GENERALIZED MODULAR FRAMEWORK FOR DISTILLATION COLUMN SYNTHESIS

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Abstract

In this work the distillation column sequencing problem is addressed through the Generalized Modular Framework, based on formal superstructure optimization techniques. The proposed method overcomes structural complexities through the use of systematically composed structural models incorporating all the feasible sequencing alternatives. The generated sequences are evaluated with respect to their cost efficiency, based on aggregated physical models, enhanced with principles of the Orthogonal Collocation technique for distillation order reduction. This allows the generation of compact optimization problems, while avoiding the use of potentially limiting simplifying assumptions. The synthesis method coupled to a formal Mixed Integer Nonlinear Programming (MINLP) solution algorithm, was applied for a number of sequencing case studies generating substantial economic savings by finding systematically and accurately the most cost efficient column sequence.

Keywords

Generalized Modular Framework, Distillation Column Sequences, Superstructure Optimization, Orthogonal Collocation.

Introduction

The synthesis of distillation column sequences is one of the most important and challenging problems in Chemical Engineering. Its importance lies in the fact that substantial energy savings can be achieved through the selection of the most appropriate distillation sequence for a particular separation. These savings can be further enhanced through the incorporation in the sequencing problem of techniques such as Heat Integration and Thermal Coupling. The challenges are related to the underlying structural and physical complexities that need to be formally addressed in order to achieve the synthesis targets. The structural complications are related to the large number of available alternative column sequences, which increase with the number of components to be separated. Moreover, these structural complications become even more intense with the incorporation of Heat Integration and Thermal Coupling possibilities. The physical complications are related to the complexity of the underlying physical phenomena, which involve simultaneous mass and heat exchange between liquid and vapour streams at the tray cascades. Due to its economic importance, the distillation column sequencing problem has been the focus of numerous approaches. The latter include evolutionary techniques (Stephanopoulos and Westerberg, 1976), heuristic methods (Seader and Westerberg, 1977), hierarchical decomposition (Douglas, 1988), thermodynamic insights (Bek-Pedersen and Gani, 2004), to name but a few. However, an approach that has been receiving considerable attention is superstructure optimization. Superstructures are, in general, superset flowsheets incorporating every feasible realization of the process in question. The generation and evaluation of the structural alternatives takes place with the solution of an optimization problem. A number of superstructure methods has been proposed for the distillation column sequencing problem, focusing on simple columns (Aggarwal and

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Floudas (1990), Yeomans and Grossmann (2000*a*)), on Heat Integrated columns (Andrecovich and Westerberg (1985), Paules and Floudas (1988), Caballero and Grossmann (1999)) and on Complex columns (Sargent and Gaminibandara (1976), Dünnebier and Pantelides (1999), Yeomans and Grossmann (2000*b*), Agrawal (2003)). The superstructure sequencing methods, are generally either based on simplifying assumptions and shortcut calculations, or employ rigorous *MESH* distillation models. An alternative approach is through the use of aggregated distillation models, which can be considered an intermediate between rigorous and shortcut models, avoiding simplifying assumptions, while generating compact optimization problems. In this work an aggregation superstructure method, enhanced with elements of distillation order reduction techniques, is proposed for the synthesis of distillation column sequences in a systematic and compact way.

GMF Distillation Column Sequencing

The proposed distillation column sequencing method is based on the Generalized Modular Framework (GMF) (Papalexandri and Pistikopoulos, 1996). The GMF is a superstructure optimization method for process synthesis at the conceptual level. Like most methods belonging to the superstructure optimization approach, the GMF consists of a Structural Model responsible for the generation of the structural alternatives and a Physical Model responsible for their evaluation with respect to the design targets. These two models are described in more detail below for the synthesis of distillation column sequences.

The GMF Structural Model

The GMF Structural Model consists of the *GMF Building Blocks* and their *Interconnection Principles*. There are two main types of superstructure building blocks in the GMF, the Mass/Heat *(M/H)* modules, where heat and mass transfer take place and the Pure Heat *(He)* modules, where the streams do not come into mass active contact and only heat exchange takes place. The GMF Interconnection Principles define the way the building blocks should be connected to each other in order to generate physically meaningful column sequences. These principles are translated mathematically through: *(i)* GMF Structural Sets and *(ii)* GMF Structural Constraints. The GMF Structural Sets provide information about the available building blocks, their topology in the superstructure and their allowed or forbidden interconnections for the specific problem examined. The GMF Structural Constraints are pure integer constraints (constraints involving only binary variables) providing the rules for the generation of distillation columns and their sequences. In the GMF, binary variables are employed denoting the existence of building blocks and of interconnection streams. This formulation is employed since it facilitates considerably the construction of the

structural models and the performance of the structural optimization. The increase of the problems' combinatorial size is compensated through tight structural modeling, guaranteeing the incorporation of all the structural alternatives and computational efficiency.

