Title: Logic hybrid simulation-optimization algorithm for distillation design.

Author: J.A. Caballero

PII: S0098-1354(14)00097-0
DOI: http://dx.doi.org/doi:10.1016/j.compchemeng.2014.03.016
Reference: CACE 4929

To appear in: *Computers and Chemical Engineering*

Received date: 15-1-2014
Revised date: 18-3-2014
Accepted date: 20-3-2014

Please cite this article as: Caballero, J. A.,Logic hybrid simulation-optimization algorithm for distillation design., *Computers and Chemical Engineering* (2014), http://dx.doi.org/10.1016/j.compchemeng.2014.03.016

Logic hybrid simulation-optimization algorithm for distillation design.

J. A. Caballero

Department of Chemical Engineering, University of Alicante. Apartado de correos 99, 03080, Alicante Spain. email: caballer@ua.es.

Highlights

- A novel Generalized Disjunctive Model is presented for rigorous distillation design.
- A logic based algorithm without MINLP reformulation is used for solving the problem
- The model is solved using a hybrid simulation-optimization approach.
- We take advantage of the best of algebraic models and process simulators.

Abstract

In this paper, we propose a novel algorithm for the rigorous design of distillation columns that integrates a process simulator in a generalized disjunctive programming formulation. The optimal distillation column, or column sequence, is obtained by selecting, for each column section, among a set of column sections with different number of theoretical trays. The selection of thermodynamic models, properties estimation etc., are all in the simulation environment. All the numerical issues related to the convergence of distillation columns (or column sections) are also maintained in the simulation environment. The model is formulated as a Generalized Disjunctive Programming (GDP) problem and solved using the logic based outer approximation algorithm without MINLP reformulation. Some examples involving from a single column to thermally coupled sequence or extractive distillation shows the performance of the new algorithm.

Keywords: Distillation; Generalized Disjunctive Programming; Simulation; Optimization.

1. Introduction

The general separation problem was defined more than 40 years ago by Rudd & Watson (1968) as the transformation of several source mixtures into several product mixtures. More than forty years later we can say that this general problem has not been completely solved. We will focus, in this work, in the more restricted problem of separating a single source mixture into several products using only distillation columns.

Distillation is likely the most important separation and purification operation in chemical process industries. Typically more than half of the process heat distributed to a plant is dedicated to supply heat in the reboilers of distillation columns (Kunesh et al., 1995). However, the energy is provided to the bottom of the column and approximately the same amount of energy removed in the top, although at lower temperature, which yields an inefficient process, but still one of the most effective for homogeneous mixtures separations. To get an idea of the importance of distillation, Humphrey (1995) estimated that distillation handles more than 90% of all the separations and purifications. Soave & Feliu (2002), using data by Mix et al (1978) estimated that distillation accounts about 3% of the total United States energy consumption. This is equivalent to $2.87 \cdot 10^{18}$ J per year (91 GW or 54 million tons of crude oil). The capital investment for these distillation systems was estimated to be around 8 billion US$.

The optimization of distillation columns involves the selection of the number of trays, the feed location and the operating conditions to minimize a performance function, usually the total annualized cost that involves investment and operating costs. Discrete decisions are related to the calculation of the number of trays and feed and products location, and continuous decisions are related to the operation conditions. Due to the discrete-continuous nature of the problem and to the complex equations involved, it is common use shortcut or aggregated models together with some rules of thumb that under some assumptions have proved to produce good results, at least in the first stages of design where a rigorous design is neither necessary nor convenient due to the large computational effort needed. Some of the most successful shortcut methods are:

Fenske – Underwood- Gilliland (FUG). (Fenske, 1932; Gilliland, 1940; Underwood, 1948). The FUG method assumes a constant molar overflow and constant relative volatilities in all the trays of the distillation column. This method considers two extreme ideal situations. a) The distillation column operates at total reflux (no feed is entering or exiting from the column), which allows calculating the minimum number of trays for a given separation of two key components, and b) when the column operates at pinch conditions, (infinite number of trays), which allows calculating the minimum reflux. The optimal situation is in some point in between these two extreme cases. *Group methods* (GM) (Edmister, 1943; Kamath et al., 2010; Kremser, 1930). GMs use approximate calculations to relate the outlet stream properties to the inlet stream specifications and number of equilibrium trays. They provide only an overall treatment of the stages in the cascade without considering detailed changes in the temperature and composition of individual stages. However, they are much easier to solve because they involve fewer variables and constraints. *Aggregated models* (AG) (Bagajewicz & Manousiouthakis, 1992; J. A.

Caballero & Grossmann, 1999). AG models are similar to group methods, they are based on mass balances and equilibrium feasibility, expressed in terms of flows, inlet concentrations, and recoveries. The *Boundary Value Method* (BVM) (Barbosa & Doherty, 1988; Fidkowski et al., 1993; Fidkowski et al., 1991; Julka & Doherty, 1990; Levy & Doherty, 1986; Levy et al., 1985). BVM can be used to determine the minimum reflux ratio and feasible design parameters for a column separating a ternary mixture. It allows to obtain the number of trays, composition profiles etc. The *Rectification Body Method* (RBM) (Bausa et al., 1998; von Watzdorf et al., 1999). RBM is used for the determination of minimum energy requirements for a specified separation. The method approximates rectification bodies by straight lines. The intersection of the rectification bodies of two sections indicates its feasibility. *Driving Force Method* (DFM) (Gani & Bek-Pedersen, 2000). The DFM is a graphical method. Its authors proved that the minimum energy requirements corresponds to a maximum in the driving force. The *Shortest Stripping Line* (SSL) (Angelo Lucia et al., 2008; A. Lucia & McCallum, 2010; Angelo Lucia & Taylor, 2006) Authors showed that the longest residue curve is related with the highest energy consumption for a given separation. Then the shortest curve should produce the minimum energy required for the same separation.

Some of the previous methods have been automated, although not all of them can be directly used with an optimization algorithm. In any case, they are valuable tools for obtaining precise initial values or reliable bounds for the rigorous optimization of distillation columns.

2. Overview of rigorous tray by tray optimization models.

As commented in the introduction section, the economic optimization of a distillation column involves continuous decisions, related to the operational conditions and energy involved in the separation, and discrete decisions related to the total number of trays, and the tray positions of each feed and product streams. A major challenge is to perform the optimization using tray by tray models that assume phase equilibrium.

The first approach to solve the above commented problem was due to Sargent & Gaminibandara (1976). In this case, the authors assumed a fixed number of trays, and the goal was to select the optimal feed location. To that end, the feed is split into as many streams as trays has the column (condenser and reboiler are excluded). Figure 1 shows the superstructure. The model can be written as a Mixed Integer Nonlinear Programming (MINLP) problem by considering the MESH equations (Mass balances, Equilibrium equations, molar fraction Summation equals one in all phases, and Enthalpy balances). However, computational experience shows that this problem is usually solved as a relaxed NLP.

The first model that considers both, the feed tray position optimization and the total number of trays was due to Viswanathan & Grossmann (1993). The authors used a superstructure that involves a variable reflux location as shown in Figure 2. The superstructure considers a fixed feed tray and a column formed by a large enough number of trays above and below the feed. The reflux (reboil) is returned to all the trays above (below) the feed. The model takes the form

of a MINLP and relies also on MESH equations. A major difficulty with this model is related to the non-existing trays. In these trays, there is a zero liquid flow (rectifying section) or a zero vapor flow (stripping section), which can produce numerical problems due to the convergence of equilibrium equations with a zero value in the flow of one of the phases..

To avoid the numerical problems in MINLP models Yeomans and Grossmann (Yeomans & Grossmann, 2000a, 2000b) proposed a Generalized Disjunctive Programming model by allowing the bypass of those trays that are not selected. Figure 3 shows the column representation for this approach. For each existing tray the mass transfer task is accounted for and modeled with the MESH equations. For a non-existing or inactive tray the task considered is simply an input-output operation with no mass transfer. 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. 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. Barttfeld et al., (2003) considered different representations for the GDP model. Numerical results studies for separation of ternary mixtures in a single column suggests that the GDP formulation requires less solution time but is more sensitive toward local optima than MINLP formulations. Even though, GDP seems to be more reliable than MINLP models both require good initial values and bounds to converge. Barttfeld & Aguirre (2002, 2003), propose to use a reversible distillation model that involves the minimum reflux conditions as well as minimum entropy production to provide a feasible initial design, and good initial values for the rigorous optimization. Their method is mainly limited by the drawbacks of this so-called “preferred separation”, because, for azeotropic mixtures, usually nonsharp splits are generated. The extension to the most common sharp split is not trivial. Kossack et al (2006) proposed to use the Rectification Body Method (RBM) that can be used in all the cases. However, the initialization procedure is rather complex.

Another option was proposed by Harwardt & Marquardt (2012): They start calculating the minimum energy demand and the concentration profile estimated based on pinch points. Based on these results a simplified model that comprises only component mole balances and equilibrium relations, but no energy balances, is solved. In subsequent solution steps the energy balance was included again and the model resolved. To solve the problem, they used the so called successive relaxed MINLP (SR-MINLP) proposed by Kraemer et al (2007), which reformulate the MINLP or GDP problems as pure continuous problems with tailored big-M constraints, where all discrete decisions are represented by continuous variables. The discrete decisions are enforced by non-convex constraints that force the continuous variables to take discrete values

Even with all these difficulties, complex problems have been successfully solved, including reactive distillation (Ciric & Gu, 1994; Jackson & Grossmann, 2001); azeotropic sequences (Mariana Barttfeld et al., 2004; Bauer & Stichlmair, 1996, 1998), Thermally coupled distillation sequences (Dunnebier & Pantelides, 1999) or hybrid membrane/distillation systems (Kookos, 2003) among others.

On other side, process simulators are commonly used tools in both academy, even by undergraduate students, and industry to accurately simulate the behavior of complex distillation systems. Process simulators include thermodynamic and transport models that allow accurately calculate properties of pure components, mixtures and state of the art algorithms to simulate distillation columns. Using optimization algorithms with these types of models is a challenging problem because some variables cannot be accessed or modified directly by the user; which sometimes introduces non-differentiabilities.

One interesting approach that makes use of available process simulators and optimization tools was proposed by Lang & Biegler (2002) These authors proposed a distributed stream-tray optimization method (DSTO) in which the reflux and the feed flow rates can be distributed and directed to a set of candidate trays according to a differentiable distribution function (DDF). Using this DDF, the location of the feed, reflux, and other side streams can be treated as continuous instead of integer variables. The drawback, however, is that the DDF function is highly non-convex, and the method can easily be trapped in local solutions.

Caballero et al. (2005) proposed a GDP model –reformulated as a MINLP- that combines the capabilities of process simulators, taking advantage of the tailored algorithms designed for distillation and property estimation implemented in these simulators. The model iterates between two problems: a NLP problem, in which the trays are divided in existing or non-existing and a specially tailored MILP master problem. The basic idea consists of modifying the master by adding to the objective function and to the constraints the 'extra' contributions due to the addition or deletion of trays to each section of the column. The model probed to be robust, but the necessity of a tailored master problem is an important drawback that avoids the inclusion of the model in general flow-sheets.

3. Problem statement.

Taking into account all the previous comments the problem we will address can be state as follows. Given a distillation column (or distillation sequence) for performing a given separation determine the best column or column sequence.

To that end the designer must specify upper and lower bounds to the total number of trays and feeds / products tray positions as well as the purity (or other requirements) of the final streams.

The «best column« is defined in economic terms (minimize the Total Annualized Cost –TAC-) although any other objective can be used.

It is assumed that all the component and thermodynamic properties are known and accurate.

The solution will include the optimal operating conditions, number of trays and feed(s) and product(s) tray positions as well as all the internal flows, temperatures, etc.

In the rest of the paper we first present a novel algorithm that combines-state of the art chemical process simulators (AspenHysys ^{TM}) with GDP model, without reformulation to MINLP, and that does not require modifications in NLP or master sub-problems, overcoming some of the difficulties of the previous approaches. Then a set of examples from single columns to complex sequences illustrates the approach and its advantages and drawbacks. Finally some conclusions and directions for future work are included.

4. GDP disjunctive model for rigorous tray by tray optimization

In this section we propose a novel algorithm for the rigorous design of distillation columns that integrates a process simulator in a generalized disjunctive programming formulation.

The basic idea consists of developing a superstructure that uses as basic unit the distillation section. The optimal distillation column, or column sequence, is obtained by selecting among a set of column sections with different number of theoretical trays. Figure 4 shows an example of superstructure for a conventional two-section column.

In order to write the detailed GDP model let us first introduce the following index sets, parameters and variables in the model

The index sets:

REB	[j | j is a reboiler in the superstructure]
COND	[j | j is a condenser in the superstructure]
Sections	[i | i is a column section]
Vessels	[*v* | v is the column vessel]
DS_i	[k | k is a candidate section formed by N_k trays in column section i]
SV_v	[Sections that form part of the column vessel *v*]

Data:

f	Charging factor for annualizing the capital cost. It was calculated by the equation (1), as recommended by Smith (2005) taking into account the fractional interest rate per year (*r*) and the horizon time (n).

$$f = \frac{r\left(r+1\right)^{n}}{(r+1)^{n}-1} \quad (1)$$

CH_j Heating utility cost in reboiler j

CC_j Cooling utility cost in condenser j

$NTS_{i,k}$ Number of trays of column section k in disjunction i

Variables

x^I Independent variables (Degrees of freedom) in the process simulator

TAC Total annualized cost

D_i Diameter of column section i

$DS_{i,k}$ Diameter of column section k, in disjunction i

NT_i Number of trays in column section i

$Property_i$ Any property of section i that must be calculated.

$Y_{i,k}$ Boolean variable. It takes the value True if the column section k is selected in disjunction i, and False otherwise.

