

Optimal Synthesis of Complex Distillation Columns Using Rigorous Models

Ignacio E. Grossmann¹, Pío A. Aguirre² and Mariana Barttfeld²

¹Department of Chemical Engineering, Carnegie Mellon University, Pittsburgh, PA 15213,
USA.

²INGAR- Instituto de Desarrollo y Diseño - Avellaneda 3657, 3000 Santa Fe, Argentina.

Abstract

The synthesis of complex distillation columns has remained a major challenge since the pioneering work by Sargent and Gaminibanadara that was reported in 1976. In this paper we first provide a review of recent work for the optimal design of distillation of individual columns using tray-by-tray models. We examine the impact of different representations and models, NLP, MINLP and GDP, as well as the importance of appropriate initialization schemes. We next provide a review of the synthesis of complex column configurations for zeotropic mixtures and discuss different superstructure representations as well as decomposition schemes for tackling these problems. Finally, we briefly discuss extensions for handling azeotropic mixtures, reactive distillation columns and integration in process flowsheets. Numerical examples are presented to demonstrate that effective computational strategies are emerging that are based on disjunctive programming models that are coupled with thermodynamic initialization models and integrated through hierarchical decomposition techniques.

1. Introduction

The optimal synthesis of distillation continues to be a major problem in the design of chemical processes due to the high investment and operating costs involved in these systems. The recent trends in this area have been to address models of increasing complexity through the use of mathematical programming. The high degree of nonlinearity and the difficulty of solving the corresponding optimization models, however, have prevented methods with rigorous models from becoming tools that can be readily used by industry. For instance, a common problem that is experienced with rigorous models is when the trays or columns are “deleted”, as then the equations describing the MESH equations become singular, which in turn produces convergence failure.

In this paper we provide a general review of the area of optimal design and synthesis of distillation columns, emphasizing recent developments in our groups at Carnegie Mellon and INGAR, particularly the Ph.D. work of Mariana Barttfeld. As will be shown, the successful solution of the optimization of individual columns and complex column systems seems to

require appropriate representations for the design alternatives, disjunctive programming formulations that are coupled to decomposition methods, and initialization schemes that are based on thermodynamics. We first present a general review of optimal distillation design. This is followed by a brief review of MINLP and Generalized Disjunctive Programming (GDP). We then examine the optimal design problem of columns and contrast the relative advantages/disadvantages of MINLP and GDP models. We also discuss the impact of various column superstructure representations as well as the importance of suitable initialization schemes. We next present a general classification of superstructures and discuss briefly several alternatives, including a thermodynamically-based superstructure. We then discuss a decomposition strategy for solving the GDP model. Finally, we describe an example problem.

2. Background

The economic optimization of a distillation column involves the selection of the number of trays and feed location, as well as the operating conditions to minimize the total investment and operation cost. Discrete decisions are related to the calculation of the number of trays and feed and products locations and continuous decisions are related to the operation conditions and energy use involved in the separation. A major challenge that remains is to perform the optimization using tray-by-tray models that assume phase equilibrium.

There are two major formulations for the mathematical representation of problems involving discrete and continuous variables: Mixed-Integer Nonlinear Programming (MINLP) and General Disjunctive Programming (GDP) where the logic is represented through disjunctions and propositions (Grossmann, 2001). Both approaches have been employed in the literature to model distillation columns.

The most common form of MINLP problems is the special case in which the 0-1 variables are linear while the continuous variables are nonlinear:

$$\begin{aligned}
 \min Z &= c^T y + f(x) \\
 \text{s.t. } h(x) &= 0 \\
 By + g(x) &\leq 0 \\
 x \in X, y &\in \{0,1\}^m
 \end{aligned} \tag{MINLP}$$

Major methods for MINLP problems include first Branch and Bound (BB) (Gupta and Ravindran, 1985; Borchers and Mitchell, 1994; Stubbs and Mehrotra, 1999), which is a direct extension of the linear case, except that NLP subproblems are solved at each node. Generalized Benders Decomposition (GBD) (Benders, 1962; Geoffrion, 1972), and Outer-Approximation (OA) (Duran and Grossmann, 1986; Yuan, Zhang, Piboleau and Domenech, 1988; Fletcher and Leyffer, 1994; Ding-Mai and Sargent, 1992), are iterative methods that solve a sequence of alternate NLP subproblems with all the 0-1 variables fixed, and MILP master problems that predict lower bounds and new values for the 0-1 variables. The difference between the GBD

and OA methods lies in the definition of the MILP master problem; the OA method uses accumulated linearizations of the functions, while GBD uses accumulated Lagrangean functions parametric in the 0-1 variables. The LP/NLP based branch and bound by Quesada and Grossmann (1992) essentially integrates both subproblems within one tree search, while the Extended Cutting Plane Method (ECP) (Westerlund and Pettersson, 1995) does not solve the NLP subproblems, and relies exclusively on successive linearizations. All these methods assume convexity to guarantee convergence to the global optimum. Nonrigorous methods for handling nonconvexities include the equality relaxation algorithm by Kocis and Grossmann (1987) and the augmented penalty version of it (Viswanathan and Grossmann, 1990). A review on these methods and how they relate to each other can be found in Grossmann (2002).

MINLP problems can be solved for instance with the computer code DICOPT (Viswanathan and Grossmann, 1990), which is an implementation of the Outer Approximation/Equality Relaxation (OA/ER) algorithm (Kocis and Grossmann, 1987). The computational expense in solving these models depends largely on the problem structure. There is also the computational difficulty that each constraint must be solved even if the stage “disappears” from the column. It would be desirable to eliminate these constraints, not only to reduce the size of the NLP subproblems, but also to avoid singularities that are due to the linearization at zero flows.

MINLP formulations have been used for optimizing individual columns and superstructures using economic objective functions (Viswanathan and Grossmann, 1990; Viswanathan and Grossmann, 1993; Bauer and Stilchmair, 1998; Aguirre et al., 2001; Dunebier and Pantelides, 1999). Two basic representations arise from this formulation according to the way the discrete decisions related to the tray optimization are modeled. In one a binary variable with a value of “1” is assigned to each tray of the column denoting its existence, and with a value of “0” its absence (Viswanathan and Grossmann, 1990). In the other representations, binary variables are used for the discrete decisions related to the location of the reflux, reboil or both (Viswanathan and Grossmann, 1993; Bauer and Stilchmair, 1998; Aguirre et al., 2001). In both cases, flows of streams of non-existing trays are driven to zero which tends to cause singularities, and hence numerical difficulties for convergence.

In order to overcome difficulties in MINLP with “disappearing streams and units,” Raman and Grossmann (1994) proposed Generalized Disjunctive Programming (GDP), which in turn provides a modeling and solution framework for formulating problems with algebraic equations and symbolic logic equations. The GDP model consists of Boolean and continuous variables that are involved in an objective function, subject to three types of constraints: (a) global inequalities that are independent of discrete decisions; (b) disjunctions that are conditional constraints involving an OR operator; (c) pure logic constraints that involve only the Boolean variables. More specifically, the problem is given as follows:

$$\begin{aligned}
\min Z &= \sum_{k \in K} c_k + f(x) \\
s.t. \quad g(x) &\leq 0 \\
&\bigvee_{j \in I_k} \left[\begin{array}{l} y_{jk} \\ h_{jk}(x) \leq 0 \\ c_k = \gamma_{jk} \end{array} \right] \quad k \in K \\
&\Omega(y) = \text{True} \\
&x \in X, y_{jk} \in \{\text{True}, \text{False}\}
\end{aligned} \tag{GDP}$$

where x are continuous variables and y are the Boolean variables. The objective function involves the term $f(x)$ for the continuous variables (e.g. operating cost) and the charges c_k that depend on the discrete choices. The equalities/inequalities $g(x) \leq 0$ must hold regardless of the discrete conditions and $h_{jk}(x) \leq 0$ are conditional constraints that must be satisfied when the corresponding Boolean variable y_{jk} is true for the j th term of the k th disjunction. The set I_k represents the number of choices for each disjunction defined in the set K . Also, the fixed charge c_k is assigned the value γ_{jk} for that same variable. Finally, the constraints $\Omega(y)$ involve logic propositions in terms of Boolean variables.

