Chemical Engineering Science 192 (2018) 306–317

Contents lists available at ScienceDirect

Chemical Engineering Science

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

Efficient calculation of constraint back-offs for optimization under uncertainty: A case study on maleic anhydride synthesis

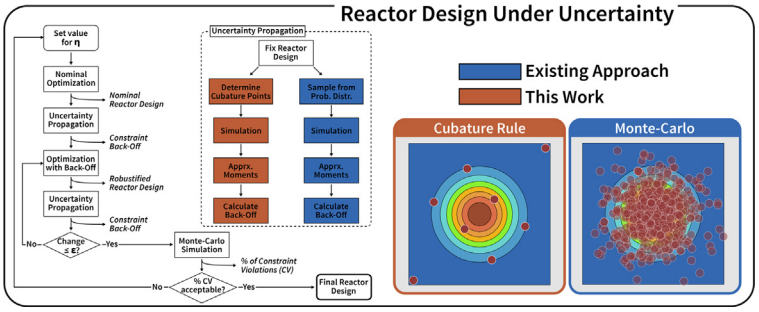
Johannes Maußner, Hannsjörg Freund *

Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), Lehrstuhl für Chemische Reaktionstechnik, Egerlandstr. 3, 91058 Erlangen, Germany

HIGHLIGHTS

- Optimization under uncertainty is increasingly important for reactor design.
- An improved algorithm for reactor design under uncertainty is proposed.
- Specialized cubature rules for uncertainty propagation are applied.
- Significant performance enhancement compared to existing approach.
- New approach demonstrated on the reactor design for maleic anhydride synthesis.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 3 May 2018
Received in revised form 15 June 2018
Accepted 30 June 2018

Keywords:
Optimization under uncertainty
Reactor optimization
Cubature rules
Maleic anhydride
Reaction engineering

ABSTRACT

In the present work we propose an efficient and general algorithm for optimization under uncertainty based on the work of Srinivasan et al. (2003). We use specialized cubature rules to speed up the uncertainty propagation step which results in a significant reduction of the overall computational effort. The approach is illustrated by studying the optimal design of a fixed bed reactor for the synthesis of maleic anhydride from raffinate II feedstock, where the amount of *n*-butane and *n*-butenes in the feed is assumed to be uncertain. Applying the algorithm results in a robustified reactor design which shows significantly less temperature constraint violations and runaway conditions while still satisfying reactor performance criteria such as minimally required conversion and maximum allowable pressure drop.

1. Introduction

The reliability of results obtained from model-based optimization highly depends on the quality of the underlying mathematical models. In chemical engineering applications, the models frequently contain a variety of model parameters which are identified from experimental data and are thus subject to uncertainty (Grossmann and Sargent, 1978; Halemane and Grossmann, 1983). Moreover, chemical processes are affected by uncertainties or fluctuations in process parameters such as, e.g., feed composition or temperature (Rooney and Biegler, 2001; Wendt et al., 2002). Neglecting these uncertainties at the design stage can lead to chemical processes which are very sensitive to parameter variations (Terwiesch and Agarwal, 1995). This sensitivity can lead to the violation of critical constraints and thus to infeasible processes. Hence, many studies have been devoted to optimization under consideration of uncertainty in the chemical engineering community (Bernardo et al., 1999, 2001; Wendt et al., 2002; Halemane and Grossmann, 1983; Shen and Braatz, 2016; Puschke et al., 2017). The present work contributes to this area of research by

* Corresponding author.
E-mail address: hannsjoerg.freund@fau.de (H. Freund).

https://doi.org/10.1016/j.ces.2018.06.079
0009-2509/

Nomenclature

Latin symbols

A	cross-sectional tube area, m^2
b_g	back-off for constraint g
b_{bta}, b_{bte}	inhibition coefficients, Pa^{-1}
$c_{p,g}$	specific heat capacity, $J\ K^{-1}\ kg^{-1}$
d	precision of cubature rule
D_t	tube diameter, m
d_p	particle diameter, m
$\Delta_r H_j^0$	molar standard reaction enthalpy of reaction j, $J\ mol^{-1}$
k	reaction rate constants, $mol\ s^{-1}\ kg^{-1}\ Pa^{-1}$
k_W	overall heat transfer coefficient, $W\ m^{-2}\ K^{-1}$
L_t	tube length, m
$\overline{M}_i$	molecular weight of component i, $kg\ mol^{-1}$
$\overline{M}$	molecular weight of gas phase, $kg\ mol^{-1}$
$\dot{m}$	mass flow rate, $kg\ s^{-1}$
$\dot{n}$	molar flow rate, $mol\ s^{-1}$
N_{MC}	number of Monte-Carlo samples
N	number of integration points for cubature rule
n_θ	number of uncertain parameters
p_i	partial pressure of component i, Pa
P_θ	variance-covariance matrix of random variable θ
$P(\theta)$	joint probability density function of θ
r	reaction rate, $mol\ kg^{-1}\ s^{-1}$
R_t	tube radius, m
R_i	species rate of formation, $mol\ kg^{-1}\ s^{-1}$
STY	space time yield, $mol\ m^{-3}\ s^{-1}$
T	temperature, K
T_W	wall temperature, K
T_{in}	inlet temperature, K
T_c	cooling temperature, K
u_s	superficial velocity, $m\ s^{-1}$
u_i	vector of integration points
w_i	weight fraction of component i or weight factor, –
w	weight function
x_i	mole fraction of component i, –
x	independent variable, e.g., time or reactor coordinate
X_{HC}	conversion of hydrocarbons, –