The GDP model can be written as follows:

$$\min : TAC$$

$$s.t.\ TAC = f\cdot\left(Capital\ cost\right) + \sum_{j\in REB} CH_j\, Qreb_j + \sum_{i\in COND} CC_i\, Qcond_i$$

$$Capital\ cost = \sum_{j\in REB}\left(Cost\ Reboiler_j\right) + \sum_{i\in COND}\left(Cost\ Condenser_i\right) + \sum_{v\in Vessels}\left(Cost\ Vessel_v\right)$$

$$(Cost\ Reboiler_j, Cost\ Condenser_i, Qreb_j, Qcond_i, \ldots) = Hy(\mathrm{x}^I)$$

$$\underline{\bigvee}_{k\in DS_i}\begin{bmatrix} Y_{i,k} \\ (DS_{i,k}, NTS_{i,k}, \ldots, PropertyS_{i,k}) = Hy(x^I_{i,k}) \\ D_i = DS_{i,k} \\ NT_i = NTS_{i,k} \\ \vdots \\ Property_i = PropertyS_{i,k} \end{bmatrix} i \in Sections$$

$$Vessel\ Cost_v = f(D_i, NT_i, \ldots)\ i \in SV_v; v \in Vessels \qquad (2)$$

$$Y \in \left[True, False\right]^m$$

where $Hy(\cdot)$ makes reference to the implicit equations solved by the process simulators. In other words, it is a call to AspenHysys ™.

Without loss of generality, the model given by equation (2) is a conceptual representation of the actual model. The particular details are case dependent. Some remarks are therefore necessary:

Even though, in the model there is not an explicit logical relationship that force that to a given section must be assigned exactly a column section, this logical relationship is implicitly forced by the set of disjunctions.

It is possible (usually necessary) to add some constraints in order to satisfice some problem specifications. For example, to force purity or recovery requirements that cannot be specified in the process simulator. However, these constraints are problem dependent.

The disjunctions are used to determine all the necessary data to calculate the cost (e.g. diameter (*D*), number of trays (*NT*)) or any other column section property. The cost of the vessel is calculated in terms of the values assigned to each of the sections that form the vessel. The sections that form a given column are controlled through the set '*Vessels*', that specifies which sections form part of a given column.

In equation (2) we do not differentiate between dependent and independent variables. In an equation based environment the solver deals with the variables and the user does not have to worry about. However, in a process simulator the selection of independent variables (degrees of freedom) is critical and problem dependent. Besides, the model given by (2), is a hybrid model, formed by implicit equations (solved by the process simulator) and explicit equations. The solver can only control the explicit variables –that appear in equation form- and the independent variables in the process simulator. A critical issue in the optimization with process simulators is that the failure in the simulator convergence produces the failure in the optimization. Although it is possible to introduce safeguards –the optimization can be recovered from a simulator convergence failure-, repeated failures make the optimization impossible. In the particular case of distillation columns there are sets of specifications that facilitate the convergence (i.e. reflux ratio and boilup ratio or the recovery of key components). However, specifications like compositions are usually most difficult to converge. In these last case a better approach consists of selecting a set of 'easy to converge' specifications and add the difficult constraints as external equations in the model.

The direct implementation of the superstructure presented in Figure 4 is not practical even for the case of a single column, at least for two reasons. First, it results in a very complex model in the process simulator –Figure 5 shows an example of how the model looks like in AspenHysys^{TM}-. The NLP optimization takes large CPU times mainly due to the time to converge the complete flow sheet each time the solver calls it.

Fortunately, it is not necessary to use that 'Brute Force Approach'. Instead, the problem can be reformulated as a Disjunctive Problem with Net Structure (only two term disjunctions appear, and the second term just state that if a Boolean variable is set to False, all the variables related to that disjunction are set to zero). It is worth noting that all disjunctive problems can be reformulated as disjunctive problems with net structure, and solved using the Logic Based Outer Approximation (LBOA) algorithm presented by Turkay & Grossmann (1996). The reformulated problem is as follows:

$$\min : TAC$$

$$s.t.\ TAC = f\cdot\left(Capital\ cost\right) + \sum_{j\in REB} CH_j\, Qreb_j + \sum_{i\in COND} CC_i\, Qcond_i$$

$$Capital\ cost = \sum_{j\in REB}\left(Cost\ Reboiler_j\right) + \sum_{i\in COND}\left(Cost\,Condenser_i\right) + \sum_{v\in Vessels}\left(Cost\,Vessel_v\right)$$

$$(Cost\ Reboiler_j, Cost\,Condenser_i, Qreb_j, Qcond_i, ...) = Hy(\mathrm{x}^I)$$

$$\begin{bmatrix} Y_{i,k} \\ (DS_{i,k}, NTS_{i,k}, ..., PropertyS_{i,k}) = Hy(x^I_{i,k}) \\ D_i = DS_{i,k} \\ NT_i = NTS_{i,k} \\ \vdots \\ Property_i = PropertyS_{i,k} \end{bmatrix} \underline{\vee} \begin{bmatrix} \neg Y_{i,k} \\ DS_{i,k} = 0 \\ NTS_{i,k} = 0 \\ \vdots \\ PropertyS_{i,k} = 0 \end{bmatrix} \begin{matrix} i \in Sections \\ k \in DS_i \end{matrix}$$

$$\underline{\vee}_{k\in DS_i} Y_{i,k} \quad \forall i \in Sections \tag{3}$$

$$Vessel\ Cost_v = f(D_i, NT_i, ...)\ i \in SV_v; v \in Vessels$$

$$Y \in \left[True, False\right]^m$$

The following remarks are important:

In the LBOA algorithm (Turkay & Grossmann, 1996), each NLP is formed by a potential flow sheet configuration, the rest of variables do not appear in the model. Note that strictly speaking those variables are set to zero –second term in the disjunctions in equation (3)- but in the practical implementation, when solving the NLP those variables are dropped from the model. In this case a potential flow sheet is simply a distillation column (or a distillation sequence). This fact is really important and what makes the algorithm really useful. Although, from a conceptual point of view the superstructure is relatively complex (see Figures 4 and 5) the LBOA algorithm decomposed the problem in feasible NLPs that, as commented, results to be regular distillation columns. The number of trays in each section at each iteration are «decided» by the algorithm without user intervention. Therefore, even though there is complex model formulation, the GDP algorithm results in a sequence of relatively easy to solve sub-problems. As a consequence the algorithm could be eventually super-imposed to any process simulator without modifying at all the simulator internal algorithms.

In a single column, with known pressure profile, there are only two degrees of freedom. Therefore the CPU time needed to solve each NLP is of a few seconds. Even in systems

involving more than one distillation column these NLPs can be efficiently solve, with the same degree of difficulty that a regular NLP optimization in a process simulator.

4.1. Implementation details

The logic based outer approximation algorithm has been implemented in Matlab (MATLAB., 2006.) and completely automatized, i.e. once the model is set there the user intervention is not necessary. For solving the NLP and Master-MILP sub-problems used in the LBOA algorithm we used the Tomlab optimization environment (Holmström, 1999) that provides a gateway for using state of the art NLP/MILP solvers. Although it is possible to use the internal NLP optimizers provided by the process simulator, our experience shows (arguable) that external NLP solvers like CONOPT (Drud, 1996) or SNOPT (Gill et al., 2002) are more robust and reliable. The MILP sub-problems were solved using CPLEX (Holmström et al., 2009).

Figure 6 shows a scheme of the actual implementation. A description step by step of the problem is as follows

Step 1. In the process simulator (AspenHysys^{TM}). Set a distillation column (or distillation sequence) the total number of trays or the feed(s) tray positions are not important at this stage. The lack of convergence of the process simulator could eventually produce that the entire algorithm fails. In order to minimize that eventuality the set of specifications must be selected to make the convergence as easy as possible. For example, usually selecting the reboil ratio and the boil-up ratio as degrees of freedom results in easy to converge columns (although this is case dependent). At this stage, purity requirements or other constrains can be violated.

The selection of thermodynamic models, properties estimation etc., are all in the simulation environment. All the numerical issues related to the convergence of distillation columns (or column sections) are also maintained in the simulation environment and must be specified in this stage.

Step 2. Initialization. In the model we specified, for each column section, a set of candidate column sections with different number of trays. In order to initialize the problem we must solve a set of NLP problems that include, at least once, each candidate column section. To this end we solve a set covering problem to determine the minimum number of feasible flow-sheets that comply with this condition (Turkay & Grossmann, 1996). For example, consider a single column with one feed, and two products (distillate and bottoms). Assume that the rectifying section must be selected among *NR* different column section, and that the stripping section must be selected among *NS* different column sections. The minimum number of initial NLP problems will be equal to the maximum of *NR* and *NS*. Each one of those initial flow-sheets is simply a distillation column with fixed number of trays and fixed tray position. In general, the number of initial problems is given by equation (4):

$$N^{o}\,Initial\,NLPs = \max\left(NTS_{i,k}\right) \tag{4}$$

If this number is too large, it is possible reduce the number of candidate sections by considering sections that differentiate in 2, 3, … or n trays. Once the problem has been solved, it is possible to refine the search by considering values around the optimal solution obtained in previous iteration.

Step 3. Generate and solve the Master problem. The MILP is generated using the approach presented in the original LBOA algorithm (Turkay & Grossmann, 1996), with equality relaxation to deal with equality constraints (Kocis & Grossmann, 1987) and the augmented penalty strategy (Viswanathan & Grossmann, 1990) to ensure that linearizations of non-convex problems yield always feasible MILP problems. The derivative information for generating the MILP Master problem is provided directly by the last NLP (or the initial sets of NLPs) and corresponds to the Jacobian and function gradient in the optimal NLP solution.

It is worth mentioning that in the Master problem depends only on independent variables (degrees of freedom in the flow-sheet) and explicit variables in the model. Those variables calculated by the flow-sheet (i.e. reboiler and condenser heat loads, compositions, etc.) cannot appear in the Master problem. In other words, the Master is defined in the reduced space of explicit variables.

The master problem includes also an integer cut (Balas & Jeroslow, 1972) to exclude configurations explored by the algorithm in previous iterations.

Step 4. Solve the new NLP problem. The NLP problem is just a flow-sheet in which all the structural parameters (number of trays, feed and products tray positions) are known. The complexity of these NLPs is equivalent to any regular optimization using a process simulator. As previously commented, we obtained good and reliable results using an external solver. A detailed discussion about this topic is out of the scope of this paper. The interested reader can found information, for example in the following references (Biegler & Cuthrell, 1985; Y.D. Lang & Biegler, 1987)

Step 5. Check convergence. Due to the high non-convexity, the lower bounding property of the Master does not always hold and therefore, the usual stopping criteria (the last MILP master problem and the best NLP upper bound cross each other or the heuristic rule: stop when in two consecutive major iterations the objective of the NLPs worsen) are likely to provide a local optimal solution. However, the experience with the outer approximation algorithm for both the MINLP and logic versions shows that the optimal solution is usually found in the first major iterations (usually in the five first and rarely after the 10^{th} major iteration). So we force the algorithm to perform at least 10 major iterations. If in these 10 iterations both, worsening of the objective function in two consecutive NLPs and crossing of the objective values of NLPs and master problems are simultaneously fulfilled, we stop. Otherwise, the iterations continue until both conditions hold.

Steps 2 to 5 are completely automatized and do not requires the user intervention.

Some final remarks are of interest:

The problem we are solving in a nonconvex discrete/continuous optimization problem. It is clear that the approach presented offers no guarantee of convergence to the global optimum. There are two issues to consider here. The first is that the NLP subproblems are nonconvex, and therefore local optimizers might get trapped in local solutions. Numerical test, however shows that most of the NLPs converges to the global optimal solution but if necessary the NLPs can be re-initialized from different starting points to increase the chances of identifying the global optimum. The second is that the master MILP might cut-off feasible solutions when adding supporting hyperplanes constructed from nonconvex functions. This limitation is handled (to some extent) through the addition of slack variables and penalty terms and forcing the stopping criteria commented in previous paragraphs.

As commented the logic based GDP approach generates only feasible NLPs that in this case result in just a sequence of columns with fixed number of trays and fixed feed position. As a consequence it is possible super-impose the algorithm in almost any process simulator without modifying the internal algorithms. Eventually it could be included as an "Add in" in any commercial process simulator.

5. Examples

Example 1. This first example deals with the separation of a mixture of Methanol, Acetone and Water (MeOH – Ac – W). Acetone and methanol forms a minimum boiling azeotrope. The objective is to obtain a mixture of acetone and methanol with a combined mole fraction greater than 0.99 and a combined mole recovery greater than 99%. All relevant data for the example are shown in Table 1.

This case study has been included to provide an example that is easy to reproduce and in which equations for calculating rigorous costs do not hide the essence of the algorithm. Instead of rigorous calculation of the total annual cost, we assume that the cost is given by a simple expression in terms of the heat loads and number of column trays:

$$\min : 0.2Qcond(kW) + QReb(kW) + 100(Total\ \ number\ \ of\ \ Trays)$$

where *Qcond* and *Qreb* make reference to the condenser and reboiler heat loads respectively.

In order to increase the robustness of the algorithm it is convenient to take as much advantage as possible from the specialized algorithms included in the process simulator (Aspen.Hysys ^{TM}). In this case the two constraints (mole fraction and recovery in distillate) can be used as specifications and therefore the problem is transformed in finding the combination of column sections with a minimum value of objective function. However, as previously commented, it is

worth remarking that transferring constraints (specifications) to the process simulator is not always possible because under some specifications the simulator convergence is difficult. Even though, the NLP solver can usually recover from a convergence failure, in general the lack of convergence in the simulator makes that all the procedure fail. In those cases a good alternative consists of selecting a set of specifications that ensure convergence for a large interval of values (i.e. reboil ratio and boilup ratio), and let the NLP solver to converge the difficult constraints. It is also convenient to carry out a preliminary study to bound the value of variables to ensure the convergence of the process simulator.

In order to ensure convergence with the current specifications, in this case study the minimum number of trays for the rectifying sections was fixed to 2; and to one in the stripping section. The maximum number of trays was fixed to the minimum plus 10 trays. For the case in which the optimal solution lies at one of these limits, the maximum number of trays is increased or the minimum number of trays is decreased.

First we solve the initialization NLPs ensuring that a particular tray in each section is selected at least once, see Table 2. The optimal solution was found in the 9^{th} major iteration with an objective value of 2578.4 (*Qreb* =1318 kW; *QCond* =1304 kW; Number of trays = 10). Table 2 shows also the results of the major iterations. Note that in the 7^{th} major iteration the MILP master problem and the best NLP crossed each other, using that stopping criterion the solution would be 2635.8, which is also a good solution because only differs from the optimal in one tray and only 2.2% in objective function value. Figure 7 shows the optimal solution.

