BUDAPEST UNIVERSITY OF TECHNOLOGY AND ECONOMICS
Department of Chemical Engineering

Ph.D. Theses

# Chemical Process Synthesis Using Mixed Integer Nonlinear Programming

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## Introduction

The target of process synthesis is to discover the best complete design to accomplish a chemical manufacturing goal. The alternatives have to be considered first in this process. Most of the alternatives can usually be ruled out according to engineering experience, but even the number of the remained alternatives is so huge that process synthesis needs a systematic study.

Mathematical programming is a widespread approach in process synthesis. This method has three main steps. (1) First a superstructure containing all the considered alternatives, and its graph representation, are generated. (2) Then a mathematical representation is formulated, based on the graph representation of the superstructure. (3) Finally, the extreme of the objective function is looked for, the mathematical model is optimised.

The most common mathematical representation which can handle discrete decisions is the mixed integer nonlinear programming (MINLP). An MINLP problem contains both continuous and integer variables; the integer variables are usually binary variables. Grossmann (1996) defined the three major guidelines of a „good" MINLP representation: (1) Keep the problem as linear as possible. (2) Develop a formulation whose NLP relaxation is as tight as possible. (3) If possible, reformulate the MINLP as a convex programming problem. However, according my experience, these criteria are not enough for efficient modelling.

Multiplicity causes serious problems in MINLP models. Multiplicity means that several solutions of the mathematical representation define the same structure. It can also be said that a structure is represented by isomorphic graphs. In this case, the objective function has the same value in several different points. It makes more difficult finding the optimum, and the search space is unnecessarily wide.

An important characteristic of an MINLP model is the number of its binary variables. With increasing number of binary variables, the complexity of the problem and the solution time increase exponentially. An MINLP model can be reformulated in a way as to decrease the number of binary variables by introducing new continuous variables and constraints.

During my PhD study, I mainly dealt (1) with the first step of the mathematical programming, i.e. the generation of the superstructure and its graph representation, and (2) with the connection of this step to the other ones.

First I studied how the earlier experience can be applied during the solution of a new synthesis problem. I applied case-based reasoning for generating proper superstructure with MINLP representation in distillation column synthesis. In case-based reasoning, first the most similar case to the actual problem is retrieved from a case library. If the solution of this most similar case cannot be used for the actual problem, then the earlier solution has to be adapted according to the actual requirements. If the problem is solved then, in the last step, it is incorporated in the case library.

Then I studied in details the connection between structures and graphs. I defined when an MINLP model represents a structure, and what the reasons of multiplicity are. I gave guidelines for how the multiplicity, and the number of binary variables of an MINLP representation, can be decreased in order to narrow the search space, to enhance the possibility of finding the global optimum, and to decrease the solution time.

Finally, I used my results and experience to develop a new superstructure and MINLP model for distillation column synthesis. This new superstructure and MINLP model was developed with respect to its applicability for synthesizing process systems containing such units.

## Approach

My work comprised the creation and solution of different MNLP models. These problems are solved with GAMS program package (Brooke et al., 1992) using DICOPT++ solver (Viswanathan and Grossmann, 1990). The GAMS program package was chosen because it is easily accessible, user friendly, and very well documented. DICOPT++ solver uses Outer Approximation method. In this method nonlinear programming (NLP) and mixed integer linear programming (MILP) subproblems are solved iteratively

Example problems and comparison models were taken from the literature. Some of the results were checked with ChemCAD 5.2 simulator.

## Major new results

1. **I proved that case-based reasoning is suitable for selecting a superstructure with an MINLP model in process synthesis using mathematical programming.** A case-based reasoning method has been developed which can retrieve the most similar case from a data base in distillation column synthesis problems of ideal mixture containing up to five components. A case library, containing 26 cases from the literature, was generated. First, a set of matching cases is retrieved using inductive retrieval. The cases are classified according to the operational attributes like sharp/non-sharp separation, heat integration, number of products and feeds. Then the cases of the set are ranked using the nearest neighbour method according to their similarities to the actual problem considering the component types, the boiling point and molar masses of the components, the feed and the product compositions. After the retrieval, the three most similar cases are reported. The superstructure can be used for solving the actual problem. The MINLP model and the solution of the selected cases can be adapted to the actual requirements, and the adapted solution can be used as an initial point during the optimisation.

