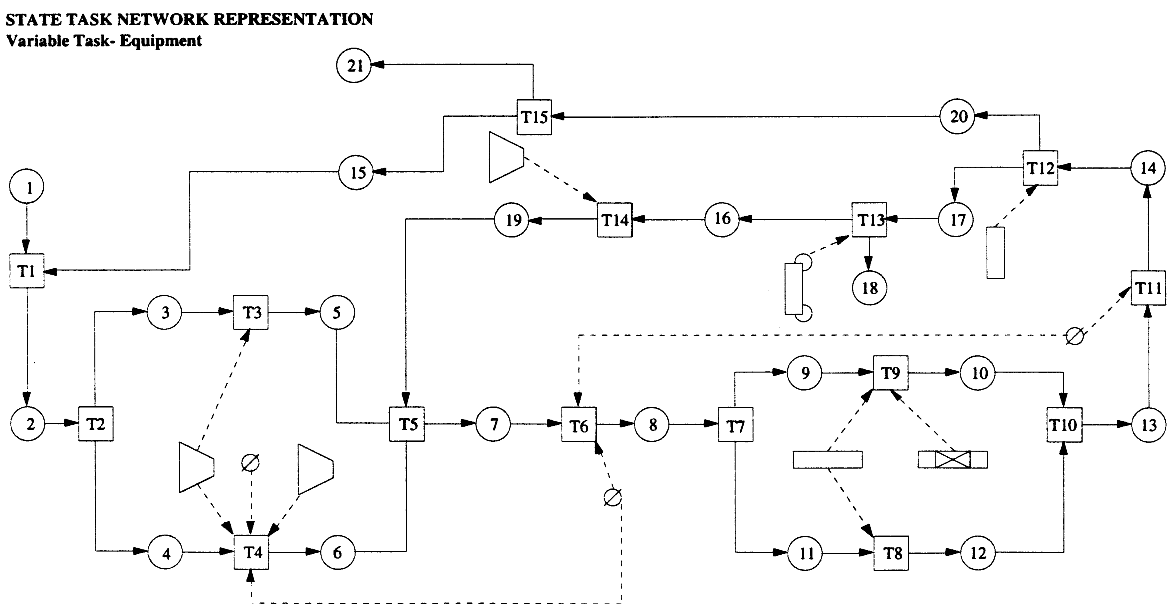

Fig. 3. VTE assignment for the STN representation.

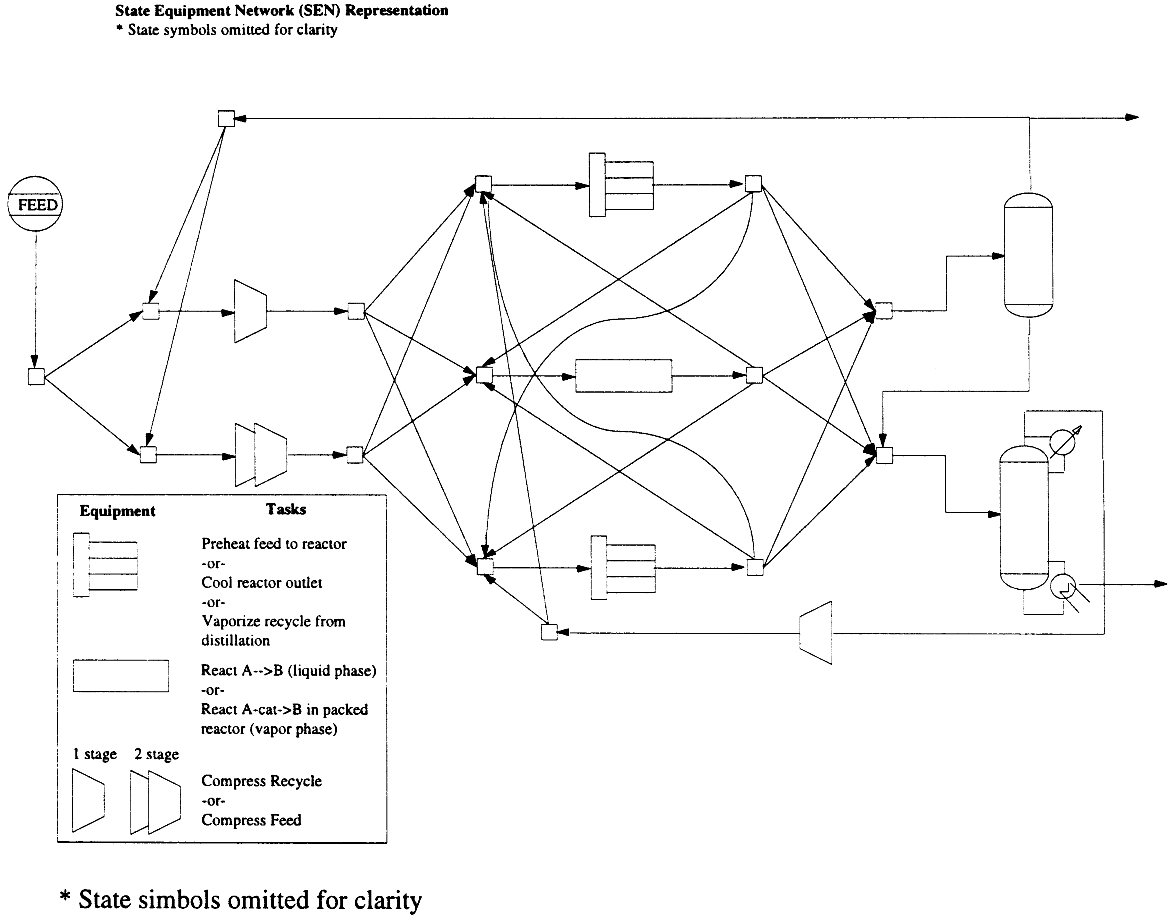

Fig. 4. SEN representation of a synthesis problem.

$$\begin{bmatrix} Y_t \\ \left.\begin{matrix} g_t(d_j, z_t, x_s, x_{s'}) \le 0 \\ c_t = f(d_j, z_t) \end{matrix}\right\} \begin{matrix} j' \in Q_t & j' \in Q \\ s \in I_t, & s' \in O_t \end{matrix} \end{bmatrix} \vee \begin{bmatrix} \neg Y_t \\ \left.\begin{matrix} d_{j'} = z_t = 0 \\ x_s = x_{s'} = 0 \end{matrix}\right\} \begin{matrix} j' \in Q_t \\ \forall\ s \in I_t, \quad s' \in O_t \end{matrix} \end{bmatrix} t \in T_c \tag{3}$$

$$\Omega(y) = \text{True} \tag{4}$$

$$d \in D, \quad z \in Z, \quad x \in X \qquad Y_t = \{\text{True, False}\}$$

Eq. (1) represents the objective function in terms of costs incurred by the selection of a task with its equipment, and variable costs associated with flows through the different states. Eq. (2) represents the mass and energy balances, as well as the design constraints of all the tasks that are permanent throughout the flowsheet. In Eq. (3), the selection of a conditional task $t \in T_C$ is represented by a Boolean variable. When the value of the variable is true (Y_t) the task is selected. When the conditional task is not selected (Y_t = False), it is assumed for ease of notation that all the corresponding variables are set to zero. Eq. (4) represents the logic relations between Boolean variables in formulation (PSTN-1). Fig. 5(a) presents qualitatively the elements of the model with those of the STN/OTOE representation for a small example.

For the case of VTE assignment, another type of disjunction must be introduced since the equipment assignment must be determined. For each task (either permanent or conditional) it is necessary to select one and only one of the equipment configurations available for the task. To model this disjunction define for every task t the set $A_t = \{j \in E \mid \text{equipment } j \text{ can be assigned to task } t\}$, and define W_{jt} as a Boolean variable that indicates whether equipment j will perform task t. The general STN/VTE model is as follows:

$$\text{(P-STN2):} \quad \min \sum_{t \in T} \sum_{j \in E} c_{tj} + \sum_{s \in S} \alpha_s x_s \tag{5}$$

$$\text{s.t.} \quad h_t(z_t, x_s, x_{s'}) \le 0 \quad t \in T_p \tag{6}$$

$$\bigvee_{j \in A_t} \begin{bmatrix} W_{tj} \\ r_j(d_j, z_t, x_s, x_{s'}) \le 0 \\ c_{tj} = f(d_j, z_t) \end{bmatrix} \quad t \in T_P \tag{7}$$

$$\begin{bmatrix} Y_t \\ h_t(z_t, x_s, x_{s'}) \le 0 \\ \bigvee_{j \in A_t} \begin{bmatrix} W_{tj} \\ r_j(d_j, z_t, x_s, x_{s'}) \le 0 \\ c_{tj} = f(d_j, z_t) \end{bmatrix} \end{bmatrix} \vee \begin{bmatrix} \neg Y_t \\ z_t = d_j = 0 \\ x_s = x_{s'} = 0 \end{bmatrix}$$

$$t \in T_C \tag{8}$$

$$\Omega(W, Y) = \text{True} \tag{9}$$

$$d \in D, \quad z \in Z, \quad x \in X \qquad Y_t = \{\text{True, False}\},$$

$$W_{tj} = \{\text{True, False}\}$$

where for simplicity we have excluded the sets of the indices $s \in \mathrm{I}_t$, $s' \in \mathrm{O}_t$ within the disjunctions. Eq. (5)

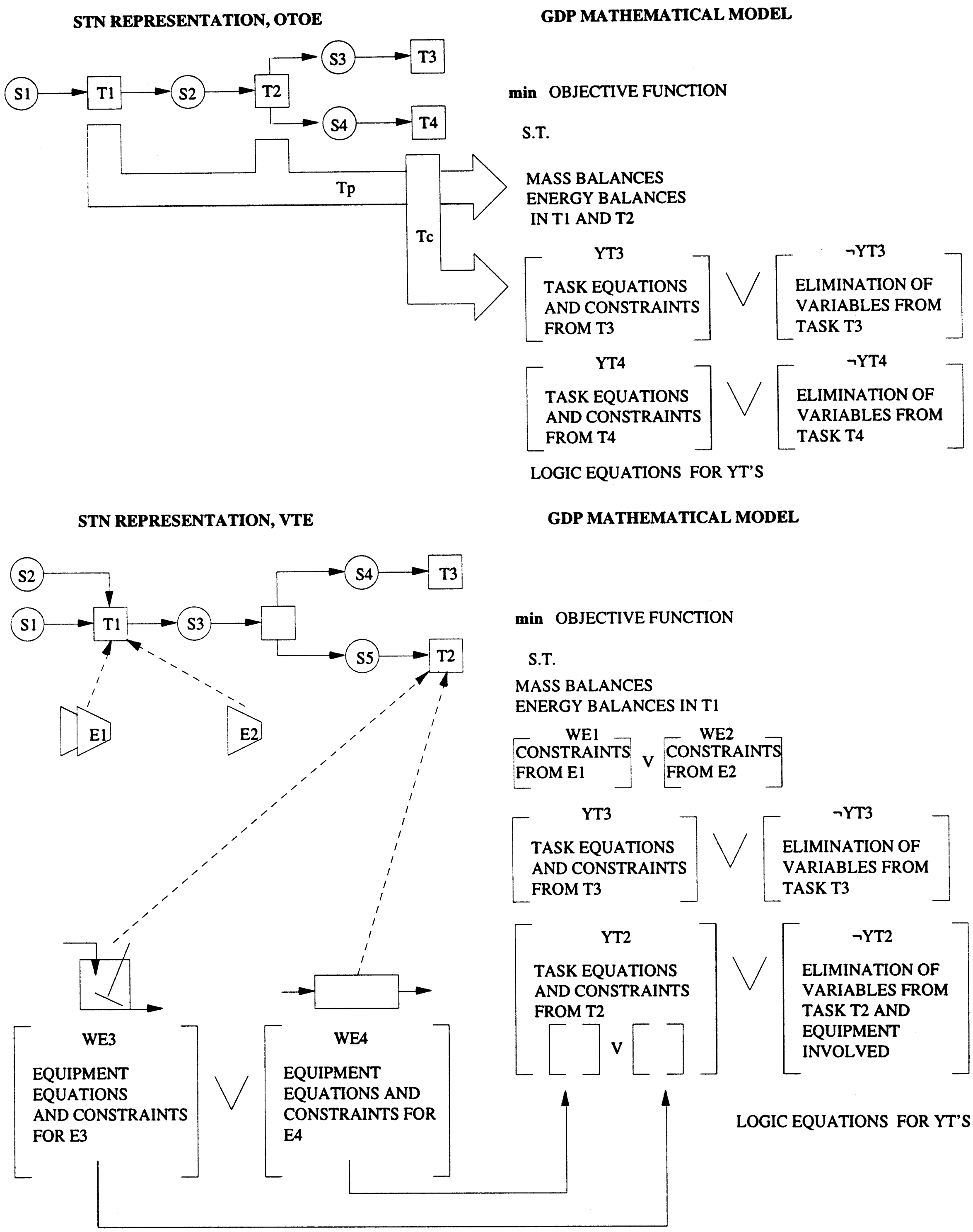

Fig. 5. STN representation and translation to GDP.