This is a small problem therefore it is possible to systematically check all the alternatives that prove that the solution obtained was also the global optimum.

Example 2. In this second example the objective is to separate ethanol from propanol and butanol, to obtain a distillate with at least 0.98 mole fraction in ethanol and at least 99.5% ethanol recovery. All relevant data for this example are in Table 1.

The objective function consists of minimizing the total annualized cost (TAC), calculated as the sum of operating and annualized investment costs. In this example and the following, the sizing of the distillation column is done following the procedure proposed by Stichlmair & Fair (1998), and the investment cost is calculated using the correlations presented by Turton et al (2003).

The annualizing factor of the capital cost (*f*) was calculated by equation (1), as recommended by Smith (2005) taking into account the fractional interest rate per year (*i*) and the horizon time (n). Of course, changing the annualizing period can lead to different optimal columns, due to the tradeoff between the capital and operating costs.

In this example the number of trays in each section ranges between 5 and 25, which gives a column with a maximum of 51 trays (25 per section plus the feed tray). With this configuration

we should solve 20 initial NLPs. To reduce the number of initial NLPs we consider only column sections that differentiate in n-stages (i.e. n = 2, 3 or 4) and then contract the column around the best solution.

In the first iteration we assume five potential sections (in both stripping and rectifying sections) that differentiate in 4 trays, the optimal solution for this first iteration, is obtained for a configuration with 13 stages in the rectifying section and 21 in the stripping section (35 trays – including the feed tray- plus condenser and reboiler) with a total annual cost of $ 564.99·10^3. In the second iteration we consider again 5 potential sections (for both rectifying and stripping sections) that differentiate in two trays centered in the optimal solution obtained in previous iteration (e.g. 9, 11, 13, 15, and 17 trays in the rectifying section). The optimal solution in this iteration was $562.8·10^3, and was formed by 15 trays in the rectifying section, and 21 in the stripping section. In the third iteration, we consider five new sections that differentiate in just one tray. The optimal solution was $562.15·10^3, and was formed by 15 trays in the rectifying section, and 20 in the stripping section (Total number of trays equal to 36).

Starting with 10 potential sections that differentiate in 2 trays or starting with different initial configurations yield solutions that differs from the one presented above in just one tray. Figure 8 shows the optimal configuration.

Example 3. In this example, instead of a single column we optimize a given sequence of partially thermally coupled distillation sequences. Information about thermally coupled distillation can be found, for example in (Agrawal, 1996; Blancarte-Palacios et al., 2003; José A. Caballero, 2009; J. A. Caballero & Grossmann, 2001, 2004; José A. Caballero & Grossmann, 2012; Gutérrez-Antonio et al., 2011; Rong et al., 2001; Shah & Agrawal, 2010):

The objective consists of determining the number of theoretical trays, feed positions in each column and operating conditions for separating a mixture of Benzene, Toluene, p-Xylene and Bi-Phenyl using the partially thermally coupled sequence of columns presented in Figure 9a. The molar fraction of each final product must be greater than 0.99. All relevant data for this example can be found in Table 1.

In order to facilitate the convergence of each column, It is convenient to transform the sequence given in Figure 9a into another thermodynamically equivalent (Agrawal & Fidkowski, 1998; J. A. Caballero & Grossmann, 2003; Hernandez et al., 2006; Rong et al., 2004) that can be associated to a sequence of conventional columns –each column with a rectifying and a stripping section- like the sequence shown in Figure 9b. The simulation of thermally coupled distillation sequences presents the problem that a thermal couple is formed by two side liquid and vapor streams connecting two column sections. In other words, each thermal couple introduces a recycle. The usual approach in modular process simulator to converge the cycles is by using fixed point methods (Biegler et al., 1997), that has only linear convergence. If we

have to converge the flow sheet each time the optimizer calls the simulator the total CPU time quickly becomes prohibitive. Besides, in slightly numerical noisy systems (J. A. Caballero & Grossmann, 2008) the recycles act as noise amplifiers. To avoid all these problems we simulate the sequence of columns using the procedure proposed by Navarro et al. (2012), that substitute the two streams forming a thermal couple by the equivalent set formed by a saturated stream and an energy stream.

For column sections 1 and 2 (referred to Figure 9) we assume a set of ten column sections ranging from 10 to 20 trays. For sections 3 to 6 we assume ten column sections ranging from 5 to 15 trays each one. And for column sections 7 and 8 we assume a set of seven column sections ranging from 1 to 8 trays each.

As independent variables we use the recoveries of key components in each separation. The optimal solution of the problem is obtained in around 3 minutes of CPU time (*Intel Core(TM)2 Quad CPU Q6600 @ 2.40GHz 2.39 GHz under Windows 7*). The optimal solution with the most relevant values is presented in Figure 10

Example 4 This example involves the optimization of an extractive distillation process. Extractive distillation is used to separate close boiling or homogeneous binary azeotropes by adding an entrainer that must be a higher-boiling component. The proposed case study is adapted from Luyben (2011). The objective is to separate an isomolar mixture of acetone and methanol using dimethyl sulfur oxide (DMSO) as entrainer. The system acetone-methanol has, at atmospheric pressure, a minimum boiling homogeneous azeotrope at 77.6 mol% acetone. The normal boiling point of acetone is 239 K and for methanol is 338 K, while for DMSO is 464 K. Relevant data for this case study is presented in Table 1.

Extractive distillation comprises two columns. The first one is the extractive column, which has two feeds. The entrainer is fed to the column above the process feed. The second column is the entrainer-recovery column. The acetone is recovered at the top of the extractive column with purity higher than 0.9995 mole fraction, while the methanol with the DMSO is obtained as bottoms product. In the second column the DMSO is separated from the methanol both with purities larger than 0.9995 mole fraction.

In extractive distillation, one of the factors that influence the most the cost is the flow of entrainer introduced in the first column. The flow must be large enough to facilitate the separation between acetone and methanol, but at the same time, large flows inside a distillation column increases the heat duties and column diameters.

The extractive distillation sequence can be divided in five column sections (see Figure 11). Due to the number of trays in each column section can be very different, and in particular due to in section 2 the number of trays is considerably larger than in the rest of the sections, we follow a sequential approach similar to that in example 2. Initially, for section 1 we select a column section among a set of five column sections that differentiate in two trays ranging from 3 to 11

trays. For sections 2 to 5 we assume a set of 5 column sections for each one that differentiate in four trays: Between 20 and 40 for section 2, between 10 and 30 for section 3 and between 2 and 18 for sections 4 and 5. Then in successive iterations we consider set of columns that differentiate in 1 or 2 trays by contracting around the optimal solution of the previous iteration. Table 3 shows the details of the three major iterations needed to obtain the optimal solution. Figure 11 shows the optimal solution obtained.

The importance of the DMSO flow rate is evident by a sensitivity analysis of the optimal solution (Figure 12). This Figure shows that the reboiler heat load in the first column decreases when the DMSO flow rate increases. However, the condenser heat load presents a minimum for values around 365 kmol/h of DMSO (DMSO to feed ratio equal to 0.73). In the second column, as expected, the reboiler and condenser heat loads increases with the DMSO flow rate. The minimum total annualized cost is obtained for a DMSO flow rate equal to 344.5 kmol/h.

6. Conclusions

This paper has proposed a systematic method for the rigorous design of distillation columns in which operational conditions (reflux ratio, internal and external flows, etc.) as well as structural parameters (number of trays in each column section and consequently location of feed and product streams, etc.) are simultaneously optimized.

The rigorous optimization of a distillation column, or column sequence, can be performed by considering a column as a succession of column sections, separated by feeds, products or heat streams. Therefore, the optimal distillation column, or column sequence, is obtained by selecting, for each column section, among a set of column sections with different number of theoretical trays. This problem is formulated as a generalized disjunctive problem, in which each m-term disjunction is related with selection among a set of m candidate column sections.

The model is solved using a Hybrid simulation optimization approach by taking advantage of the effective and reliable numerical methods included in process simulators for converging distillation columns as well as the thermodynamic packages, property estimation etc., and at the same time the flexibility of an equation based environment. Difficult constraints can be transferred to the explicit equation part increasing the robustness of the optimization process.

The model is solved using the Logic Based Outer Approximation algorithm. The major advantages of this algorithm are

- It allows the use of commercial process simulators to perform the rigorous design of distillation columns or column sections without the necessity of special algorithms but standard logic based GDP solvers.

- Due to the fact that the LBOA algorithm or its modifications solves a series of NLPs that must correspond to feasible alternatives, and that in the context of distillation columns/sequences this is a column/sequence with fixed number of trays and fixed feeds positions, these NLPs correspond to the continuous optimization of a regular distillation column that can be done by internal (included in the simulator) or external solvers –in this paper we follow this second approach-. The complex superstructure is reduced, from the point of view of the final user, to a single column or column sequence in the process simulator.

- As a consequence the implementation of this algorithm in a process simulator is relatively easy because there is no needed any modification, or MINLP reformulation.

- The examples presented, show that the approach is robust and reliable with CPU times lower than 5 minutes in the worst case. However, due to the extremely non-convex nature of the problem, we can ensure just an optimal solution. Although the experience shows that usually, if not the global optimal, very good solutions are obtained.

- The simplicity, from the point of view of a final user, make that the algorithm can be used by designers without an optimization background.

Two drawbacks should also be mentioned:

- Even though the model usually get near global optimal solutions, this fact cannot be guaranteed and correspond to the designer the critical analysis of the final solution.

- Some knowledge about the system is needed. In general it would not be a good idea try to perform a 'blind optimization' from scratch. Adequate bounds on the number of trays in each section, reasonable initial values and feasible constraints on purity should be provided. In that sense this algorithm can be considered an excellent tool to complement other approaches (conceptual design, shortcut methods or any other of those outlined in the introduction), and can be used to 'get a rigorous design' from a preliminary design.

Future directions include the extension to the synthesis of distillation sequences where a full column can completely disappear in a superstructure, and the integration in a general superstructure synthesis framework.

Acknowledgements

The author wish to acknowledge financial support from the Spanish Ministry of Science and Innovation (CTQ2012-37039-C02-02).

References

Agrawal, R. (1996). Synthesis of distillation column configurations for a multicomponent separation. Industrial & Engineering Chemistry Research, 35, 1059-1071.

Agrawal, R., & Fidkowski, Z. T. (1998). More operable arrangements of fully thermally coupled distillation columns. AIChE Journal, 44, 2565-2568.

Bagajewicz, M. J., & Manousiouthakis, V. (1992). Mass/heat-exchange network representation of distillation networks. Aiche Journal, 38, 1769-1800.

Balas, E., & Jeroslow, R. (1972). Canonical Cuts On Unit Hypercube. Siam Journal On Applied Mathematics, 23, 61-69.

Barbosa, D., & Doherty, M. F. (1988). Design and minimum-reflux calculations for single-feed multicomponent reactive distillation columns. Chemical Engineering Science, 43, 1523-1537.

Barttfeld, M., & Aguirre, P. A. (2002). Optimal Synthesis of Multicomponent Zeotropic Distillation Processes. 1. Preprocessing Phase and Rigorous Optimization for a Single Unit. Industrial & Engineering Chemistry Research, 41, 5298-5307.

Barttfeld, M., & Aguirre, P. A. (2003). Optimal Synthesis of Multicomponent Zeotropic Distillation Processes. 2. Preprocessing Phase and Rigorous Optimization of Efficient Sequences. Industrial & Engineering Chemistry Research, 42, 3441-3457.

Barttfeld, M., Aguirre, P. A., & Grossmann, I. E. (2003). Alternative representations and formulations for the economic optimization of multicomponent distillation columns. Computers & Chemical Engineering, 27, 363-383.

Barttfeld, M., Aguirre, P. o. A., & Grossmann, I. E. (2004). A decomposition method for synthesizing complex column configurations using tray-by-tray GDP models. Computers & Chemical Engineering, 28, 2165-2188.

Bauer, M. H., & Stichlmair, J. (1996). Superstructures for the mixed integer optimization of nonideal and azeotropic distillation processes. Computers & Chemical Engineering, 20, S25-S30.

Bauer, M. H., & Stichlmair, J. (1998). Design and economic optimization of azeotropic distillation processes using mixed-integer nonlinear programming. Computers & Chemical Engineering, 22, 1271-1286.

Bausa, J., von Watzdorf, R., & Marquardt, W. (1998). Shortcut Methods for Nonideal Multicomponent Distillation: 1. Simple Columns. AIChE Journal, 44, 2181-2198.

Biegler, L. T., & Cuthrell, J. E. (1985). Improved infeasible path optimization for sequential modular simulators—II: the optimization algorithm. Computers & Chemical Engineering, 9, 257-267.

Biegler, L. T., Grossmann, I. E., & Westerberg, A. W. (1997). Systematic methods of chemical process design. Upper Saddle River, N.J.: Prentice Hall PTR.

Blancarte-Palacios, J. L., Bautista-Valdes, M. N., Hernandez, S., Rico-Ramirez, V., & Jimenez, A. (2003). Energy-efficient designs of thermally coupled distillation sequences for four-component mixtures. Industrial & Engineering Chemistry Research, 42, 5157-5164.

Caballero, J. A. (2009). Thermally Coupled Distillation. In C. A. O. d. N. Rita Maria de Brito Alves & B. Evaristo Chalbaud (Eds.), Computer Aided Chemical Engineering (Vol. Volume 27, pp. 59-64): Elsevier.

Caballero, J. A., & Grossmann, I. E. (1999). Aggregated models for integrated distillation systems. Industrial and Engineering Chemistry Research, 38, 2330-2344.

Caballero, J. A., & Grossmann, I. E. (2001). Generalized disjunctive programming model for the optimal synthesis of thermally linked distillation columns. Industrial & Engineering Chemistry Research, 40, 2260-2274.

Caballero, J. A., & Grossmann, I. E. (2003). Thermodynamically equivalent configurations for thermally coupled distillation. AIChE Journal, 49, 2864-2884.

Caballero, J. A., & Grossmann, I. E. (2004). Design of distillation sequences: from conventional to fully thermally coupled distillation systems. Computers & Chemical Engineering, 28, 2307-2329.