For the nonlinear case of problem (GDP), Lee and Grossmann (2000) have developed reformulations and algorithms that rely on obtaining the algebraic description of the convex hull of the nonlinear convex inequalities. The reformulations lead to tight MINLP problems, while the algorithms generally involve branch and bound methods where branching is performed on disjunctions. For the case of process networks, Türkay and Grossmann (1996) proposed a logic-based Outer-Approximation algorithm. This algorithm consists of solving NLP subproblems in reduced space, in which constraints that do not apply in the disjunctions are disregarded, with which both the efficiency and robustness can be improved. In this method the MILP master problems correspond to the convex hull of the linearization of the nonlinear inequalities. Also, several NLP subproblems must be solved to initialize the master problem in order to cover all the terms in the disjunctions. Penalties can also be added to handle the effect of nonconvexities as in the method by Viswanathan and Grossmann (1990).

The logic-based Outer-Approximation algorithm has been successfully applied for solving GDP models of individual distillation columns and superstructures (Yeomans and Grossmann, 2000a; 2000b), as well as to reactive distillation columns (Jackson and Grossmann, 2001). Different approaches can be used with this formulation depending on which trays are defined as permanent in the configuration. It is this issue that has been analyzed in depth by Barttfeld et al. (2003).

A major difficulty that arises in the MINLP and GDP approaches is dealing with the nonlinearities that are involved in distillation models, which complicates the convergence of

solvers and often leads to infeasible solutions. Therefore, developing methods for the initialization and bounding of the variables involved in the problem is an essential part for the successful application of optimization formulations and algorithms for distillation columns.

Fletcher and Morton (2000) examined the infinite reflux case for generating good initial values for the NLP solution of general distillation columns. Buggemann and Marquardt (2001) have proposed a short cut method based on the Rectification Body Method (RBM) that provides qualitative insights for rigorous simulations. The method gives information on the minimum energy demand involved in a separation by a trial and error procedure. Given the products and feed compositions as well as the operating pressure, an estimate of the energy demand is determined to calculate the pinch points to construct the rectification bodies related to both column sections. The energy involved in the separation under minimum reflux is achieved when the bodies intersect in exactly one point. An automatic initialization scheme based on the successive solution of NLP and MINLP optimization problems was presented by Barttfeld and Aguirre, (2002). These authors developed rigorous and robust optimization models that approach reversible conditions in order to initialize and bound zeotropic distillation models. No external parameters have to be tuned in the model to achieve convergence.

3. Optimization of single columns

3.1 MINLP Models

The simplest type of distillation design problem is the one where there is a fixed number of trays, and the goal is to select the optimal feed tray location. Figure 1 shows that a superstructure that can be postulated is one where simply the feed is split into as many streams as there are trays, excluding condenser and reboiler. This is in essence the superstructure that was proposed by Sargent and Gaminibandara (1976). The model can easily be written as an MINLP model by considering all the mass and enthalpy balances, and phase equilibrium equations (MESH equations), in addition to the following mixed-integer constraints. Let z_i , $i \in LOC$, denote the *binary variable* associated with the selection of i as the feed tray; i.e., $z_i = 1$ if i is the feed tray. Let F_i , $i \in LOC$ denote the amount of feed entering tray i .

$$\begin{aligned} \sum_{i \in LOC} z_i &= 1 \\ \sum_{i \in LOC} F_i &= F \\ F_i - F z_i &\leq 0, \quad i \in LOC \end{aligned} \tag{1}$$

The last constraint in (1) expresses the fact that if tray $i \in LOC$ is selected as the feed tray, then, the amount of feed entering other candidate locations is zero. This follows from the fact $z_j = 0$, $j \neq i$, $i \in LOC$. In addition, there may be constraints on purity, recovery, or reflux ratio.

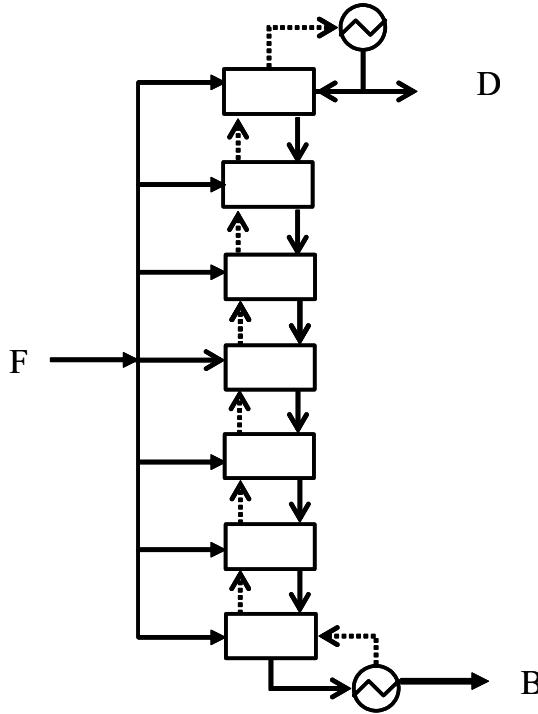


Figure 1: Superstructure for feed tray location.

The MINLP problem, then, is to minimize (or maximize) a given objective function (e.g. minimize energy cost). Note that in this model, the variables z_i are binary, while all other variables are continuous.

An interesting property of the MINLP for fixed number of trays is that computational experience has shown that this problem is solved almost always as a relaxed NLP. The physical explanation is that one can expect the optimal distribution to be one where the feed is all directed into a single tray where the tray composition matches closely the composition of the feed. Our computational experience has supported this observation many times (e.g. see Viswanathan and Grossmann, 1990, 1993; Barttfeld et al., 2003).

When the objective is to optimize not only the feed tray, but also the number of trays, the complexity of the model greatly increases. One possible configuration that was proposed by Viswanathan and Grossmann (1993) involving variable reflux location is depicted in Figure 2. The basic idea here is to consider a fixed feed tray with an upper bound of trays specified above and below the feed. The reflux is then returned to all trays above the feed, and the reboil returned to all trays below the feed. In essence this representation determines the “optimal

feed” of the reflux and reboils streams. In order to assign the actual number of trays 0-1 variables are assigned to the existence of each of the reflux and reboil returns. The problem then leads to an MINLP mode, which has as constraints the MESH equations, and mixed-integer constraints for the return of reflux and reboil streams. While in principle this model is suitable for optimizing the feedtray location and number of trays, it has the difficulty that trays not selected above the feed only handle vapor flow since the liquid flow is zero, rendering the phase equilibrium equations redundant. A similar situation arises with trays not selected below the feed. This means that the vapor liquid equilibrium (VLE) conditions have to be satisfied in non-existing trays where no mass transfer takes place. This feature clearly showed in the work by Viswanathan and Grossmann (1993) a marked increase in computation time versus the case of fixed number of trays.

