J. Hasenauer[a,*] · **P. Rumschinski**[b,*] · **S. Waldherr**[a] · **S. Borchers**[b] · **F. Allgöwer**[a] · **R. Findeisen**[b]

Guaranteed Steady State Bounds for Uncertain (Bio-)Chemical Processes using Infeasibility Certificates

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[a]Institute of Systems Theory and Automatic Control, University of Stuttgart,
Pfaffenwaldring 9, 70569 Stuttgart/ Germany
{hasenauer,waldherr,allgower}@ist.uni-stuttgart.de
www.ist.uni-stuttgart.de

[b]Institute for Automation Engineering, Otto-von-Guericke-University Magdeburg,
Universitätsplatz 2, 39106 Magdeburg/ Germany
{philipp.rumschinski,steffen.borchers,rolf.findeisen}@ovgu.de
http://ifatwww.et.uni-magdeburg.de/syst

*Both authors contributed equally.

Abstract Analysis and safety considerations of chemical and biological processes require complete knowledge of the set of all feasible steady states. Nonlinearities, uncertain parameters, and discrete variables complicate the task of obtaining this set. In this paper, the problem of outer-approximating the region of feasible steady states, for processes described by uncertain nonlinear differential algebraic equations including discrete variables and discrete changes in the dynamics, is addressed.

The calculation of the outer bounds is based on a relaxed version of the corresponding feasibility problem. It uses the Lagrange dual problem to obtain certificates for regions in state space not containing steady states. These infeasibility certificates can be computed efficiently by solving a semidefinite program, rendering the calculation of an outer bounding set computationally feasible. The derived method guarantees globally valid outer bounds for the feasible steady states.

The method is exemplified by the analysis of a simple chemical reactor showing parametric uncertainties and large variability due to the appearance of bifurcations characterising the ignition and extinction of a reaction.

Keywords feasible steady state · uncertain nonlinear differential algebraic systems · semidefinite programming · infeasibility certificates · CSTR

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Stuttgart Research Centre for Simulation Technology (SRC SimTech)
SimTech – Cluster of Excellence
Pfaffenwaldring 7a
70569 Stuttgart
publications@simtech.uni-stuttgart.de
www.simtech.uni-stuttgart.de

1 Introduction

Chemical and biochemical processes are often subject to large modeling uncertainties and process disturbances. Precise reaction mechanisms and kinetic parameters might be unknown and operating conditions, e.g. feed flowrate, or feed temperature, can vary. Moreover, used substances in chemical plants are potentially dangerous, e.g. inflammable or explosive; reactions can lead to the release of large amounts of thermal energy. Also stationary temperature and pressure have to stay below critical values and for instance in pharmaceutical processes the variability within the drug production has to be restricted. This makes an in-depth analysis of process uncertainties and their influences on the steady state behavior of the processes essential.

In this paper, we address the problem of computing the set of all feasible steady states of a process described by uncertain hybrid nonlinear differential algebraic equations. Using the set of feasible steady states the stationary process uncertainty can be upper bounded. Furthermore, it can be used to check whether the process operates within previously defined constraints for all possible disturbances, parameter variations, and operating conditions. One exemplary question to be asked is whether thermal runaway of a chemical reactor under specific failure situations can be avoided or if product specifications are met.

Physical processes taking place in chemical plants are mostly continuous. There are, however important discrete phenomena like changes in the physical system, e.g. phase transitions, imposed qualitative changes caused by limitation of the equipment, such as limited tank capacity, discontinuous input signals and process faults (7). To capture continuous as well as discrete phenomena, regime based approaches are used to model the process behavior (21; 14; 13). Often, one refers to this kind of models as hybrid models, because they contain both discrete and continuous dynamical components and an interface describing the interaction between them.

For most nonlinear systems an analytical calculation of the set of feasible states is impossible. Therefore, several methods have been developed for approximating this set. In the context of analysis of trajectories one refers to this research as reachability analysis. Methods developed in this research field are rather efficient if the considered system is linear time-invariant (9) and also for uncertain linear systems some results exist (8). However, if the system under consideration is nonlinear, the approximation of the feasible set is more difficult. Asarin and coworkers developed an approach for two-dimensional systems based on piecewise linear approximation (1) and Ramdani and coworker (18) proposed a method for high dimensional uncertain nonlinear systems using guaranteed set integration, which yields good results for cooperative systems. Nevertheless, the performance of these methods strongly depends on the particular structure of the nonlinear system and in many cases the results are very conservative.