Caballero, J. A., & Grossmann, I. E. (2008). An algorithm for the use of surrogate models in modular flowsheet optimization. Aiche Journal, 54, 2633-2650.

Caballero, J. A., & Grossmann, I. E. (2012). Synthesis of complex thermally coupled distillation systems including divided wall columns. Aiche Journal, 59, 1139-1159.

Caballero, J. A., Milan-Yanez, D., & Grossmann, I. E. (2005). Rigorous Design of Distillation Columns: Integration of Disjunctive Programming and Process Simulators. Industrial & Engineering Chemistry Research, 44, 6760-6775.

Ciric, A. R., & Gu, D. (1994). Synthesis of nonequilibrium reactive distillation processes by MINLP optimization. Aiche Journal, 40, 1479-1487.

Drud, A. S. (1996). CONOPT: A System for Large Scale Nonlinear Optimization. Reference Manual. Bagsvaerd, Denmark: ARKI Consulting and Development A/S.

Dunnebier, G., & Pantelides, C. C. (1999). Optimal design of thermally coupled distillation columns. Industrial & Engineering Chemistry Research, 38, 162-176.

Edmister, W. C. (1943). Design of hydrocarbon absorption and stripping. . Industrial & Engineering Chemistry, 35, 837-839.

Fenske, M. R. (1932). Fractionation of Straight-Run Pennsylvania Gasoline. Industrial & Engineering Chemistry, 24, 482-485.

Fidkowski, Z. T., Doherty, M. F., & Malone, M. F. (1993). Feasibility of separations for distillation of nonideal ternary mixtures. Aiche Journal, 39, 1303-1321.

Fidkowski, Z. T., Malone, M. F., & Doherty, M. F. (1991). Nonideal multicomponent distillation: Use of bifurcation theory for design. Aiche Journal, 37, 1761-1779.

Gani, R., & Bek-Pedersen, E. (2000). Simple new algorithm for distillation column design. Aiche Journal, 46, 1271-1274.

Gill, W. P. E., Murray, W., & Saunders. (2002). SNOPT: an SQP algorithm for large-scale constrained optimization. SIAM J. Optimization, 979-1006.

Gilliland, E. R. (1940). Multicomponent Rectification Estimation of the Number of Theoretical Plates as a Function of the Reflux Ratio. Industrial & Engineering Chemistry, 32, 1220-1223.

Gutérrez-Antonio, C., Briones-Ramírez, A., & Jiménez-Gutiérrez, A. (2011). Optimization of Petlyuk sequences using a multi objective genetic algorithm with constraints. Computers & Chemical Engineering, 35, 236-244.

Harwardt, A., & Marquardt, W. (2012). Heat-integrated distillation columns: Vapor recompression or internal heat integration? Aiche Journal, 58, 3740-3750. .

Hernandez, S., Gabriel Segovia-Hernandez, J., & Rico-Ramirez, V. (2006). Thermodynamically equivalent distillation schemes to the Petlyuk column for ternary mixtures. Energy, 31, 2176.

Holmström, K. (1999). The Tomlab Optimization Environment in Matlab. Adv. Model Optim., 1, 47-69.

Holmström, K., Anders, O. G., & Edwall, M. (2009). User's guide for TOMLAB/CPLEX v.11.2. In http://tomopt.com/docs/TOMLAB_CPLEX.pdf (Ed.).

Humphrey, J. (1995). Separation processes: playing a critical role. Chemical Engineering Progress, 91, 43-54.

Jackson, J. R., & Grossmann, I. E. (2001). A disjunctive programming approach for the optimal design of reactive distillation columns. Computers & Chemical Engineering, 25, 1661-1673.

Julka, V., & Doherty, M. F. (1990). Geometric behavior and minimum flows for nonideal multicomponent distillation. Chemical Engineering Science, 45, 1801-1822.

Kamath, R. S., Grossmann, I. E., & Biegler, L. T. (2010). Aggregate models based on improved group methods for simulation and optimization of distillation systems. Computers & Chemical Engineering, 34, 1312-1319.

Kocis, G. R., & Grossmann, I. E. (1987). Relaxation Strategy For The Structural Optimization Of Process Flow Sheets. Industrial & Engineering Chemistry Research, 26, 1869-1880.

Kookos, I. K. (2003). Optimal Design of Membrane/Distillation Column Hybrid Processes. Industrial & Engineering Chemistry Research, 42, 1731-1738.

Kossack, S., Kraemer, K., & Marquardt, W. (2006). Efficient Optimization-Based Design of Distillation Columns for Homogenous Azeotropic Mixtures. Industrial & Engineering Chemistry Research, 45, 8492-8502.

Kraemer, K., Kossack, S., & Marquardt, W. (2007). An efficient solution method for the MINLP optimization of chemical processes. . Computer Aided Chemical Engineering, 105-110.

Kremser, A. (1930). Theoretical analysis of absortion process. . Natural Petroleum News, 22, 43-49.

Kunesh, J., Kister, H., Lockett, M., & Fair, J. R. (1995). Distillation: still towering over other options. Chemical Engineering Progress, 91.

Lang, Y. D., & Biegler, L. T. (1987). A unified algorithm for flowsheet optimization. Computers & Chemical Engineering, 11, 143-158.

Lang, Y. D., & Biegler, L. T. (2002). Distributed stream method for tray optimization. AIChE Journal, 48, 582-595.

Levy, S. G., & Doherty, M. F. (1986). Design and synthesis of homogeneous azeotropic distillations. 4. Minimum reflux calculations for multiple-feed columns. Industrial & Engineering Chemistry Fundamentals, 25, 269-279.

Levy, S. G., Van Dongen, D. B., & Doherty, M. F. (1985). Design and synthesis of homogeneous azeotropic distillations. 2. Minimum reflux calculations for nonideal and azeotropic columns. Industrial & Engineering Chemistry Fundamentals, 24, 463-474.

Lucia, A., Amale, A., & Taylor, R. (2008). Distillation pinch points and more. Computers & Chemical Engineering, 32, 1342-1364.

Lucia, A., & McCallum, B. R. (2010). Energy targeting and minimum energy distillation column sequences. Computers & Chemical Engineering, 34, 931-942.

Lucia, A., & Taylor, R. (2006). The geometry of separation boundaries: I. Basic theory and numerical support. Aiche Journal, 52, 582-594.

Luyben, W. L. (2011). Principles and Case Studies of Simultaneous Design. Hoboken, New Yersey, USA.: John Wiley & Sons.

MATLAB. (2006.). The Language of Technical Computing. In: The Mathworks Inc.

Mix, T., Dweck, J., Weinberg, M., & Armstrong, R. (1978). Energy conservation in distillation. Chemical Engineering Progress, 74.

Navarro, M. A., Javaloyes, J., Caballero, J. A., & Grossmann, I. E. (2012). Strategies for the robust simulation of thermally coupled distillation sequences. Computers & Chemical Engineering, 36, 149-159.

Rong, B. G., Kraslawski, A., & Nystrom, L. (2001). Design and synthesis of multicomponent thermally coupled distillation flowsheets. Computers & Chemical Engineering, 25, 807-820.

Rong, B. G., Kraslawski, A., & Turunen, I. (2004). Synthesis and optimal design of thermodynamically equivalent thermally coupled distillation systems. Industrial & Engineering Chemistry Research, 43, 5904-5915.

Rudd, D. F., & Watson, C. C. (1968). Strategy of process engineering. . New York: Wiley.

Sargent, R. W. H., & Gaminibandara, K. (1976). Optimum Design of Plate distillation Columns. In L. W. C. Dixon (Ed.), Optimization in Action (pp. 267): Academic Press, London.

Shah, V. H., & Agrawal, R. (2010). A matrix method for multicomponent distillation sequences. AIChE Journal, 56, 1759-1775.

Smith, R. (2005). Chemical Process Design and Integration. Chichester: John wiley & Sons, Ltd.

Soave, G., & Feliu, J. A. (2002). Saving energy in distillation towers by feed splitting. Applied Thermal Engineering, 22, 889.

Stichlmair, J. G., & Fair, J. R. (1998). Distillation: Principles and Practice. New York.: Wiley-VCH.

Turkay, M., & Grossmann, I. E. (1996). Logic-based MINLP algorithms for the optimal synthesis of process networks. Computers & Chemical Engineering, 20, 959-978.

Turton, R., Bailei, R. C., Whiting, W. B., & Shaeiwitz, J. A. (2003). Analysis Synthesis and Design of Chemical Processes. New York: McGraw-Hill.

Underwood, A. (1948). Fractional distillation of multicomponent mixtures. Chemical Engineering Progress, 44, 603-614.

Viswanathan, J., & Grossmann, I. E. (1990). A combined penalty function and outer-approximation method for MINLP optimization. Computers & Chemical Engineering, 14, 769-782.

Viswanathan, J., & Grossmann, I. E. (1993). An Alternate MINLP Model for Finding the Number of Trays Required for a Specified Separation Objective. Computers & Chemical Engineering, 17, 949-955.

von Watzdorf, R., Bausa, J., & Marquardt, W. (1999). Shortcut Methods for Nonideal Multicomponent Distillation: 2. Complex Columns. AIChE Journal, 45, 1615-1628.

Yeomans, H., & Grossmann, I. E. (2000a). Disjunctive programming models for the optimal design of distillation columns and separation sequences. Industrial & Engineering Chemistry Research, 39, 1637-1648.

Yeomans, H., & Grossmann, I. E. (2000b). Optimal design of complex distillation columns using rigorous tray-by-tray disjunctive programming models. Industrial & Engineering Chemistry Research, 39, 4326-4335.

Figure Captions

Figure 1. Superstructure by Sargent and Gaminibandara (left) and a possible solution (right).

Figure 2. Superstructure by Viswanathan and Grossmann (left) and a possible solution (right).

Figure 3. Superstructure by Yeomans and Grossmann.

Figure 4. Superstructure for a single distillation column. Each column section must be selected among a set of column sections that differentiate in the number of trays.

Figure 5. Direct Implementation of the proposed superstructure in Hysys™. The complexity of the resulting superstructure makes the direct implementation difficult for more than a single column.

Figure 6. Scheme of the general modeling framework and the hybrid simulation-optimization solution algorithm.

Figure 7. Optimal solution for example 1.

Figure 8. Optimal solution for example 2.

Figure 9. Sequence of partially thermally coupled distillation sequence and its thermodynamically equivalent configurations using to section columns.

Figure 10. Optimal solution for example 3.

Figure 11. Optimal solution for example 4.

Figure 12. Results of the sensitivity analysis for the optimal solution of example 4 in terms of DMSO flow rate introduced to the first columns.

Table 1. Data for the Examples

Heat Exchangers

Reboiler: U = 820 W/(m^2 K)

Condenser: U = 1000 W/(m^2 K)

Utilities

Low Pressure Steam	(254 ºC)	17.70 $/GJ
High Pressure Steam	(130 ºC)	14.05 $/GJ
Cold Water	(20-40 ºC)	0.354 $/GJ

Columns

Calculated based on sieve trays.

Stainless steel.

Tray separation 0.609 m.

Sizing following the procedure by Stichlmair & Fair (1998).

Economics

Annual interest rate (i) 10%

Time horizon (n) 8 years

Capital cost correlations from Turton et al (2003)

Example 1		**Example 2**	
Feed	100 kmol /h	Feed	100 kmol/h
Composition (m.f.)		Composition (m.f.)	
Methanol	0.4	Ethanol	0.3
Acetone	0.3	1-propanol	0.4
Water	0.3	1-butanol	0.3
Pressure	101.325 kPa	Pressure	101.325 kPa
Thermal state	Saturated liquid	Thermal state	Saturated liquid
Thermodynamics	NRTL Hysys default	Thermodynamics	NRTL Hysys default
Specifications		Specifications	
Acetone + Methanol mole fractions	>0.99	Ethanol mole fraction	>0.98
Acetone + Methanol recovery	>99%	Ethanol recovery	>99.5%
Example 3		**Example 4**	
Feed	200 kmol/h	Feed	
Composition (m.f.)		Composition (m.f.)	
Benzene	0.25	Acetone	0.5
Toluene	0.25	Methanol	0.5
p-Xylene	0.25	Entrainer	
Bi-phenyl	0.25	DMSO	1
Pressure	101.325 kPa	Pressure	100 kPa
Thermal state	Saturated liquid	Thermal state	Saturated liquid
Thermodynamics	Peng Robinson	Thermodynamics	NRTL Hysys default
Specifications		Specifications	
Mole fraction of each pure component	>0.99	Mole fraction of each pure component	>0.9995

Table 2. Step by step iterations of example 1.