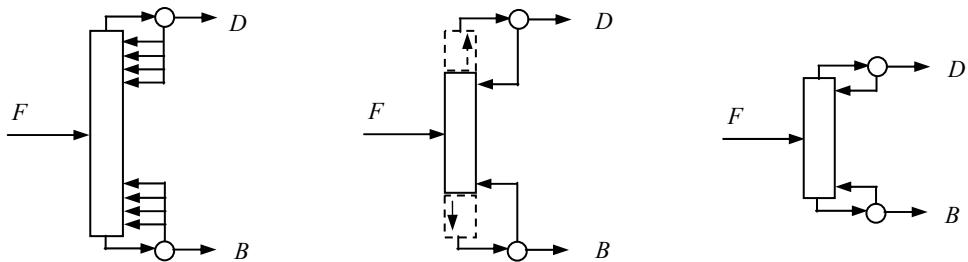


Figure 2: Variable reflux and reboil location with fixed heat exchangers.

3.2. Disjunctive Model for Single Column

Yeomans and Grossmann (2000a) have proposed a Generalized Disjunctive Programming (GDP) model that overcomes difficulties of the MINLP models by allowing the “by-pass” of those trays that are not selected. Figure 3 shows the column representation for this approach. Consider the conditional trays. For each existing tray the mass transfer task is accounted for and modeled with the MESH equations: the component mass balances, the tray energy balance, the equilibrium equations and the summation of liquid and vapor mole fractions to 1. For a non-existing or inactive tray the task considered is simply an input-output operation with no mass transfer, which gives rise to trivial mass and energy balance equations (inlet and outlet flows and enthalpies are same for the liquid flows and the vapor flows). Because the MESH equations include the solution for trivial mass and energy balances, the only difference between existing and non-existing trays is the application of the equilibrium equations. As for the permanent trays, all the equations for an existing tray apply.

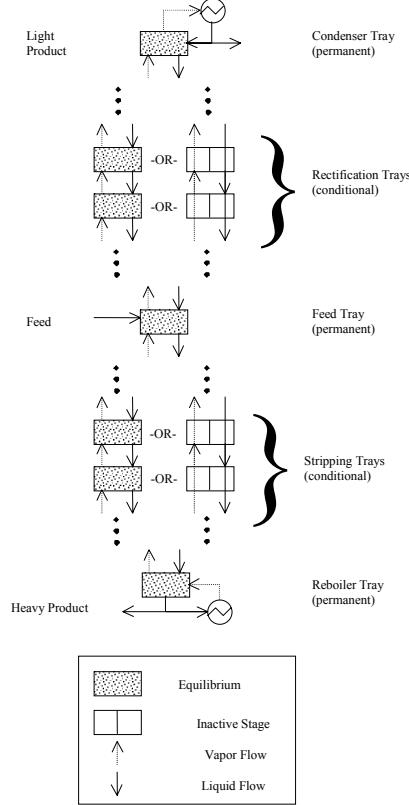


Figure 3: Structure of disjunctive model with permanent and conditional trays.

The general form of the GDP model is given by equation (GDP-C) where a disjunction is postulated for each conditional tray.

$$\begin{aligned}
 & \text{Min Cost} \\
 \text{s.t.} \\
 & \text{MESH equations for permanent trays} \\
 & \text{Mass / energy balances for conditional trays} \\
 & \left(\begin{array}{c} Y_n \\ \text{Equilibrium} \\ \text{equations} \\ \text{Conditional trays} \end{array} \right) \vee \left(\begin{array}{c} \neg Y_n \\ \text{Bypass} \\ \text{equations} \\ \text{Conditional trays} \end{array} \right) \quad (\text{GDP-C})
 \end{aligned}$$

The advantage of the disjunctive modeling approach is that the MESH equations of the non-existing trays do not have to be converged, and no flows in the column are required to take

values of zero, making the convergence of the optimization procedure more reliable. Also, by using Generalized Disjunctive Programming (GDP) as the modeling tool, the computational expense of solving the problem can be reduced.

3.3. Different Representations for MINLP and GDP Models

Barttfeld et al., (2003) have recently studied the impact of different representations and models that can be used for the optimization of a single distillation column. General models comprising different column configurations were presented for the MINLP and GDP formulations. Figure 4 shows two possible representations that are different from Fig. 2 to determine optimal feed tray and number of trays with the MINLP formulation. In part (a) of this picture, a condenser and a reboiler are placed in all candidate trays for exchanging energy. This means that a variable reflux (reboil) stream is considered by moving the condenser (reboiler). Otherwise, in the representation of variable reflux location (Figure 4 (b)), the condenser and reboiler are fixed equipments in both column extremes. The reflux (reboil) flow location is variable and not the condenser (reboiler) itself. These two alternatives are the same if one fixed equipment is considered at each column ends. However, when heat exchange variable locations are modeled as part of the tray optimization procedure as seen in Fig. 4 (c), some differences arise. In one case, the problem consists in finding the optimal location for the energy exchanged, while in the other the optimal location for a “secondary” feed stream (reflux) is considered. The variable heat exchange representation has an important advantage. The energy can be exchanged at intermediate trays temperatures, possibly leading to more energy efficient designs.

The results by Barttfeld et al., (2003) have shown that the most efficient MINLP representation involves variable reboiler and feed tray location (Fig. 4 (a)). In addition these authors also found that the most convenient formulation involves the use of total flows and compositions, and variable energy demand for the variable reboiler location representation. In a similar way as in the case of the MINLP models, Barttfeld et al., (2003) considered different representations for the GDP model, with fixed and variable feeds as shown in Figure 5. The computational results showed that the most effective structure is the one with fixed feed (Fig. 5 (a)), which was the original representation used by Yeomans and Grossmann (2000a).

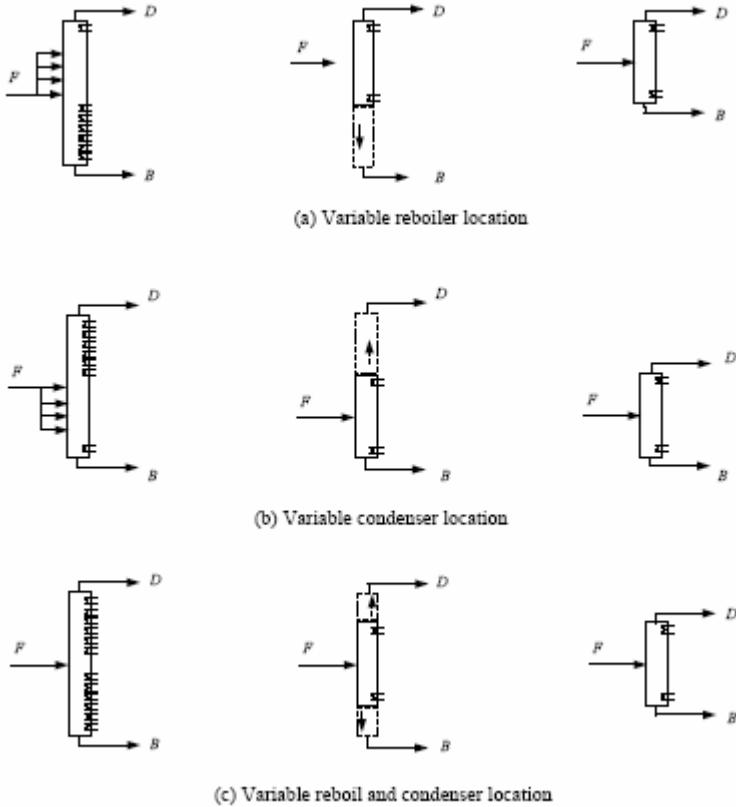


Figure 4: MINLP distillation column representations.

3.4 Initialization Procedures

Due to the complexity, nonlinearities and nonconvexities involved in both, the MINLP and GDP models, good initial values and bounds are essential in order to achieve convergence. Barttfeld et al., (2002) proposed a preprocessing phase to generate a good initial solution. The column topology in this phase corresponds to the one used for the economic optimization, except that the number of trays is fixed to the maximum specified. This means that the same upper bound on the number of trays has to be employed as well as the potential feed and product location. The initial design considered is the one that involves minimum reflux conditions as well as minimum entropy production. This reversible separation provides a feasible design and hence a good initial guess to the economic optimization.

