Model-based Conceptual Design and Tool Support for the Development of Continuous Chemical Processes

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Abstract
Despite many efforts to formalize the early phase of conceptual process design, no tool-supported systematic method for the conceptual design of chemical processes has been developed to date. This contribution presents a methodology and a software tool for the early conceptual design phase, which is characterized by limited and uncertain information on the available process units. The proposed methodology supports the graphical modeling of hierarchical superstructures which are subsequently evaluated by optimization. Techniques for the analysis of the effect of uncertainties are included.

Keywords: Conceptual design, flowsheet optimization, uncertainty, superstructure

1. Introduction
The early phases of process design commonly suffer from lack of information, widespread uncertainties which are difficult to quantify and a large number of possible design choices. Several approaches to the solution of process flowsheet design problems have been presented in the last decades. A successful method in the situation where unit models are available is the application of mixed-integer nonlinear programming [Grossmann et al., 1999]. This approach has been applied to the synthesis of reactor networks, distillation sequences and mass and heat exchanger networks [Grossmann et al., 2000]. If precise models are not available, heuristic methods for the identification of promising design candidates can be employed [Douglas, 1988]. This however does not exploit the potential of optimization. [Li and Kraslawski, 2004] concluded that a key topic for the advancement of conceptual process design is the “improvement of optimization and simulation techniques as well as of information management tools in order to handle more information and knowledge from various sources”.

The aim of this contribution is to apply mathematical optimization methods to the early phase of process design where the information on the performance of the different building blocks of the flowsheet is available in different forms, e.g. experimental data, short-cut models or rigorous unit models. The goal is to detect promising process concepts as early as possible and to single out the crucial parameters for the performance of the process such that experimental and modeling work can be focused on those areas that most likely will lead to success, i.e. an economic process design.
2. Case Study

Before the modeling method is presented in section 3, the process design problem considered later is presented. Our work is part of the DFG Collaborative Research Center SFB-TR 63 InPROMPT. The goal of the SFB-TR InPROMPT is the development of novel process concepts for integrated processes in liquid multiphase systems. Currently, the focus is on the hydroformylation of long-chain alkenes. The key challenges of this process are to bring syngas, a polar rhodium-based catalyst and the non-polar reactant in close contact to enable the reaction, and the retention of the very expensive homogenous catalyst. For the recycling of the catalyst and to overcome mass transfer limitations two technologies are investigated: liquid multiphase systems (LMS) and thermomorphic solvent systems (TSS). LMS employ the complex phase behavior of aqueous solutions of surfactants to reduce mass transfer limitations [Milano-Brusco et al., 2010]. TSS employ mixtures of polar and non-polar components that show biphasic behavior at low temperatures and homogenous behavior at higher temperatures where the reaction takes place. The catalyst is then recycled by phase separation at lower temperatures [Behr et al., 2008].

A basic process flowsheet that depicts the main steps of such a hydroformylation process is shown in fig. 1.

![Basic flowsheet of the hydroformylation process](image)

Figure 1: Basic flowsheet of the hydroformylation process

The case study considered in this contribution features a small subset of the possible designs. It is limited to two possible variants of a TSS reactor for which experimental data from batch experiments is available. The chemical components involved are n-dodecene (C12), hydrogen (H2), carbon monoxide (CO), n-tridecanal (C13), 2-methyl-dodecanal (iso-C13), decane (C10) and di-methyl-formamide (DMF). In the case study, dodecen, decane and DMF form a thermomorphic solvent system. After phase separation, the catalyst is retained in the polar DMF phase, while the product C13 in the non-polar C10 phase is separated further. A reduced chemical pathway of one main reaction and one side reaction is presented in equations (1) and (2).

\[
C_{12}H_{10} + H_2 + CO \xrightarrow{Rh} C_{13}H_{16}O \quad (1)
\]

\[
C_{12}H_{10} + H_2 + CO \xrightarrow{Rh} iso-C_{13}H_{16}O \quad (2)
\]

The following set of separation technologies is considered: Vacuum distillation, crystallization (for the separation of the isomers), evaporation (for the recovery of H2 and CO), distillation (for the recovery of educts and solvent) and decanting (for the recovery of the polar DMF phase that contains the catalyst).

3. Modeling of flowsheet superstructures

3.1. Modeling of information

The goal of the proposed method is the support of the development process of chemical processes from the first idea to the final process under permanent consideration of the resulting production cost. The selection of alternatives starts as soon as the first indicative information on the performance of the units is available.
In a first step the user specifies the components and the reactions considered in the chemical process. In a second step the user defines a set of possible reactors and separators. Reactors can be modeled by data obtained at a fixed operating point or using kinetic information once it is available. The separators can either be defined by heuristic rules (e.g. “it is possible to separate syngas with an evaporator”), or by short-cut models (e.g. using relative volatilities). The uncertainty of the information is accounted for by defining upper and lower bounds for the parameter values in addition to experimental or estimated values.