represents the objective function in terms of investment cost incurred by the assignment of an equipment to a selected task, and variable costs generated by the states. Eq. (6) represents the equations and constraints that are valid for all the flowsheet due to the tasks that are permanent. Eq. (7) represents disjunctions corresponding to the different equipment for which the permanent tasks in (Eq. (6)) can be performed. The Boolean variables W_{tj} control the application of the constraints for equipment ($j \in A_t$) for the given permanent task $t \in T_P$. Eq. (8) represents the constraints and equations for the conditional tasks $t \in T_C$. If the conditional task exists ($Y_t =$ True), the outermost brackets of the left hand side enforce the corresponding equations and constraints. If a conditional task does not exist ($Y_t =$ False), the right hand side brackets include the variables to be set to zero. If a conditional task is selected, there is a choice of equipment $j \in A_t$ where it can be performed, given by the innermost bracket that contains the corresponding equations and constraints. Finally, Eq. (9) represents the logic relations between the Boolean variables in the formulation (PSTN-2). Fig. 5(b) shows qualitatively the relation between the STN/VTE representation and its GDP model.

4.5. GDP model for the SEN representation

Let the equipment in a flowsheet be divided into two sets: $E = E_P \cup E_C$, where E_P represents the equipment that is permanent for all synthesis alternatives, and E_C represents the conditional equipment. Also, define the set $B_j = \{t \in T \mid \text{task } t \text{ can be performed in equipment } j\}$. The Boolean variable U_j represents whether conditional equipment j exists, while the Boolean variable V_{tj} represents whether task t is performed in equipment j. The equations and constraints that are activated when task t is performed in equipment j is defined by $p_{tj}(d_j, z_t, x_s, x_{s'})$. The GDP model for the SEN representation is as follows,

$$\text{(P-SEN):} \quad \min \sum_{j \in E} \sum_{t \in T} c_{jt} + \sum_{s \in S} \alpha_s x_s \tag{10}$$

$$\text{s.t.} \bigvee_{t \in B_j} \begin{bmatrix} V_{jt} \\ p_{tj}(d_j, z_t, x_s, x_{s'}) \le 0 \\ c_{jt} = f(d_j, z_t) \end{bmatrix} \quad j \in E_P \tag{11}$$

$$\begin{bmatrix} U_j \\ \bigvee_{t \in B_j} \begin{bmatrix} V_{jt} \\ p_{tj}(d_j, z_t, x_s, x_{s'}) \le 0 \\ c_{tj} = f(d_j, z_t) \end{bmatrix} \end{bmatrix} \vee \begin{bmatrix} \neg U_j \\ z_t = d_j = 0 \\ x_s = x_{s'} = 0 \end{bmatrix} \quad j \in E_C \tag{12}$$

$$\Omega(V, U) = \text{True} \tag{13}$$

$$d \in D, \quad z \in Z, \quad x \in X, \quad V_{jt} = \{\text{True, False}\}, \quad U_j = \{\text{True, False}\}$$

Eq. (10) is the objective function that includes the fixed cost for task t performed in equipment j, and the variable cost from the flow of materials through the different states in the flowsheet. Eq. (11) corresponds to the disjunctions that apply for the permanent equipment. Each disjunction is used to model the selection of tasks $t \in B_j$ for each equipment $j \in E_P$ for which the Boolean variable V_{jt} is used. Eq. (12) has two nested disjunctions because it is necessary to determine if a conditional equipment will exist ($U_j =$ True), and once determined it is necessary to select a task $t \in B_j$ that the equipment $j \in E_C$ can perform ($V_{jt} =$ True). Notice that if an equipment is not selected, the variables that participate on it are set to zero, as it happens in (PSTN-1) and (PSTN-2). Fig. 6 shows for a small example the relation between the SEN representation and its model.

4.6. Remarks

It is clear from the models (P-STN1), (P-STN2) and (P-SEN) that their mathematical structure is quite different. Not surprisingly, (P-STN1) for the STN/OTOE representation, has the simplest structure with disjunctions involving only two terms. In contrast, both (P-STN2) and (P-SEN) involve embedded disjunctions with multiple terms.

Two interesting theoretical questions on the above models, are first whether they become equivalent under some limiting conditions, and second whether any of them are inherently 'tighter', and thereby produce models that are in principle easier to solve.

On the first question, it is clear that for the OTOE assignment, (P-STN2) reduces to (P-STN1) since in that case $Q_t = A_t \ \forall t \in T$, with which the Boolean variables W_{tj} can be eliminated. Furthermore, if OTOE assignments are considered for the SEN network, then model (P-SEN) is also reduced to model (P-STN1) because the set B_j has only one task element given by the one in the set Q_j and $E = T$. Hence, the Boolean variables V_{jt} can be eliminated, leading to the structure of (P-STN1) by setting $p_{tj} = g_t$. For the VTE assignment case, both (P-STN2) and (P-SEN) lead to different models. It is also possible to model a physically equivalent problem with the (P-STN1) and (P-SEN) models, as will be shown in the distillation example later in the paper.

On the question about tightness, the numerical results will show that in the simpler case (distillation sequencing with no heat intergration) the SEN model is not always tighter than the STN model, while in the more complex case (see problem with heat integration)

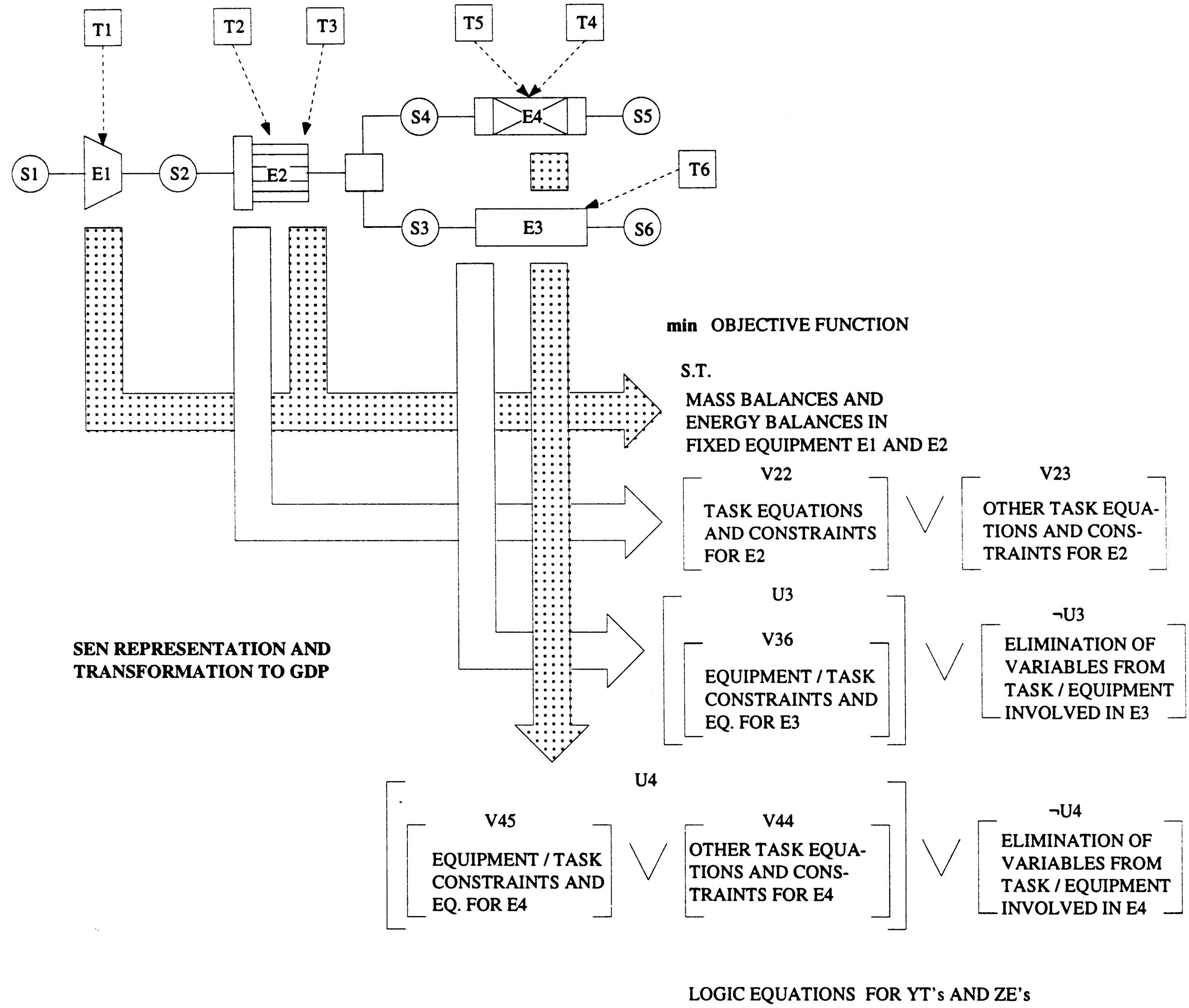

Fig. 6. Transformation of the SEN Representation into GDP a model.

the STN model was tighter. Therefore, unless specific models are considered, it appears that no general property of tightness can be established between the (P-STN2) and (P-SEN) models.

4.7. Transformation of GDP models into MILP problems

Several strategies have been developed to solve mathematical programming problems in disjunctive form. For instance, Beaumont (1991), Raman and Grossmann (1994) and Hooker and Osorio (1996) have developed algorithms to solve disjunctive linear problems, while Turkay and Grossmann (1996a) developed an algorithm to solve disjunctive non-linear problems. These methods involve algorithms that are still in the early stages of development and implementation, and therefore, are generally not yet comparable in terms of speed and ease of solution, particularly in the case of MILP models. Since the problems presented in this paper are linear, we will focus on the reformulation of the GDP problems of the previous sections into MILP problems.

Because of the nature of GDP, it is possible to transform in a systematic way the synthesis models (P-STN1), (P-STN2) and (P-SEN) into MILP form using the convex hull formulation of the disjunctions (Balas, 1985). The convex hull formulation is based on the disaggregation of variables that gives the tightest continuous relaxation of the disjunctions. In particular, consider the linear disjunction:

$$\bigvee_{i\in D}\left[\sum_{j\in N} a_{ij}x_j \le b_i\right] \tag{14}$$

The convex hull formulation of Eq. (14) is given by the following constraints, where z_j^i are the disaggregated variables for x_j and N is the index set of the continuous variables x,

$$x_j = \sum_i z_j^i, \quad j \in N \tag{15}$$

$$\sum_{j \in N} a_{ij} z_j^i \le b_i y_i, \quad i \in D, \tag{16}$$

$$0 \le z_j^i \le U_j y_i, \quad i \in D, \tag{17}$$

$$\sum_i y_i = 1, \quad y_i = 0, 1 \tag{18}$$

Eq. (16) represents each term of the disjunction with disaggregated variables z_j^i and right hand side multiplied by the binary variable y_j. Eq. (18) ensures that only one disjunction holds, while the inequalities in Eq. (17) ensure that disaggregated variables for terms in the disjunction that do not apply be set to zero. A direct derivation of these constraints is given in Turkay and Grossmann (1996b). The application of the convex hull of disjunctions will be illustrated with the synthesis problem described in the next section.