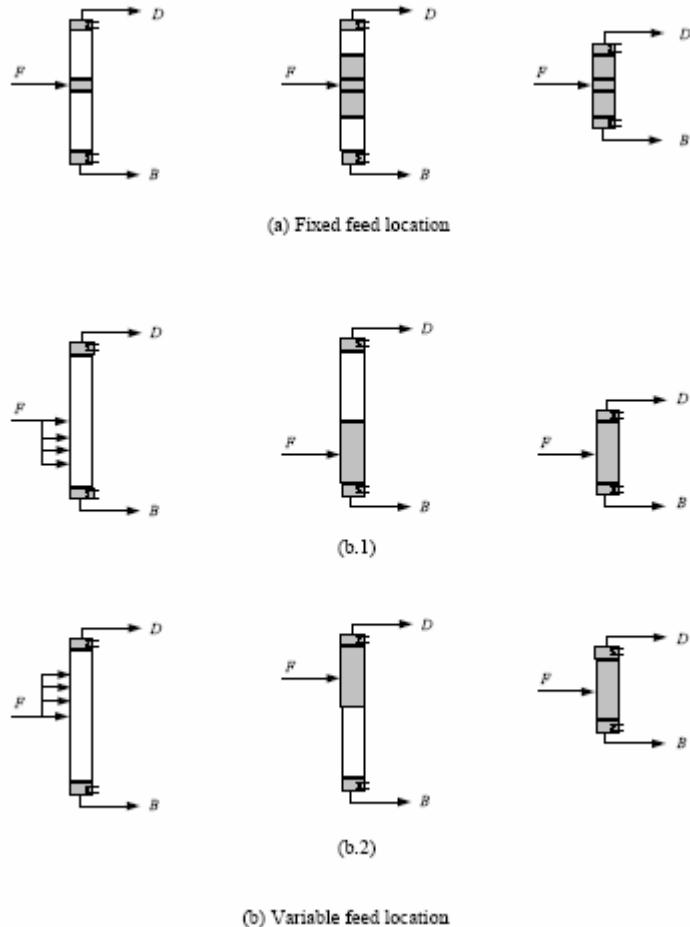


Figure 5: GDP distillation column representations

In the preprocessing phase, for the case of zeotropic columns, overall mass and energy balances are formulated as an NLP problem to compute the reversible products. This preliminary formulation is a well-behaved problem that provides initial values and bounds for the rigorous NLP tray-by-tray preprocessing formulation. Barttfeld et al., (2003) have shown that convergence is greatly enhanced including the preprocessing procedure. Also, these authors have described an initialization procedure for azeotropic columns.

It is also interesting to note that the MINLP formulation can be solved with a reduced number of binary variables. The reason is that the NLP relaxation yields a number of trays that is often very close to the integer optimal design. This relaxation also provides a good lower bound on the objective function value. Therefore, the solution of the relaxed problem can be employed to

reduce the domain of the variable tray location such that they contain few additional trays compared to the ones at the relaxation solution. In the case of the GDP model one cannot take advantage of the relaxation since only NLP subproblems with fixed number of trays are solved.

3.5 Numerical performance

Barttfeld et al., (2003) solved several example problems to evaluate the robustness and performance of the MINLP and GDP models for the optimal design of single columns. For the examples studied, the MINLP formulation with preprocessing and domain reduction yields designs involving lower total costs. In the azeotropic example, the distillate composition achieved in the economic solution crosses the distillation boundary. In all cases, the MINLP solution times were considerably longer than the ones of the GDP models. The robustness of the MINLP formulations was observed to depend very much on the solution scheme. If a good initial guess is generated with the preprocessing phase and the domain reduction for the binary variables is applied, an integer solution is often obtained in few iterations. However, the total solution time is long because the convergence of the NLP subproblems is usually very difficult to achieve. Also, the MILP subproblems include constraints, which were generated by linearizing the original constraints of the problem at zero flows.

On the other hand, the GDP formulations were found to be more robust and faster than the MINLP model. It was also observed that the GDP formulation is not as strongly dependent of the initial guess as the MINLP formulation. If a good initial solution guess is provided, the convergence of the initial NLP problems is guaranteed without tuning external parameters and also, better solutions can be found. It should be noted that the relaxed solution of the GDP formulation does not provide a useful distribution of trays as it was the case of the relaxed MINLP solution. Interestingly, despite the greater robustness of the GDP models, solutions with about 1% lower cost were found with the MINLP models when they converged. As an example in an equimolar mixture of butane, toluene and xylene, with minimum purity of 98% of butane at the distillate and a minimum recovery of 98%, and an upper bound of 60 for the number of trays, the results were as follows:

Table 1. Comparison for butane/toluene/xylene mixture

MINLP model	GDP Model
26 trays	19 trays
Feed tray: 13	Feed tray: 8
Cost: \$79,962/yr	Cost: \$80,720/yr
648 CPU sec	211 CPU sec

4. Classification Superstructures

In the application of mathematical programming techniques to design and synthesis of distillation systems it is necessary to postulate a superstructure of alternatives. This is true whether one uses a high level aggregated model, or a fairly detailed model. (1999a) have

characterized two major types of superstructure representations for process synthesis. The first is the State-Task Network (STN) which is motivated by the work in scheduling by Kondili *et al.*, (1993). The basic idea here is that the representation makes use of two types of nodes: states and tasks. The assignment of different pieces of equipment is usually assumed for each separation task. Figure 6 provides an example of an STN superstructure for the sharp separation of 4 components.

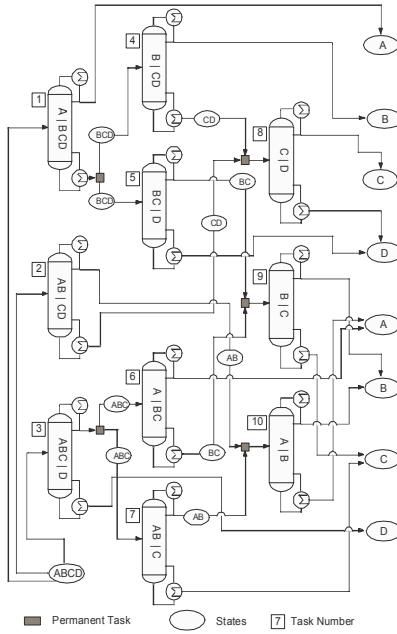


Figure 6: State Task Network superstructure representation for distillation sequence.

It is clear from that figure that using detailed tray-by-tray models in such a superstructure leads to a problem of large dimensionality.

The second representation is the State Equipment Network (SEN) which is motivated by work of Smith and Pantelides (1995), and where the basic idea is to work with two types of nodes: states and equipment. The tasks in this case are treated implicitly through the model. Figure 7 shows the example again for the 4 component system. It is clear that if rigorous tray-by-tray models are to be used SEN superstructures should lead to much more compact formulations. In fact Yeomans and Grossmann (1999a) have developed generic GDP models for each of the two different types of representations. These can then be used for solution with a GDP algorithm, or they can be used for reformulation as MILP or MINLP problems, depending on the complexity of the model.

