Optimization for the flexibility analysis of processes: Application to the acetone-ethanol-butanol producing process

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Abstract

The design and operation of sustainable biorefineries is an important subject of research since the current environmental context makes urgent the development of robust methodologies able to design innovative industries. In this context, flexibility of distillation sequences plays a very important role. The design is based on nominal production data may imply operation infeasibilities, unsustainable operation and an energy overconsumption. This work proposes a new methodology for the flexibility analysis of chemical processes based on rigorous thermodynamic models and optimizations tools with special emphasis on purification processes in the biorefining field.

Keywords: Nonlinear Programming, Optimization, Distillation sequences flexibility, Phase equilibrium, Biorefinery

1. Introduction

In the last decades, biomass has been identified as a potential renewable resource for producing biofuels, chemicals and other high value-added products using diverse processing technologies. However, the changing conditions within the energy sector, associated to variability of biomass quality have forced biorefineries to comply with new requirements. In this context, flexibility of distillation sequences to purify final product(s) plays a very important role, since the design of distillation sequences based on nominal production data may imply operation infeasibilities (e.g. failure to achieve desired product purity) and moreover, unsustainable operation and an energy overconsumption (Hoch et al., 1995). Swaney and Grossmann (1985) defined flexibility as the ability to operate over a range of conditions while satisfying performance specifications. In a biorefinery context, it is implied that flexibility-wise most concerned unit operations are distillation columns. Paules and Floudas (1992) had already studied the question of flexible distillation sequences, for example. However, distillation columns are normally modeled through shortcut methods, or aggregate methods (Kamath et al. (2010)), not taking into account rigorous evaluation of highly non-ideal mixture distillation sequences that demand rigorous thermodynamic models. For instance, biorefineries are usually involved in the production of fuels, which exhibit a strong nonlinear behavior in the liquid phase. In consequence, rigorous models have to be included in the design and simulation of these kinds of sequences.

Rigorous models based in numerical optimization have been demonstrated as a reliable methodology in order to optimally design and operate distillation columns and distillation sequences by extension. For instance, (Caballero and Grossmann, 2004) have successfully studied the synthesis of distillation sequences by using rigorous methods in a MINLP-modeling context. Nevertheless, it has to be noted that rigorous models of distillation columns exhibit a large degree of non-linearity, fact that makes the optimization procedure a non-trivial process. Moreover, in a biorefinery context, highly non-ideal phase equilibrium in the liquid phase the model non-linearity. Thus, in a MINLP-modeling context this issue that could limit the procurement of optimal solutions. Recently, Ramos et al. (2014) studied the optimal design and control of non-ideal extractive distillation columns using a rigorous equilibrium model and developed an efficient Mathematical Program with Complementary Constraints (MPCC)-based methodology in order to deal with the discrete decisions related to column design. This MPCC model is used to study the flexibility of a three distillation columns sequence for the production of bioethanol by maximizing/minimizing the biobutanol throughput, subject to column structural and hydraulics constraints as well as purity constraints. All models are solved with GAMS® using IPOPT as the nonlinear solver.

2. Mathematical programming based methodology

The mathematical programming-based methodology is composed of two parts: first, an MPCC model to design the columns and second, an NLP model, which studies the flexibility of the previously designed columns. Note that the columns were not designed to maximize flexibility, but to operate optimally at nominal conditions.

2.1. Nominal operation design through a MPCC model

For distillation column, we adapted MPCC dynamic model to steady-state operation presented by Ramos et al. (2014). The MPCC model has at its core an equilibrium-based model, so-called the MESH model (Mass, equilibrium, fraction summations and enthalpy equations). Additionally, it contains complementarity constraints, which model discrete decisions, i.e. the number of stages (modeled through the stage where the condenser reflux is fed) and the feed location for each column. Heat exchangers between columns energy balances are also part of the model. The assumptions taken into account on this research, to develop a steady-state model for the aforementioned distillation system, are the following: (i) Thermodynamic equilibrium at each stage, (ii) Adiabatic operation, (iii) Ideal vapor phase, (iv) NRTL model represents the behavior of the liquid phase, (v) Total condenser and partial reboiler; (vi) Residual thermodynamic properties are negligible.