5. General synthesis framework for optimal distillation sequences

The objective of the problem considered here is to separate a multicomponent mixture into its individual components at minimum cost using sharp separators. According to the systematic framework proposed in this paper, the first step is to determine the representation of design alternatives. If the STN representation is used, one can specify either a OTOE assignment, or a VTE assignment. The STN/OTOE representation will be used, and compared with an SEN model in which columns can perform multiple separation tasks.

5.1. STN representation

The first step to generate a STN representation is to identify the tasks and states for the problem. The tasks for this problem correspond to all the possible cuts that can be performed on the mixtures of two or more components. Mixing and splitting tasks are also needed. The states will be identified quantitatively by their flow, and qualitatively by the components that appear in the streams of the process. The phase of all the streams is assumed to be liquid, and there is no heat transfer among streams (no heat integration). Fig. 7 shows the STN/OTOE representation for a problem involving the separation of a mixture of four components.

From Fig. 7, the following sets can be defined: C = {k | k is a separation task with an associated column}, FS_i = {ℓ | state ℓ is a feed into splitter i}, OS_i = {j | state j is output from splitter i}, FM_i = {j | state j is feed to mixer i}, OM_i = {ℓ | state ℓ is output from mixer i}, TF_k = {i | state i is feed to task k}, OTT_k = {ℓ | state ℓ is the output at top of task k} and OBT_k = {ℓ' | state ℓ' is the output at bottoms of task k}. NM is the number of mixers and NS is the number of splitters.

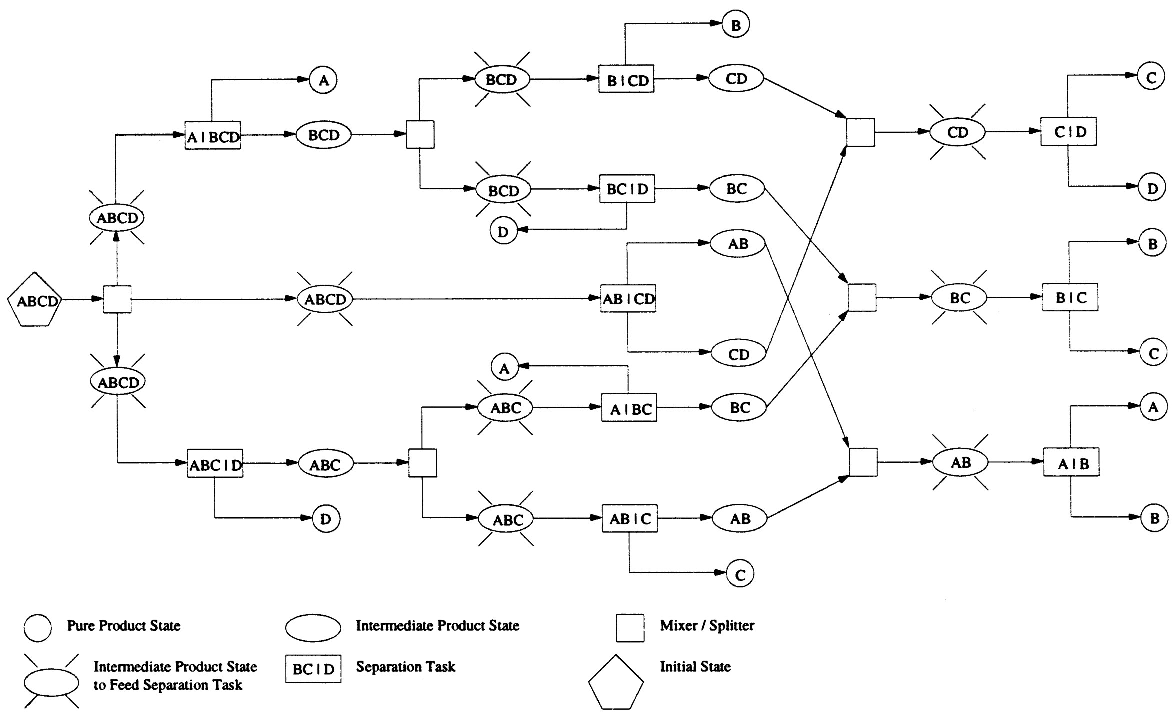

Fig. 7. STN/OTOE representation, four component separation.

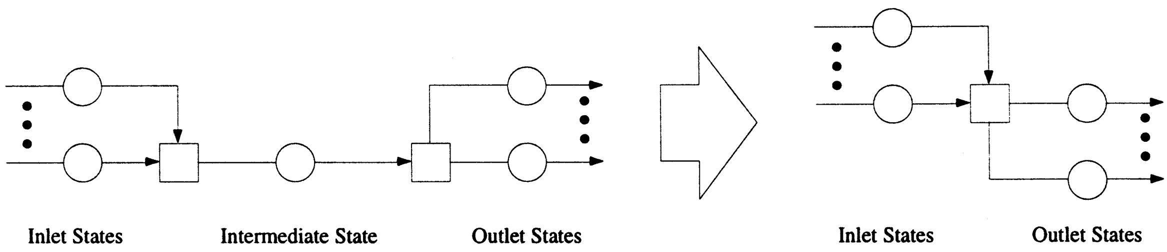

Fig. 8. Elimination of streams by joining mixers and splitters.

The GDP model for this synthesis problem can be derived from model (P-STN1). The variables are defined as follows: F_ℓ are the total mass flows in each of the corresponding streams, Q_k are the heat loads in the condenser and reboiler which are assumed to be the same, VC_k and FC_k are respectively the variable and fixed costs of a separation task, HK_k is the energy balance coefficient for each of the separation tasks, and C_U is the sum of the costs of utilities, considering that the heat loads of both reboiler and condenser are the same. ξ_k represents the split fraction of a column, top or bottom. α_k and β_k are the fixed and variable cost coefficients of a task assigned to a column k. $\Omega(Y_k)$ represents the set of logic propositions that specify how the tasks are interconnected (see Raman & Grossmann, 1993). The GDP formulation is given by:

$$\text{(P1):}\quad \min \sum_{k\in C} (FC_k + VC_k + C_U Q_k) \tag{19}$$

$$\text{s.t.}\quad F_\ell = \sum_{j\in OS_i} F_j \quad \ell\in FS_i \quad i = 1, 2, \ldots, NS \tag{20}$$

$$\sum_{j\in FM_i} F_j = F_\ell \quad \ell\in OM_i \quad i = 1, 2, \ldots, NM \tag{21}$$

$$\begin{bmatrix} Y_k \\ \left.\begin{matrix} F_\ell = \xi_k^{\text{top}} F_i \\ F_{\ell'} = \xi_k^{\text{bot}} F_i \end{matrix}\right\} \begin{matrix} \ell\in OTT_k \\ \ell'\in OBT_k \\ i\in TF_k \end{matrix} \\ FC_k = \alpha_k \\ VC_k = \beta_k F_i \\ Q_k = HK_k F_i \end{bmatrix} \vee \begin{bmatrix} \neg Y_k \\ F_\ell = 0, \quad \ell\in OTT_k \\ F_{\ell'} = 0, \quad \ell'\in OBT_k \\ F_i = 0, \quad i\in TF_k \\ FC_k = 0 \\ VC_k = 0 \\ Q_k = 0 \end{bmatrix}$$

$$k\in C \tag{22}$$

$$\Omega(Y_k) = \text{True} \tag{23}$$

$$F, FC, VC, Q \geq 0, \quad Y_k \in \{\text{True, False}\}$$

Eq. (19) represents the total cost given by the investment and utility cost. Eqs. (20) and (21) represent the mass balances on splitters and mixers, respectively, and are analogous to Eq. (2). The conditional tasks for the distillation sequence problem represent the different cut locations to separate a stream into distillate and bottoms. Each separator is considered to have only top and bottom outlet streams. The mass balances for these streams are included on the left hand bracket in Eq. (22). When a conditional task is selected, the mass balances, fixed and variable costs are applied. When the conditional task does not exist, the output flows and the corresponding costs of the given task are set to zero, as is shown in the second term of the disjunction in Eq. (22). Eq. (23) represents the logic constraints that relate the existence of a given task with a previous or preceding task (see Raman & Grossmann, 1993). These logic relations involve only the Boolean variables Y_k.

The GDP model (P1) includes variables and equations that can be eliminated without changing the objective function value. If we define the subset of states $\text{IP} = \{m \mid m \text{ is an intermediate product with 2 to } n \text{ components}\}$, it can be noted that

$$IP = \left[\bigcup_{i=1,\ldots,NS} FS_i\right] \cup \left[\bigcup_{i=1,\ldots,NM} OM_i\right]$$

since none of the streams that contain pure products lead to splitters or come from mixers. It is possible then to eliminate a stream F_ℓ if it is not intermediate, because it does not affect the task selection process.

The GDP model (P1) can be simplified by the elimination of variables and equations for streams that are not intermediate products. The new sets are defined as follows: $\text{MST} = \{i \mid i \text{ is a mix or split task}\}$, $\text{OT}_i = \{j \mid j \text{ is an outlet from a mix or split task } i\}$, $\text{IT}_i = \{\ell \mid \ell \text{ is inlet to mix/split task } i\}$ and $\text{SP}_k = \{\ell\in\{\text{top, bot}\} \mid \ell \text{ is intermediate product flow from task } k\}$. It is important to note that if two mixing and/or splitting tasks are following each other, they can be joined in a single task that involves mixing and splitting by elimination of the stream that joins the tasks, as is shown in Fig. 8. This is the reason for the existence of the summation of streams on both sides of Eq. (25). Model (P2) is then given by:

$$\text{(P2):}\quad \min \sum_{k\in C} (FC_k + VC_k + C_U Q_k) \tag{24}$$

$$\text{s.t.}\quad \sum_{\ell\in IT_i} F_\ell = \sum_{j\in OT_i} F_j \quad i\in MST \tag{25}$$

$$\begin{bmatrix} Y_k \\ F_\ell = \xi_k^m F_i \\ FC_k = \alpha_k \\ VC_k = \beta_k F_i \\ Q_k = HK_k F_i \end{bmatrix} \vee \begin{bmatrix} \neg Y_k \\ F_\ell = 0, F_i = 0 \\ FC_k = 0 \\ VC_k = 0 \\ Q_k = 0 \end{bmatrix} \quad k \in C \qquad (26)$$

$$\Omega(Y_k) = \text{True} \qquad (27)$$

$$F, FC, VC, Q \geq 0, \quad Y_k \in \{\text{True, False}\}$$

where for simplicity we are not showing the following indices inside the disjunctions: $i \in \text{TF}_k$, $\ell \in \text{SP}_k$, and $m \in \text{IP}$. To transform model (P2) into MILP form, the convex hull formulation of Eqs. (15)–(18) is applied to the disjunctive program shown above. After reordering the equations and replacing the general mass balances with variables in the disjunctions, model (P3) is derived in terms of the binary variables $y_k = \{0, 1\}$, which are associated to the existence of each column, and hence separation task. The new sets are as follows: $\text{FIS} = \{k \mid k$ is the split after initial state$\}$, $\text{IPS}_\text{m} = \{\ell \mid \ell$ is product stream from task $k\}$, $\text{IFS}_\text{m} = \{j \mid j$ is intermediate feed state for task $k\}$, $\text{IP} = \{$intermediate products $m\}$.

$$\text{(P3):} \quad \min \sum_{k \in C} (\alpha_k y_k + \beta_k F_k + C_U Q_k) \qquad (28)$$

$$\text{s.t.} \quad \sum_{k \in FIS} F_k = F_{\text{total}} \qquad (29)$$

$$\sum_{\ell \in IPS_m} F_\ell - \sum_{j \in IFS_m} \xi_j^m F_j = 0 \quad m \in IP \qquad (30)$$

$$Q_k - HK_k F_k = 0 \quad k \in C \qquad (31)$$

$$F_k - U y_k \leq 0 \quad k \in C \qquad (32)$$

$$Ay \leq a \qquad (33)$$

$$F_k, Q_k \geq 0, \quad y_k = 0, 1 \quad k \in C$$

Eq. (29) represents the mass balance for the initial splitter in terms of the total flow, F_{total}, to be processed. Eq. (30) represents the substitution of the disaggregated equations from the convex hull of the conditional task disjunctions into the permanent mixer and splitter equations. Eqs. (31) and (32) are equations for heat loads and flows from the disjunctions for conditional tasks. Eq. (33) represents the logic constraints for interconnection of tasks in (Eq. (26)), now transformed into algebraic equations with binary variables (see Raman & Grossmann, 1993).

