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R-graph-based distillation column superstructure and MINLP model

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Abstract

A new model has been developed for distillation column synthesis and optimization based on R-graph represented superstructure. A GDP model is generated first; this is transformed into an MINLP model which represents only the considered structures, and uses minimum number of binary variables to make distinctions between the different structures. The MINLP model is compared to literature models (Viswanathan and Grossmann, 1993, and Yeomans and Grossmann, 2000). The new model uses less number of binary variables, and is significantly faster.

Keywords: MINLP, distillation, superstructure, process synthesis

1. Introduction

Synthesis of optimal process structures including distillation columns involves optimal design of each column as part of the structure. Designing of a column as part of process or even in itself is a synthesis task characterised with both continuous and integer variables. The design and the optimization of a column can be executed in one step, simultaneously optimizing all the design and operation parameters, with structure oriented process synthesis. Here first a superstructure which includes all the considered structures is generated. Then a mathematical model of the superstructure, usually a Mixed Integer NonLinear Programming (MINLP) problem, is formulated. Finally, this mathematical model is optimized.

Two MINLP models have already been published for distillation column synthesis: Viswanathan and Grossmann (1993), and Yeomans and Grossmann (2000). In the latter paper a Generalized Disjunctive Programming (GDP) model is given but an MINLP model is applied in the solution procedure. In these papers the superstructure contains by-pass streams or multifunctional units in order to maintain the possibility of representing the distillation column with different stage numbers. How easy the solution of an MINLP model of a superstructure is has usually been analyzed according to the

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shape (linearity, convexity, relaxation, scaling) of the equations, see e.g. Grossmann (1996). If these characteristics do not deteriorate, it is worth to decrease the number of binary variables in the model.
Use of supergraphs instead of process networks decreases structural multiplicity. For example, Friedler et al, 1992 defined P-graphs for synthesizing structures. For representing structures, here we apply R-graphs (Rev et al, 2004), which are extended bipartite graphs in mathematical sense. The nodes are identified as the input and output ports of the units; the edges are identified as the streams always connecting output ports to input ports. Additionally, Farkas et al. (2004) defined the binarily minimal MINLP representation as that which applies a minimum number of binary variables used to make distinction between structures. In the case of *k* different structures, the smallest whole number *n* of binary variables has to be used that satisfies $n \geq \log_2 k$.
A new superstructure and MINLP model for distillation column synthesis are presented in our pape. The model uses minimum number of binary variables in each column section for representing the different structures. R-graph superstructure representation is applied for generating the MINLP model.

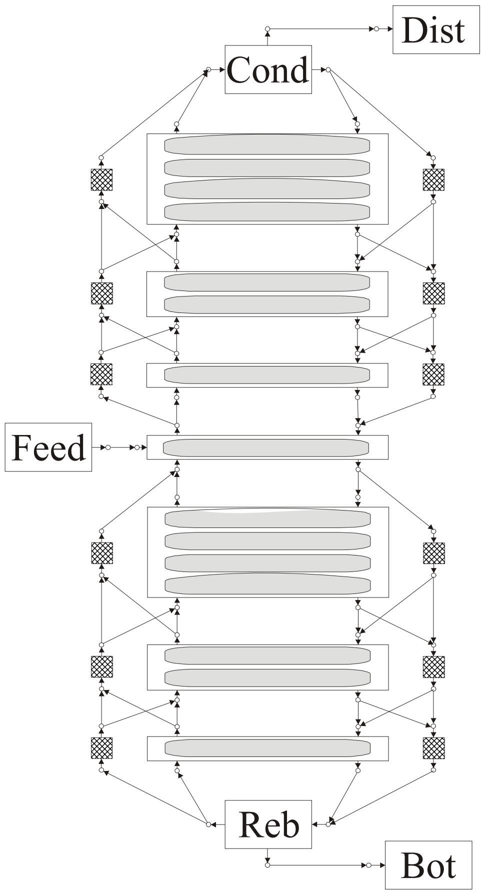

Figure 1. R-graph representation of the superstructure

2. Superstructure and R-graph representation

The R-graph superstructure of a conventional distillation column containing maximum 15 equilibrium trays is shown in Figure 1. The equilibrium stages are represented by shaded ovals, and the units by rectangles. The small circles represent input or output unit ports, connected by stream edges. Feed (source unit), Dist and Bot (sink units), Cond (condenser), Reb (boiler), and the feed stage are permanent units; all the others are conditional. The number of the equlibrium stages in the k^{th} unit of a column section (calculated bottom up) is 2^{k-1}. This arrangement is applied in order to facilitate forming binarily minimal ideal MINLP representation, as explained in section 4.
There are a vapor transport unit and a liquid transport unit beside each conditional unit that contains equlibrium stages. A stage-containing conditional unit and the transport units on its sides are called conjugate to each other. With superstructure any conditional stage-containing unit can be partially or perfectly by-passed. In the MINLP model formulation we will constraint the search space for structures where each stage-containing unit is either perfectly by-passed (i.e. omitted) or not by-passed at all.