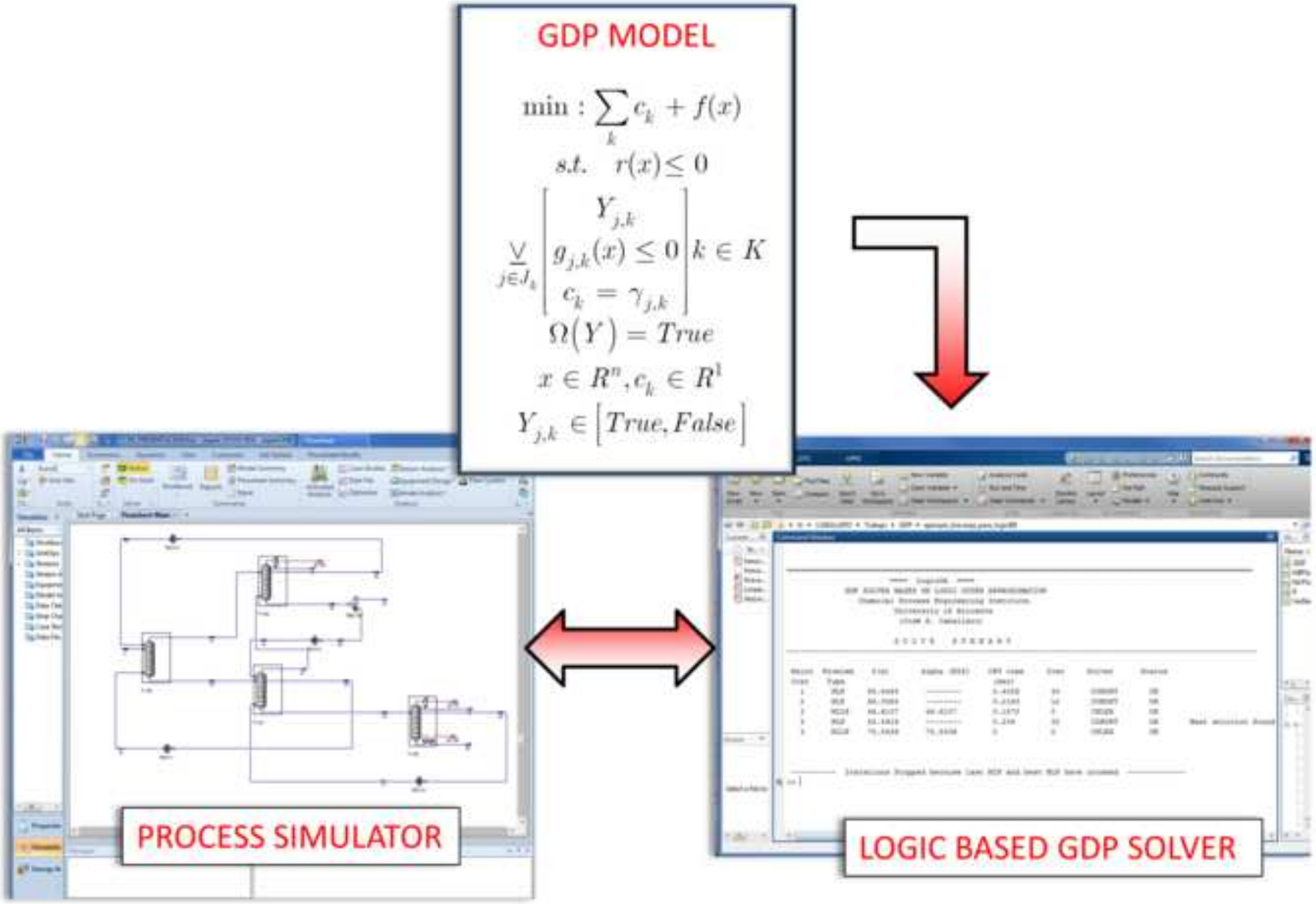
	Sub-problem Type	Nº of trays	Feed tray	Objective function	CPU time (s)++	Solver
Initialization						
1	NLP	6	4	4920.7	0.998	CONOPT
2	NLP	8	5	2857.8	0.374	CONOPT
3	NLP	10	6	2688.8	0.328	CONOPT
4	NLP	12	7	2739.4	0.265	CONOPT
5	NLP	14	8	2857.7	0.296	CONOPT
6	NLP	16	9	3006.0	0.218	CONOPT
7	NLP	18	10	3170.4	0.218	CONOPT
8	NLP	20	11	3344.5	0.312	CONOPT
9	NLP	22	12	3525.0	0.187	CONOPT
10	NLP	24	13	3709.8	0.176	CONOPT
Major Iterations						
1	*Master MILP*	----	----	*2625.8*	*0.078*	*CPLEX*
1	NLP	10	5	2930.4	0.296	CONOPT
2	*Master MILP*	----	----	*2650.3*	*0.280*	*CPLEX*
2	NLP	9	5	2862.1	0.280	CONOPT
3	*Master MILP*	----	----	*2657.7*	*0.062*	*CPLEX*
3	NLP	11	5	3019.7	0.218	CONOPT
4	*Master MILP*	----	----	*2714.6*	*0.078*	*CPLEX*
4	NLP	12	5	3115.9	0.203	CONOPT
5	*Master MILP*	----	----	*2785.0*	*0.156*	*CPLEX*
5	NLP	13	5	3214.6	0.687	CONOPT
6	*Master MILP*	----	----	*2863.4*	*0.093*	*CPLEX*
6	NLP	14	5	3314.1	0.328	CONOPT
7	*Master MILP*	----	----	*2896.3*	*0.094*	*CPLEX*
7	NLP	9	6	2635.8	0.468	CONOPT
8	*Master MILP*	----	----	*2947.1*	*0.062*	*CPLEX*
8	NLP	15	5	3414.0	0.468	CONOPT
9	*Master MILP*	----	----	*2971.4*	*0.109*	*CPLEX*
9	NLP	10	7	**2578.4****	0.250	CONOPT
10	*Master MILP*	----	----	*2998.0*	*0.156*	*CPLEX*
10	NLP	11	6	2773.0	0.234	CONOPT

** Optimal solution.

++Intel Core(TM)2 Quad CPU Q6600 @ 2.40GHz 2.39 GHz under Windows 7

Table 3. External iterations in example 4.

Iteration 1	Nº of trays in candidate sections	Nº of trays in optimal solution		
Column 1				
Section 1	3, 5, 7, 9, 11	5	DMSO Feed Tray	6
Section 2	19, 23, 27, 31, 35	31	Feed Tray	37
Section 3	14, 18, 22, 26, 30	18	Nº of total trays	56
Column 2				
Section 1	2, 6, 10, 14, 18	6	Feed Tray	7
Section 2	2, 6, 10, 14, 18	6	Nº of total trays	13
TAC ($\$10^6$/yr)	*4.827*			
Iteration 2	Nº of trays in candidate sections	Nº of trays in optimal solution		
Column 1				
Section 1	3, 4, 5, 6, 7	3	DMSO Feed Tray	4
Section 2	27, 29, 31, 33, 35	29	Feed Tray	33
Section 3	14, 16, 18, 20, 22	16	Nº of total trays	50
Column 2				
Section 1	2, 4, 6, 8, 10	6	Feed Tray	7
Section 2	2, 4, 6, 8, 10	6	Nº of total trays	13
TAC ($\$10^6$/yr)	*4.818*			
Iteration 3	Nº of trays in candidate sections	Nº of trays in optimal solution		
Column 1				
Section 1	1, 2, 3, 4, 5	3	DMSO Feed Tray	4
Section 2	27, 28, 29, 30, 31	29	Feed Tray	33
Section 3	14, 15, 16, 17, 18	16	Nº of total trays	50
Column 2				
Section 1	4, 5, 6, 7, 8	6	Feed Tray	7
Section 2	4, 5, 6, 7, 8	6	Nº of total trays	13
TAC ($\$10^6$/yr)	*4.818* (same as in iteration 2)			


GDP MODEL
$\min : \sum_k c_k + f(x)$
$s.t. \quad r(x) \leq 0$
$\underset{j \in J_k}{\vee} \begin{bmatrix} Y_{j,k} \\ g_{j,k}(x) \leq 0 \\ c_k = \gamma_{j,k} \end{bmatrix} k \in K$
$\Omega(Y) = True$
$x \in R^n, c_k \in R^1$
$Y_{j,k} \in [True, False]$
PROCESS SIMULATOR
LOGIC BASED GDP SOLVER

Figure(s)