Let *n* denote the maximum number of stages in a column and $NS = \{1,2,...n\}$ the corresponding set of the stages. Let *nc* denote the number of components in the system and $C = \{1,2,...nc\}$ denote the set of components. Let *nf* denote the number of feeds to the column and $FE = \{1,2,...nf\}$ denote the corresponding set of feeds. The subsets $REB = \{n\}, COND = \{1\} \in NS$ denote the reboiler (stage *n*) and the condenser (stage 1), respectively. Additionally, let $COL = \{2,3,...n-1\} \in NS$ denote the subset of stages between the condenser and the reboiler. The approach deals with finding the optimal number of trays in the column by determining the optimal location of the reflux.

To represent the steady-state behavior of the superstructure, the model includes the following algebraic equations for each one of the subsets in the column:

-Total mass balance:

$$0 = V_{j+1} + L_{j-1} - V_j - L_j + \sum_{f=1}^{nf} F_{j,f} + LR_j, \ \forall j \in COL \tag{1}$$

$$0 = V_{j+1} - D\left(1 + R_R\right), \forall j \in COND \tag{2}$$

$$0 = L_{j-1} - L_j - V_j, \forall j \in REB \tag{3}$$

-Partial mass balance:

$$0 = V_{j+1} y_{c,j+1} + L_{j-1} x_{c,j-1} - V_j y_{c,j} - L_j x_{c,j} + \sum_{f=1}^{nf} F_{j,f} z_{c,f} + LR_j x_{c,1}, \forall j \in COL, \ \forall c \in C \tag{4}$$

$$0 = V_{j+1} y_{c,j+1} - D\left(1 + R_R\right) x_{c,j}, \forall j \in COND, \forall c \in C \tag{5}$$

$$0 = L_{j-1} x_{c,j-1} - L_j x_{c,j} - V_j y_{c,j}, \forall j \in REB, \forall c \in C \tag{6}$$

-Thermodynamic equilibrium governing equations:

$$y_{c,j} = \beta_j K_{c,j} x_{c,j}, \forall j \in NS, \ \forall c \in C \tag{7}$$

$$1 - \beta_j = sl_j, \forall j \in COL \tag{8}$$

$$0 \le L_j \perp sl_j \ge 0, \ \forall j \in COL \tag{9}$$

Note that in the stages located above the stage where the reflux is fed, there is only vapor flow, and consequently, each stage model must include complementarities that allow for disappearance of the liquid phase (9). Thus, the equilibrium relation (7) is enforced only when liquid and vapor are present on the stage and relaxed when there is only vapor flow, modeled through the variable β_j. This formulation is derived as the reformulation of Gibbs free energy minimization on each tray *j*.

-Phase equilibrium error:

$$\sum_{c=1}^{nc} y_{c,j} - \sum_{c=1}^{nc} x_{c,j} = 0, \forall j \in NS \tag{10}$$

-Energy balance:

$$0 = V_{j+1} h_{j+1}^v + L_{j-1} h_{j-1}^l - V_{j-1} h_{j-1}^v - L_j h_j^l + \sum_{f=1}^{nf} F_{j,f} h_f + LR_j h_1^l, \forall j \in COL \tag{11}$$

$$0 = V_{j+1} h_{j+1}^v - D\left(1 + R_R\right) h_j^l - Q_c, \forall j \in COND \tag{12}$$

$$0 = L_{j-1} h_{j-1}^l - L_j h_j^l - V_j h_j^v + Q_R, \forall j \in REB \tag{13}$$