In the case study, the decanter is modeled as an ideal component splitter for C10, C12, C13 and iso-C13 from DMF. The split factor of the catalyst-carrying DMF phase is assumed to be uncertain and may take values from an interval that was determined from early laboratory experiments. The flash is modeled as an ideal splitter that separates syngas from the other components. The distillation column is regarded as an ideal splitter between C10, C12 and the other components, while the vacuum distillation and the crystallizer are modeled as ideal component splitters with uncertain recoveries of C13. The reactors are modeled as PFRs with the stoichiometry from equations (1) and (2) and partial conversions obtained from laboratory batch experiments.

3.2. Algorithm for the generation of flowsheet superstructures

Using the information defined above, the program generates a superstructure that contains all feasible combinations of all process steps. Recycle streams are not explicitly modeled in the superstructure at this stage because it is assumed that only educts are recycled. Then recycle streams will only result in lower feed flows which is included in the cost evaluation by using the same price for buying and selling of educts.

The superstructure that was generated for the case study is depicted in fig. 2. The algorithm generates a directed bipartite graph expressed as a state-task-network as introduced by Kondili and Pantelides [Kondili et al., 1993]. Each process unit is modeled as a task and streams are modeled as states. The superstructure starts at a single feed state and branches on stream states which have multiple compatible separators.

The algorithm consists of two stages. In the first stage all possible reactors are added to the superstructure and the resulting open streams are calculated based on the component balances. The second stage searches for compatible separators for each open stream and generates two open output stream states for each compatible separator. The algorithm responsible for finding compatible separators compares the molar composition window of the open stream to the specified molar composition window of the input stream of a separator. A separator is considered compatible if the operating windows overlap. If the pressure or temperature windows of two units do not overlap a virtual temperature or pressure change unit is inserted into the flowsheet. This process is repeated until no open streams
exist anymore or until a predefined maximum number of separation steps in a branch have been reached. Non-terminated branches are subsequently eliminated from the superstructure.

3.3. Optimization of flowsheet superstructures
For small problems, all design alternatives contained in the superstructure are enumerated. The uncertain parameters in each flowsheet are identified and subsequently represented by a three-level discretization. From all combinations of the parameter levels for each uncertainty a set of scenarios results. Then a NLP problem is automatically constructed for each flowsheet containing only continuous variables. The continuous variables are separated into design variables and control variables. The control variables, e.g. pressures, temperatures and flows, are optimized for each scenario while the design variables apply to all scenarios of one design (two-stage optimization). The resulting distribution of the costs is visualized and analyzed.

4. Results for the example
The superstructure modeled in section 3 that consists of 20 designs was investigated by the optimization algorithm. As the computation was based on split factors only, no continuous design parameters were present. The cost intervals constructed from the maximum and minimum costs of all scenarios for each design are shown in figure 3. From figure 3, it is not yet possible to identify one process alternative as the best one, as many intervals overlap. Four alternatives can be identified as promising. It is advisable to focus the further research on the uncertain parameters that have the largest impact on the spread of the costs for the designs 121, 1221, 221 and 2221 to reduce the cost intervals and identify the best design. Figure 4 shows the sensitivities of the cost function for the uncertain parameters for each of the most promising designs. Of all the uncertainties, the split factor of DMF in the decanter has the largest impact on the spread of the costs for all four alternatives. The sensitivity of the split factor of C13 in the crystallizer is zero for all promising designs, since the crystallizer was not utilized in any of those designs. Even considering uncertainties, vacuum distillation alone will suffice to yield the desired product purity. The logical next step is to reduce the uncertainty of the catalyst retention in the decanter by laboratory experiments and more accurate modeling, if at all possible.

5. Future Developments
Since the number of alternatives in the superstructure of the case study was small, it is possible to perform an optimization of each individual design for every scenario. When the number of alternatives grows, more sophisticated methods for the selection of designs must be employed. [Urselmann et al., 2011] have shown the applicability of
hybrid evolutionary (memetic) algorithms to superstructure optimization problems which seem well-suited for the problem at hand. [Tometzki and Engell, 2011] applied hybrid multi-objective evolutionary algorithms to the solution of optimization problems under uncertainty, including a pipeline layout problem. We will therefore follow this line of development and design a tailored memetic algorithm for the screening of the superstructures including parametric uncertainties in a two-stage setting.

6. Conclusion

An integrated development environment for the conceptual design of chemical process flowsheets was developed and applied to a real world design problem. The effect of uncertain parameters can be evaluated and used for the comparison of different design alternatives and for the visualization of the trade-offs inherent in each design. This shows the potential of this novel technique as it is possible to identify promising process alternatives in the early phase of process design using only basic information that is available from the literature and laboratory results.

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