Due to this drawback of set-based approaches, for the analysis of nonlinear systems, often simple Monte-Carlo type methods are employed (20). However, such approaches only provide the complete set of possible steady states in the limit of infinite many samples. Important solutions might be left out, especially for highly nonlinear systems.

In contrast to reachability analysis in this paper only the steady state behavior is studied. The derived method follows the idea presented in the work of Waldherr et al. (25). There, recent advances in the field of semidefinite programming (SDP) (16; 5) are employed to compute certificates that a given set in state space cannot contain a steady state for any feasible model parameterization. A very similar approach was earlier proposed by Küpfer et al. (12) for parameter estimation and later extended to dynamical systems by Borchers and coworkers (2). However, these methods are restricted to systems described by polynomial or rational vector fields, which is rarely the case for chemical processes. Furthermore, discrete variables or parameters have not been considered.

In the following, an approach will be presented, which overcomes this shortcoming and allows the outer approximation of the set of all feasible steady states of a process described by uncertain hybrid nonlinear differential algebraic equations with non-polynomial vector fields. Thus, systems combining continuous dynamics with logic or discrete components can be studied. Furthermore, in comparison with (25) a more elaborate algorithm is proposed to obtain a more precise approximation of the set of feasible steady states, for systems with steady state multiplicity.

The derived method can be used as a structured approach to obtain the bifurcation diagram of uncertain systems. Bifurcation analysis is of particular interest in the analysis of uncertain nonlinear systems and can help to obtain a better understanding of effects which may be observed , e.g. steady

state multiplicity or saddle-node bifurcations (24). Calculating the bifurcation diagram shows the potential of the derived method since up to now to the best knowledge of the authors no method exists which can be employed to obtain the envelop of bifurcation diagrams of uncertain hybrid differential systems.

The remainder of this paper is structured as follows: In Section 2 the problem of bounding the set of steady states for processes described by non-polynomial hybrid differential algebraic equations is presented. Section 3 contains a formalization of the problem. In Section 4 the problem is restated as a feasibility problem, which is relaxed to a semidefinite program. Given this formulation an algorithm is developed which can be used to determine an outer approximation of the set of feasible steady states. In Section 5 we provide as an example the analysis of a uncertain CSTR, before in Section 6 we conclude the paper.

Mathematical notation: The space of real symmetric $n \times n$ matrices is denoted as $\mathcal{S}^n$. The positive semidefiniteness of a symmetric matrix $X \in \mathcal{S}^n$ is denoted $X \succeq 0$ and the trace of X by $\mathrm{tr}(X)$. Furthermore, $\mathcal{I}_\delta^x$ denotes the discrete set $\{1, \ldots, n_\delta^x\}$, where n_δ^x is the dimension of a vector x for a particular discrete variable δ.

2 Problem statement

For the purpose of this work, we consider the set of all feasible steady states of processes described by hybrid differential algebraic systems. This system class is quite general and able to describe both continuous and discrete dynamical behavior. Classical chemical and biochemical reaction networks belong to this system class, but also more complicated systems. Well known examples are chemical reactors which allow for phase transitions and systems with discrete variables/inputs, e.g. opening of a valve, or on/off switching of a heater.

In many cases using a hybrid description of a process is more natural than deriving a single ordinary differential equation model, holding for all possible operating conditions at once. For every operating condition a specifically tailored model can be employed, and the transitions between the models are defined by switching surfaces.

According to Branicky et al. (4) hybrid differential algebraic systems can be modeled as 4-tuples

$$\left[\mathcal{I}^\delta,\ \mathbf{\Sigma},\ \mathbf{A},\ \mathbf{G}\right], \tag{1}$$

in which $\mathcal{I}^\delta$ is the set of discrete states. $\mathbf{\Sigma} = \{\Sigma_\delta\}_{\delta \in \mathcal{I}^\delta}$ denotes the collection of controlled dynamical systems with $\Sigma_\delta = [F_\delta, X_\delta, U_\delta, P_\delta]$, with the continuous state space $X_\delta \subseteq \mathbb{R}^{n_\delta^x}$, the continuous input space $U_\delta \subseteq \mathbb{R}^{n_\delta^u}$, the continuous parameter space $P_\delta \subseteq \mathbb{R}^{n_\delta^p}$, and continuous dynamics F_δ. The collection of jump sets is denoted by $\mathbf{A} = \{A_\delta\}_{\delta \in \mathcal{I}^\delta}$, with $A_\delta \subset \mathcal{X}_\delta$. $\mathbf{G} = \{G_\delta\}_{\delta \in \mathcal{I}^\delta}$ is the jump transition map.