3. Basic GDP Representation

The Basic GDP Representation (BGR, see Farkas et al., 2004) was generated according to the R-supergraph (R-graph of the superstructure). It contains the unit relations, the balances of the ports, and the objective function. No additional relations are added to the GDP model, therefore it represents all the feasible sub-R-graphs of the R-supergraph, and it uses distinct logical variables for each conditional units.
The unit relations of Feed, Bot, and Dist contain only the trivial positiveness constraints. The unit relations of Reb and Cond include the heat and component balances. The purity and recovery constraints are specified here, and the variable cost of the column are also calculated here.
Unit relations of the units containing equilibrium trays include the mass and heat balances, the calculation of flow rates and molar enthalpies from the concentrations, and the equations of chemical equilibrium. The fix cost of the feed tray is also determined.
The full list of the modelling equations is not presented here because of the lack of space; only examples for the unit relations of the conditional units are detailed. The following set of definitions are used. S: set of column sections, K: set of conditional units in a column section, I: set of components, Jn: set of equilibrium trays in a unit containing n trays. Jn^{first}, Jn^{last}, and Jn^{inner} are subsets of Jn, containing the first, the last, and the inner trays of the unit, respectively.
Logical variables $Z^{U}_{s,k}$, $Z^{V}_{s,k}$, and $Z^{L}_{s,k}$ are applied to represent the existence of the conditional tray containing units and existence their conjugate vapor and liquid transport units, respectively ($s \in S$, $k \in K$).
Equation 1 is applied to the unit relations of a unit containing 4 equilibrium trays. This unit is the third in a section; thus, k=3 ($k \in K$). In this equation V is molar vapor flowrate; L is molar liquid flowrate; VAP is total vapor flowrate; LIQ is total liquid flowrate; hV is molar vapor enthalpy; hL is molar liquid enthalpy; x is liquid mole fraction; y is vapor mole fraction; T is temperature; f is fugacity; P is pressure; c is cost

of the unit; DC is column diameter. Superscripts *in* and *out* denote the input and output variables of the units; superscrits V and L refer to the vapor and liquid flows.