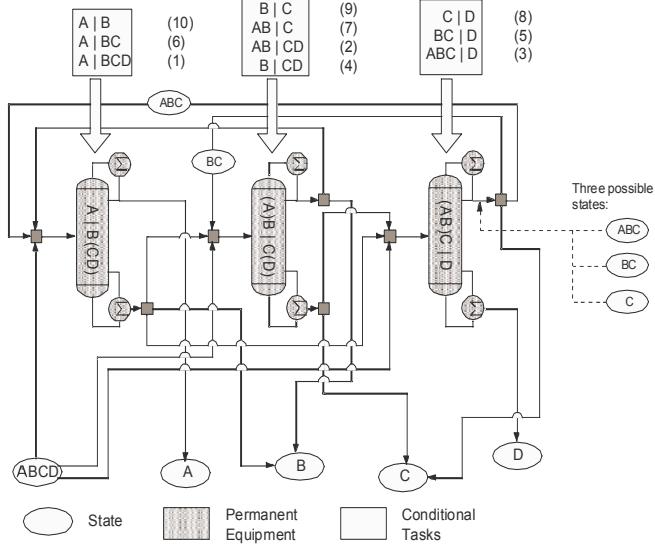


Figure 7: State Equipment Network superstructure representation for distillation sequences.

5. Superstructure Optimization of Complex Columns

5.1 The Sargent Gaminibandara Superstructure

In the previous section we presented superstructures for the case of sharp splits. The separation of more than two components by continuous distillation is often accomplished by simply arranging columns in such systems. However, even under the assumption of minimum reflux, past work has shown that complex column arrangements can yield significant savings in the operating costs. Most of the effort in the field of distillation synthesis has been applied to develop short-cut and simplified methods (Fidkowski and Krolikowski, 1986; Glinos and Malone, 1988; Triantafyllou and Smith, 1992; Annakou and Mizsey, 1996), mainly because of the convergence difficulties of rigorous formulations. As an example of recent work, Caballero and Grossmann (2003) have presented a systematic approach for generating all the thermodynamic equivalent structures for a given sequence. If our objective is to be able to synthesize complex columns possibly Petlyuk columns, columns with side strippers and side rectifiers, it is clear that more complex superstructures are needed compared to the ones in Figures 6 and 7.

The generation of complex column configurations has been principally developed by Sargent and Gaminibandara (1976), Agrawal (1996) and Fidkowski and Agrawal (1995, 1996). Other superstructures include for instance the one by Koehler et al. (1992) who consider thermodynamic aspects. However, the problem for systematically obtaining the optimal design out of superstructure was not addressed by these authors. Some recent work has applied mathematical programming tools to rigorously solve the distillation design problem. The superstructure most commonly used in the literature is based on the one proposed by Sargent

and Gaminibandara (1976) for ideal mixtures (see Figure 8) and later extended for azeotropic cases (Sargent, 1998). It is interesting to note that the superstructure in Figure 8 can be derived from the functional State-Task-Network shown in Figure 9, which in fact corresponds to a zeotropic mixture (see Sargent, 1998). A different superstructure that is not so commonly used is the one proposed by Bauer and Stichlmair (1998) that uses thermodynamic information in the representation itself. These authors applied this representation in the design of azeotropic sequences.

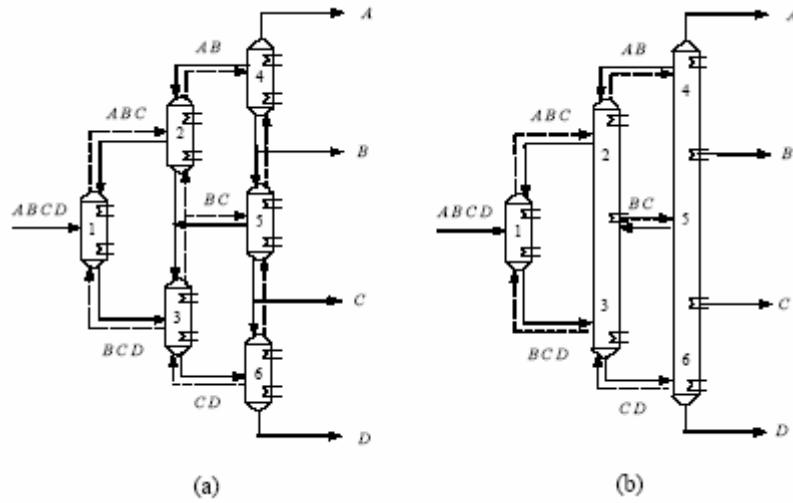


Figure 8: Sargent-Gaminibandara superstructure for complex columns.

As for other superstructures, Dunnebier and Pantelides (1999) have considered the optimal design of thermally coupled distillation columns and dividing wall columns for ideal mixtures using detailed distillation models and mathematical optimization. Yeomans and Grossmann (1999b) presented the rigorous synthesis of heat integrated sequences applying disjunctive programming techniques to formulate the problem. These authors have also developed a modeling disjunctive programming procedure for the optimal design of single ideal and nonideal single distillation units and separation sequences (Yeomans and Grossmann, 2000a) as well as complex sequences (Yeomans and Grossmann, 2000b). In these two methods, the major challenge is that the optimal design of distillation columns configurations involves the solution of large, highly nonlinear nonconvex optimization problems.

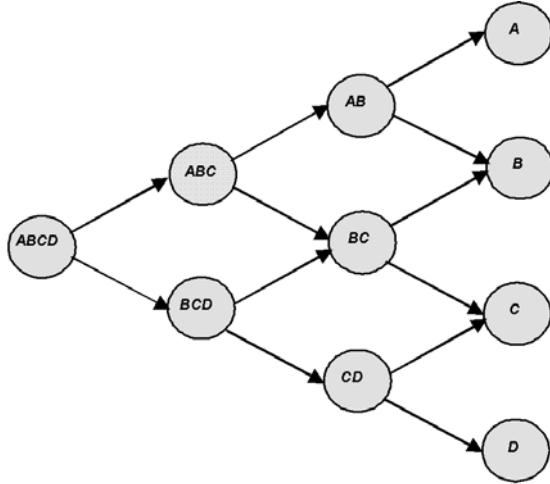


Figure 9: Sargent-Gaminibandara STN representation for a four component zeotropic mixture.

5.2. Reversible Distillation Sequence Model Superstructure

The superstructure considered by Barttfeld et al. (2004) is based on the Reversible Distillation Sequence Model (RDSM) proposed by Fonyó (1974), which allows the introduction of thermodynamic aspects in the design (for details of the RDSM theory see Koehler et al., 1992; Barttfeld and Aguirre, 2003). The motivation in using such a superstructure is that it is tied closely to a robust initialization scheme similar to the one that was described for single columns. The RDSM superstructure can be automatically generated for zeotropic as well as for azeotropic mixtures. In the latter case, a composition diagram of the mixture is assumed to be available. The RDSM-based superstructure can also be generated using the STN representation of Sargent (1998). For the RDSM-based superstructure the states are defined in the same way as in the Sargent-Gaminibandara superstructure, but the tasks in this representation are different as seen in Figure 10 for a four component zeotropic mixture. In order to approximate reversibility conditions, only products having the same composition can be represented in one state. As an example, in the RDSM STN, two different states are defined for the mixture, BC , as shown in the representation of Figure 10. These states come from states, ABC , or BCD , and do not necessarily have the same composition. As a consequence of this fact, for separating a NC -zeotropic mixture, the RDSM-based superstructure has the same number of levels as the Sargent-Gaminibandara representation, but a larger number of columns, given by 2^{NC-1} .

The representation of the equipment for the RDSM-based superstructure for a four component zeotropic mixture is shown in Figure 11a. Note that in this representation, columns 2 and 3 (second level) cannot be coupled. However, other representations are possible for the RDSM superstructure (see Koehler et al., 1992).