-Enthalpy definition:

$$h_{c,j}^v = \int_{T_0}^{T_j} Cp_c^{ig} dT, \forall j \in NS, \forall c \in C \tag{14}$$

$$h_{c,j}^l = h_{c,j}^v - h_{c,j}^{vap}, \forall j \in NS, \forall c \in C \tag{15}$$

$$h_j^l = \sum_{c=1}^{nc} h_{c,j}^l x_{c,j}, \forall j \in NS \tag{16}$$

$$h_j^v = \sum_{c=1}^{nc} h_{c,j}^v y_{c,j}, \forall j \in NS \tag{17}$$

It is important to remark that $L_1 = 0$ for the equations above, since the reflux rate is not necessarily entering in stage 2. For instance, the reflux rate feed (to a stage *j*) is being accounted with the term LR_j.

The additional equations are the following :

-*Constraints on the amount of reflux and their locations:*

$$LR_j = (R_R)(D)(yr_j), \forall j \in COL \tag{18}$$

$$\sum_{j=2}^{n-1} yr_j = 1 \tag{19}$$

-*Constraints on feeds and their locations:*

$$F_{j,f} = (yf_{j,f})(Feed_f), \forall j \in COL, \forall f \in FE \tag{20}$$

$$\sum_{j=2}^{n-1} yf_{j,f} = 1, \forall f \in FE \tag{21}$$

-*Logical relations between the locations of the feeds and the reflux (note that the feeds must be located in the same or below the stage where the reflux is located):*

$$yf_{j,f} - \sum_{j'=2}^{j} yr_{j'} \leq 0, \forall j \in COL, \forall f \in FE \tag{22}$$

-*If feed two (2), has to be located below feed one (1):*

$$yf_{j,2} - \sum_{j'=2}^{j} yf_{j',1} \leq 0, \forall j \in COL \tag{23}$$

In order to obtain a feasible design of the column, it should be inferred that the variables $yf_{j,f}, yr_j, \forall j \in COL, \forall f \in F$ must have binary values. As it was mentioned before, the traditional approach was to consider these variables as binary and solve the problem as a MINLP. Instead, in the present research, these variables are treated as continuous and the 0-1 decision is modelled with complementarity constraints, as follows:

-*Complementarity constraints for determining feed and reflux location*:

$$0 \leq (1 - yr_j) \perp sr_j^a \geq 0, \forall j \in COL \tag{24}$$

$$0 \leq yr_j \perp sr_j^b \geq 0, \forall j \in COL \tag{25}$$

$$sr_j^a + sr_j^b = 1, \forall j \in COL \tag{26}$$

$$0 \leq (1 - yf_{j,f}) \perp sf_{j,f}^a \geq 0, \forall j \in COL, \forall f \in FE \tag{27}$$

$$0 \leq yf_{j,f} \perp sf_{j,f}^b \geq 0, \forall j \in COL, \forall f \in FE \tag{28}$$

$$sf_{j,f}^a + sf_{j,f}^b = 1, \forall j \in COL, \forall f \in FE \tag{29}$$

24- 29 in combination with 19 and 21 ensure the unique possible values of the integer decision variables (0 or 1). It is also ensured that there is only one possible reflux and feed location (for each feed). The methodology to formulate the complementarities is not an easy task and a wrong formulation can lead to disjoint regions, and no solution can be obtained. To formulate the complementarities in such a way that the selection of the values of the slack variables is not conditioned by a disjoint region is important to follow the guidelines of (Baumrucker et al., 2008).