$$
\left(\begin{bmatrix}
Z_{s,3}^{U} \\
\wedge \\
V_i^{in,s,3} + L_{i,j+1}^{s,3} = V_{i,j}^{s,3} + L_{i,j}^{s,3} \quad j \in J4^{first} \\
V_{i,j-1}^{s,3} + L_{i,j+1}^{s,3} = V_{i,j}^{s,3} + L_{i,j}^{s,3} \quad j \in J4^{inner} \\
V_{i,j-1}^{s,3} + L_i^{in,s,3} = V_{i,j}^{s,3} + L_{i,j}^{s,3} \quad j \in J4^{last} \\
L_i^{out,s,3} = L_{i,j}^{s,3} \quad j \in J4^{first} \\
V_i^{out,s,3} = V_{i,j}^{s,3} \quad j \in J4^{last} \\
L_{i,j}^{s,3} = LIQ_j^{s,3} \cdot x_{i,j}^{s,3} \quad j \in J4 \\
V_{i,j}^{s,3} = VAP_j^{s,3} \cdot y_{i,j}^{s,3} \quad j \in J4 \\
V_i^{in,s,3} \cdot hV_i^{in,s,3} + L_{i,j+1}^{s,3} \cdot hL_{i,j+1}^{s,3} \\
= V_{i,j}^{s,3} \cdot hV_{i,j}^{s,3} + L_{i,j}^{s,3} \cdot hL_{i,j}^{s,3} \quad j \in J4^{first} \\
V_{i,j-1}^{s,3} \cdot hV_{i,j-1}^{s,3} + L_{i,j+1}^{s,3} \cdot hL_{i,j+1}^{s,3} \\
= V_{i,j}^{s,3} \cdot hV_{i,j}^{s,3} + L_{i,j}^{s,3} \cdot hL_{i,j}^{s,3} \quad j \in J4^{inner}
\end{bmatrix}
\wedge
\begin{bmatrix}
V_{i,j-1}^{s,3} \cdot hV_{i,j-1}^{s,3} + L_i^{in,s,3} \cdot hL_i^{in,s,3} \\
= V_{i,j}^{s,3} \cdot hV_{i,j}^{s,3} + L_{i,j}^{s,3} \cdot hL_{i,j}^{s,3} \quad j \in J4^{last} \\
hL_{i,j}^{s,3} = f\left(T_j^{s,3}\right) \quad j \in J4 \\
hV_{i,j}^{s,3} = f\left(T_j^{s,3}\right) \quad j \in J4 \\
hL_i^{out,s,3} = hL_{i,j}^{s,3} \quad j \in J4^{first} \\
hV_i^{out,s,3} = hV_{i,j}^{s,3} \quad j \in J4^{last} \\
\sum_{i \in I} x_{i,j}^{s,3} = 1 \quad j \in J4 \\
\sum_{i \in I} y_{i,j}^{s,3} = 1 \quad j \in J4 \\
f_{i,j}^{L,s,3} = f_{i,j}^{V,s,3} \quad j \in J4 \\
f_{i,j}^{L,s,3} = f\left(x_{i,j}^{s,3}, T_j^{s,3}, P_j^{s,3}\right) \quad j \in J4 \\
f_{i,j}^{V,s,3} = f\left(x_{i,j}^{s,3}, T_j^{s,3}, P_j^{s,3}\right) \quad j \in J4 \\
c^{s,3} = f\left(DC\right) \quad ;\text{for all}: \quad i \in I,\ s \in S
\end{bmatrix}\right)
\vee
\left(\begin{bmatrix}
\neg Z_{s,3}^{U} \quad s \in S \\
\wedge \\
V_i^{in,s,3} = 0 \quad s \in S, i \in I \\
L_i^{in,s,3} = 0 \quad s \in S, i \in I \\
L_i^{out,s,3} = 0 \quad s \in S, i \in I \\
V_i^{out,s,3} = 0 \quad s \in S, i \in I \\
L_{i,j}^{s,3} = 0 \quad s \in S, i \in I, j \in J4 \\
V_{i,j}^{s,3} = 0 \quad s \in S, i \in I, j \in J4
\end{bmatrix}
\wedge
\begin{bmatrix}
LIQ_j^{s,3} = 0 \quad s \in S, j \in J4 \\
VAP_j^{s,3} = 0 \quad s \in S, j \in J4 \\
hL_{i,j}^{s,3} = 0 \quad s \in S, i \in I, j \in J4 \\
hV_{i,j}^{s,3} = 0 \quad s \in S, i \in I, j \in J4 \\
hL_i^{out,s,3} = 0 \quad s \in S, i \in I \\
hV_i^{out,s,3} = 0 \quad s \in S, i \in I \\
c^{s,3} = 0 \quad s \in S
\end{bmatrix}\right) \tag{1}
$$

Equation 2 is applied to a vapor transport unit:

$$
\begin{bmatrix}
Z_{s,k}^{V} \quad s \in S, k \in K \\
\wedge \\
tV_i^{in} = tV_i^{out} \quad i \in I
\end{bmatrix}
\vee
\begin{bmatrix}
\neg Z_{s,k}^{V} \quad s \in S, k \in K \\
\wedge \\
tV_i^{in} = 0 \quad i \in I \\
tV_i^{out} = 0 \quad i \in I
\end{bmatrix} \tag{2}
$$

where tV is the molar vapor flowrate in the transport units.

4. MINLP Representations

The Basic MINLP Representation (BMR) can automatically be generated from the BGR (given in section 3) by substituting the logical variables with binary ones (Farkas et al., 2004). BMR uses distinct binary variables for all the conditional units, and represents all the sub-R-graph of the R-supergraph. Thus, all the feasible structures, including those with partial by-pass, are respresented.

Our intention here, however, is that either the conditional unit or its conjugate transport units should be included in the solution, but not both. This can be achieved by inserting a logical constraint:

$$Z^U_{s,k} \oplus \left(Z^V_{s,k} \wedge Z^L_{s,k}\right) \qquad s \in S, k \in K \tag{3}$$

This logical constraint can also be formulated with binary variables $z^U_{s,k}$, $z^V_{s,k}$, and $z^L_{s,k}$:

$$z^U_{s,k} + z^V_{s,k} = 1 \qquad s \in S, k \in K \tag{4}$$

$$z^U_{s,k} + z^L_{s,k} = 1 \qquad s \in S, k \in K \tag{5}$$

Adding these constraints to the BMR, it represents the considered graphs only; i.e. it becomes an Ideal MINLP Reprepresentation (IMR).

Solvability of the model can be enhanced by decreasing the number of binary variables. According to the logic expressed by Equation 3, the binary variables $z^V_{s,k}$, and $z^L_{s,k}$ are redundant if binary variables $z^U_{s,k}$ is already in use; (1- $z^U_{s,k}$) can be substituted for them.