A hybrid differential algebraic system (1) can be seen as an automaton. Each node of this automaton is a dynamical system described by Σ_δ,

$$0 = F_\delta(\dot{x}, x, u, p), \quad x(0) = x_0 \tag{2}$$

in which $x \in X_\delta$ denotes the vector of continuous state variables, $u \in U_\delta$ is the vector of continuous control inputs, and $p \in P_\delta$ the vector of process parameters. The index δ is the label of the node. The different nodes are connected by edges which represent possible transition between dynamical systems Σ_δ, labeled by the appropriate transition condition and the update of the continuous state (4). The interconnection from node δ to all other nodes and the update rules are defined in the jump transition map G_δ. A jump to another node can only happen if the states of the dynamical system Σ_δ are contained in the jump set $A_\delta \subseteq X_\delta$.

Note that for simplicity we do not distinguish here between controlled and uncontrolled jumps, as this is not required for the rest of this work. For a more detailed discussion of hybrid dynamical systems we refer to (4).

For the remainder of this paper, we are only interested in the steady states of (1). The steady states have to lie in one of the continuous state spaces X_δ. Therefore, the interconnection structure $\mathbf{A}$ and $\mathbf{G}$

do not have to be considered, which considerably simplifies the analysis of (1). The system can thus be written as

$$\left\{ \begin{array}{l} 0 = F_\delta(\dot{x}, x, u, p),\, x(0) = x_0 \\ 0 \leq k_\delta(x, u, p), \end{array} \right\}_{\delta \in \mathcal{I}^\delta} \tag{3}$$

in which the constraint $x \in X_\delta$ is realized via the vector-valued function k_δ. k_δ is positive if and only of $x \in X_\delta$. Note that here and in the following the dependence of x, u, and p on δ is not indicated, but implicitly assumed.

Given this description of the model class, the problem of finding all feasible steady states of system (3) can be formalized as:

Problem 1 *(Set of feasible steady states): Given the sets $\mathcal{I}^\delta$, $\mathcal{P}_\delta \subseteq P_\delta$, $\mathcal{U}_\delta \subseteq U_\delta$, compute the set $\mathcal{X}^*$, which contains all feasible steady states of* (3)*.*

The sets $\mathcal{P}_\delta$ and $\mathcal{U}_\delta$ are hereby the sets of considered feasible parameters and inputs, in case of uncertainties, and $u \in \mathcal{U}_\delta$ is assumed to be stationary.

Note that the set of feasible steady states for a given decision variable $\delta \in \mathcal{I}^\delta$ is defined by

$$\begin{aligned} 0 &= f_\delta(x, u, p) \\ 0 &\leq k_\delta(x, u, p), \end{aligned} \tag{4}$$

with $f_\delta(x, u, p) := F_\delta(0, x, u, p)$. Hence, problem (1) can be split into n^δ subproblems. For each subproblem one obtains a set of feasible steady states

$$\mathcal{X}_\delta^* = \{x \in \mathbb{R}^{n_\delta^x} \mid \exists\, p \in \mathcal{P}_\delta, u \in \mathcal{U}_\delta : f_\delta(x, u, p) = 0 \wedge k_\delta(x, u, p) \geq 0\}. \tag{5}$$

The whole set of feasible steady states is given by the union of all steady states

$$\mathcal{X}^* = \bigcup_{\delta \in \mathcal{I}^\delta} \mathcal{X}_\delta^* \times \{\delta\}. \tag{6}$$

Unfortunately, the set $\mathcal{X}^*$ is in general highly complex and cannot be determined precisely. Therefore, we address in the following the problem of computing an outer-approximation $\hat{\mathcal{X}}^*$ of $\mathcal{X}^*$, such that $\mathcal{X}^* \subseteq \hat{\mathcal{X}}^*$. This was previously done by Waldherr et al. (25) for ordinary differential equations with polynomial right hand sides. The main contribution of this paper is the generalization of this result to hybrid non-polynomial DAE systems, what for instance enables us to apply the method for the analysis of chemical reactors.