Greek symbols

α_W	wall heat transfer coefficient, $W\ K^{-1}\ m^{-2}$
ϵ_{bed}	catalyst bed void fraction, –
ϵ	tolerance used in algorithm
μ	expected values
$\nu_{i,j}$	stoichiometric coefficient of component i in jth reaction, –
ρ_{cat}	catalyst density, $kg\ m^{-3}$
η_g	dynamic viscosity of gas phase, Pa s
η	scalar to control conservatism
θ	vector of uncertain parameters
Θ	space of uncertain parameters
λ_{bed}	heat conductivity of catalyst bed, $W\ m^{-1}\ K^{-1}$
λ_f	heat conductivity of fluid, $W\ m^{-1}\ K^{-1}$
λ_r	radial heat conductivity, $W\ m^{-1}\ K^{-1}$
Ω	integration region

Abbreviations

$CU_{3,1}$	cubature rule of precision 3 (version 1)
$CU_{3,2}$	cubature rule of precision 3 (version 2)
$CU_{5,1}$	cubature rule of precision 5 (version 1)
$CU_{5,2}$	cubature rule of precision 5 (version 2)
UT	unscented transformation
HC	hydrocarbon

proposing an improvement to an existing and frequently applied approach to handle inequality constraints under uncertainty.

1.1. Background on case study

The development of the new approach is motivated by the maleic anhydride synthesis from raffinate II feedstock. Maleic anhydride (MA) is an important intermediate in the chemical industry. Modern plants produce maleic anhydride by partial oxidation of *n*-butane in multi-tubular fixed bed reactors (Lesser et al., 2017; Becker, 2002). Brandstädter and Kraushaar-Czarnetzki (2005, 2007) and Brandstädter (2008) studied the synthesis of MA from raffinate II, i.e., the C_4-cut of the steamcracker effluent. Raffinate II contains about 16 mol% *n*-butane, 75 mol% *n*-butenes and 9 mol% of isobutane, isobutene and isopentenes (Arpe, 2007). Raffinate II could be an attractive alternative to a pure *n*-butane feedstock for maleic anhydride synthesis. The advantage is that the costly separation of the C_4-mixture can be avoided when raffinate II is directly converted to MA in a one step process (Brandstädter and Kraushaar-Czarnetzki, 2005). A reactor simulation of an industrial scale multi-tubular fixed bed reactor for the synthesis of MA from raffinate II is provided by Brandstädter and Kraushaar-Czarnetzki (2007) to illustrate its feasibility and potential. An important point to note is that the mole fractions of *n*-butane and *n*-butenes in the raffinate II mixture can vary depending on the cracking process (Arpe, 2007). This uncertainty in the feed composition can lead to unexpected temperature hot spots due to the different reactivities of *n*-butane and *n*-butenes. This results in a higher catalyst deactivation and thus in a loss of productivity. Therefore, the uncertainty in the raffinate II feed composition has to be considered already at the reactor design stage. The synthesis of maleic anhydride from raffinate II thus represents a suitable case study for optimization under uncertainty.

1.2. Scope of the present work

A frequently applied strategy to ensure that critical constraints are satisfied under uncertainty is to introduce a back-off to the respective constraints (Visser et al., 2000; Rossner et al., 2010; Steiner et al., 2005; Telen et al., 2015). The constraint back-off is related to the variance, i.e., the second statistical moment of the respective constraint. Srinivasan et al. (2003) proposed an iterative procedure to efficiently determine the constraint back-off. After solving an initial nominal optimization problem, an uncertainty propagation step is performed to calculate a first back-off value. Subsequently, the optimization is repeated with the modified constraint to yield a robustified reactor design. This procedure is repeated until a suitable back-off value is obtained. In the uncertainty propagation step, a Monte-Carlo simulation is used to determine the variance of the respective constraint. This iterative strategy has been applied by Shi et al. (2016) to the optimization of a solution polymerisation process and by Aydin et al. (2018) to the nonlinear model predictive control of a semi-batch hydroformylation reactor. Recently, Koller et al. (2018) extended and applied the procedure to the simultaneous design, control and scheduling of a multi-product continuous stirred tank reactor system.

A drawback of using a Monte-Carlo approach in the uncertainty propagation step is that possibly a large number of samples is required in order to obtain an accurate approximation of the con-