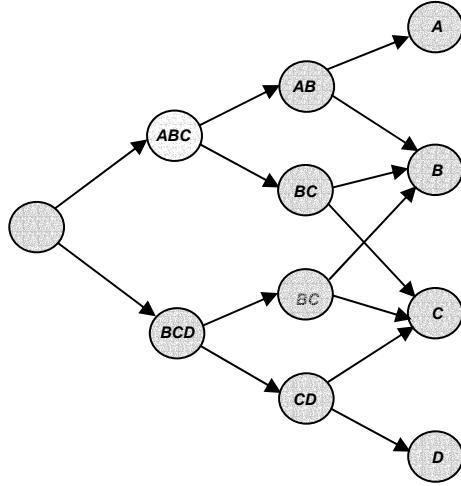


Figure 10: STN representation of the RDSM-based superstructure.

In the RDSM representation considered, column coupling is only possible in those columns that yield pure products, that is, in the last level of the superstructure. Note that columns 4 and 5 are integrated to produce product, B as well as columns 6 and 7 to produce pure product C (see Figure 11b). Therefore, in the proposed superstructure it is not possible to represent in a level all the columns by one single unit as in the representation of Sargent and Gaminibandara of Figure 8. Only 2^{NC-3} columns integrations (single columns) can be found in the last level of the superstructure. Each column in the superstructure of Figure 11 is represented by an adiabatic unit, and with one condenser and one reboiler. The trays in each unit can be classified as intermediate or permanent trays (see Figure 3). This representation is the one that has been found to be the most effective to model distillation columns with GDP formulations (Barttfeld et al., 2003). Those trays that can disappear in the superstructure optimization are the intermediate trays. Note that the column sections contain intermediate trays and each section is located between two permanent trays. An upper bound on the number of trays is assigned to each section of a column. The columns in the superstructure are interconnected by feeds and products streams. The columns where multicomponent separations take place (columns 1, 2 and 3, Figure 11 (a)), are coupled by the feeds and products streams. Each column can be fed by primary and secondary feeds. Compared to the Sargent and Gaminibandara superstructure, the RDSM representation excludes certain configurations that involve mixing of streams as would be the case of a Petluk column. However, if desired additional streams can be added to the RDSM superstructure in order to account for the same alternatives as in the Sargent and Gaminibandara superstructure.

It should also be noted that the RDSM superstructure can also be extended for azeotropic distillation. Due to the existence of distillation boundaries, the order of the relative volatility of

the components cannot be predefined. Therefore, a composition diagram showing the distillation boundaries is needed to define the feasible states that can be achieved from a given feed (see Barttfeld et al., 2004).

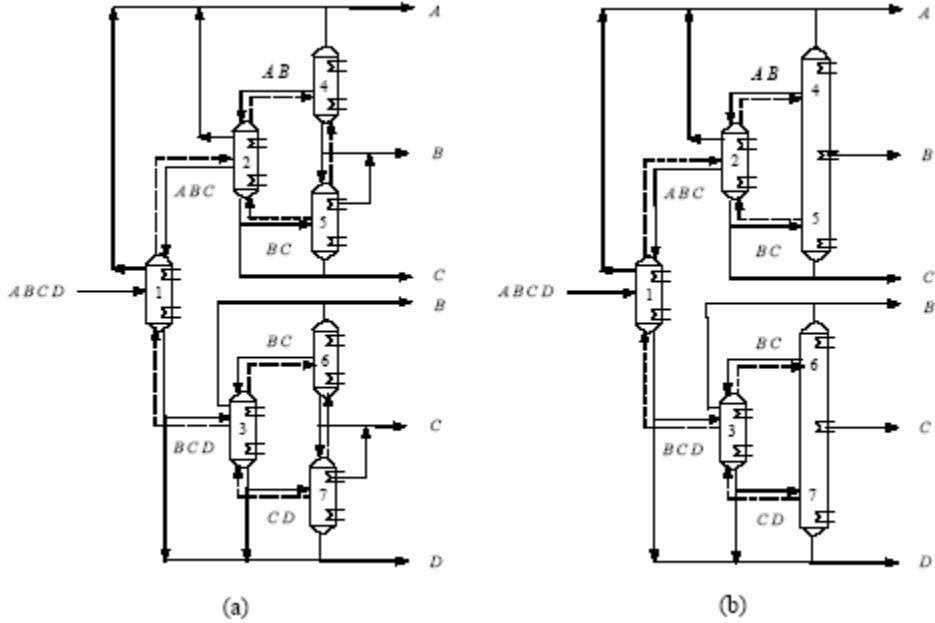


Figure 11: RDMS-based superstructure for a four component mixture.

5.3 Decomposition Strategy

Tray-by-tray distillation synthesis models are very difficult to optimize due to the highly nonlinear and nonconvex equations that are involved, as well as to the large size of the corresponding formulations. Furthermore, formulating and solving a single optimization problem to simultaneously establish the existence of columns as well as the feed tray location generally leads to a very difficult problem that often fails to converge. Convergence problems are often found when solving complex MINLP models (Dunnebier and Pantelides, 1999; Bauer and Stichlmair, 1998). Also, although the disjunctive formulation increases the robustness, it is still quite difficult to solve these problems as was reported by Yeomans and Grossmann (2000a, b). Barttfeld et al., (2004) have developed a computational strategy that exploits the nature of the decisions involved in the GDP model in order to yield robust and computationally effective models.

Barttfeld et al., (2004) formulated the synthesis problem as a GDP problem that does not have to be solved simultaneously and is amenable to decomposition. Specifically, the GDP of the RDSM superstructure in Fig. 11 can be formulated in the following general form:

$$\begin{aligned}
 & \text{Min Cost} \\
 \text{s.t.} \\
 & \text{MESH equations for permanent trays} \\
 & \text{Mass / energy balances for conditional trays} \\
 & \left(\begin{array}{c} Y_s \\ \text{Equilibrium} \\ \text{equations} \\ \text{Conditional trays} \\ st_n = 1 \end{array} \right) \vee \left(\begin{array}{c} n_s = st_n \\ -Y_n \\ \text{Bypass} \\ \text{equations} \\ \text{Conditional trays} \\ st_n = 0 \end{array} \right) \vee \left(\begin{array}{c} \neg Y_s \\ n_s = 0 \end{array} \right) \quad (\text{GDP-S})
 \end{aligned}$$

Note that model (GDP-S) involves embedded disjunctions. At the outer level the Boolean variables Y_s determine the selection of the sections in the columns (rectifying or stripping), while at the inner level the Boolean variables Y_n determine the existence or non-existence of the trays that are postulated in each section.

Based on the embedded disjunctions, Barttfeld et al., (2004) proposed an iterative decomposition strategy that exploits two major levels of decisions in the problem (see Fig. 12). In the first level, a configuration is derived by making the decision related to the selection of column sections (i.e. with the Boolean variables Y_s). In this level each section is assigned a maximum number of trays in order to produce a bounding solution. In the second level, the feed tray location and the number of trays of the selected sections are optimized (i.e. with the Boolean variables Y_n). The algorithm solves the disjunctive programming model by iteratively solving an MILP for selecting the sections, an MILP for selecting the trays of that configuration and an NLP subproblem for optimizing the particular design. Integer cuts are only added to the MILP for trays and not the one for the sections in order to ensure proper optimization of the number of trays. Similarly as in the single column case, a thermodynamic based NLP is solved for the initialization of this decomposition strategy.

5.4. Numerical experience

Numerical examples were solved by Barttfeld et al., (2004) to test the performance of the formulations. Two zeotropic examples were solved and nontrivial configurations were found, which include column coupling. In the azeotropic example, the influence of the product purity specification was analyzed with respect to the azeotrope recycle. Also, the influence of including intercondensers in the first column was analyzed. In all the examples, the solutions were obtained with the proposed method, are non trivial and require reasonable solution times.