3 Bounding by piece-wise polynomial functions

The computational method we propose allows to handle uncertain systems that are described by polynomial equations. Therefore, (4) has to be transformed to a set of uncertain polynomial equations. In the case that f_δ is rational, this can be achieved by multiplication with the denominator. However, if non polynomial/rational terms appear, such as the Arrhenius term, then the system has to be rewritten or approximated by a polynomial system.

Ohtsuka (15) showed that any system with smooth non-polynomial nonlinearities can be converted to a polynomial system of larger state dimension, which is restricted via equality constraints to a manifold of the dimension of the original system. Unfortunately, in many cases the equality constraints are non-polynomial. To overcome this limitation, we apply a different method, which achieves a comparable result without enlarging the state space.

Piece-wise polynomial functions:

In case that f_δ is piece-wise polynomial, e.g. piece-wise linear, the state space can be partitioned into different intervals. This leads to an increase in the number of decision variables, but no approximation is required, Figure 1 illustrates this for the saturation function, which appears for instance if a process contains flow limiting valves. It has to be emphasized that in cases like this, the partitioning depends on the state, which induces constraints $k_\delta(x, u, p) \geq 0$.

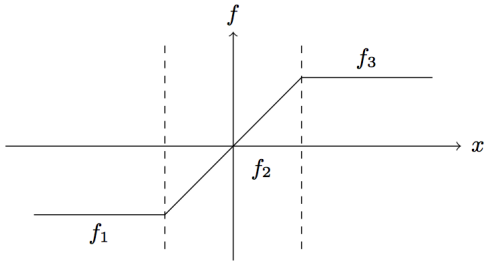

Fig. 1 Saturation function as example for the partitioning of piece-wise polynomial functions.

General nonlinear functions:

For functions which are not piece-wise polynomial, e.g. the exponential term in the Arrhenius like rate constant, polynomial lower and upper bounds can be introduced as

$$g^{(1)}_{\delta,i}(x,u,p) \leq f_{\delta,i}(x,u,p) \leq g^{(2)}_{\delta,i}(x,u,p),\ \forall x \in \mathcal{X}_\delta, u \in \mathcal{U}_\delta, p \in \mathcal{P}_\delta, i \in \mathcal{I}^x_\delta \tag{7}$$

where the index i corresponds to the i^{th} element of the respective vector function. Using these bounds it can be shown that

$$\begin{aligned}\mathcal{X}^*_\delta \subseteq \{\ x \in \mathbb{R}^{n^x_\delta} | \exists\ p \in \mathcal{P}_\delta, u \in \mathcal{U}_\delta, c \in [0,1]^n :\\ c_i g^{(1)}_{\delta,i}(x,u,p) + (1-c_i) g^{(2)}_{\delta,i}(x,u,p) = 0, \forall i \in \mathcal{I}^x_\delta \Big\}.\end{aligned} \tag{8}$$

Hence, the steady state constraint $f_\delta(x,u,p) = 0$ can be substituted by the polynomial constraint

$$c_i g^{(1)}_{\delta,i}(x,u,p) + (1-c_i) g^{(2)}_{\delta,i}(x,u,p) = 0, \quad c_i \in [0,1], i \in \mathcal{I}^x_\delta \tag{9}$$

where c has to be appended to p. This step corresponds to a constraint relaxation and $||f_{\delta,i}(x,u,p) - g^{(j)}_{\delta,i}(x,u,p)|| \ll 1$ should be enforced to keep the difference between $\mathcal{X}^*_\delta$ and the set of solutions of the relaxed problem small.

Combinations of the methods, e.g. rational, polynomial and nonlinear functions are possible, see Section 5 for an example.

In the following, we assume that the system is polynomial or already approximated by a polynomial system as described in this section.