It is interesting to note that the MILP formulation (P3) corresponds exactly to the one proposed by Andrecovich and Westerberg (1985) with the addition of the logic constraints in (Eq. (32)). Thus, this example shows that the proposed framework can be used to systematically derive models reported in the literature.

The STN model (P1) for the synthesis of sharp distillation sequences is based on the OTOE assignment. It is possible to generate a model for the VTE assignment, which will include costs that are different if a task is performed in a different column. The appendix includes the general model for the VTE approach.

5.2. SEN representation

To construct the SEN representation, it is necessary to define the states involved in a problem, as well as the number and characteristics of the equipment that are available. The states for the problem considered are the same as the ones defined for the STN representation. Engineering criteria are used to fix the number of columns and the tasks they can perform, which will be implicitly included in the representation. Since four components are involved in the original mixture, we can select three columns to perform the separation into pure components. This is provided that we allow each column to perform multiple separation tasks, in contrast to the OTOE assignment that we considered for the STN model.

The SEN representation for the separation of a four component mixture is shown in Fig. 9. Note that three columns are considered for the four component mixture (i.e. $N-1$ for N components), and the split C/D is considered in columns 2 and 3 in order to accommodate the sequence where split AB/CD is performed in the first column. Note also that the states cannot be defined independently of the task to be performed in the equipment, since the selection of a task in the equipment will determine the proper value of a state. There can be logic constraints that will eliminate or activate streams depending on the task selected for a given equipment.

From (P-SEN), the GDP model for the SEN representation in Fig. 9 can be derived by introducing the Boolean variables Y_{ik} to select task i for column k. Note that since the number of columns is known, there is no need to introduce Boolean variables for the units. Defining the sets $\text{D}_k = \{\ell \mid \ell$ is distillate stream out of column $k\}$, $\text{B}_k = \{\ell' \mid \ell'$ is bottom stream out of column $k\}$, $\text{TC}_k = \{i \mid i$ is a task allowable for column $k\}$, leads to the following model:

$$\text{(P4):} \quad \min \sum_{k \in C} (FC_k + VC_k + C_U Q_k) \qquad (34)$$

$$\text{s.t.} \quad F_\ell = \sum_{j \in OS_i} F_j \; \ell \in FS_i \quad i = 1, \ldots, NS \qquad (35)$$

$$\sum_{j \in FM_i} F_j = F_\ell \quad \ell \in OM_i \quad i = 1, \ldots, NM \qquad (36)$$

$$\bigvee_{\forall i \in TC_k} \left[\begin{array}{c} Y_{ik} \\ F_{\ell} = \xi_i^{\text{top}} F_k, \quad \ell \in D_k \\ F_{\ell'} = \xi_i^{\text{bot}} F_k, \quad \ell' \in B_k \\ FC_k = \alpha_i \\ VC_k = \beta_i F_k \\ Q_k = HK_i F_k \end{array} \right] \quad k \in C \tag{37}$$

$$\Omega(Y_{ik}) = \text{True} \tag{38}$$

$$F, FC, VC, Q \geq 0, \quad Y_{ik} \in \{\text{True, False}\}$$

Eq. (34) is the objective function identical to the one in (P1). Eqs. (35) and (36) represent the mass balances in splitters and mixers, respectively, where the mixer/splitter is permanent equipment with only one task and no associated fixed or variable cost. Eq. (37) represents the disjunction for the selection of the task to be performed in each of the three columns. The terms in the brackets represent the equations that will be applied when the task associated with the Boolean variable Y_{ik} is selected. Eq. (38) represents the logic relations for the existence of different tasks in the different columns.

A detailed analysis of the formulation (P4) and the representation for distillation sequences allows us to simplify its GDP model. Since the SEN representation has a fixed number of columns for the number of component in a mixture, it is implicit in the superstructure that only one separation path can be performed. This fact is not implicit in the STN representation, but it was imposed through the logic constraints. Because only one separation sequence can be used, it can be shown that the mixers and splitters in the SEN representation will have only one active inlet or outlet respectively. This fact allows to move their equations inside the disjunctions for the discrete tasks. It is also possible to change the definition of the split fraction of each separation, ξ_{ik}, so that they are referred to the total flow that inputs to the sequence. Problem (P5) shows the resulting GDP model.

$$\text{(P5):} \quad \min \sum_{k \in C} (FC_k + VC_k + C_U Q_k) \tag{39}$$

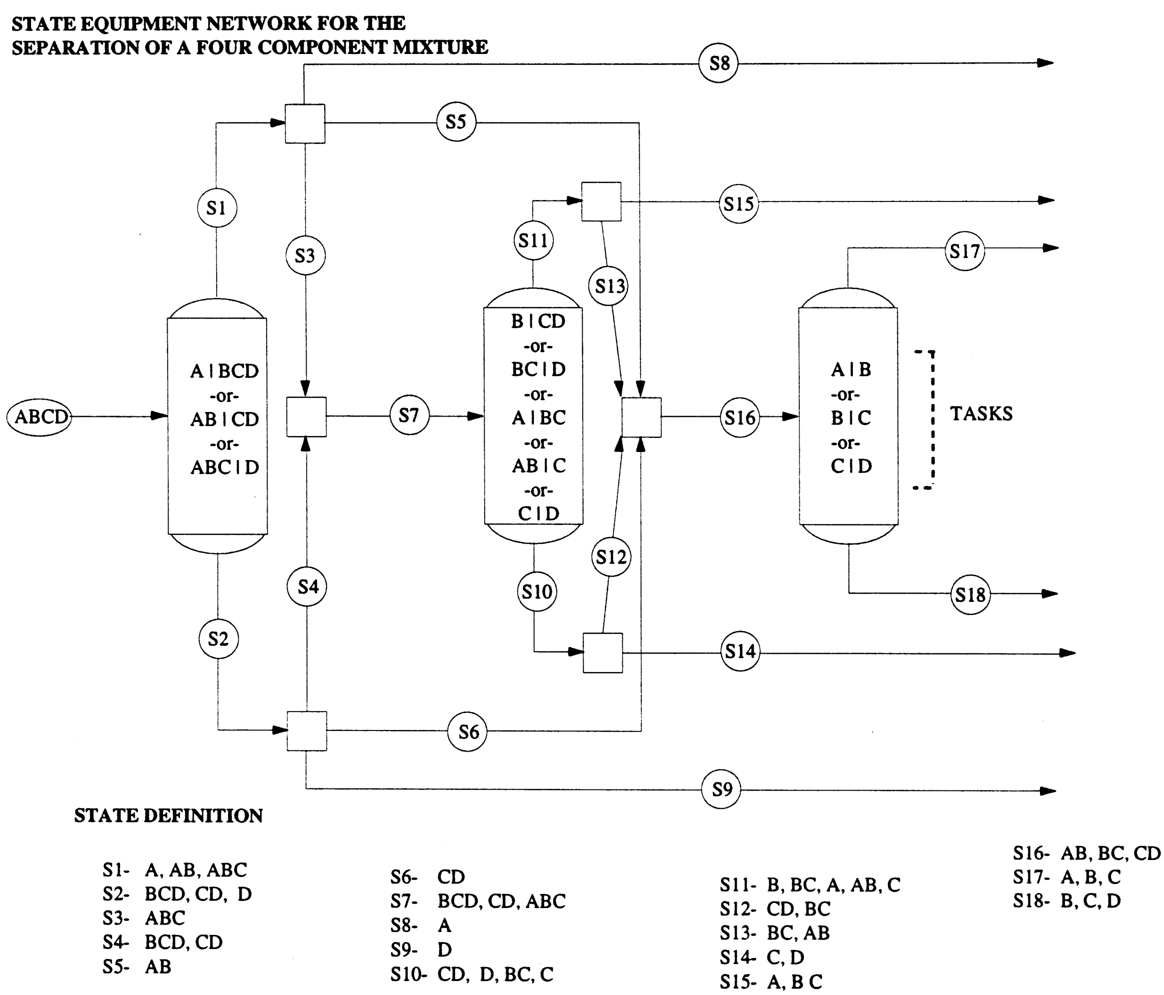

Fig. 9. SEN representation for the separation of a four component mixture.

Table 1
Comparison between STN and SEN representation for distillation sequences.

Criteria	4 Component separation		5 Component separation		6 Component separation	
	STN MILP (P3)	SEN MILP (P6)	STN MILP (P3)	SEN MILP (P6)	STN MILP (P3)	SEN MILP (P6)
Constraints	43	75	88	181	157	344
Variables	31	68	61	157	106	302
Binary vars.	10	11	20	28	35	46
Relaxed sol.	3623.81	3625.81	4278.34	4304.00	18161.56	18030.18
Integer sol.	3625.81	3625.81	4304.08	4304.00	18170.35	18170.26
B&B nodes	2	1	2	1	10	2
Iterations	30	17	53	52	165	120
CPU time [a]	0.14	0.13	.14	0.20	0.47	0.43

[a] Using GAMS/OSL in a HP 9000/712.

$$\text{s.t.} \quad \bigvee_{i\in TC_k} \left[\begin{array}{c} Y_{ik} \\ \left.\begin{array}{c} F_\ell = \xi_{ik}F_{\text{total}} \\ FC_k = \alpha_{ik} \\ VC_k = \beta_{ik}F_\ell \\ Q_k = HK_{ik}F_\ell \end{array}\right\} \ell \in IC_k \end{array} \right] \quad k\in C \tag{40}$$

$$\Omega(Y_{ik}) = \text{True} \tag{41}$$

$$F, FC, VC, Q \geq 0, \quad Y_{ik}\in\{\text{True, False}\}$$

where $IC_k = \{\ell \mid \ell \text{ is the inlet flow to column } k\}$ and ξ_{ik} is the split fraction of the intermediate mixture referred to the total mass flow when task i is performed in column k. The reduced GDP formulation (P5) can be transformed into an MILP model (P6) by means of the convex hull formulation of disjunctions introduced by Eqs. (15)–(18) and by redefining F_k as the inlet flow to column k.

$$\text{(P6):} \quad \min \sum_{k\in C} (FC_k + VC_k + C_U Q_k) \tag{42}$$

$$\left.\begin{array}{l} s.t. \quad F_k = \sum_{i\in TC_k} f_{ki} \\ FC_k = \sum_{i\in TC_k} fc_{ki} \\ VC_k = \sum_{i\in TC_k} vc_{ki} \\ Q_k = \sum_{i\in TC_k} q_{ki} \end{array}\right\} \quad k\in C \tag{43}$$

$$\left.\begin{array}{l} f_{ki} = \xi_{ki}F_{\text{total}}y_{ik} \\ fc_{ki} = \alpha_{ik}y_{ik} \\ vc_{ki} = \beta_i f_{ki} \\ q_{ki} = HK_i f_{ki} \end{array}\right\} \quad \begin{array}{c} k\in C \\ i\in TC_k \end{array} \tag{44}$$

$$\sum_{i\in TC_k} y_{ik} = 1 \quad k\in C \tag{45}$$

$$Ay \leq b \tag{46}$$

$$F, f, FC, fc, VC, vc, Q, q \geq 0, \quad y_{ik}\in\{0, 1\}$$

Eqs. (45) and (46) represent the logic relations between tasks and equipment expressed in algebraic form with binary variables.

