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Reactor network synthesis with guaranteed robust stability

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ABSTRACT

This paper proposes a systematic approach to design reactor networks with guaranteed robust stability. The approach is based on the superstructure approach for reactor network synthesis. A structured dynamic model for reactor network modeling is formulated and embedded in a MINLP with robust eigenvalue constraints. Design parameters, structural alternatives and parametric uncertainty are considered simultaneously as design degrees of freedom. Structural alternatives result from decisions on the existence of reactors and flow connections in the superstructure. Parametric uncertainty may either result from model uncertainties such as reaction kinetic constants or heat transfer coefficients, or from process uncertainties including slow disturbances in load or quality of raw materials. A tailored two-step solution strategy is proposed to tackle the robust mixed-integer optimization problem. A case study with five continuous stirred-tank reactors (CSTR) and five plug flow reactors (PFR) is presented for illustration.

1. Introduction and motivation

Reactor network synthesis is a classical problem in process systems engineering, which is defined as follows Biegler et al. (1997): "*For given reaction stoichiometry, rate laws, a desired objective, and system constraints, what is the optimal reactor network structure and its flow pattern? Where should mixing, heating, and cooling be introduced into the network?*" Solution approaches can be classified as heuristic, attainable region and rigorous optimization methods Peschel et al. (2010). The most common optimization methods rely on superstructure approaches, which have been under investigation for more than 20 years Achenie and Biegler (1986, 1990), Kokossis and Floudas (1990). An economical objective function is minimized subject to a process model representing steady states of a variety of reactor networks to find the best design parameters, operating point and network structure. Although the superstructure approach has shown great success in reactor network synthesis, dynamic properties such as stability, operability, or controllability are typically not considered Biegler et al. (1997). Consequently, the designed network may not be operated without a stabilizing controller. Thus it is important to consider dynamic properties already during reactor network synthesis Kokossis and Floudas (1994).

In this paper, we readdress the reactor network synthesis problem, but aim at designs with guaranteed open-loop robust stability. This approach will typically result in a conservative and not necessarily most economical design. However, we avoid this way the problem that a stabilizing feedback controller for an open-loop unstable reactor network may not exist. Furthermore, an open-loop stable reactor network is often preferred in practice because of its inherent safety. The extension of the presented synthesis method to the integrated design of a reactor network and its stabilizing control system in a simultaneous approach is possible and will be treated in future work.

Reactor network synthesis with guaranteed robust stability has only rarely been addressed in literature Dangelmayr and Stewart (1985), Feinberg (1987), Garhyan et al. (2003), Kubicek et al. (1979). Kokossis and Floudas (1994) proposed a methodology based on matrix measures to bound the eigenvalues of the system's Jacobian matrix in order to guarantee stability. Mohideen et al. (1996, 1997) developed an iterative procedure for optimal process design based on the concepts of feasibility and flexibility introduced by Grossmann and coworkers Halemane and Grossmann (1983), Swaney and Grossmann (1985) to achieve robust stability of the designed flowsheet.

The present paper proposes a structured dynamic model for reactor networks consisting of continuous stirred-tank reactors (CSTR) and plug flow reactors (PFR), which defines the superstructure of the network. Stability of the reactor network with possibly idle reactors in the superstructure is guaranteed in the final design under parametric uncertainty by tailored superstructure optimization. Hence, the novel approach presented in this paper extends

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the work of Kokossis and Floudas (1994) in two ways: it is not restricted to non-idle reactors in the superstructure and guarantees robust stability at nominal points. This way, a less conservative approach to design reactor networks with guaranteed robust stability is obtained.

In Section 2, the dynamics of a network of N reactors is modeled in a structured manner by means of $N+2$ interconnected subsystems. In Section 3, a stability constraint involving the Jacobian matrix of the network is introduced and its smoothness is analyzed afterwards. The introduced stability constraint is at the core of this work. Smoothness analysis shows that it is a discontinuous constraint. In Section 4, we first use the introduced stability constraint to directly formulate a design optimization problem. However, because of the difficulties to treat the discontinuities in the introduced stability constraint by numerical optimization methods, the problem is then reformulated by introducing integer variables. This procedure results at the end in a mixed-integer nonlinear program (MINLP) with a robust eigenvalue constraint.

Because of many interrelated issues, the obtained MINLP is still difficult to solve. Firstly, it is an eigenvalue optimization problem of a non-symmetric matrix, which belongs to the class of non-smooth optimization problems. Although some methods for eigenvalue optimization problems are available, e.g., algorithms originating from non-smooth optimization techniques Burke et al. (2002), Overton (2013), Bonnans et al. (2006), Lewis and Overton (2013), methods based on eigenvalue measures Kokossis and Floudas (1994), Mohideen et al. (1997) and methods based on Hurwitz criterion or related theory Blanco and Bandoni (2003), Chang and Sahinidis (2011), it is still a challenging task to solve eigenvalue optimization problems properly. Secondly, uncertain parameters result in a semi-infinite program (SIP), where the inner problem must be solved to global optimality (i.e. global minimums/maximum) in order to guarantee feasibility. Although some results on finding the global minimums of smooth SIP are available, e.g., Mitsos and Tsoukalas (2014), Mitsos et al. (2009) and the references therein, to the authors' knowledge, SIP with a non-smooth inner problem is rarely addressed in the literature.