For the examined sequencing problems the construction of the GMF Structural Models is carried out in a systematic *3*-Stage procedure. In the *1ˢᵗ* Stage the GMF Structural Constraints are initially posed as Propositional Logic Expressions. In the latter, the GMF binary variables are replaced, for consistency, by their corresponding Boolean variables (logical variables taking values of true or false) expressed over the GMF Structural Sets. According to their function, the logical expressions are categorized into Superstructure Basis, Intra-Column and Inter-Column logical expressions.

The Superstructure Basis Logical Expressions are general constraints providing information about the maximum or minimum number of *M/H* modules and fix the existence of building blocks that participate in all the structural alternatives. *Illustrating Example: M/H* module *1* always exists:

$$y_1 \quad : \quad true \tag{1}$$

The Intra-Column Logical Expressions provide the rules that arrange the connectivity of the GMF Building Blocks for the systematic representation of *each* simple column of the superstructure. *Illustrating Example:* If a Lower *M/H* module exists, then its immediate Upper *M/H* module should also exist:

$$y_e \iff y_{e+1} \qquad \forall\, e \in E^L \tag{2}$$

In the above, E^L is the GMF Structural Set of all Lower (odd numbered) *M/H* modules in the superstructure.

The Inter-Column Logical Expressions are employed in order to provide the rules that arrange the connectivity between building blocks belonging to *different* columns, thus generating the feasible column sequences for the examined problem. *Illustrating Example:* If a liquid interconnection leaves a Lower Inner *M/H* module, then the destination module also exists:

$$y_{e,ea}^{LL} \implies y_e \wedge y_{ea} \qquad \forall\, e \in E^{LI}, ea \in E_e^{LLo} \tag{3}$$

Similarly as before, in Equation (3) E^{LI} and E_e^{LLo} are the GMF Structural Sets of the Lower Inner *M/H* modules and of the allowed liquid interconnections from *M/H* module *e*, respectively.

In the *2ⁿᵈ* Stage, the Logical Expressions are transformed into their conjunctive normal form, *i.e.* to a set of clauses which include OR operators linked by AND operators. This is achieved by following the systematic rules presented in Biegler *et al.* (1997). Then the logical expressions in their conjunctive form are translated into their respective pure integer structural constraints, by replacing the Boolean with their corresponding binary

variables and by following the rules presented in Raman and Grossmann (1991). For illustration, the above equations become:

$$y_1 = 1 \tag{4}$$

$$y_e - y_{e+1} = 0, \qquad \forall\, e \in E^L \tag{5}$$

$$1 - y_{e,ea}^{LL} + y_e \geq 1, \qquad \forall\, e \in E^{LI},\, ea \in E_e^{LLo} \tag{6}$$

$$1 - y_{e,ea}^{LL} + y_{ea} \geq 1, \qquad \forall\, e \in E^{LI},\, ea \in E_e^{LLo} \tag{7}$$

In the 3rd Stage, the GMF Structural Sets, utilized for the generation of the above constraints, are appropriately assigned in order to construct the GMF Structural Models for each examined problem.

This *3*-Stage procedure is followed throughout the GMF as it has been found that the GMF Structural Models generated are always accurate and tight. This is particularly important for the GMF which is based on building blocks of high levels of structural abstraction (where distillation columns are not represented *a-priori*). Moreover, the tightness of the generated problems enhanced considerably the computational efficiency of the proposed methods since the solution of each structural problem in the employed decomposition MINLP algorithm required only minimum computational effort for its solution.

The GMF Physical Model

The GMF Physical Model is employed for the representation of the underlying physical phenomena of the structures generated by the GMF Structural Model. Each GMF Building Block is accompanied by its physical model which depends on the block's function in the framework. In this work, the aggregated GMF Physical Models of the *M/H* models which are based on fundamental mass and heat exchange principles at the blocks' boundaries are coupled with principles of Collocation techniques for distillation order reduction. This is the only method which can guarantee the safeguarding of the GMF physical representation advantages (no use of simplifying assumptions and size compactness) and can still achieve the GMF Physical Model enhancement, providing intra-module and detailed column design information.