Farkas et al. (2004) defined the Binarily Minimal MINLP Representation (BMMR) as that which applies a minimum number of binary variables applied to make distinction between structures. In the case of *k* different structures, the smallest whole number *n* of binary variables has to be used that satisfies $n \geq \log_2 k$. The k^{th} conditional unit contains 2^k equilibrium trays in our superstructure (Figure 1); each such unit occupy 1 binary variable, and only these binary variables are used in our model. If, for example, the number of stages in a column section is constrained to 7 then 8 different alternative sub-structures, including an empty section, should be distinguished. In this case, 3 binary variables are used in BMMR, because $3=\log_2 8$. Thus, our representation is ideal and binarily minimal in the same time (BMIMR).

Some redundant variables are also discarded by taking into account component balances around each transport unit, so that not each stream variables has to be dealt with.

5. Example

The new model (BMIMR) is compared to two models from the literature (Viswanathan and Grossmann, 1993; Yeomans and Grossmann, 2000) on a two-component separation example.

The task is separating 100 kmol/h equimolar benzene / toluene system into pure products (0.98 minimum purity in the distillate and also in the bottom product).

Constant molar overflow and equilibrium stages are assumed. The hold-up on the stages, and the pressure drop in the column, are neglected. The system is considered as ideal mixture; the vapor-liquid equilibrium is calculated according to the Raoult-Dalton equation; pure component vapor pressure is calculated with Antoine equation.

The objective of Luyben and Floudas (1994) divided by 1000 for scaling was applied:

$$Cost = \frac{\beta_{tax}(C_{LPS}\Delta H_{vap} + C_{CW}\Delta H_{cond})V + \dfrac{12.3[615 + 324D^2 + 486(6 + 0.76N)D] + 245N(0.7 + 1.5D^2}{\beta_{pay}}}{1000} \quad (6)$$

β_{tax} is tax factor; C_{LPS} is cost of LP steam; C_{CW} is CW cost; ΔH_{vap} and ΔH_{cond} are latent heat; V is vapor flowrate; β_{pay} is payback period; N is number of trays, D is diameter. A superstructure with at most 63 equilibrium stages (31 above, and 31 below the feed stage) was used. A stop criterion of maximum 150 main iteration steps was applied. The characteristics of the MINLP models are shown in Table 1.

Table 1. Characteristics of the MINLP representations, and solutions

Model	No. of eqs.	No. of non-linear eqs.	No. of vars.	No. of bin. vars.	No. of iters.	N	D	R	Obj.	Sol. time (CPU s)
Visw.	1138	388	1105	60	14	20	1.11	1.46	74.07	482
Yeom.	2051	514	1272	60	150	13	1.38	2.83	83.15	79,565
New	1592	519	1449	10	150	16	1.18	1.76	73.12	13,643

The MINLP representations were solved on a Sun Sparc workstation, using GAMS (Brooke et al., 1992). The MINLP solver was DICOPT++, the NLP subproblems were solved by CONOPT, the MILP subproblems by CPLEX. The solution of the model of Viswanathan et al. was stopped after the 14^{th} iteration because of a solver failure. As is also shown in Table 1, our new model was solved in a significantly shorter time than the model of Yeomans et al., although the number of variables were higher. This must be a consequence of decreasing the number of binary variables.

Conclusions

A new model has been developed for distillation column synthesis and optimization based on R-graph representation of the superstructure. First, a GDP model is generated from the supergraph. This GDP is transformed into an MINLP model which represents the considered structures only, and uses minimum number of binary variables to distinguish different structures. The MINLP model is compared to two other model form the literature (Viswanathan and Grossmann, 1993; Yeomans and Grossmann, 2000). The new model results in shorter computation time.

References

Brooke, A., D. Kendrick and A. Meeraus, 1992, GAMS. User's Guide. Scientific Press, USA.

Farkas, T., E. Rev and Z. Lelkes, 2004, submitted to Comp. Chem. Eng.

Friedler, F., K. Tarjan, Y.W. Huang and L.T. Fan, 1992, Chem. Eng. Sci., 47, 1973-1988.

Grossmann, I.E., 1996, Adv. Chem. Eng., 23, 171.

Luyben, M.L. and C.A. Floudas, 1994, Comp. Chem. Eng., 18, 933.

Rev, E., T. Farkas and Z. Lelkes, 2004, submitted to Int. J. Comput. Math.

Viswanathan, J. and I.E. Grossmann, 1993, Comp. Chem. Eng., 17, 949-955.

Yeomans, H. and I.E. Grossmann, 2000, Ind. Eng. Chem. Res., 39, 1637-1648.

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