The SEN representation can be useful for retrofit problems if Boolean variables and logic constraints are added to eliminate or activate streams that are associated with the repiping and layout of the given system. This might be done in the form of new disjunctions, or in the form of extra constraints within the existing disjunctions.

5.3. Comparison between STN and SEN in numerical examples

To compare the performance of the SEN representation with that of the STN/OTOE, distillation sequence problems with four, five and six components were solved. The problem data can be found in Raman and Grossmann (1993). Table 1 shows the results of both models, (P3) and (P6), respectively.

Model (P6) solves the four and five component problem at the root node of the B&B tree, and model (P3) requires two nodes for the same problems. However, when the number of components increases, the STN B&B requires a larger number of nodes than the SEN, even though the solution times are comparable. This behavior suggests that the computational complexity of the SEN problems is higher.

Finally, it is worth noting that the differences between the number of equations and variables for the STN and SEN models are due to the different type of disjunction used. When the disjunctions of the SEN are disaggregated they introduce a larger number of continuous and binary variables in proportion to the number of tasks each equipment can perform. However, this disadvantage is offset by the specific knowledge that is used in the SEN representation (number of columns, tasks they perform).

6. Synthesis of sharp distillation sequences with heat integration

The synthesis of distillation sequences that has been considered in the previous sections takes into account only the mass balances. The SEN and STN representations, however, can also include tasks that perform energy transfer. In this section, we will extend the STN and SEN representations for the synthesis of sharp distillation sequences to include energy balances and possibilities of heat integration.

In order to keep the models linear, the problem will be simplified as in Raman and Grossmann (1993), who proposed a heat integrated model for distillation sequences where the cost of columns operating at different pressures is represented through a linear function of the temperature in the condenser. To perform the heat integration it is necessary to include the temperatures of reboilers and condensers as variables to verify the feasibility of heat exchange.

6.1. STN representation and GDP model

In order to consider heat integration, the following approach will be used in this work. First, heat transfer tasks will be introduced in the STN/OTOE network in which the only change considered is the heat content of the corresponding streams. The heat transfer tasks can be reduced to source or sink nodes depending on whether they release or absorb heat. To account for the heat integration, all the heat transfer tasks will be integrated in a block for heat recovery as in Raman & Grossmann (1994). For the case of isothermal streams, such as is the case of this distillation problem, the block may consist of all possible matches for heat exchange (see Fig. 10). Alternatively, one might use the aggregated models by Duran and Grossmann (1986), Grossmann, Yeomans and Kravanja (1998) or the detailed HEN superstructure by Yee, Grossmann and Kravanja (1990).

In Fig. 10, note that each distillation task that was introduced in the previous section has four outlet states. Two of them involve mass flows, while the other two are for heat transfer. The heat exchanger network is based on stream splitting and has no equipment in series.

To model the problem, it was considered that the columns have a linear cost relation with the temperature of the condenser, and that the costs of the heat exchangers are not included. The Boolean variable (Y_k) is defined for the distillation tasks $k \in$C, while the Boolean variable Z_{kj} represents the match between the heat source (QC_k) from distillation task k (condensers at temperature TC_k) and the heat sink (QR_j) of distillation task j (reboiler at temperature TR_j). ZC_{kn} and ZS_{km} represent the match of heat source in distillation task k with cold utility n, and the match of heat sink from task k with hot utility m, respectively. The continuous variables QX_{kj}, QW_{kn} and QS_{km} represent the corresponding exchanges of heat. The parameter DRC_k represents the temperature difference between reboiler and condenser if separation task k takes place, TWL is the temperature of the coldest cooling utility, EMAT is the minimum approach temperature that is required for feasible heat exchange, U is an upperbound for the temperatures, and U_Q is an upperbound on the heat exchanges. The model is as follows:

(P7):

$$\min \sum_{k\in C}\left[DP_k+\beta_k F_k+\sum_{m\in HU}(pu_m QS_{km}) + \sum_{n\in CU}(pu_n QW_{kn})\right] \tag{49}$$

$$\text{s.t.} \quad \sum_{\ell\in IT_k} F_\ell = \sum_{j\in OT_k} F_j \quad k\in MST \tag{50}$$

$$QC_k = \sum_{h\in CU} QW_{nk} + \sum_{j\in C\setminus k} QX_{kj} \quad k\in C \tag{51}$$

$$QR_k = \sum_{m\in HU} QS_{mk} + \sum_{j\in C\setminus k} QX_{jk} \quad k\in C \tag{52}$$

$$\begin{bmatrix} Y_k \\ F_\ell = \xi_k^m F_i, \quad \forall i\in TF_k, \quad \forall \ell\in IP_k \\ QC_k = HK_k F_k \\ QR_k = HK_k F_k \\ TC_k = TR_k - DRC_k \\ DP_k = \dfrac{\alpha_k TC_k}{TWL+EMAT} \end{bmatrix} \vee \begin{bmatrix} \neg Y_k \\ F_\ell = 0, \quad \forall \ell\in IP_k \\ QC_k = QR_k = 0 \\ TC_k = TR_k = 0 \\ DP_k = 0 \end{bmatrix} \quad k\in C \tag{53}$$

$$\begin{bmatrix} Z_{kj} \\ TC_k \geq TR_j + EMAT \\ QX_{kj} \leq U_Q \end{bmatrix} \vee \begin{bmatrix} \neg Z_{kj} \\ TC_k \leq U \\ TR_j \leq U \\ QX_{kj} = 0 \end{bmatrix} \quad \begin{matrix} k, j\in C \\ k\neq j \end{matrix} \tag{54}$$

$$\begin{bmatrix} ZC_{kn} \\ TC_k \geq TW_n + EMAT \\ QW_{kn} \leq U_Q \end{bmatrix} \vee \begin{bmatrix} \neg ZC_{kn} \\ TC_k \leq U \\ QW_{kn} = 0 \end{bmatrix} \quad \begin{matrix} k \in C \\ n \in CU \end{matrix} \tag{55}$$

$$\begin{bmatrix} ZS_{km} \\ TR_k \leq TS_m - EMAT \\ QS_{km} \leq U_Q \end{bmatrix} \vee \begin{bmatrix} \neg ZS_{km} \\ TR_k \leq U \\ QS_{km} \leq 0 \end{bmatrix} \quad \begin{matrix} k \in C \\ m \in HU \end{matrix} \tag{56}$$

$$\Omega(Y, Z, ZS, ZC) = \text{True} \tag{57}$$

$$TC, TR, F, QX, DP, QS, QW \geq 0,$$

$$Y, Z, ZC, ZS = \{\text{True, False}\}$$

The objective function in (Eq. (49)) is expressed in terms of the investment cost, given by the fixed charge DP_k and variable cost term $\beta_k F_k$, and in terms of the hot and cold utility costs with corresponding prices, pu_m and pu_h. Eqs. (51) and (52) represent the energy balances for each condenser and heat exchanger, and equations Eqs. (54)–(56) enforce feasible temperature

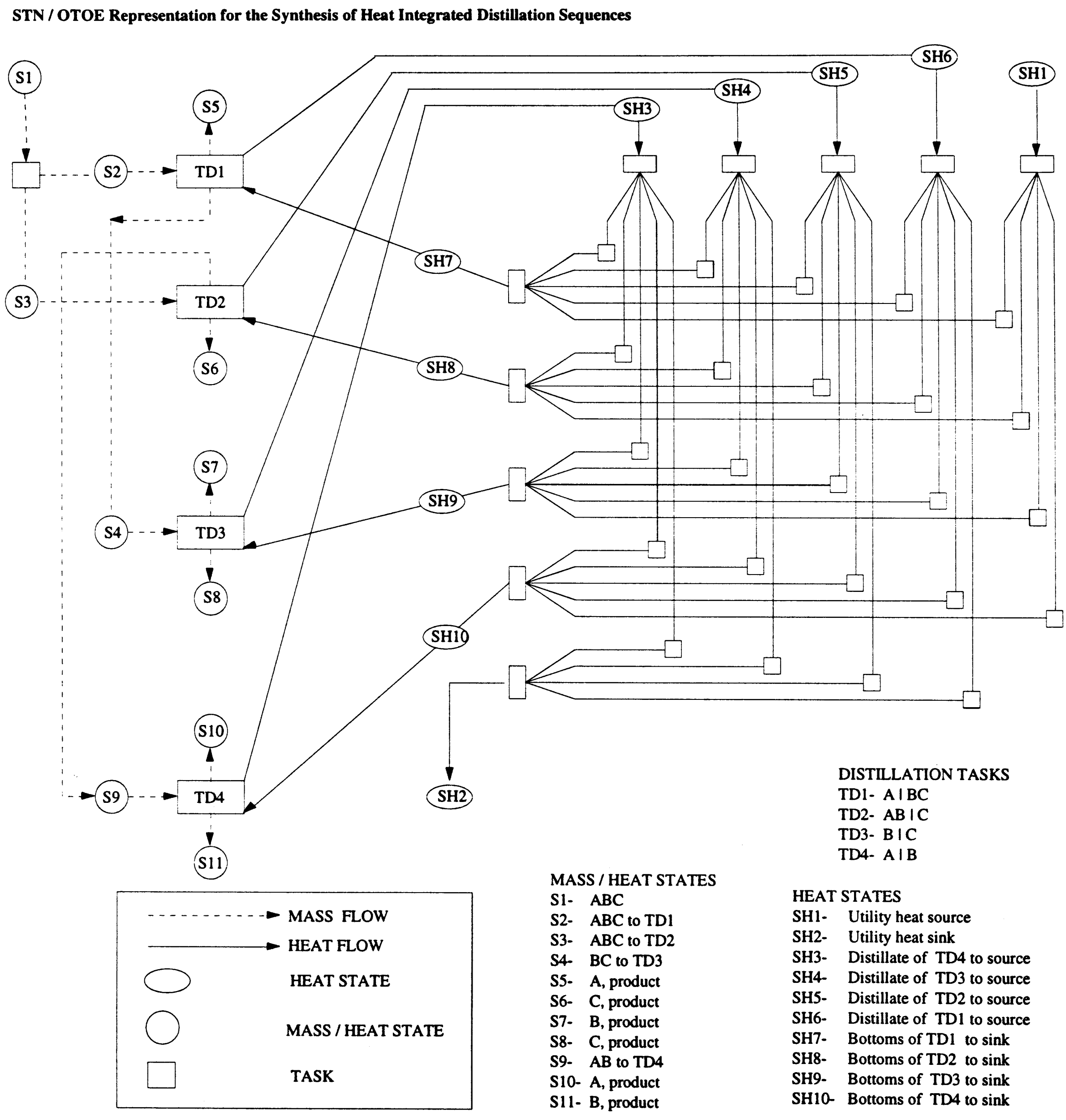

Fig. 10. STN representation for the synthesis of heat integrated distillation sequences.

differences if the corresponding match is performed. Eq. (50) corresponds to Eq. (25) in the STN model (P1), while equation Eq. (53) is similar to equation Eq. (26) plus the temperature difference for reboiler and condenser, as well as the fixed cost expressed as a function of the temperature in the condenser. The logic constraints in (Eq. (57)) include the relations that permit heat transfer only if the distillation column connected to it is active. These relations are important because heat and mass transfer are treated separately. Notice that the model includes the possibility of using multiple utilities, represented by the sets CU and HU for cooling and heating utilities, respectively.