2. **I developed an automated procedure for the generation of Basic MINLP Representation (BMR) which can serve as a reference to study whether an MINLP Representation (MR) represents a supergraph or not.** First, the Basic Generalized Disjunctive Programming Representation (BGR) is formulated based on the R-graph representation of the superstructure. BGR contains the constraints of the units in logical disjunctive form, the balances of input and output ports, and the cost function. BGR does not include any additional logical constraints but the unit relations; therefore, it represents all the subgraphs of the supergraph (the graph representation of the superstructure). Then BMR is generated from BGR using binary variables instead of logical ones, and transforming logical relations into algebraic ones. The generation of BGR and BMR is performed automatically from the R-supergraph; therefore, BMR can serve as a reference in checking whether an MINLP Representation (MR)

represents the supergraph or not. **An MR represents the supergraph if a bijective mapping can be given between a subset of the feasible region of MR and the feasible region of BMR.**

3. **I defined a new criterion of a "good" MINLP Representation (MR): MR should represent only the considered graphs of the supergraph. In this case, MR is called Ideal MINLP Representation (IMR).** Considered structures can be preliminary selected in an implicit or explicit way. Considered graphs represent the considered structures in a way that there are not isomorphic graphs among them. IMR can always be constructed by excluding the representation of the non-considered graphs. The representation of non-considered graphs can be excluded by extending the MR with algebraic transformations of the pure logical constraints. Computational results show (see Figs. 1 and 2) that the solution time is smaller with using IMR, and the maximum solvable size of the problem (represented by $n_{max}$) is greater than with using Conventional MINLP Representation (CMR) or Non-considered Structures eXcluded MINLP Representation (NSXMR).

4. **I defined a new criterion of a "good" MINLP Representation (MR): MR should use minimum number of binary variables to make distinction between different structures. In this case, MR is called Binarily Minimal MINLP Representation (BMMR).** This means the use of $n_{bv}$ number of binary variables in case of $n_g$ number of represented graphs, where $n_{bv}$ is the smallest whole number that satisfies $n_{bv} \geq \log_2 n_g$. **I proved that BMMR can always be generated.** I developed a general method for the generation of BMMR in which new binary variables are introduced instead of the old ones, and the equations are transformed. Computational results show (see Figs. 1 and 2) that the solution time is smaller with using Binarily Minimal and Ideal MINLP Representation (BMIMR), and the maximum solvable size of the problem (represented by $n_{max}$) is greater than with using IMR, CMR, or NSXMR.