4 Bounding steady states

In this section a method to compute an outer approximation of the state space region containing all steady states is derived. For this purpose we define the feasibility problem,

$$(P): \begin{cases} \text{find} & \delta \in \mathcal{I}^\delta,\ x \in \mathbb{R}^{n^x}, u \in \mathbb{R}^{n^u}, p \in \mathbb{R}^{n^p} \\ \text{subject to} & f_\delta(x,u,p) = 0 \\ & k_\delta(x,u,p) \geq 0 \\ & x \in \mathcal{X}_\delta,\ u \in \mathcal{U}_\delta,\ p \in \mathcal{P}_\delta, \end{cases}$$

which is in the following used for the classification of the set $\mathcal{X} = \bigcup_{\delta \in \mathcal{I}^\delta} \mathcal{X}_\delta \times \{\delta\}$ in the state space. If (P) is infeasible, $\mathcal{X}$ cannot contain any equilibrium points. (P) is called a mixed integer nonlinear program. Unfortunately, the feasibility problem (P) is in general non-convex and NP-hard.

Küpfer et al. (12) proposed a framework for relaxing a polynomial non-convex feasibility problem to a semidefinite program (SDP). Due to inherent convexity of SDPs, these problems can be solved computationally efficient, e.g. via primal-dual interior point methods. In the following, we present an approach which is based on the work of Küpfer et al. (12) and has been used for analysis of the set of feasible steady states in the case of biochemical reaction networks in (25).

For the relaxation of (P) to a SDP, the original feasibility problem is at first rewritten as a quadratic feasibility problem (QP), for each δ. Therefore, the vector $\xi \in \mathbb{R}^{n_\delta^\xi}$ are introduced, which consists of the monomials of the model equation (4), i.e.

$$\xi = (1,\ x_i,\ u_j,\ p_k,\ x_i u_j,\ x_i p_k,\ u_j p_k, \ldots)^T \tag{10}$$

for all $x \in \mathcal{X}_\delta, u \in \mathcal{U}_\delta, p \in \mathcal{P}_\delta, i \in \mathcal{I}_\delta^x$, $j \in \mathcal{I}_\delta^u$ and $k \in \mathcal{I}_\delta^p$. Using this monomial vectors ξ, the vector of equality constraints $f_\delta(x,u,p) = 0$ can be transformed to

$$0 = f_{\delta,i}(x,u,p) = \xi^T Q_i \xi, \quad i \in \mathcal{I}_\delta^x, \tag{11}$$

in which $f_{\delta,i}$ is the ith row of f_δ, and $Q_i \in \mathcal{S}^{n_\delta^\xi}$. Note that for higher order polynomial terms, additional constraints have to be introduced. For instance if ξ contains the second order term $x_1 p_1$, the constraint $x_1 p_1 = x_1 \cdot p_1$ must be introduced to express the dependency of the higher order monomial on the first order monomials. Note that there are in general multiple possibilities to decompose a specific term into monomials, which provides additional degrees of freedom. The decomposition leads to additional constraints of the form

$$\xi^T Q_i \xi = 0, \quad i \in \mathcal{I}_\delta^{ac}, \tag{12}$$

in which $Q_i \in \mathcal{S}^{n_\delta^\xi}$, $\mathcal{I}_\delta^{ac} = \{n_\delta^x + 1, \ldots, n_\delta^x + n_\delta^{ac}\}$, and n_δ^{ac} is the number of such dependencies. To simplify the overall notation we define $\mathcal{I}_\delta^{eq} = \mathcal{I}_\delta^x \cup \mathcal{I}_\delta^{ac}$.

Besides the equality constraints, also the inequality constraints $x \in \mathcal{X}_\delta$, $u \in \mathcal{U}_\delta$, $p \in \mathcal{P}_\delta$, and $k_\delta(x,p,u) \geq 0$ are transformed. To simplify the notation we require $\mathcal{X}_\delta$, $\mathcal{U}_\delta$, and $\mathcal{P}_\delta$ to be given as convex polytopes. In this case, there exists a matrix $B \in \mathbb{R}^{n_\delta^b \times n_\delta^\xi}$ and a choice of the monomial vector ξ such that

$$\begin{pmatrix} x \in \mathcal{X}_\delta \\ u \in \mathcal{U}_\delta \\ p \in \mathcal{P}_\delta \\ k_\delta(x,p,u) \geq 0 \end{pmatrix} \Leftrightarrow B\xi \geq 0. \tag{13}$$

Hereby, n_δ^b is the total number of constraints. Note that also the sets $\mathcal{X}_\delta$, $\mathcal{U}_\delta$, and $\mathcal{P}_\delta$ whose boundaries are determined by polynomial or rational constraints in x, p, and u can be handled in this framework.