If the convex hull formulation is applied to each disjunction in model (P7) (see Eqs. (15)–(18)), and appropriate simplifications are made, the following MILP model is obtained:

(P8):

$$\min \sum_{k\in C}\left[DP_k+\beta_kF_k+\sum_{m\in HU}(pu_mQS_{km}) + \sum_{n\in CU}(pu_nQW_{kn})\right] \tag{58}$$

$$\text{s.t.}\quad \sum_{i\in FIS}F_i=F_{\text{total}} \tag{59}$$

$$\sum_{\ell\in IPS_m}F_\ell-\sum_{j\in IFS_m}\xi_j^mF_j=0\quad m\in IP \tag{60}$$

$$HK_kF_k=\sum_{h\in CU}QW_{hk}+\sum_{j\in DT\backslash k}QX_{kj}\quad k\in C \tag{61}$$

$$HK_kF_k=\sum_{m\in HU}QS_{mk}+\sum_{j\in DT\backslash k}QX_{jk}\quad k\in C \tag{62}$$

$$TC_k=TR_k-DRC_ky_k\quad k\in C \tag{63}$$

$$DP_k=\frac{\alpha_kTC_k}{TWL+EMAT}\quad k\in C \tag{64}$$

$$\left.\begin{array}{l}F_k\le F_{\text{total}}y_k\\ TC_k\le Uy_k\\ TR_k\le Uy_k\end{array}\right\}\quad k\in C \tag{65}$$

$$\begin{array}{l}\left.\begin{array}{l}TC_k=TC1_{kj}^X+TC2_{kj}^X\\ TR_k=TR1_{kj}^X+TR2_{kj}^X\end{array}\right\}\quad j,k\in C,\quad k\neq j\\ TC_k=TC1_{kn}^W+TC2_{kn}^W\quad k\in C,\quad n\in CU\\ TR_k=TR1_{km}^S+TR2_{km}^S\quad k\in C,\quad m\in HU\end{array} \tag{66}$$

$$\left.\begin{array}{r}TC1_{kj}^X\ge TR1_{jk}^X+EMATz_{kj}\\ QX_{kj}\le U_Qz_{kj}\\ TC1_{kj}^X\le Uz_{kj}\\ TR1_{jk}^X\le Uz_{kj}\\ TC2_{kj}^X\le U(1-z_{kj})\\ TR2_{jk}^X\le U(1-z_{kj})\end{array}\right\}\quad \begin{array}{c}k,j\in C\\ k\neq j\end{array} \tag{67}$$

$$\left.\begin{array}{r}TR1_{km}^S\le(TS_m-EMAT)zs_{km}\\ QS_{km}\le U_Qzs_{km}\\ TR1_{km}^S\le Uzs_{km}\\ TR2_{km}^S\le U(1-zs_{km})\end{array}\right\}\quad \begin{array}{c}k\in C\\ m\in HU\end{array} \tag{68}$$

$$\left.\begin{array}{r}TC1_{kn}^W\ge(TW_n+EMAT)zc_{kn}\\ QW_{kn}\le U_Qzc_{kn}\\ TC1_{kn}^W\le Uzc_{kn}\\ TC2_{kn}^W\le U(1-zc_{kn})\end{array}\right\}\quad \begin{array}{c}k\in C\\ n\in CU\end{array} \tag{69}$$

$$[y^T,z^T,zs^T,zc^T]^TA^T\le a^T \tag{70}$$

$$TC,TR,F,QX,DP,QS,QW\ge 0,\quad y,z,zc,zs=\{0,1\}$$

$$TC1,TC2,TR1,TR2\ge 0$$

In the equations above, recall that FIS = $\{i \mid i$ is a split of the initial stream$\}$, IP = $\{m \mid m$ is an intermediate product$\}$, $\text{IPS}_n=\{\ell \mid \ell$ is product stream from task $k\}$, and $\text{IFS}_n=\{j \mid j$ is intermediate feed state for task $k\}$. The Boolean variables in (P7) are replaced by their corresponding binary variables in lowercase. Eqs. (59) and (60) were derived in similar fashion to the ones in (P3), after the same variable and equation reduction. Eqs. (61) and (62) are also derived after applying the convex hull to (Eq. (53)) and substituting variables in Eqs. (51) and (52). Eqs. (63) and (64) correspond to those equations inside disjunction (Eq. (53)) that could not be reduced. Eq. (65) includes the binary variables that will control the selection of a given task, and will therefore affect Eqs. (59)–(64). Eq. (66) includes the disaggregation of variables for the disjunction sets (Eqs. (54)–(56)), after the application of the convex hull formulation. Eqs. (67)–(69) correspond to the rest of the convex hull formulation, for exchange between reboiler and condenser (Eq. (67)), condenser and cooling water (Eq. (68)), and reboiler and steam (Eq. (69)). Finally, Eq. (70) represents the logic relations between separation and heat exchange tasks, given in algebraic expressions.

It is possible to generate an STN/VTE representation for this problem, but it will not be shown here because the interest of this section is to reproduce existing models in order to demonstrate the representation and modeling framework presented in this paper.

6.2. SEN representation and modeling of heat integrated distillation sequences

For the SEN representation of the synthesis of heat integrated sharp distillation sequences, the heat exchange part will be based on similar superstructure criteria used for the STN model (e.g. stream splitting and parallel exchangers). The part corresponding to the distillation will be treated in the same way as in non-integrated distillation sequences.

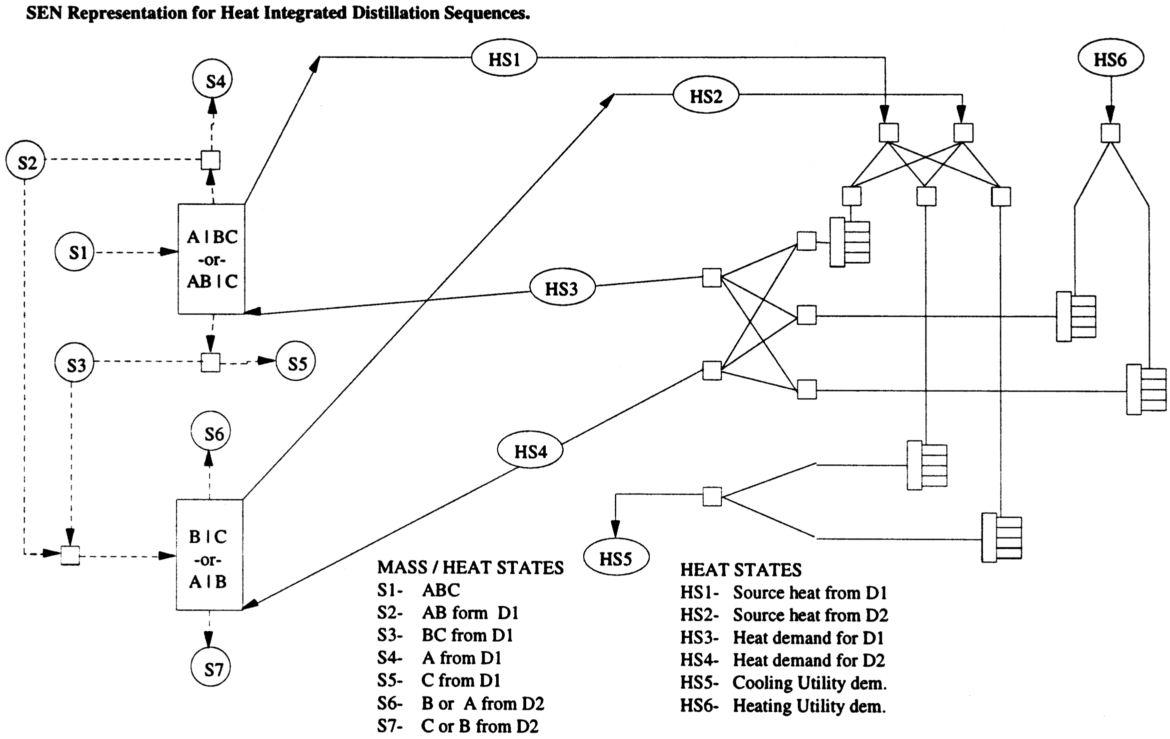

Fig. 11. SEN representation for the separation of a three component mixture with heat integration.

Since the SEN representation requires first to determine the equipment and states involved, it is necessary to select the number of distillation columns and heat exchangers that will be involved in the superstructure. For the representation shown in Fig. 11, the minimum number of columns and exchangers was chosen (number of columns $= N - 1$, where N is number of components to separate).

The same consideration of heat flows that are treated independently of the mass flows for the STN representation can be applied to the SEN. The heat flows will now be related to equipment units instead of tasks.

The intermediate states before and after the heat transfer operations have been omitted in the representation for clarity purposes. Note that the representation in Fig. 11 covers all the possible equipment matches for heat transfer, but this is performed by postulating several exchangers for which no specific heat exchange task is pre-assigned (see also Yee & Grossmann, 1991). Note that it is possible to only have a match among either the first column reboiler and the second column condenser or vice versa, but not both. Also, the possibility of both reboilers in Fig. 10 being operated with steam, and both condensers with cooling water is considered. This is an example of the application of engineering knowledge when constructing the SEN representation.

Let $C = \{k \mid k$ is an available column$\}$, $E = \{j \mid j$ is an exchanger$\}$, $TC_k = \{i \mid i$ is a task to be performed by column $k\}$, $EC_k = \{i \mid i$ is a heat exchanger that can remove heat from condenser in column $k\}$, $ER_k = \{j \mid j$ is a heat exchanger that can provide heat to reboiler in column $k\}$, $EE = \{\ell \mid \ell$ is the set of exchanger equipment$\}$, $SO_k = \{i \mid i$ is a heat source for exchanger $\ell\}$, $SI_\ell = \{j \mid j$ is a heat sink for exchanger $\ell\}$.

The variable definition is as follows: $QR_{k'}$ represents the amount of heat required by the reboiler in column k'. QC_k is the amount of heat to be removed in the condenser of column k. $QX_{kk'}$ is the amount of heat exchanged between condenser of column k and reboiler of column k'. QXW_{kn} is the heat exchanged between condenser k and cooling utility n. $QXS_{k'm}$ is the heat exchanged between reboiler k' and heating utility n. The GDP model (P9) is then as follows:

(P9):

$$\min \sum_{k\in C}\left[(DP_k + VC_k) + \sum_{m\in HU}(pu_m QXS_{k'm}) + \sum_{n\in CU}(pu_n QXW_{kn})\right] \tag{71}$$

$$\text{s.t.}\quad F_\ell = \sum_{j\in OS_i} F_j \quad \forall \ell \in FS_i, \quad i = 1, \ldots, NS \tag{72}$$

$$\sum_{j\in FM_i} F_j = F_\ell \quad \forall \ell \in OM_i, \quad i = 1, \ldots, NM \tag{73}$$

$$QC_k = \sum_{k'\neq k} QX_{kk'} + \sum_{n\in CU} QXW_{kn} \quad k\in C \tag{74}$$

$$QR_{k'} = \sum_{k \neq k'} QX_{kk'} + \sum_{n \in HU} QXS_{k'n} \quad k' \in C \tag{75}$$

$$TC_i = TR_i - DTC_i \quad i \in T \tag{76}$$

$$\bigvee_{i \in TC_k} \begin{bmatrix} Y_{ik} \\ F_\ell = \xi_i^{\text{top}} F_k, \quad \ell \in D_k \\ F_j = \xi_i^{\text{bot}} F_k, \quad j \in B_k \\ DP_k = \dfrac{\alpha_i TC_k}{TWL + EMAT} \\ VC_k = \beta_i F_k \\ QC_k = QR_k = HK_i F_k \\ TC_k = TC_i \\ TR_k = TR_i \end{bmatrix} \quad k \in C \tag{77}$$

$$\begin{bmatrix} Z_j \\ \bigvee_{\substack{k,k' \in C \\ k \neq k'}} \begin{bmatrix} W_{jkk'} \\ Q_j = QX_{kk'} \\ TC_k \geq TR_{k'} + EMAT \end{bmatrix} \\ \bigvee_{\substack{n \in CU \\ k \in C}} \begin{bmatrix} W_{jkn} \\ Q_j = QXW_{kn} \\ TC_k \geq TR_{k'} + EMAT \end{bmatrix} \\ \bigvee_{\substack{m \in HU \\ k \in C}} \begin{bmatrix} W_{jkm} \\ Q_j = QXS_{k'm} \\ TS_m \geq TR_k + EMAT \end{bmatrix} \end{bmatrix} \vee \begin{bmatrix} \neg Z_j \\ Q_j = 0 \end{bmatrix} \quad j \in E \tag{78}$$

$$\Omega(Y, W, Z) = \text{True} \tag{79}$$

$TC, TR, F, QXC, QXR, QC, QR, DP, QC^{so}, QR^{si}, FC, VC \geq 0,$

$Y_{ik} = \{\text{True, False}\}, \quad Z_j = \{\text{True, False}\},$

$W_{ij} = \{\text{True, False}\}$

In model (P9), Eq. (71) is the objective function in terms of fixed and variable costs. Note that HU is a subset of SO_ℓ and CU is a subset of SI_ℓ. Eqs. (72) and (73) represent the mass balances in all the mass splitters and mixers, while Eqs. (74) and (75) represent the energy balances in the heat mixers and splitters. Eq. (76) represents the thermodynamic constraint for temperature difference in every column. Eq. (77) is the disjunction of tasks for the permanent columns, and include mass balances, energy balances and thermodynamic constraints for the column. Eq. (78) represents the disjunction for the conditional heat exchangers, where the task that represents the match of hot and cold streams in the exchanger is given by the innermost disjunction, selected by the Boolean variable W_{ij}. When a heat exchanger is not used ($Z_j = \text{False}$), the heat exchange variables are set to zero, and the thermodynamic temperature difference constraint is not enforced. Finally, Eq. (66) represents the logic connectivity between columns, exchangers and heat exchange matches.