For these reasons, a tailored two-step solution strategy is proposed at the end of Section 4. In the first step, we consider that there are no parametric uncertainties and assume that the eigenvalue constraint in the MINLP is locally smooth near a local minimum. In this setting, the resulting MINLP has no uncertain parameters and it is locally smooth. Hence, one can apply directly standard MINLP methods Floudas (1995), if the starting points are chosen sufficiently close to the local minimum. In a second step, we consider uncertainties and solve the resulting SIP by fixing all integer variables to those determined in the first step. There exist several algorithms to solve SIP problems Stein and Still (2000, 2003) to a local minimum. Here, we use the normal vector approach (NVA) Mönnigmann and Marquardt (2002), which is related to the local reduction method Diego and Marquardt (2013). A great advantage of this tailored two-step strategy is that, the normal vector approach can be properly initialized using the solutions of step 1 such that good local solutions can be found.

In Section 5, a case study of allyl chloride production with reactor networks including up to five CSTR and up to five PFR is presented. The case study is introduced at the beginning of this paper and used continuously to demonstrate important building blocks of the proposed method.

2. Structured modeling of reactor networks

In this section, we present a structured dynamic model of reactor networks in order to allow for stability considerations during process synthesis. The superstructure consists of N non-isothermal reactors, which are either CSTR or PFR (Fig. 1). The approach firstly models each individual reactor in the network as separate subsystem and then builds their flow connections. An illustrating example is introduced first, which will be used throughout the paper.

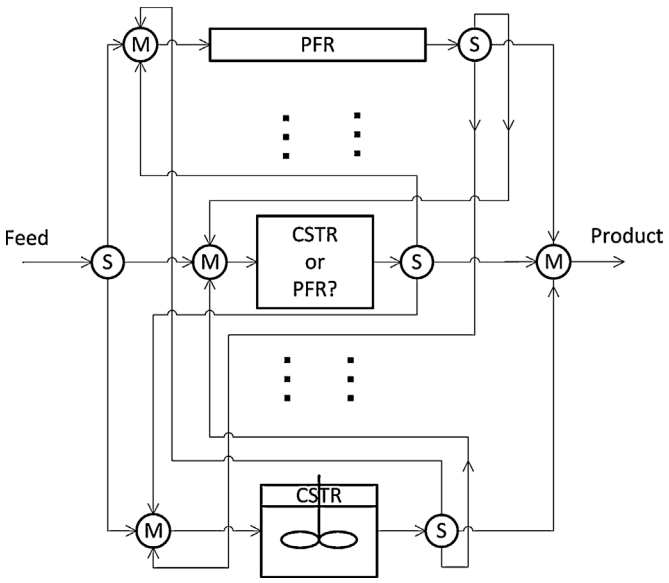

Fig. 1. A superstructure of N-reactor network Kokossis and Floudas (1994). M and S refer to mixing and splitting units. PFR and CSTR refer to reactors.

Example. Allyl chloride can be produced by means of non-catalytic chlorination of propylene in the vapor phase Pahor et al. (2001). The reaction mechanism is as follows:

$$\underbrace{Cl_2}_{C} + \underbrace{CH_2-CHCH_3}_{A} \xrightarrow{k_1} \underbrace{CH_2-CHCH_2Cl}_{B} + HCl,$$

$$\underbrace{Cl_2}_{C} + \underbrace{CH_2-CHCH_2Cl}_{B} \xrightarrow{k_2} ClCH-CHCH_2Cl + HCl,$$

$$\underbrace{Cl_2}_{C} + \underbrace{CH_2-CHCH_3}_{A} \xrightarrow{k_3} CH_2ClCHClCH_3.$$

We use A, B, C to denote propylene, allyl chloride and chlorine. A, C are raw materials, B is the main product, while 1,3-dichloropropene and 1,2-dichloropropane are side products. The reaction kinetics is modeled as

$$\begin{aligned} &r_1(c_A,T)=k_1c_Ac_C, \quad r_2(c_B,T)=k_2c_Bc_C, \quad r_3(c_A,T)=k_3c_Ac_C,\\ &k_1=a_1e^{-15840/RT}, \quad k_2=a_2e^{-23760/RT}, \quad k_3=a_3e^{-7920/RT}. \end{aligned} \tag{1}$$

T [K] denotes temperature, while c_A, c_B, c_C [mol/m^3] denote concentrations of A, B, C, respectively. All parameters can be found in Table 1. The reaction rates for component A, B, C are

$$R_A=-r_1-r_3,\ R_B=r_1-r_2,\ R_C=-r_1-r_2-r_3.$$

□

Table 1
Constants and reaction parameters for allyl chloride production Pahor et al. (2001).

Parameter	Value		Units
a_1	1.5×10^6	Reaction constant	1/s
a_2	4.4×10^8	Reaction constant	1/s
a_3	1.0×10^2	Reaction constant	l/mol s
R	1.987	Gas constant	cal/mol K
H_1	118.82	Reaction heat	kJ/mol
H_2	114.79	Reaction heat	kJ/mol
H_3	183.03	Reaction heat	kJ/mol