The original feasibility problem (P) can then be restated as

$$(QP): \begin{cases} \text{find} & \xi \in \mathbb{R}^{n_\delta^\xi},\ \delta \in \mathcal{I}^\delta \\ \text{subject to} & \xi^T Q_i \xi = 0,\ i \in \mathcal{I}_\delta^{eq} \\ & B\xi \geq 0 \\ & \xi_1 = 1. \end{cases}$$

Using the method suggested by Parrilo (17), (QP) is subsequently relaxed to a SDP, for each δ, by introducing the matrix

$$\Xi = \xi \xi^T, \tag{14}$$

with $\Xi \in \mathcal{S}^{n_\delta^\xi}$ and substituting the appearing non-convex constraint $\text{rank}(\Xi) = 1$ with the convex constraint $\Xi \succeq 0$, which is implicitely satisfied for all Ξ satisfying (14). This leads to the relaxed feasibility problem

$$(RP): \begin{cases} \text{find} & \Xi \in \mathcal{S}^{n_\delta^\xi},\ \delta \in \mathcal{I}^\delta \\ \text{subject to} & \text{tr}(Q_i \Xi) = 0,\ i \in \mathcal{I}_\delta^{eq} \\ & B\Xi e_1 \geq 0 \\ & B\Xi B^T \geq 0 \\ & \text{tr}(e_1 e_1^T \Xi) = 1 \\ & \Xi \succeq 0, \end{cases}$$

in which $e_1 = (1, 0, \ldots, 0)^T \in \mathbb{R}^{n_\delta^\xi}$. Note that the relaxation may induce additional solutions. To reduce conservatism, the redundant constraint $B\Xi B^T \geq 0$ is added, which is fulfilled by every solution of the problem (QP) (12).

From (RP) one can derive the set of Lagrange dual problems $\{(DP_\delta)\}_{\delta\in\mathcal{I}^\delta}$, with

$$(DP_\delta): \begin{cases} \text{maximize } \nu_1 \\ \text{subject to } e_1\lambda_1^T B + B^T\lambda_1 e_1^T + B^T\lambda_2 B \\ \qquad +\lambda_3 + \nu_1 e_1 e_1^T + \sum_{i\in\mathcal{I}_\delta^{eq}} \nu_{2,i} Q_i = 0 \\ \qquad \lambda_1 \geq 0,\ \lambda_2 \geq 0,\ \lambda_3 \succeq 0, \end{cases}$$

in which the Lagrangian multipliers are $\lambda_1 \in \mathbb{R}^{n_\delta^b}$, $\lambda_2 \in \mathcal{S}^{n_\delta^b}$, $\lambda_3 \in \mathcal{S}^{n_\delta^\xi}$, $\nu_1 \in \mathbb{R}$, and $\nu_2 \in \mathbb{R}^{n_\delta^{eq}}$ (25). Using the dual problem, one can obtain an infeasibility certificate for the original problem.

Lemma 1 *Let $\nu_1^{\delta,*}$ be the optimal cost of (DP_δ). If*

$$\inf\left\{\nu_1^{\delta,*} \mid \delta\in\mathcal{I}^\delta\right\} = \infty, \tag{15}$$

then the original feasibility problem (P) is infeasible.

This follows directly from weak duality and the conservatism of the made relaxations. Only if the Lagrangian dual problem is unbounded from above for all $\delta\in\mathcal{I}^\delta$ the infeasibility of (P) can be guaranteed.

The advantage of the formulation using the Lagrange duals is that all subproblems are convex and if no solution is found, this is a certificate that no solution exists.

4.1 Algorithm

Using the Lagrangian dual problems $\{(DP_\delta)\}_{\delta\in\mathcal{I}^\delta}$, certificates for the infeasibility of (P) can then be computed. This allows to exploit $\{(DP_\delta)\}_{\delta\in\mathcal{I}^\delta}$ to determine an outer approximation $\hat{\mathcal{X}}^*$ of $\mathcal{X}^*$. In this work, this is done using a multi-dimensional bisection algorithm (11).