The SEN model (P9) can be transformed in MILP form using the convex hull formulation of disjunctions, leading to model (P10).

(P10):

$$\min \sum_{k \in C} \left[(DP_k + VC_k) + \sum_{m \in HU} (SP_m QXS_{km}) + \sum_{n \in CU} (WP_n QXW_{kn}) \right]$$

$$\text{s.t.} \quad F_\ell = \sum_{j \in OS_i} F_j \quad \ell \in FS_i, \quad i = 1, \ldots, NS \tag{80}$$

$$\sum_{j \in FM_i} F_j = F_\ell \quad \ell \in OM_i, \quad i = 1, \ldots, NM \tag{81}$$

$$QC_k = \sum_{k' \neq k} QX_{kk'} + \sum_{n \in CU} QXW_{kn} \quad k \in C \tag{82}$$

$$QR_{k'} = \sum_{k \neq k'} QX_{kk'} + \sum_{n \in HU} QXS_{k'n} \quad k' \in C \tag{83}$$

$$F_\ell = \sum_{i \in TC_k} FF_{i\ell} \quad \ell \in D_k, \quad k \in C \tag{84}$$

$$F_j = \sum_{i \in TC_k} FF_{ij} \quad j \in B_k, \quad k \in C \tag{85}$$

$$\left. \begin{aligned} F_k &= \sum_{i \in TC_k} FF_{ik} \\ DP_k &= \sum_{i \in TC_k} DDP_{ik} \\ TC_k &= \sum_{i \in TC_k} TTC_{ik} \\ TR_k &= \sum_{i \in TC_k} TTR_{ik} \\ VC_k &= \sum_{i \in TC_k} VVC_{ik} \\ QC_k &= \sum_{i \in TC_k} QQC_{ik} \\ QR_k &= \sum_{i \in TC_k} QQR_{ik} \end{aligned} \right] \quad k \in C \tag{86}$$

$$FF_{i\ell} = \xi_{ik}^{\text{top}} FF_{ik} \quad \ell \in D_k, \quad i \in TC_k, \quad k \in C \tag{87}$$

$$FF_{ij} = \xi_{ik}^{\text{bot}} FF_{ik} \quad j \in B_k, \quad i \in TC_k, \quad k \in C \tag{88}$$

$$\left. \begin{aligned} DPP_{ik} &= \frac{\alpha_{ik} TTC_{ik}}{TWL + EMAT} \\ VC_{ik} &= \beta_{ik} FF_{ik} \\ TTC_{ik} &= TTR_{ik} - DTC_{ik} y_{ik} \\ QQC_{ik} &= QQR_{ik} = HK_{ik} FF_{ik} \end{aligned} \right\} \quad \begin{matrix} k \in C \\ i \in TC_k \end{matrix} \tag{89}$$

$$FF_{i\ell} \leq F_{\text{total}} y_{ik} \quad \ell \in D_k, \quad i \in TC_k, \quad k \in C \tag{90}$$

$$FF_{ij} \leq F_{\text{total}} y_{ik} \quad j \in B_k, \quad i \in TC_k, \quad k \in C \tag{91}$$

$$\left.\begin{array}{l} FF_{ik} \leq F_{\text{total}} y_{ik} \\ TTC_{ik} \geq TWL y_{ik} \\ TTR_{ik} \leq TSU y_{ik} \end{array}\right\} \begin{array}{c} k \in C \\ i \in TC_k \end{array} \tag{92}$$

$$\sum_{i \in TC_k} y_{ik} = 1 \quad k \in C \tag{93}$$

$$\left.\begin{array}{l} TC_k = XTCE^1_{kk'j} + XTCE^2_{kk'j} \\ TC_k = XTCU^1_{knj} + XTCU^2_{knj} \\ TR_k = XTRE^1_{kk'j} + XTRE^2_{kk'j} \\ TR_k = XTRU^1_{k'mj} + XTRU^2_{k'mj} \end{array}\right\} \begin{array}{c} j \in E \\ k, k' \in C, k \neq k' \\ n \in CU \\ m \in HU \end{array} \tag{94}$$

$$QX_{kk'} = \sum_{j \in E} QQX_{jkk'} \quad k, k' \in C, k \neq k'$$

$$QXW_{kn} = \sum_{j \in E} QQXW_{jkn} \quad k \in C, n \in CU$$

$$QXS_{kn} = \sum_{j \in E} QQXS_{jk'm} \quad k \in C, m \in HU \tag{95}$$

$$Q_j = \sum_k \sum_{k', k \neq k'} QQX_{jkk'} + \sum_k \sum_n QQXW_{jkn} + \sum_{k'} \sum_m QQXS_{jk'm} \quad j \in E \tag{96}$$

$$XTCE^1_{kk'j} \geq XTRE^1_{kk'j} + EMAT w_{jkk'} \quad j \in E, \quad k, k' \in C, \quad k \neq k' \tag{97}$$

$$XTCU^1_{knj} \geq (TW_n + EMAT) w_{jkn} \quad j \in E, \quad k \in C, \quad n \in CU \tag{98}$$

$$XTRU^1_{k'mj} \leq (TS_m - EMAT) w_{jk'm} \quad k' \in C, \quad j \in E, \quad m \in HU \tag{99}$$

$$\left.\begin{array}{l} XTCE^1_{kk'j} \leq U w_{jkk'} \\ XTRE^1_{kk'j} \leq U w_{jkk'} \end{array}\right\} \begin{array}{c} j \in E \\ k', \quad k \in C, \quad k \neq k' \end{array}$$

$$XTCU^1_{knj} \leq U w_{jkn} \quad j \in E, \quad k \in C, \quad n \in CU$$

$$XTRU^1_{k'mj} \leq U w_{jk'm} \quad j \in E, \quad k \in C, \quad m \in CU \tag{100}$$

$$\left.\begin{array}{l} QQX_{jkk'} \leq U w_{jkk'} \quad k, k' \in C, \quad k \neq k' \\ QQXW_{jkn} \leq U w_{jkn} \quad n \in CU \\ QQXS_{jk'm} \leq U w_{jk'm} \quad m \in HU \end{array}\right\} j \in E \tag{101}$$

$$XTCE^2_{kk'j} \leq U(1 - w_{jkk'}) \quad j \in E, \quad k,k' \in C$$

$$XTRE^2_{kk'j} \leq U(1 - w_{jkk'}) \quad j \in E, \quad k,k' \in C$$

$$XTCU^2_{knj} \leq U(1 - w_{jkn}) \quad j \in E, \quad k \in C, \quad n \in CU$$

$$XTRU^2_{kmj} \leq U(1 - w_{jkm}) \quad j \in E, k \in C, \quad m \in HU \tag{102}$$

$$Q_j \leq U z_j \quad j \in E \tag{103}$$

$$\sum_k \sum_{k', k \neq k'} w_{jkk'} + \sum_k \sum_n w_{jkn} + \sum_{k'} \sum_m w_{jk'm} = z_j \quad j \in E \tag{104}$$

$$[y^T, z^T, w^T]^T A^T \leq a^T \tag{105}$$

F_k, FF_{ik}, QC_k, QR_k, TC_k, TR_k, TTC_{ik}, TTR_{ik}, DDP_{ik}, DP_{ik}, QQC_{ik}, QQR_{ik}, $QXS_{k'm} \geq 0$

$QQX_{jkk'}$, $QQXW_{jkn}$, $QQXS_{jk'm}$, VC_k, VVC_{ik}, XTC_{kj}, XTR_{kj}, $QX_{kk'}$, $QXW_{kn} \geq 0$

$y_{ik} = \{0, 1\}, \quad z_\ell = \{0, 1\}, \quad w_{ij} = \{0, 1\}$

As in model (P8), Eqs. (80)–(83) represent the mass balances in permanent mass transfer equipment with a single task, and energy balances in permanent energy transfer equipment with a single task. Eqs. (84)–(86) represent the disaggregation of variables for each term of the disjunction in (Eq. (77)), while Eqs. (87)–(88) enforce the equations for each task permissible in column k. Eqs. (90)–(93) are the rest of the constraints generated by the application of the convex hull: bounding constraints and a logic constraints that indicate exactly one task has to be selected for each column, and each task can be selected only for one column. Eqs. (94) and (95) represent the disaggregation of variables needed to model conditional equipment with multiple tasks (the heat exchangers). Eqs. (96)–(99) represent the application of the constraints inside the disjunction (Eq. (78)), plus the extra equations introduced by the convex hull formulation (Eqs. (100)–(103)). Eq. (104) represents the logic constraint that indicates that each heat exchanger in the superstructure can perform at most one task. Finally, Eq. (105) represents the logic constraints for connectivity of equipment in algebraic form.

6.3. Numerical results for heat integrated distillation sequences

The problem of determining the optimal separation sequence for a three component mixture with heat integration was solved using the MILP models from the STN/OTOE and SEN representations. The data for this problem can be found in Raman and Grossmann (1993).

Table 2 presents the results of the STN and SEN models compared to the solution of the Raman and Grossmann (1993) model. It shows that the STN model is more efficient than the model by Raman and Grossmann, in terms of the size of the B&B tree and of the CPU time. The SEN model in this case required considerably more time due to its greatly increased size. The relaxations of all the models are rather weak, although it is somewhat better in the case of the STN model (P8). The explanation for the poor relaxations comes from the fact that the disjunctions for the conditional terms require the use of Eq. (17), whose upper bounds cannot be very tight. On the other hand, the Raman and Grossmann (1993) model makes use of a big-M constraint for the cost function, while the SEN and STN models do not. An explanation for the poor behavior of the SEN is the handling of the heat exchanger network with exchangers that can perform multiple matches. Yee and Grossmann (1991) used a similar model and also reported large computational times for solving this type of problems.

7. Conclusions

The objective of this paper has been to introduce a general framework to represent, model and solve process synthesis problems that are described by linear models. To achieve this goal, we introduced a systematic modeling framework consisting of three stages: superstructure representation, modeling of the problem with GDP, and the reformulation as an MILP problem.

The basic elements of superstructures in synthesis problems were identified as states, tasks and equipment. The relationship that each of these elements can have, leads to two representation approaches: the STN, and the SEN. It was shown that these representations are complementary to each other, although they generally give rise to different optimization models. It was also shown that the STN representation can have two major cases for the assignment of equipment: OTOE and VTE. Once the representation is specified, the problem is modeled as a Generalized Disjunctive Program (GDP). Finally, using the convex hull formulation for each disjunction, the GDP problem is transformed into an MILP problem.

The systematic framework was tested in the derivation of models for linear synthesis problems involving sharp distillation sequences. It was demonstrated that the MILP model developed from the STN representation in the OTOE case reduces to the model developed by Andrecovich and Westerberg (1985). The model developed from the SEN representation showed that it is simple to add knowledge on the number of units and their interconnections, which can help to reduce the number of alternatives for the search of the optimal design. Numerical results showed that the proposed systematic models offered a comparable or even improved performance compared to the existing models.