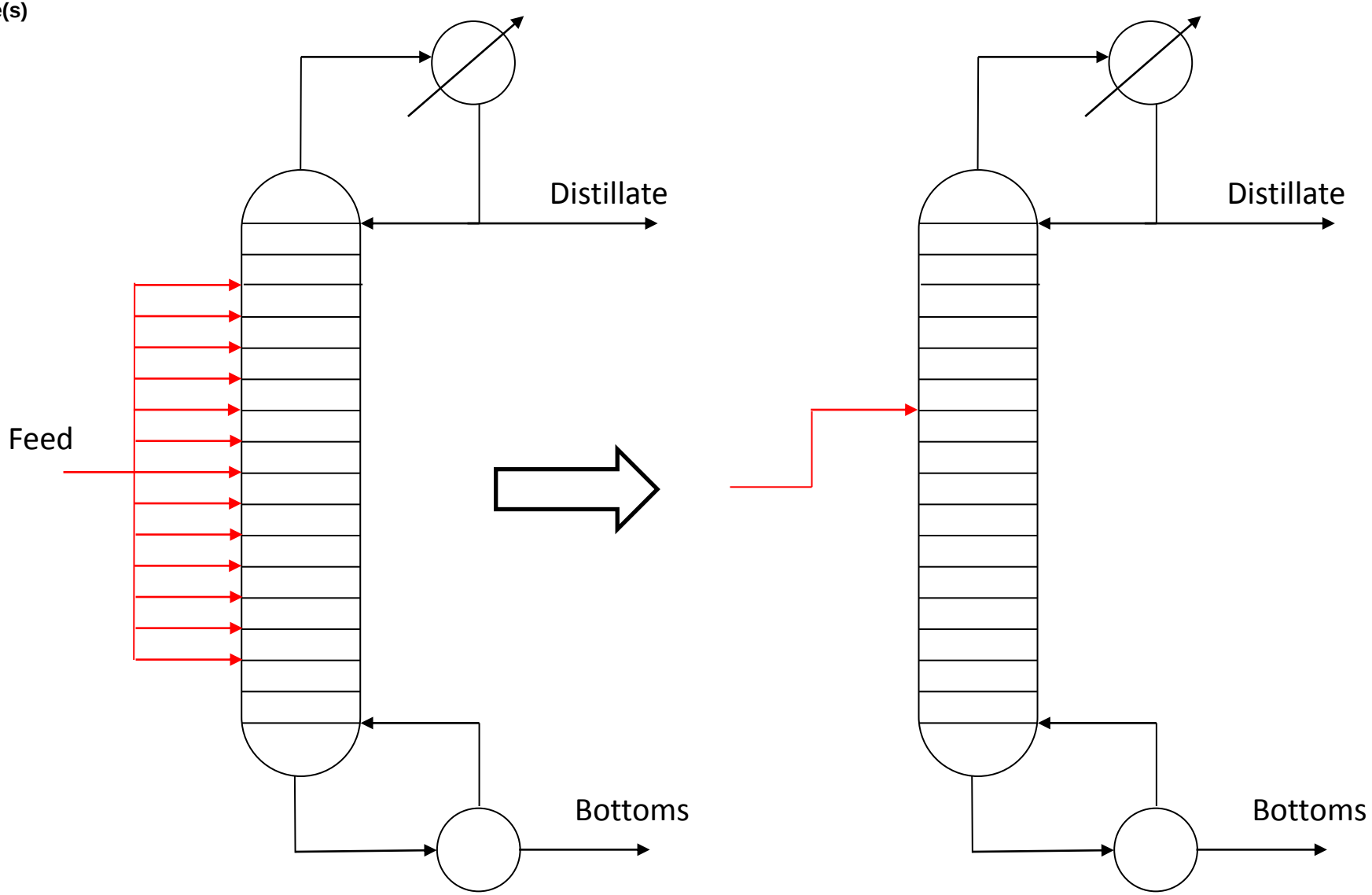


Figure 1

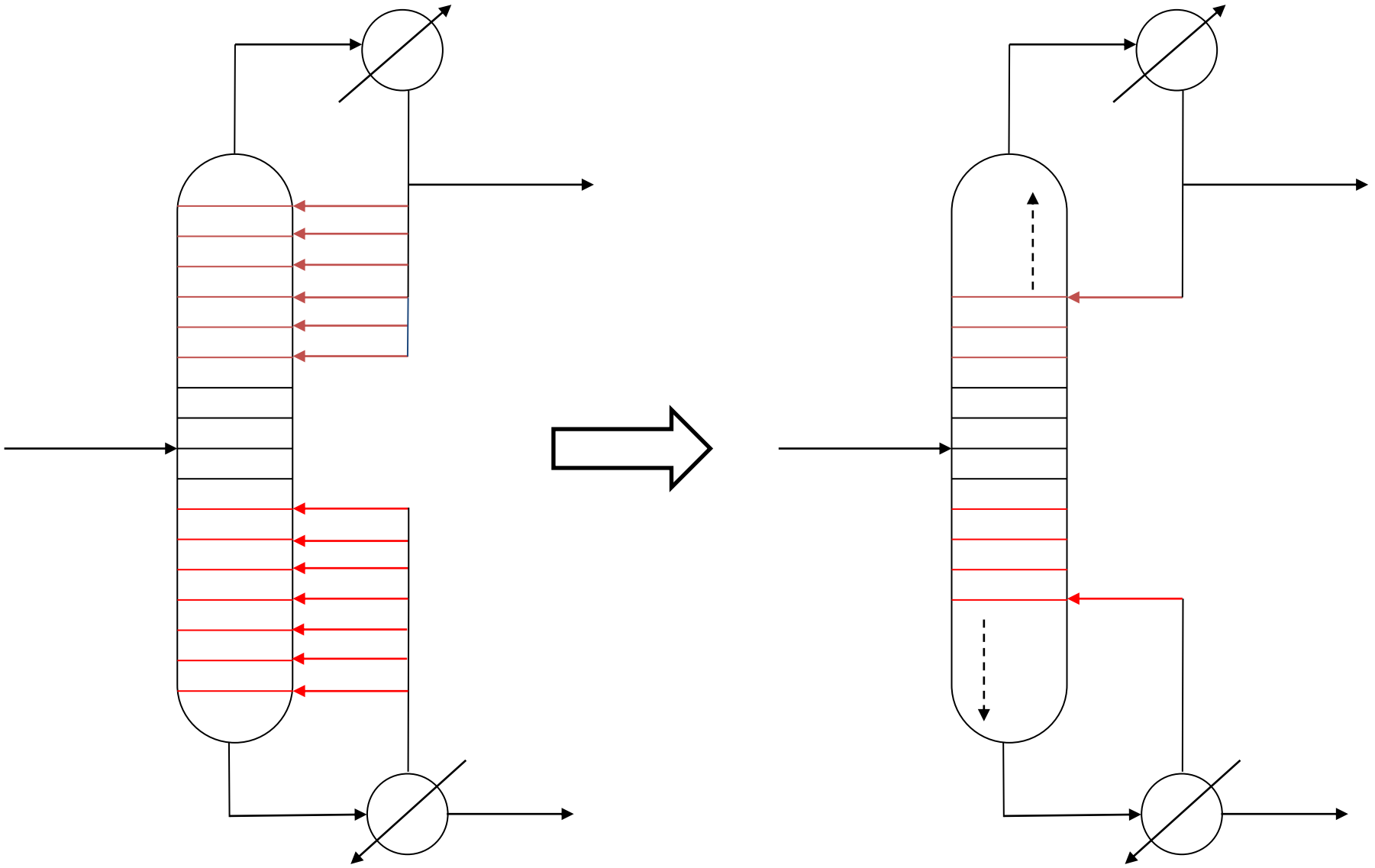


Figure 2

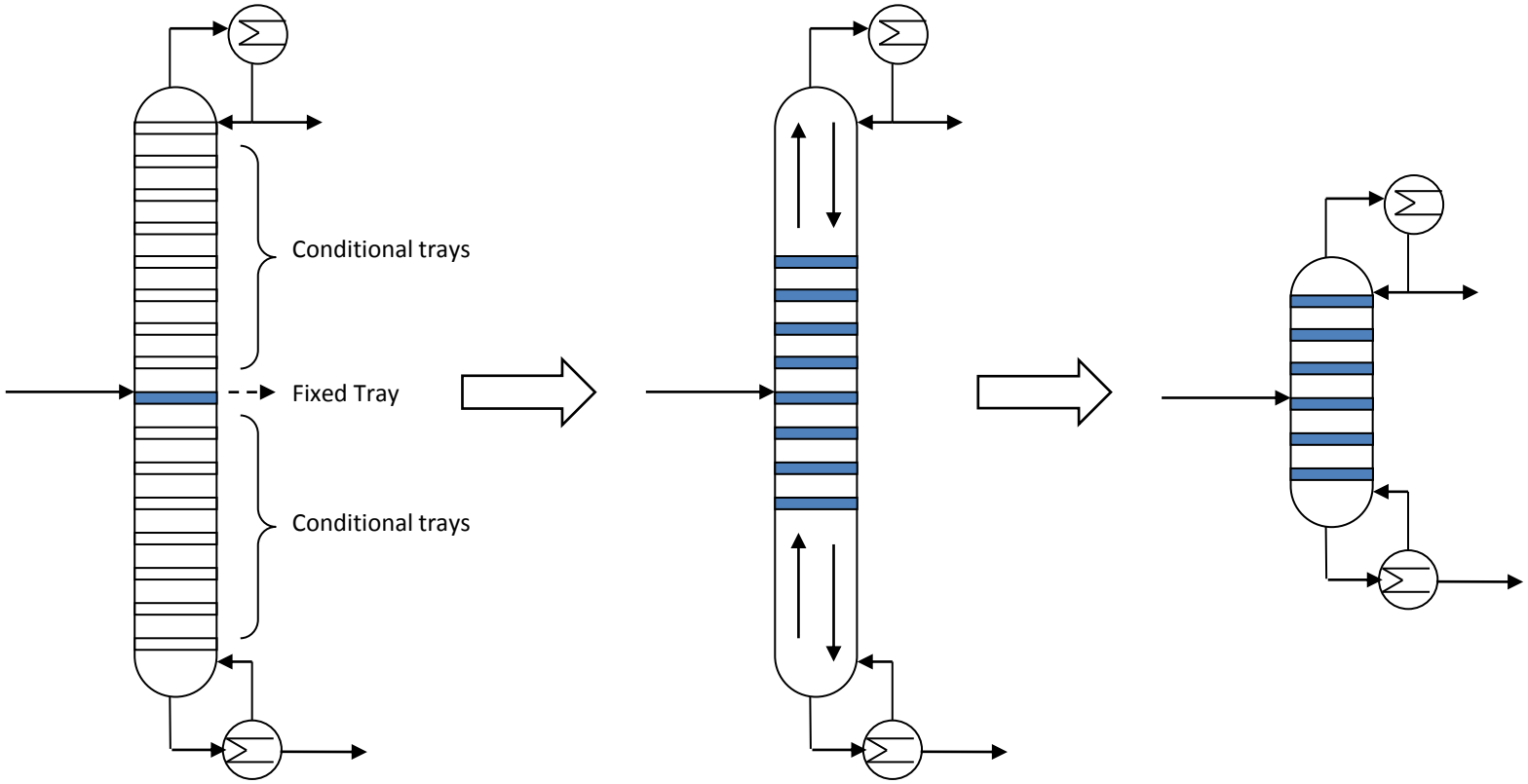


Figure 3

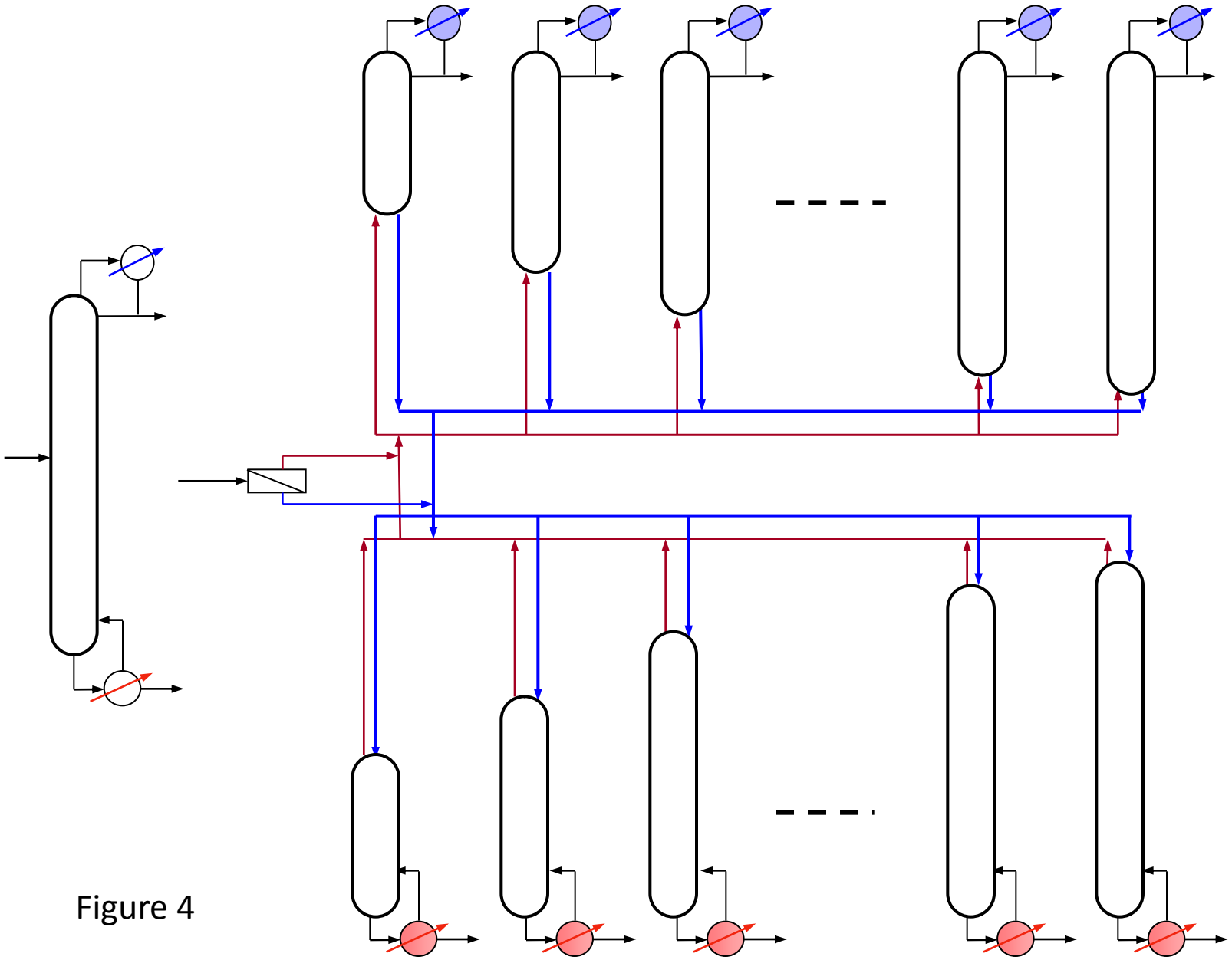


Figure 4

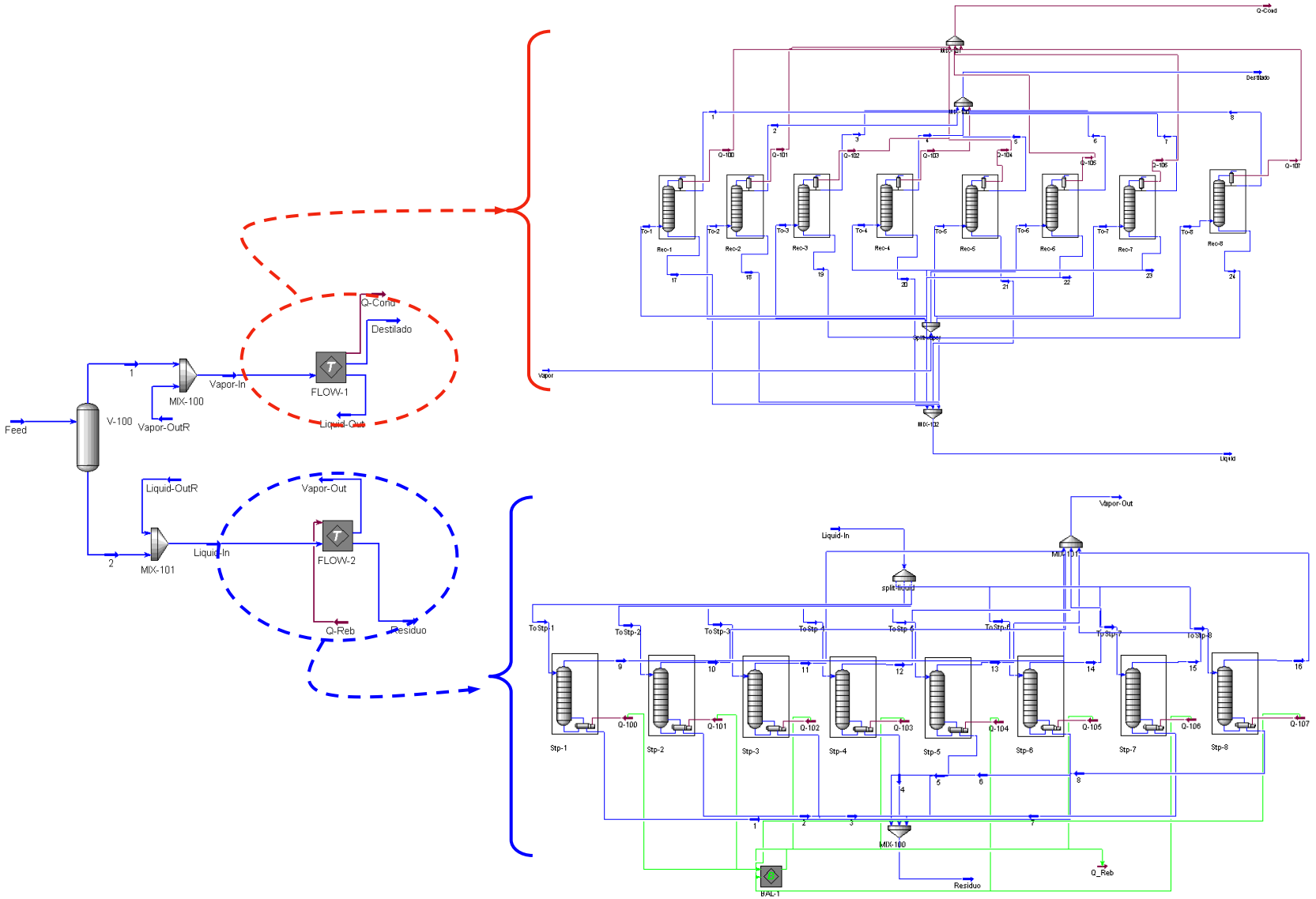

Q-Cond
Destilado
1
MIX-100
Vapor-In
FLOW-1
Feed
V-100
Vapor-OutR
Liquid-OutR
Vapor-Out
2
MIX-101
Liquid-In
FLOW-2
Q-Reb
Residuo
Vapor
Liquid
split-liquid
split-vapor
MIX-102
MIX-103
BAL-1
Stp-1
Stp-2
Stp-3
Stp-4
Stp-5
Stp-6
Stp-7
Stp-8
Rec-1
Rec-2
Rec-3
Rec-4
Rec-5
Rec-6
Rec-7
Rec-8
Q_Reb

