Computers and Chemical Engineering 81 (2015) 260–271

Contents lists available at ScienceDirect

Computers and Chemical Engineering

journal homepage: www.elsevier.com/locate/compchemeng

A unifying framework for optimization-based design of integrated reaction–separation processes

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ARTICLE INFO

Article history:
Received 13 November 2014
Received in revised form 11 February 2015
Accepted 19 March 2015
Available online 1 April 2015

Keywords:
Conceptual design
Reaction–separation process synthesis
Shortcut evaluation
ETBE production process

ABSTRACT

The determination of the best flowsheet and the development of a conceptual design of a chemical reaction–separation process is a complex task. Although model-based methods for process design have been proposed in the last decades, only a few contributions address the simultaneous design and optimization of integrated reaction–separation processes. This paper presents a systematic and optimization-based approach for the design of such processes. Shortcut methods are utilized to screen alternative flowsheet structures. For the most promising alternatives a rigorous optimization of the entire flowsheet is executed to determine the best alternative. The incremental refinement of this framework allows for the systematic generation and numerically robust evaluation of every reaction–separation process. Consequently, it helps to shorten the time for developing new and innovative processes and improves the chances to find the best process design. The suggested methodology is illustrated by means of a case study considering ethyl tert-butyl ether (ETBE) production.

1. Introduction

Depleting natural resources, increased international competition, and the desire for sustainability foster the chemical industry to pursue more efficient processes. However, new processes are often still developed in a creative design process, where new process variants – often generated based on expert-knowledge and heuristics – are routinely evaluated by means of commercial process simulators. Systematic and optimization-based design tools can increase the efficiency within the conceptual design phase and allow for the development of improved and innovative processes. In order to improve the conceptual design of separation processes, Marquardt et al. (2008) and Dowling and Biegler (2015) have proposed process design frameworks for equation-oriented optimization of hybrid separation processes. However, the optimization-based design of integrated reaction–separation processes did not get appropriate attention, because the simultaneous optimization of the flowsheet structure, unit specifications and operating points is still a hard computational problem (Kallrath, 2000). This is due to the size and complexity of these problems, since several nonlinear reaction and separation unit models are embedded in the flowsheet, recycle loops have to be optimized, and nonlinearities of the underlying non-ideal thermodynamic and reaction kinetic models complicate numerical evaluation.

The systematic and optimization-based framework for the design of hybrid separation processes proposed by Marquardt et al. (2008) has been used and demonstrated for conceptual design of various processes by our group (see, e.g., Kraemer et al., 2011). This framework combines thermodynamically sound shortcut methods and rigorous mixed-integer nonlinear programming (MINLP). The design task is separated into three distinctive steps: In the first step, different flowsheet structures are generated. In the second step, these alternative flowsheets are evaluated with shortcut methods to screen for the most promising process alternatives, which are finally modeled rigorously and optimized in the third step to calculate operating point and sizing information and finally to determine the cost-optimal process structure. While this framework is in principle applicable to all kinds of processes, its application was until now restricted to separation processes only. In this work we introduce the necessary extensions and present the application to an integrated reaction–separation process.

At first, Section 2 presents an overview on different approaches to reactor network and reaction–separation process design. In Section 3 extensions to the framework of Marquardt et al. (2008) are presented to account for systematic design of reaction–separation

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http://dx.doi.org/10.1016/j.compchemeng.2015.03.014
0098-1354/

processes. The necessary extensions of both, the shortcut evaluation methodology for the preselection of flowsheets and the rigorous optimization procedure integrate existing and new approaches. The application of the framework is illustrated with the design of an ethyl tert-butyl ether (ETBE) process in Section 4 before we close with a summary in Section 5.

2. Design methods – a brief review

2.1. Design methods for reactor networks

Existing reactor network design methods can be grouped into four main areas: heuristics, attainable region concepts, dynamic optimization, and superstructure optimization. Since this contribution can only give a broad overview on existing methods, only key features are outlined below, while a more comprehensive presentation of the technical details can be found in the cited literature.

Reactor selection heuristics can be classified into criteria based on experience and model evaluation (Schembecker et al., 1995; Till et al., 2004). In many cases, the heuristics are easy to apply, easy to use, and may result in an appropriate choice of a reactor network for common reaction classes. However, these criteria may hinder the development of innovative reactor concepts and the potential of their application to complex systems is limited.