The proposed framework was also tested in the synthesis of sharp distillation sequences with heat integration. The STN and SEN representations and their models where derived for this problem, and numerical tests showed that the performance of the STN model is more efficient than the model developed by Raman and Grossmann (1993). The SEN model proved in this case to be much more expensive to solve.

In summary, the proposed modeling framework using either the STN or SEN representations offers the capability of systematically deriving models for the solution of process synthesis problems. Work is under way to extend this framework to more complex models, that involve non-linear short-cut and tray by tray models. Also, we intend to tackle a broader type of synthesis problems, where mass, heat and momentum transfer operations are involved.

Table 2
Comparative results for the synthesis of heat integrated distillation sequences.

Criteria	Raman and Grossmann	STN OTOE (P8)	SEN (P10)
Constraints	130	282	861
Variables	88	184	597
Binary vars.	32	32	121
Relaxed sol.	311.67	313.99	292.35
Integer sol.	1040.45	1040.45	1040.45
B&B nodes	226	101	1326
Iterations	461	285	4586
CPU time [a]	3.13	2.46	34.33

[a] Using GAMS/OSL in a HP 9000/712.

Acknowledgements

The authors will like to thank the support received for this project from Consejo Nacional de Ciencia y Tecnologia (CONACYT) México, and partial support from the National Science Foundation under grant CTS-9710303.

Appendix A. VTE model for sharp distillation synthesis

The VTE model for the synthesis of sharp distillation sequences can be derived directly from (P1), by identification of the permanent tasks with a single equipment assignment (Eq. (6)), permanent tasks with multiple equipment assignment (Eq. (7)), and conditional tasks with multiple equipment assignment (Eq. (8)). For the distillation sequences problem, it is possible to consider mixers and splitters as permanent tasks with only one possible equipment assignment. The separation tasks, however, are conditional tasks that can have multiple equipment assignment. Consider that each separation task k can be performed in a subset of the equipment available $EC_k = \{i \mid \text{column } i \text{ can perform task } k\}$. The STN/VTE model is then as follows:

$$\text{(P-1A):} \quad \min \sum_{k \in C} (FC_k + VC_k + U_C Q_k) \tag{a1}$$

$$\text{s.t.} \quad F_\ell = \sum_{j \in OS_i} F_j \quad \ell \in FS_i \quad i = 1, 2, \ldots, NS \tag{a2}$$

$$\sum_{j \in FM_i} F_j = F_\ell \quad \ell \in OM_i \quad i = 1, 2, \ldots, NM \tag{a3}$$

$$\begin{bmatrix} Y_k \\ \left.\begin{matrix} F_\ell = \xi_k^{\text{top}} F_i \\ F_{\ell'} = \xi_k^{\text{bot}} F_i \\ Q_k = HK_{ki} \end{matrix}\right\} \begin{matrix} \ell \in OTT_k \\ \ell' \in OBT_k \\ i \in TF_k \end{matrix} \\ \bigvee_{i \in EC_k} \begin{bmatrix} W_{ki} \\ FC_k = \alpha_{ki} \\ VC_k = \beta_{ki} F_k \end{bmatrix} \end{bmatrix} \vee \begin{bmatrix} \neg Y_k \\ F_\ell = 0, \quad \ell \in OTT_k \\ F_{\ell'} = 0, \quad \ell' \in OBT_k \\ F_i = 0, \quad i \in TF_k \\ FC_k = 0 \\ VC_k = 0 \\ Q_k = 0 \end{bmatrix}$$

$$k \in C \tag{a4}$$

$$\Omega(Y_k, W_{ki}) = \text{True} \tag{a5}$$

$$F, FC, VC, Q \geq 0, \quad Y_k \in \{\text{True, False}\}$$

Eq. (a1) represents the objective function, which does not change with respect to the one in problem (P1), because problem (P-1A) is a formulation also based on tasks. Eqs. (a2) and (a3) represent the mass balances in mixers and splitters. Eq. (a4) represents the constraints that need to be enforced when a separation task is selected. Note that the mass balances are not equipment dependent, and this is because the separations are sharp. The cost functionalities are dependent on the equipment i that is selected for task k. Note that if the task is selected ($Y_k = \text{True}$), then exactly one of the available equipment has to be assigned to the task ($W_{ki} = \text{True}$). If a task does not exist ($Y_k = \text{False}$), then no equipment or task constraint are applied. Eq. (a5) states the logic relations that establish the task sequences and the assignment of equipment to each task.

Model (P-1A) can be transformed into a MILP by means of the convex hull of each disjunction. Reduction in the number of variables and constraints can be performed in a similar way as in the simplification of (P1) into (P2). The resulting MILP formulation is given by:

$$\text{(P-2A):} \quad \min \sum_{k \in C} (FC_k + VC_k + U_C Q_k) \tag{a6}$$

$$\text{s.t.} \quad \sum_{k \in FIS} F_k = F_{\text{total}} \tag{a7}$$

$$\sum_{\ell \in IPS_m} F_\ell - \sum_{j \in IFS_m} \xi_j^m F_j = 0 \quad m \in IP \tag{a8}$$

$$F_k - U y_k \leq 0 \quad k \in C \tag{a9}$$

$$\left.\begin{matrix} FC_k = \sum_{i \in EC_k} DFC_{ik} \\ VC_k = \sum_{i \in EC_k} DVC_{ik} \end{matrix}\right\} \begin{matrix} k \in C \\ i \in EC_k \end{matrix} \tag{a10}$$

$$\left.\begin{matrix} DVC_{ik} = \beta_{ik} F_k \\ DQ_{ik} = HK_{ik} F_k \end{matrix}\right\} \begin{matrix} k \in C \\ i \in EC_k \end{matrix} \tag{a11}$$

$$\left.\begin{matrix} DVC_{ik} \leq U_M w_{ik} \\ DFC_{ik} = \alpha_{ik} w_{ik} \end{matrix}\right\} \begin{matrix} k \in C \\ i \in EC_k \end{matrix} \tag{a12}$$

$$\sum_{i \in EC_k} w_{ik} = y_k \quad k \in C \tag{a13}$$

$$[y_k^T, w_{ik}^T]^T A^T \leq a^T \tag{a14}$$

$$F_k, Q_k, VC_k, FC_k, DQ_{ik}, DVC_{ik}, DFC_{ik}, \geq 0,$$

$$y_k = \{0, 1\}, \quad w_{ik} = \{0, 1\}$$

Eqs. (a6), (a7), (a8) and (a9) correspond exactly to Eqs. (28), (30) and (32). Eq. (a10) represents the disaggregation of variables needed for the second level of disjunctions. Eq. (a11) along with the last line of (Eq. (a12)) are the equations that need to be enforced when selecting a certain equipment for a given task. The first line of Eq. (a12) represent the bounding constraints introduced by the convex hull formulation. Eq. (a13) is used to consider the case when a task is not selected, and therefore no equipment is needed to be assigned to that task. Finally, Eq. (a14) is the algebraic representation of the logic relations between the tasks and equipment.

References

Aggrawal, A., & Floudas, C. A (1990). Synthesis of general distillation sequences—nonsharp separations. *Computers and Chemical Engineering*, *14*, 631.

Andrecovich, M. J., & Westerberg, A. W (1985). An MILP formulation for heat-integrated distillation sequence synthesis. *AIChE Journal*, *31*, 363.

Bagajewicz, M. J., & Manousiothakis, V. (1992). Mass/heat-exchange network representation of distillation networks. *AIChE J.*, *38*, 1769.

Balakrishna, S., & Biegler, L. T (1992). Constructive targeting approaches for the synthesis of chemical reactor networks. *Industrial Engineering and Chemical Research*, *31*, 300.

Balas, E. (1979). Disjunctive programming. *Annals of Discrete Mathematics.*, *5*, 3.

Balas, E. (1985). Disjunctive programming and a hierarchy of relaxations for discrete optimization problems. *SIAM Journal Alg Discrete Methods*, *6*, 466.

Beaumont, N. (1991). An algorithm for disjunctive programs. *European Journal of Operation Research*, *48*, 362.

Ciric, A. R., & Floudas, C. A. (1991). Heat exchanger network synthesis without decomposition. *Computers and Chemical Engineering*, *15*, 385.

Douglas, J. M (1988). *Conceptual design of chemical processes*. New York: McGraw-Hill Chemical Engineering Series.

Duran, M. A., & Grossmann, I. E. (1986). Simultaneous optimization and heat integration of chemical processes. *AIChE Journal*, *32*, 123.

Floudas, C. A., & Paules, G. E. (1988). A mixed-integer nonlinear programming formulation for the synthesis of heat-integrated distillation sequences. *Computers and Chemical Engineering*, *12*, 531.

Gaminibandara, K. & Sargent, R. W. H. (1976). Optimal design of plate distillation columns. In L. C. W. Dixon, *Optimization in action*. Academic Press.

Grossmann, I.E. (1996). Mixed-integer optimization techniques for algorithmic process synthesis. In J. L Anderson, *Advances in chemical engineering* vol. 23 (pp. 172–239). Academic Press.

Grossmann, I. E., Yeomans, H., & Kravanja, Z. (1998). A rigorous disjunctive optimization model for simultaneous flowsheet optimization and heat integration. *Supplement to Computers and Chemical Engineering*, *22*, S157.

Hooker, J. N. & Osorio, M. A. (1996). Mixed logical/linear programming. In *Engineering Design and Research Center Technical Report*, Carnegie Mellon University.

Kocis, G. R., & Grossmann, I. E. (1989). A modeling and decomposition strategy for the MINLP optimization of process flowsheets. *Computers and Chemical Engineering*, *13*, 307.

Kokossis, A. C., & Floudas, C. A. (1991). Synthesis of isothermal reactor-separator-recycle systems. *Chemistry and Engineering Science*, *46*, 1361.

Kondili, E., Pantelides, C. C., & Sargent, R. W. H. (1993). A general algorithm for short-term scheduling of batch operations-I. MILP formulation. *Computers and Chemical Engineering*, *17*, 211.

Papalexandri, K. P., & Pistikopoulos, E. N. (1996). Generalized modular representation framework for process synthesis. *AIChE Journal*, *42*, 1010.

Raman, R., & Grossmann, I. E. (1993). Symbolic integration of logic in mixed-integer linear programming techniques for process synthesis. *Computers and Chemical Engineering*, *17*, 909.

Raman, R., & Grossmann, I. E. (1994). Modelling and computational techniques for logic based integer programming. *Computers and Chemical Engineering*, *18*, 563.

Sargent, R. W. H. (1998). A functional approach to process synthesis and its application to distillation systems. *Computers and Chemical Engineering*, *22*, 31.

Siirola, J. J., & Rudd, D. F. (1971). Computer-aided synthesis of chemical process designs. *Industrial Engineering and Chemistry Fundamentals*, *10*, 353.

Smith, E. M. (1996). On the optimal design of continuous processes, Ph.D. Dissertation, under supervision of C. Pantelides. Imperial College of Science, Technology and Medicine, London, UK.

Turkay, M., & Grossmann, I. E. (1996a). Logic-based MINLP algorithms for the optimal synthesis of process networks. *Computers and Chemical Engineering*, *20*, 959.

Turkay, M., & Grossmann, I. E. (1996b). Disjunctive programming techniques for the optimization of process systems with discontinuous investment costs-multiple size regions. *Industrial Engineering and Chemistry Research*, *35*, 2611.

Yee, T. F., & Grossmann, I. E. (1990). Simulaneous optimization models for heat integration-II. Heat exchanger network synthesis. *Computers and Chemical Engineering*, *14*, 1165.

Yee, T. F., Grossmann, I. E., & Kravanja, Z. (1990). Simultaneous optimization models for heat integration- III. Process and heat exchanger network optimization. *Computers and Chemical Engineering*, *14*, 1185.

Yee, T. F., & Grossmann, I. E. (1991). A screening and optimization approach for the retrofit of heat-exchanger networks. *Industrial Engineering and Chemistry Research*, *30*, 146.