Figure 5

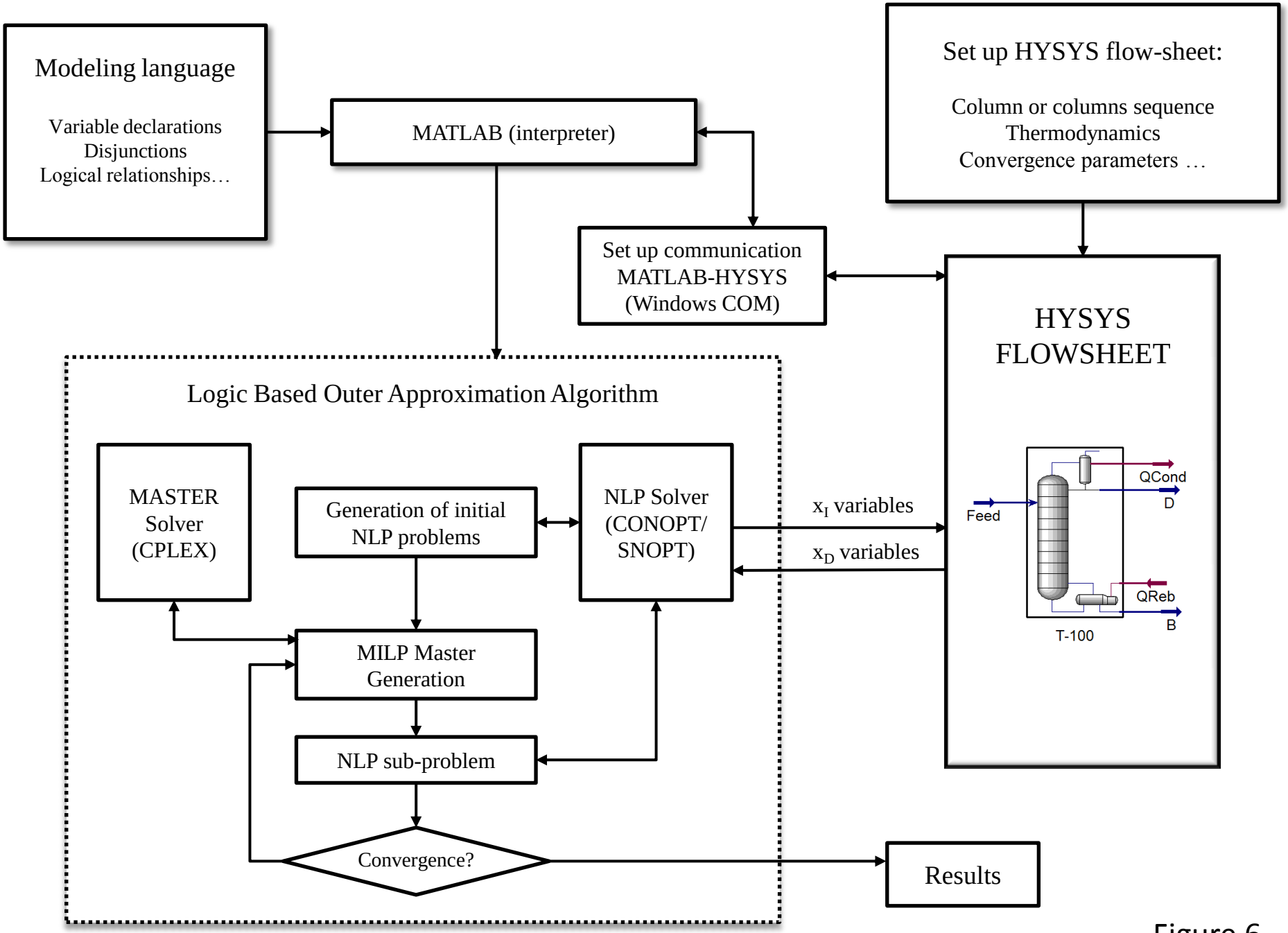
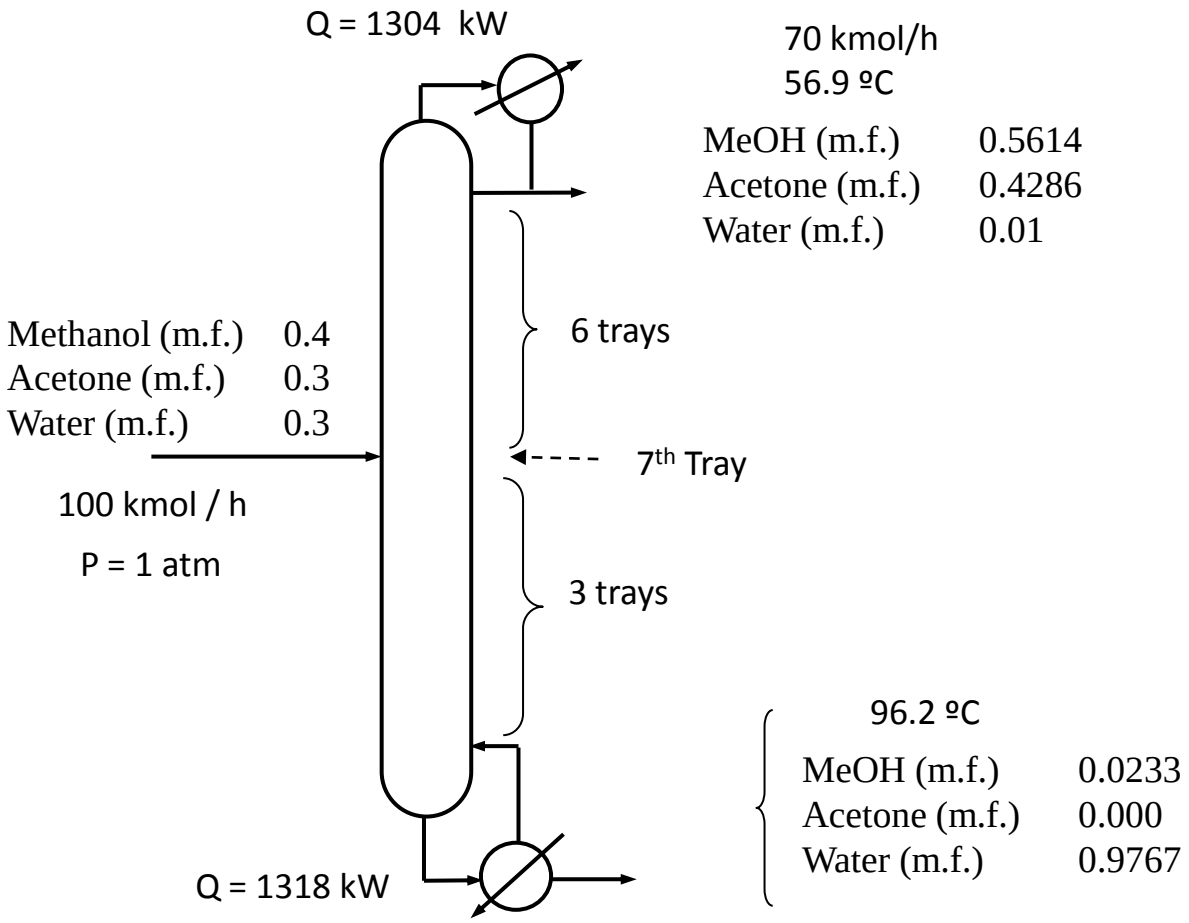

Modeling language
Variable declarations
Disjunctions
Logical relationships…
MATLAB (interpreter)
Set up HYSYS flow-sheet:
Column or columns sequence
Thermodynamics
Convergence parameters …
Set up communication
MATLAB-HYSYS
(Windows COM)
HYSYS
FLOWSHEET
Logic Based Outer Approximation Algorithm
MASTER
Solver
(CPLEX)
Generation of initial
NLP problems
NLP Solver
(CONOPT/
SNOPT)
x_I variables
x_D variables
MILP Master
Generation
NLP sub-problem
Convergence?
Results
Feed
QCond
D
QReb
B
T-100

Figure 6

$$\min : QReb(kW) + 0.2QCond(kW) + 100(N^{o}of\ Trays)$$



$$x_{MeOH} + x_{Acetone} \geq 0.99(mol\ fraction)$$
$$recovery(MeOH + Acetone) \geq 0.99$$

Figure 7

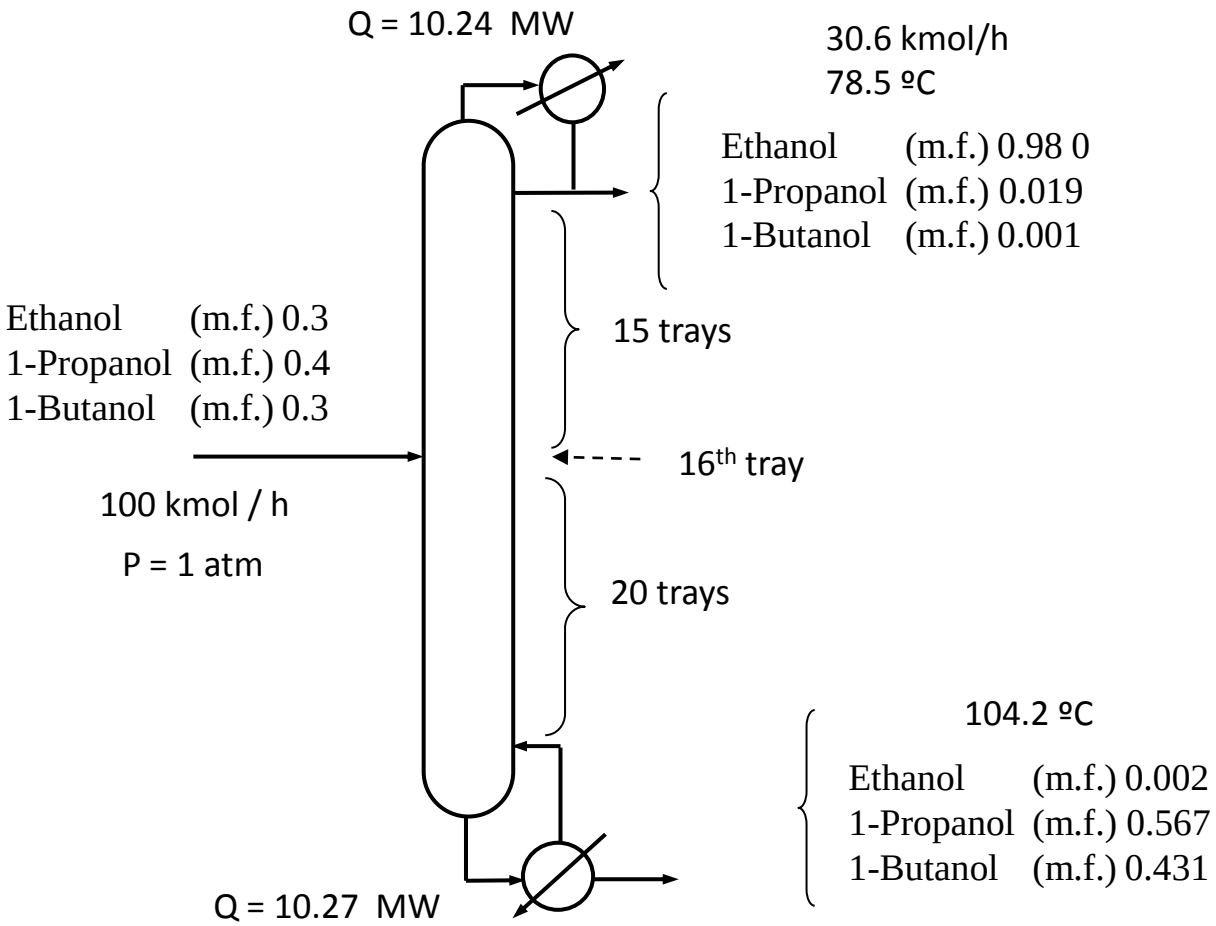

Q = 10.24 MW
30.6 kmol/h
78.5 ºC
Ethanol (m.f.) 0.98 0
1-Propanol (m.f.) 0.019
1-Butanol (m.f.) 0.001
Ethanol (m.f.) 0.3
1-Propanol (m.f.) 0.4
1-Butanol (m.f.) 0.3
15 trays
16th tray
100 kmol / h
P = 1 atm
20 trays
104.2 ºC
Ethanol (m.f.) 0.002
1-Propanol (m.f.) 0.567
1-Butanol (m.f.) 0.431
Q = 10.27 MW