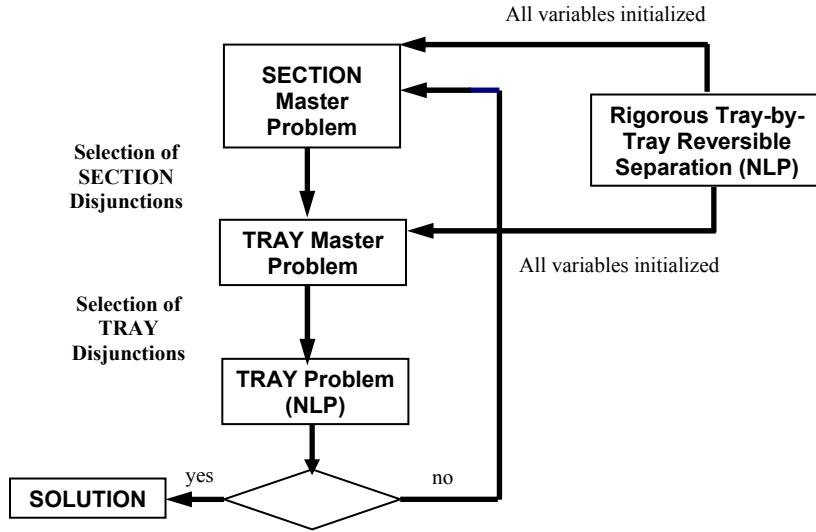


Figure 12; Decomposition strategy.

A specific example is presented in Figure 13, where it can be seen that even for an ideal system such as n-pentane, n-hexane and n-heptane a significantly improved design in the form of a complex column can be obtained (\$140,088/yr) versus a standard direct sequence (\$145,040/yr). Figure 14 shows the liquid profiles of the optimal design, and Table 2 shows the computational results.

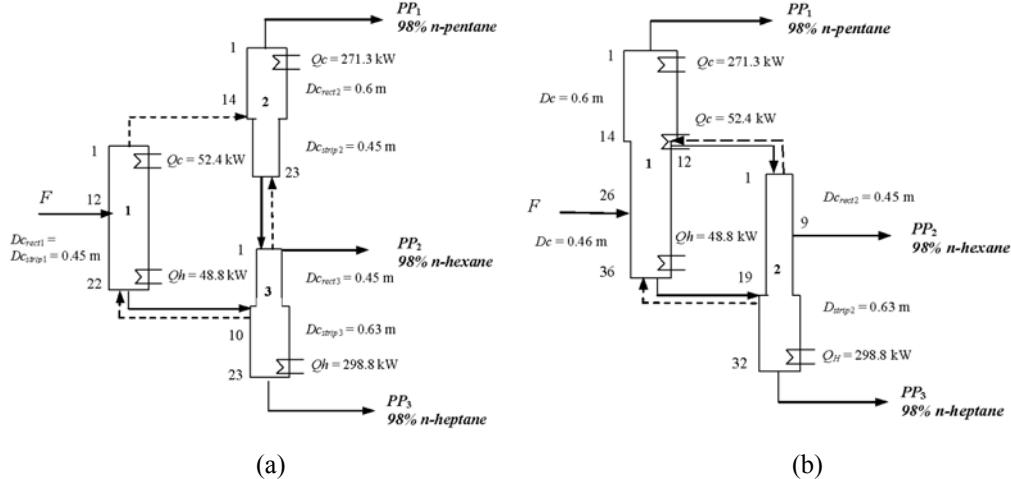


Figure 13: pentane/hexane/heptane example: (a) Superstructure with selected sections. (b) Optimal configuration in terms of two columns.

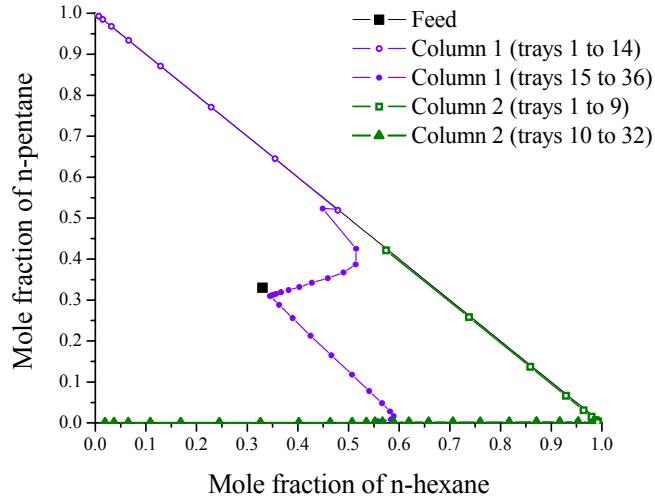


Figure 14: Liquid composition profiles of the optimal configuration.

Table 2: Computational results for pentane/hexane/heptane mixture.

Preprocessing Phase: NLP Tray-by-Tray Models	
Continuous Variables	3297
Constraints	3225
CPU time [min.]	2.20
Model Description	
Continuous Variables	3301
Discrete Variables	96
Constraints	3230
Nonlinear nonzero elements	3244
Number of iterations	5
NLP CPU time [min.]	6.97
MILP CPU time [min.]	2.29
CPU time [min.]	9.25
Objective Value [\$/year]	140,880
Total CPU time [min.]	11.46

6. Concluding remarks and future work

We hope that this paper has shown that despite its great difficulty, there has been significant progress in the optimal synthesis of complex column configurations using tray-by-tray models. As has been seen the combination of novel representations for individual columns and superstructures, combined with disjunctive programming and robust initialization schemes has made it possible to solve with reasonable computational efficiency these problems.

While the results reported in this paper have shown that there has been significant progress in the optimal design of complex distillation columns, it is clear that there is still scope for further progress in this area. For instance, while the approach proposed by Barttfeld et al., (2004) has been applied to azeotropic mixture (e.g. methanol, ethanol and water mixture), the extension for generating the superstructure to azeotropic systems of more than three components remains an open question. Also, while an extension of the GDP model for the case of reactive distillation columns has been proposed by Jackson and Grossmann (2001), the integration of such a model that is as part of a system of complex columns has not been addressed. Similarly, the heat integration or incorporation of rigorous distillation synthesis models that are part of a flowsheet superstructure has not been accomplished. At this point this has only been performed with short cut models (e.g. see Yeomans and Grossmann, 1999b). Finally, a major challenge that remains is the rigorous global optimization. The only work reported in this regard is the one by Smith (1996).

Acknowledgments

Pío Aguirre and Mariana Barttfeld would like to thank for the financial support from CONICET (Consejo Nacional de Investigaciones Científicas y Técnicas), ANPCYT (Agencia Nacional de Promoción Científica y Técnica), UNL (Universidad Nacional del Litoral) from Argentina and the Center for Advanced Process Decision-making at Carnegie Mellon. Ignacio Grossmann would like to thank the Ministerio de Educación, Cultura y Deporte in Spain for their financial support.

References

- Agrawal, R., 1996, Synthesis of Distillation Column Configurations for Multicomponent Separations. 1996. Ind. Eng. Chem. Res. 35, 1059.
- Aguirre P. A., G. Corsano and M. Barttfeld, 2001, Un Método Híbrido para la Síntesis Optima de Procesos de Destilación Multicomponente. CIT (Información Tecnológica) 12 (1), 119.
- Annakou O. and P. Mizsey P., 1996, Rigorous Comparative Study of Energy-Integrated Distillation Schemes. Ind. Eng. Chem. Res. 35, 1877.
- Barttfeld M. and P. A. Aguirre, 2002, Optimal Synthesis of Multicomponent Zeotropic Distillation Processes. Part I. Pre-processing Phase and Rigorous Optimization for a Single Unit, Ind. Chem. Eng. Res., 41, 5298.