2.2. MPCC solution strategy

MPCCs introduce an inherent non-convexity in the model as well as linear dependence of constraints, which make the non-linear program very hard to solve. It is important to note that the solution of a MPCC is always a local solution due to non-convexities, and

the modeler should be satisfied with this kind of solution. To solve a MPCC, a reformulation of the problem has to be made to efficiently solve it with a standard NLP solver. Among the MPCC reformulations to allow standard NLP tools to be applied, the most efficient and robust according to is PF (ρ), *penalty* formulation. In the penalty formulation the complementarities are moved to the objective function and the resulting problem is solved for a particular value of ρ or by a series of problems with increasing ρ. In the present research, the PF is solved with $\rho = 10^5$. In this way, the solution of the original MPCC is achieved and the complementarities are satisfied within the tolerance (10^{-6}). Baumrucker et al., (2008) report a full comparison between the MPCC reformulations to NLPs. Their conclusion is that the more reliable way to reformulate MPCC into NLPs is the PF formulation, because: (i) the complementarities are not included as constraints but only in the objective function, maintaining the problem size despite the complementarities and (ii) the problem can be solved a single time instead of multiple times, if a good value of ρ is chosen.

2.3. Flexibility analysis methodology

The flexibility analysis consists on determining the optimal feed and its composition when maximizing and minimizing independently the production, subjected to structural constraints. These constraints enforce that in either cases, the nominal operating conditions are respected within certain bounds. The latter ensures that the modified operating conditions with the columns nominal design, will not lead to real-world infeasible operation. For instance, these constraints consist on:

$$\alpha \bar{V}_{sect}^{nom} \le \bar{V}_{sect} \le \beta \bar{V}_{sect}^{nom}, \ \forall sect \in \{\text{rect, strip}\} \tag{30}$$

$$\alpha \bar{L}_{sect}^{nom} \le \bar{L}_{sect} \le \beta \bar{L}_{sect}^{nom}, \ \forall sect \in \{\text{rect, strip}\} \tag{31}$$

$$\alpha \bar{ff}_{sect}^{nom} \le \bar{ff}_{sect} \le \beta \bar{ff}_{sect}^{nom}, \ \forall sect \in \{\text{rect, strip}\} \tag{32}$$

In 30- 32, $\alpha \le 1, \beta \ge 1$ are scalars to bound the average vapor flow ($\bar{V}_{sect}$), liquid flow ($\bar{L}_{sect}$) and flood factor ($\bar{ff}_{sect}$) for the rectifying and stripping section of the columns, between the averages of the same values when the distillation sequence is operating at nominal process conditions. As such, vapor and liquid flows, and flood factor can vary between the latter bounds in order to evaluate flexibility of the designed columns. The flood factor is calculated as follows:

$$ff_i = \frac{L_i}{V_i}\sqrt{\frac{\rho_i^{vap}}{\rho_i^{liq}}}, \forall i \in COL \tag{33}$$

3. Case study

The case study consists on a quaternary-component distillation sequence in order to separate the system water (W)-acetone (A)-ethanol (E)-butanol (B) (S. Belletante et al., 2016).

The solution of the optimization problem yields the optimal design of the distillation sequence, which maximizes butanol production under nominal operating conditions, shown in Table 1.

Table 1. Nominal operating parameters, Minimum and maximum production feed conditions comparison.

Parameter (unit)	Nominal Case	Minimum production	Maximum production
Feed (kmol/hr)	323.048	290.74	371.505
z (%mol)			
Water	13.9	12.51	12.51
Acetone	13.5	15.53	12.15
Ethanol	4.3	4.94	3.87
Butanol	68.3	67.02	71.47

The flexibility analysis consists on solving the NLPs, to determine the feed composition and flowrate, which minimize and maximize the amount of butanol that can be produced, given that the number of stages and the feed location of the columns have been fixed on their optimal value for nominal operation. Note that results obtained in both cases (i.e. the design and the flexibility) correspond to local optima, since both models involve a big degree of non-convexity and local solvers are used.

4. Conclusion

In support of a framework for biorefinery flexibility, we have presented a generic methodology to optimize a distillation sequence with a rigorous thermodynamics approach. Using this methodology, we can determine, the range of the distillation sequence, but it is necessary to combine this approach with a dynamic approach in order to appreciate the controllability of the process.

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