Figure 8

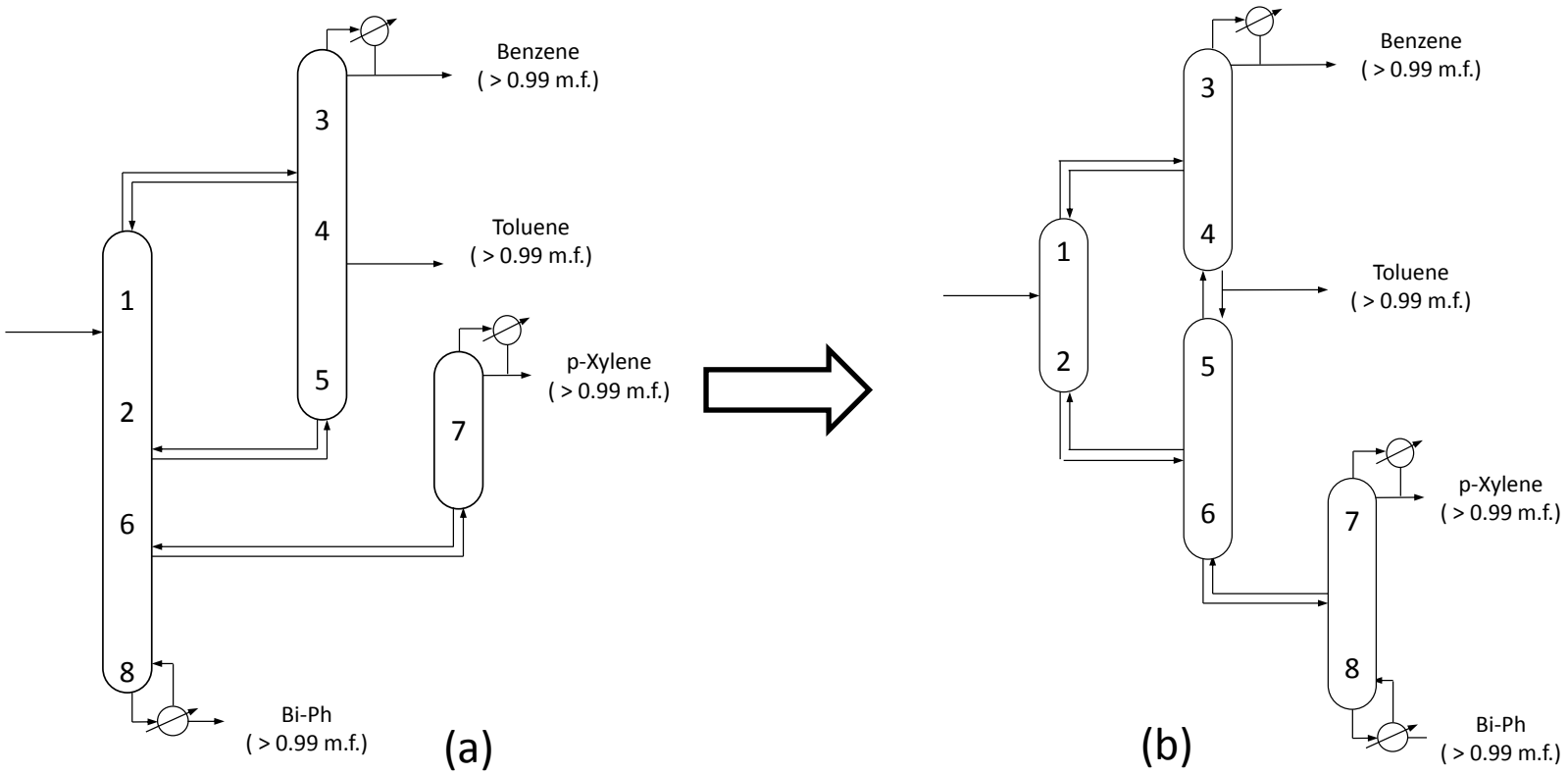
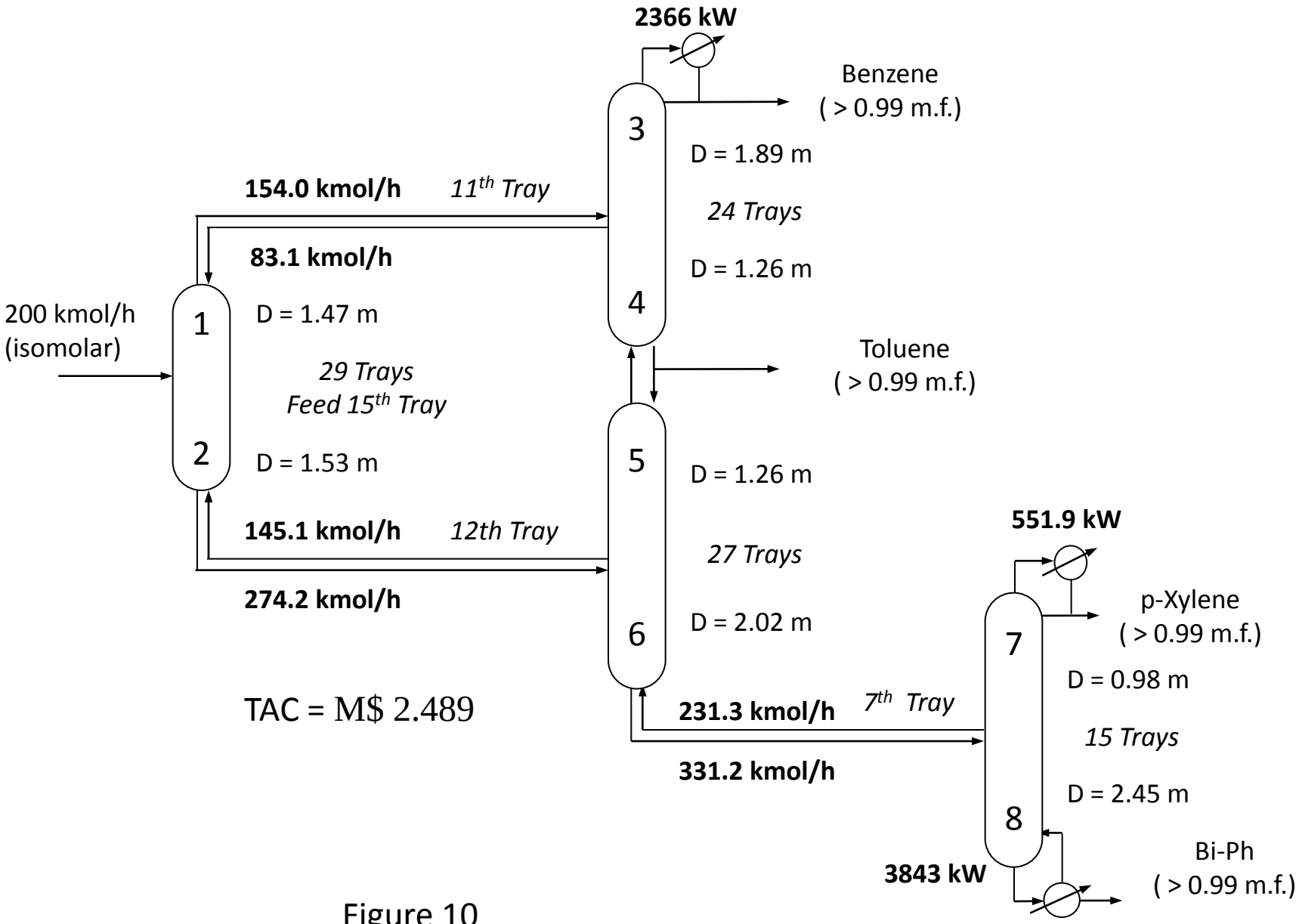

Benzene
(> 0.99 m.f.)
Toluene
(> 0.99 m.f.)
p-Xylene
(> 0.99 m.f.)
Bi-Ph
(> 0.99 m.f.)
1
2
3
4
5
6
7
8
(a)
Benzene
(> 0.99 m.f.)
Toluene
(> 0.99 m.f.)
p-Xylene
(> 0.99 m.f.)
Bi-Ph
(> 0.99 m.f.)
1
2
3
4
5
6
7
8
(b)

Figure 9



TAC = M$ 2.489

Figure 10

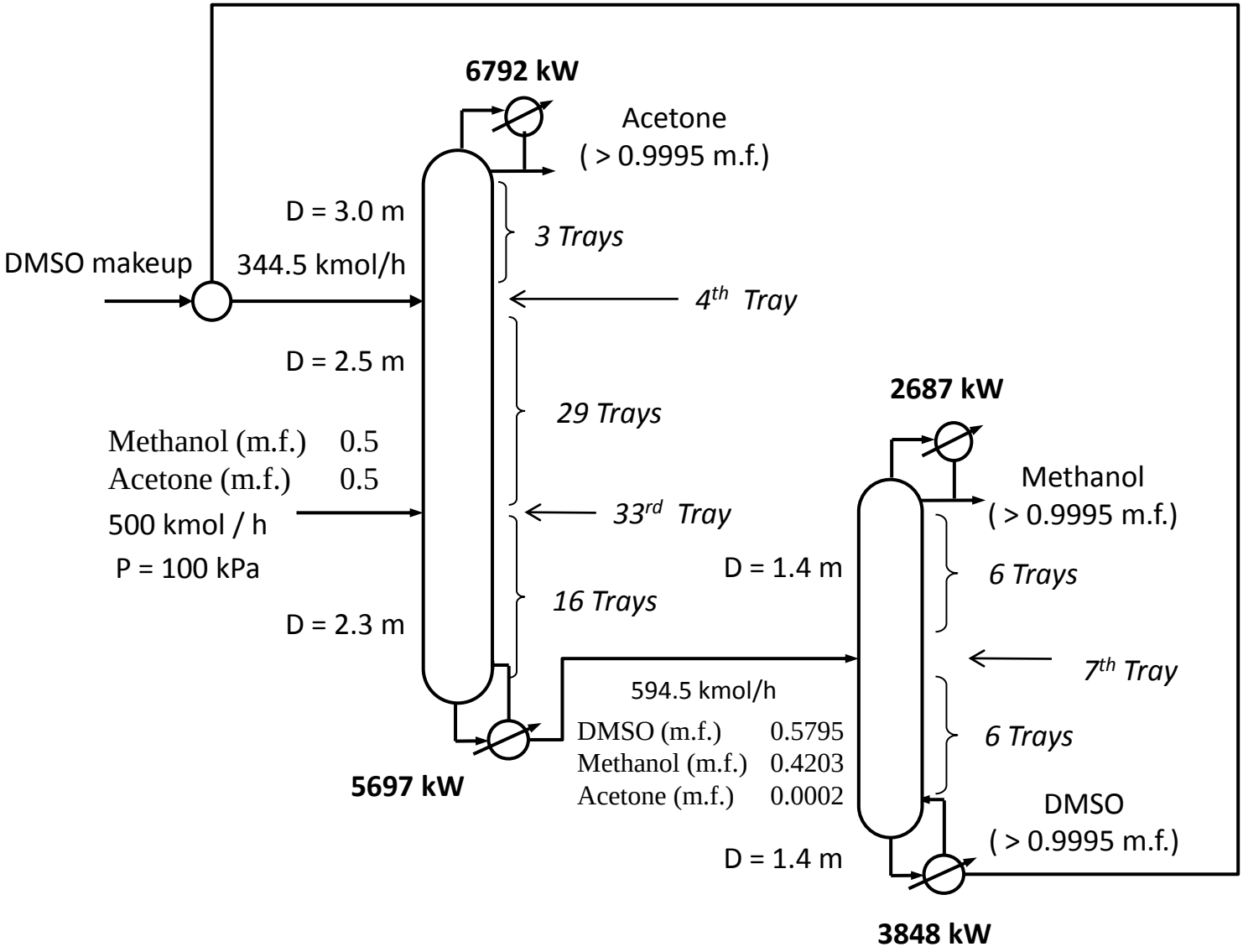
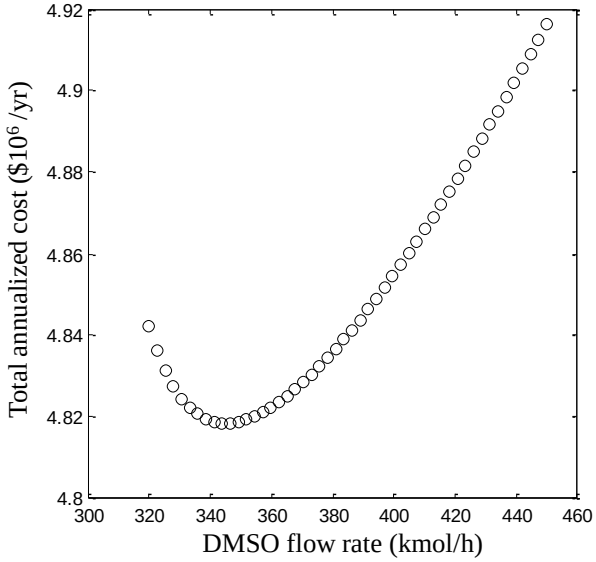
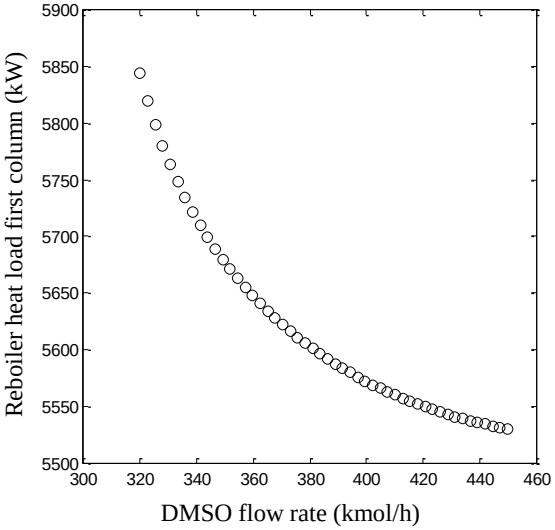
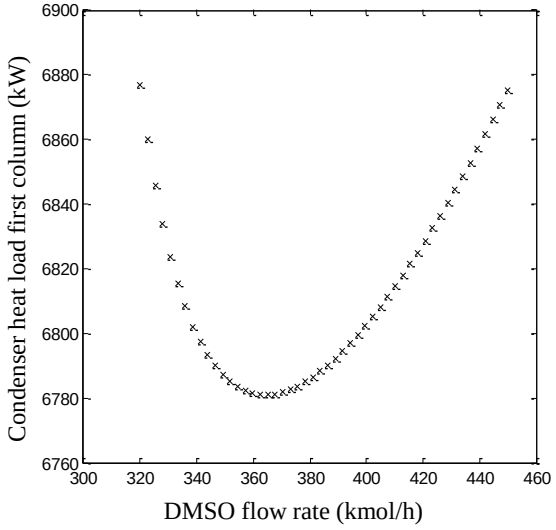

6792 kW
Acetone
(> 0.9995 m.f.)
D = 3.0 m
3 Trays
DMSO makeup
344.5 kmol/h
4th Tray
D = 2.5 m
29 Trays
2687 kW
Methanol (m.f.) 0.5
Acetone (m.f.) 0.5
500 kmol / h
P = 100 kPa
33rd Tray
Methanol
(> 0.9995 m.f.)
D = 1.4 m
6 Trays
16 Trays
D = 2.3 m
7th Tray
594.5 kmol/h
DMSO (m.f.) 0.5795
Methanol (m.f.) 0.4203
Acetone (m.f.) 0.0002
6 Trays
DMSO
(> 0.9995 m.f.)
D = 1.4 m
5697 kW
3848 kW

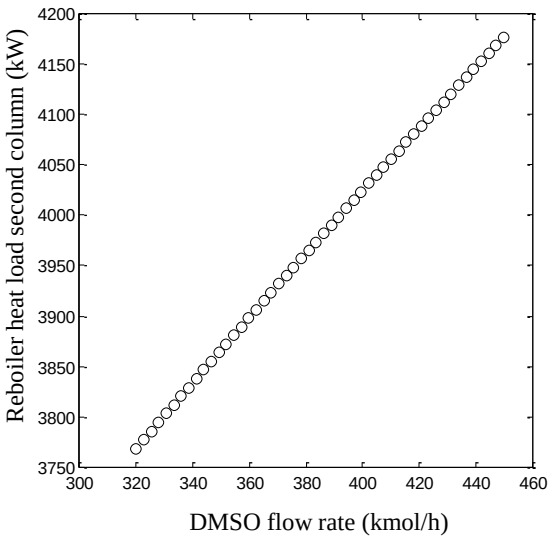
Figure 11

Figure(s)









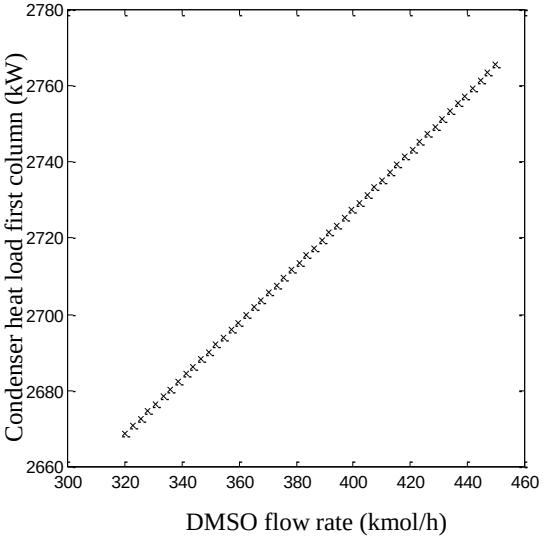


Figure 12