- Barttfeld M. and P. A. Aguirre, 2003, Optimal Synthesis of Multicomponent Zeotropic Distillation Processes. Part II. Pre-processing Phase and Rigorous Optimization of Efficient Sequences, *Ind. Chem. Eng. Res.*, 42 (14), 3441.
- Barttfeld M., P. A. Aguirre and I. E. Grossmann, 2003, Alternative Representations and Formulations for the Economic Optimization of Multicomponent Distillation Columns, *Computers and Chemical Engineering*, 27, 363.
- Barttfeld, M., P. A. Aguirre and I. E. Grossmann, 2004, A Decomposition Method for Synthesizing Complex Column Configurations Using Tray-by-Tray GDP Models, to appear in *Computers and Chemical Engineering*.
- Bauer M. H. and J. Stilchmair, 1998, Design and Economic Optimization of Azeotropic Distillation Processes Using Mixed Integer Nonlinear Programming, *Computers and Chemical Engineering*, 22, 1271.
- Benders J. F., 1962, Partitioning procedures for solving mixed-variables programming problems, *Numeri. Math.*, 4, 238.
- Borchers, B. and J. E. Mitchell, 1994, An Improved Branch and Bound Algorithm for Mixed Integer Nonlinear Programming, *Computers and Operations Research*, 21, 359).
- Bruggemann S. and W. Marquardt, Shortcut Methods for Nonideal Multicomponent Distillation: Providing Initials and Bounds for Rigorous Optimization, Internal report, May 2001.
- Caballero J. A. and I. E. Grossmann, 2003, Thermodynamically Equivalent Configurations for Thermally Coupled Distillation Columns, *AIChE Journal*, 49, 2864.
- Ding-Mei and R.W. H. Sargent, 1992, A Combined SQP and Branch and Bound Algorithm for MINLP Optimization", Internal Report, Centre for Process Systems Engineering, London.
- Dunnebier G. and C. C. Pantelides, 1999, Optimal Design of Thermally Coupled Distillation Columns, *Ind. Eng. Chem. Res.*, 38, 162.
- Duran M. A. and I. E. Grossmann, 1986, An Outer-Approximation Algorithm for a Class of Mixed-integer Nonlinear Programs, *Math Programming* 36, 307.
- Fidkowski Z. and L. Krolkowski, 1986, Thermally Coupled System of Distillation Columns: Optimization Procedure, *AICHE J.*, 32, 537.
- Fidkowski Z. and R. Agrawal, 1995, Utilization of Waste Heat Steam in Distillation, *Ind. Eng. Chem. Res.*, 34, 1287.
- Fidkowski Z. and R. Agrawal, 1996, On the Use of Intermediate Condensers in the Rectifying Section and Reboilers in the Stripping Section, *Ind. Eng. Chem. Res.*, 35, 2801.
- Fletcher, R. and S. Leyffer, 1994, Solving Mixed Integer Nonlinear Programs by Outer Approximation, *Math Programming*, 66, 327.
- Fletcher R. and W. Morton, 2000, Initializing Distillation Column Models, *Computers. Chemical Engineering*, 23, 1811.
- Fonyó Z., 1974, Thermodynamic Analysis of Rectification I. Reversible Model of Rectification, *Int. Chem. Eng.*, 14 (1), 18.
- Geoffrion A. M., 1972, Generalized Benders Decomposition, *Journal of Optimization Theory and Applications*, 10 (4), 237.

- Glinos K. and F. Malone, 1988, Optimality Regions for Complex Column Alternatives in Distillation Systems, *Chem. Eng. Res. Des.*, 66, 229.
- Grossmann I. E., 2002, Review of Nonlinear Mixed-Integer and Disjunctive Programming Techniques, *Optimization and Engineering*, 3, 227.
- Gupta O. K. and V. Ravindran, 1985, Branch and Bound Experiments in Convex Nonlinear Integer Programming, *Management Science*, 31(12), 1533.
- Jackson J. and I. E. Grossmann, 2001, A Disjunctive Programming Approach for the Optimal Design of Reactive Distillation Columns, *Computers and Chemical Engineering*, 25, 1661.
- Koehler J., P. A. Aguirre and E. Blass, 1992, Evolutionary Thermodynamic Synthesis of Zeotropic Distillation sequences, *Gas Separation and Purification*, 6 (3), 153.
- Kocis G. R. and I. E. Grossmann, 1987, Relaxation Strategy for the Structural Optimization of Process Flowsheets, *Ind. Eng. Chem. Res.*, 26, 1869.
- Kondili E., C. C. Pantelides and R. W. H. Sargent, 1993, A General Algorithm for Short-Term Scheduling of Batch Operations - I: MILP Formulation, *Computers and Chemical Engineering*, 17, 211.
- Lee S. and I. E. Grossmann, 2000, New Algorithms for Nonlinear Generalized Disjunctive Programming, *Computers and Chemical Engineering*, 24, 2125.
- Raman R. and I. E. Grossmann, 1994, Modelling and Computational Techniques for Logic Based Integer Programming, *Computers and Chemical Engineering*, 18, 563.
- Sargent R. W. H. and K. Gaminibandara, 1976, Optimal Design of Plate Distillation Columns, in *Optimization in Action*; Dixon, L. C. W., Ed; Academic Press: New York.
- Sargent R. W. H. A, 1998, Functional Approach to Process Synthesis and its Application to Distillation Systems, *Computers and Chemical Engineering*, 22, 31.
- Smith E. M. B. and C. C. Pantelides, 1995, Design of Reaction/Separation Networks using Detailed Models, *Supplement of Computer and Chemical Engineering*, 19, S83.
- Smith E. M., 1996, On the Optimal Design of Continuos Processes, PhD. Dissertation Under supervision of C. Pantelides, Imperial College of Science, Technology and Medicine, London U.K.
- Stubbs R. and S. Mehrotra, 1999, A Branch-and-Cut Method for 0-1 Mixed Convex Programming, *Mathematical Programming*, 86(3), 515.
- Türkay M. and I. E. Grossmann, 1996, Logic-Based MINLP Algorithms for the Optimal Synthesis of Process Networks, *Computer and Chemical Engineering*, 20 (8), 959.
- Viswanathan J. and I. E. Grossmann, 1990, A Combined Penalty Function and Outer-Approximation Method for MINLP Optimization, *Computer and Chemical Engineering*, 14 (7), 769.
- Viswanathan J. and I. E. Grossmann, 1993, An Alternative MINLP Model for Finding the Number of Trays Required for a Specified Separation Objective, *Computer and Chemical Engineering*, 17 (9), 949.
- Westerlund T. and F. Pettersson, 1995, A Cutting Plane Method for Solving Convex MINLP Problems, *Computers and Chemical Engineering*, 19, S131.

- Yeomans H. and I. E. Grossmann, 1999a, A Systematic Modeling Framework for Superstructure Optimization in Process Synthesis, *Computers and Chemical Engineering*, 23, 709.
- Yeomans H. and I. E. Grossmann, 1999b, Nonlinear Disjunctive Programming Models for the Synthesis of Heat Integrated Distillation Sequences, *Computers and Chemical Engineering*, 23, 1135.
- Yeomans H. and I. E. Grossmann, 2000a, Disjunctive Programming Models for the Optimal Design of Distillation Columns and Separation Sequences, *Ind. Eng. Chem. Res.*, 39 (6), 1637.
- Yeomans H. and I. E. Grossmann, 2000b, Optimal Design of Complex Distillation Columns Using Rigorous Tray-by-Tray Disjunctive Programming Models, *Ind. Eng. Chem. Res.*, 39 (11), 4326.
- Yuan X., S. Zhang, L. Piboleau and S. Domenech, 1988, Une Methode d'optimisation Nonlineare en Variables Mixtes pour la Conception de Procedes, *RAIRO*, 22, 331.