In order to assess attainable performance targets of a complete family of reactor networks Horn (1964) introduced attainable regions (AR), a geometrical concept which can be directly used for synthesizing optimal reactor configurations for a given feed and known reaction kinetics. In some appropriately chosen state space the AR refers to the set of all product compositions which can be reached by any possible reactor network employing only reaction and mixing. The boundary of the AR is of special interest, as it represents the maximum achievable product compositions. In fact, any reachable reactor product can be realized by parallel, serial or serial-parallel configurations of plug flow reactors (PFRs) (Feinberg and Hildebrandt, 1997), continuous stirred-tank reactors (CSTRs) (Feinberg, 2000a), or differential sidestream reactors (DSRs) (Feinberg, 2000b). Since the set of all attainable product compositions is derived graphically, the classical AR approach can only be applied to problems which can be reduced to at most three dimensions (Rooney et al., 2000). In order to overcome this limitation, computational techniques were developed to apply the AR concept to higher dimensional problems (Abraham and Feinberg, 2004). Kauchali et al. (2002) formulate a linear program (LP) to analyze candidate ARs by discretization of the concentration space into an arbitrarily large number of points. This model represents a large network of completely connected CSTRs which can be used to approximately determine the optimal operating point of arbitrary reactor networks consisting of CSTRs, PFRs and DSRs. However, due to the discretization of the concentration space, the number of equations and variables grows exponentially with the number of components. Thus, problems with more than three components can easily lead to models with thousands of equations and possibly millions of variables (Kauchali et al., 2002). Clearly, modern LP solvers can handle large-scale problems, but reaction processes with a typical, industrially relevant number of raw materials, intermediates, and (by-)products will result in extremely large problems, which scale with the number of chemical components involved.

A different approach of assessing attainable performance targets of reactor networks can be traced back to Denbigh (1944) and has been extended by several authors (see e.g., Bilous and Amundson, 1956; Aris, 1961; Horn, 1961). It uses the concept of dynamic optimization of an elementary process function. A fluid element is tracked on its way along a (spatially or temporally) resoluted reaction coordinate. During its evolution, the state of the fluid element defined by concentrations, temperature, and pressure can be manipulated by reaction as well as heat and mass fluxes. Thus, an optimal state trajectory of the element in thermodynamic state space can be decided. An analysis of this trajectory provides insight into the choice of promising reactor networks and can help to develop innovative reactor concepts (Peschel et al., 2010) but it does not provide detailed information about reactor sizing.

Such sizing information can be determined by means of superstructure optimization, for which flowsheet structure, operating conditions and equipment sizes are simultaneously optimized. A superstructure for reactor network synthesis is generally composed of a set of pre-defined reactor types, e.g., isothermal (Kokossis and Floudas, 1990) or non-isothermal (Kokossis and Floudas, 1994; Pahor et al., 2001) CSTRs and PFRs, or CSTRs and cross flow reactors (Schweiger and Floudas, 1999). In contrast to the AR, the possible connections between the reactors must be specified a priori, though the best connection as well as the existence of the specified reactors are identified as a result of superstructure optimization, where the possible connections between reactors are represented by integer variables in the model. Additionally, the performance characteristics of many reactor networks are similar. For example, a cascade of CSTRs may approximate a PFR very well. Therefore, finding the best combination from a technical and economic point of view can become very difficult. This problem can be addressed by means of simple superstructures, which only include the most promising combinations. However, such simplifications may result in a loss of the optimal reactor network in the superstructure. Hence, modeling for superstructure optimization requires always a trade-off between the number of combinations included in the superstructure, the uniqueness of the solution, and the solvability of the problem.

All of the available design methods for reactor networks only exhibit their strengths and weaknesses when applied to industrially relevant problems. Such design problems typically involve reaction–separation processes. Therefore, design methods for reaction–separation processes are indispensable.

2.2. Design methods for integrated reaction–separation processes

In industrial practice, the structure of a flowsheet is often pragmatically fixed using heuristics from literature, solutions of similar problems, and experience. Common heuristics for reaction–separation processes, e.g., Douglas (1988), Smith and Linnhoff (1988) or Upadhye et al. (2011), usually start with designing a suitable reactor network, before the separation and recycle system and, if appropriate, the heat exchanger network are designed in subsequent steps. The feasibility and the cost of the proposed process candidates are then determined by repetitive simulation studies, where the operating points and unit specifications of feasible alternatives are often determined by spreadsheet calculations and rules of thumbs (Luyben, 2010, 2011).

Graphical approaches such as GH-plots (Fox et al., 2013) can help to facilitate the design of efficient process variants. Enthalpy (H) and Gibbs free energy (G) depicting the flows of heat and work respectively, are determined for all unit processes and are graphically represented as vectors in the GH-space. By manipulating the vectors in the graph, the overall material, energy, and work balances can be determined, before any flowsheet exists. Furthermore, these vectors can be used to construct a flowsheet which represents a thermodynamically efficient process. Based on such an initial flowsheet design, phenomena-based synthesis methods (Lutze et al., 2012; Babi et al., 2014) can be used to derive further process variants. These methods identify the phenomena involved in each task and manipulate and recombine them to generate