In accord with the Orthogonal Collocation (OC) method principles of Seferlis and Hrymak (1994), each *M/H* module is discretized into *nc* interior and *2* exterior collocation points, as can be seen in Figure *1*. Within the *M/H* module, Lagrange polynomials are used for the approximation of liquid and vapour component flowrates and enthalpies. The roots of the orthogonal Hahn polynomials are the locations of the collocation points, $s_{e,j}$, over the *M/H* module and are explicit functions of the number of trays, M_e, of the represented column section. The location of each collocation point corresponds to a tray number, as shown in Figure *1*.
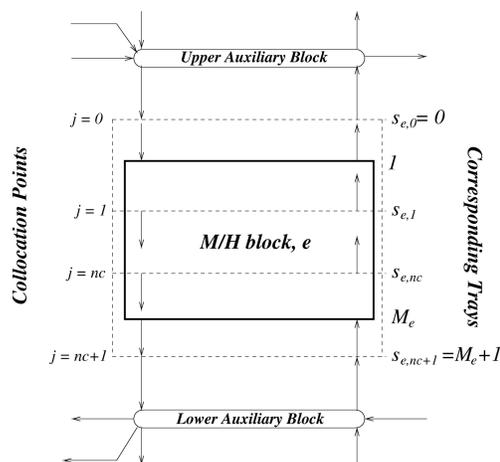

Figure 1. Discretized GMF M/H Module.

The column *MESH* equations are enforced on the *interior* collocation points, in accord with the OC principles, thus generating a compact problem to solve. An illustration is given for the component balances in each discretized *M/H* module *e*:

$$\tilde{v}_{e,j+1} + \tilde{l}_{e,j-1} - v_{e,j} - l_{e,j} = 0, \ \forall\, e \in E,\, j = 1,...,nc_e \tag{8}$$

In the above equation, *E* is the GMF Structural Set of all available *M/H* modules. The component flowrates with the tilde do not correspond to actual collocation points and are interpolated using the Lagrange polynomials at the locations $s_{e,j+1}$ and $s_{e,j-1}$. These will provide explicit functions of the number of trays per *M/H* module, to be included in the optimization problem. It must be noted, that the incorporation of OC principles within the GMF is achieved due to the modular formalism of both the order reduction and aggregation methods for distillation.

The GMF Structural and Physical Models were employed for the systematic generation and evaluation of Simple, Heat Integrated and Complex distillation column sequences for cost efficiency. In the following section, the proposed method is illustrated for the solution of a motivating sequencing example.

Illustrating Example

The GMF was employed for the complex column sequencing problem for the separation of Benzene, Toluene and o-Xylene. The problem statement was taken from Chavez *et al.* (1986). For the construction of the GMF Structural Model, the necessary assignments of the GMF Structural Sets (Stage *3*) were initially carried out. Then for the formulation of the GMF Physical Model each *M/H* module was discretized into *6* collocation points (*2* exterior and *4* interior). This discretization scheme was found adequate for the accurate representation of the underlying physical phenomena.
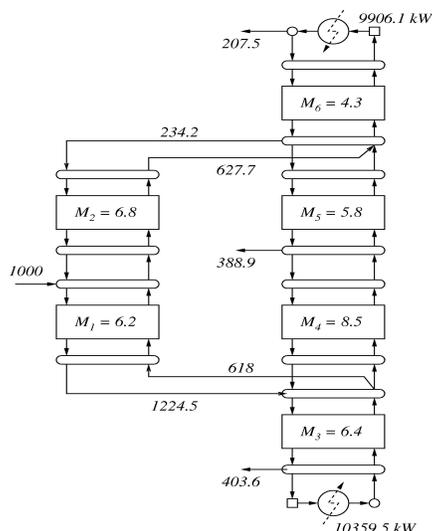
Figure 2. Optimal Complex Column Sequence.

The generated MINLP synthesis problem was implemented in GAMS based on the APROS/GBD methodology (Paules and Floudas, 1989). The optimal solution was found, in *4* GBD iterations and corresponded to the Petlyuk Column (Figure *2*). The Total Annualized Cost (TAC) of the sequence was found equal to *$2,095,900/yr*, consisting of an Operating Cost of *$1,681,500/yr* and of an Annualized Capital Cost of *$414,400/yr*. The GMF captured the effects of full thermal coupling of this sequence which led to *28%* annual savings when compared to the optimal simple column sequence. The GMF Physical Model captured simultaneously the interactions between capital and operating cost in the continuous problem. It must be noted that a rounding-off of the non-integer tray numbers, although possible, was not enforced due to its minimal impact on the system's TAC.

Conclusions

In this work the distillation column sequencing problem was systematically addressed through the Generalized Modular Framework (GMF). Appropriate GMF Structural Models were developed, based on logical modelling, generating efficiently the sequencing alternatives. The latter were evaluated with respect to their cost efficiency through an enhanced GMF Physical Model, coupling aggregation and orthogonal collocation techniques. This model allowed for the generation of compact optimization problems, while avoiding the use of simplifying and potentially limiting assumptions, thus providing a *'best of both worlds approach'* between shortcut and rigorous methods for distillation. The proposed method was illustrated over the solution of a complex column sequencing problem, obtaining accurately and efficiently the most cost efficient distillation column sequence and leading to substantial annual savings.

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