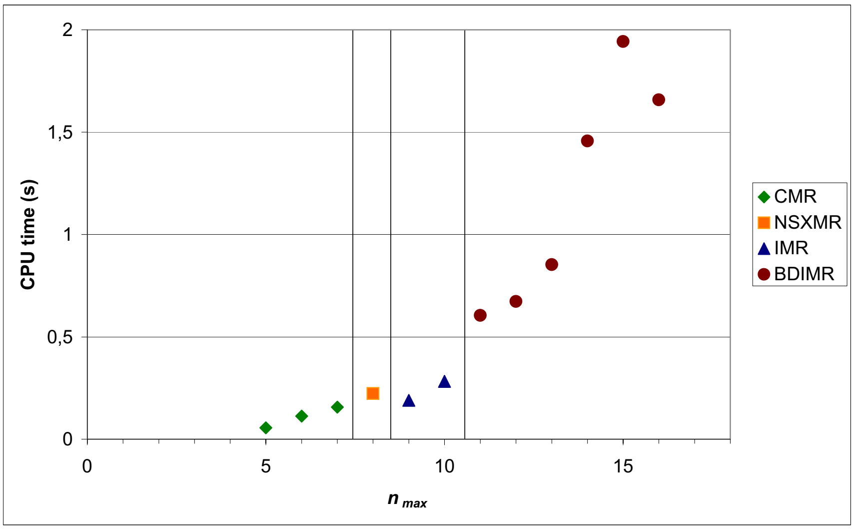


Figure 1. NLP solution time per iterations

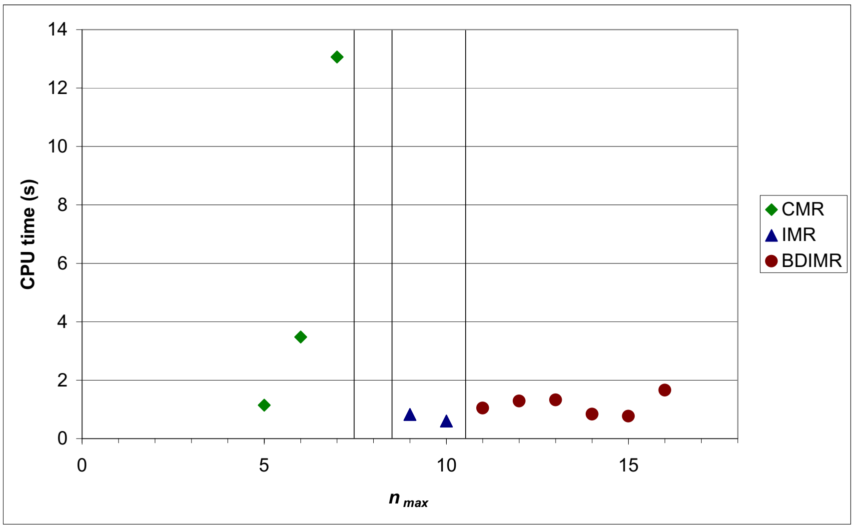


Figure 2. MILP solution time per iterations

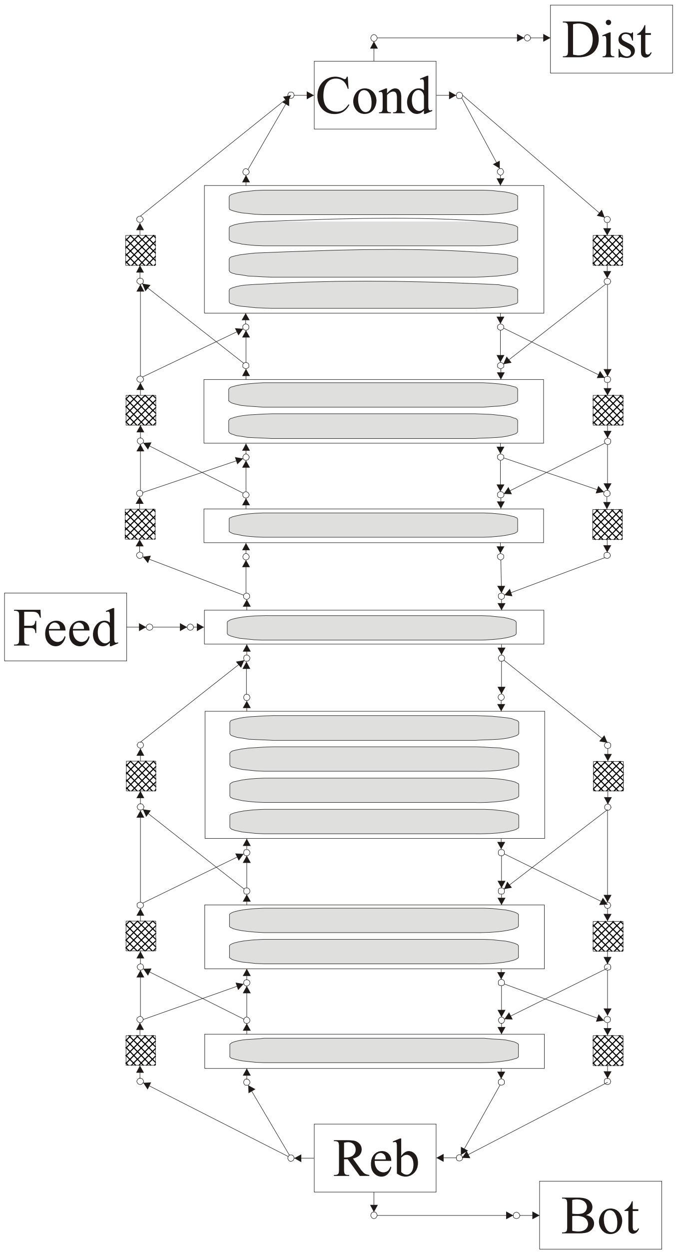

Figure 3. R-graph representation of the superstructure

5. **I developed a new, R-graph based, superstructure, and corresponding MINLP model, for the synthesis of a single distillation column.** The superstructure (see Fig. 3) is generated in a way that makes possible an easy generation of the Binarily Minimal and Ideal MINLP Representation. In the superstructure, the $k^{th}$ conditional unit (represented by rectangle, calculated bottom up) contains $2^{k-1}$ number of equilibrium stages (represented by shaded ovals). In the new MINLP model, binary variables denote the existence of each conditional unit containing equilibrium stages. The existence of the transport units (represented by hatched squares) is expressed with using the binary variables of the units containing equilibrium stages. The relations of the permanent units (source unit, Feed; sink units, Dist and Bot; reboiler, Reb; condenser, Cond; and the feed stage) do not include binary variables. **The developed MINLP model is Binarily Minimal and Ideal, i.e. it uses minimum number of binary variables to make distinction between structures, and it represents considered structures only.** The new MINLP model finds better local optima in shorter computational time than the MINLP model based on the GDP model of Yeomans and Grossmann (2000).

## Significance of the new results

In the framework of my dissertation I proved that case-based reasoning is a proper method for the generation of the superstructure with MINLP model for distillation column synthesis. In this way, the earlier experience can be used in case of a new distillation synthesis problem.

Based on my results the relations between structures, graphs, and MINLP representations, became clearer. Using my guidelines the solution time of MINLP problems can be significantly decreased, and the maximum size of solvable problem can be increased.

Using the new superstructure and MINLP model, which uses the above guidelines, distillation column synthesis problems can be solved significantly faster.

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