Starting from the initial set $\mathcal{X}_0$ a recursive bisection of $\mathcal{X}_0 = \bigcup_{\delta\in\mathcal{I}^\delta} \mathcal{X}_{\delta,0}\times\{\delta\}$ is performed. For each of the resulting subsets $\mathcal{X}_i$ arising in the bisection, the corresponding set of dual problems $\{(DP_\delta)\}_{\delta\in\mathcal{I}^\delta}$ are analyzed and it is tried to compute infeasibility certificates for $\mathcal{X}_i$. Successful computation of an infeasibility certificate assures that $\mathcal{X}_i$ does not intersect the set of feasible steady states $\mathcal{X}^*$. If no certificate can be obtained, $\mathcal{X}_i$ is bisected, and it is tried to obtain an infeasibility certificate for the subsets. An approximation $\hat{\mathcal{X}}^*$ of the set of feasible steady states is finally given by

$$\hat{\mathcal{X}}^* = \mathcal{X}_0 \setminus \bigcup_I \mathcal{X}_I \tag{16}$$

where $\mathcal{X}_I$ are the sets for which an infeasibility certificate could be obtained. The basic implementation can be summarized as follows:

Algorithm: $\hat{\mathcal{X}}^* = \text{Analyze-}\mathcal{X}(\mathcal{X},\mathcal{U},\mathcal{P})$

1. If $\mathrm{V}(\mathcal{X}) < \epsilon$, return $\hat{\mathcal{X}}^* = \mathcal{X}$
2. Check feasibility of $DP_\delta(\mathcal{X}_\delta,\mathcal{U}_\delta,\mathcal{P}_\delta)$, $\forall\delta\in\mathcal{I}^\delta$
3. If $\inf\left\{\nu_1^{\delta,*} \mid \delta\in\mathcal{I}^\delta\right\} = \infty$, return $\hat{\mathcal{X}}^* = \emptyset$
4. If $\inf\left\{\nu_1^{\delta,*} \mid \delta\in\mathcal{I}^\delta\right\} \neq \infty$:
   4.1. Bisection of $\mathcal{X}$ in $\mathcal{X}_1$ and $\mathcal{X}_2$
   4.2. $\hat{\mathcal{X}}_1^* = \text{Analyze-}\mathcal{X}(\mathcal{X}_1,\mathcal{U},\mathcal{P})$
   4.3. $\hat{\mathcal{X}}_2^* = \text{Analyze-}\mathcal{X}(\mathcal{X}_2,\mathcal{U},\mathcal{P})$
   4.4. Return $\hat{\mathcal{X}}^* = \hat{\mathcal{X}}_1^* \cup \hat{\mathcal{X}}_2^*$

This algorithm is called recursively until the volume, $V(\mathcal{X}) = \int_{\mathcal{X}} dp$, of a test set $\mathcal{X}$ is smaller than a tolerance ϵ. For a more detailed discussion of this bisection algorithm we refer to (11). The algorithm is implemented in `Matlab`. For solving the dual problems $\{(DP_\delta)\}_{\delta\in\mathcal{I}^\delta}$ the open source software toolbox `SeDuMi` is used (22).

Remark 1 *Note that for the application of this algorithm an initial set $\mathcal{X}_0$ must be chosen. If a guarantee is needed that an outer approximation of $\mathcal{X}^*$ containing all feasible equilibrium points is found, $\mathcal{X}^* \subseteq \mathcal{X}_0$ must hold. In typical applications this is not a restriction because a suitable $\mathcal{X}_0$ can often easily be determined from physical insight into the problem.*

4.2 Reduction of computational effort

The advantage of the formulation in terms of the Lagrangian dual is the inherent convexity property of $\{(DP_\delta)\}_{\delta\in\mathcal{I}^\delta}$, due to which $\{(DP_\delta)\}_{\delta\in\mathcal{I}^\delta}$ can be solved in polynomial time. The original problem (P) on the other hand is NP-hard and infeasibility of (P) cannot be certified directly.

Despite the fact that (DP_δ) can be solved in polynomial time the computational effort might still become intractable. Already for medium scale systems the number of associated optimization variables n_o is large. It grows in the order of $\mathcal{O}^2(n^\xi)$ with the number of uncertain variables, e.g. states, parameters and inputs. Furthermore, the dominating time for solving these problems is the cost for solving a linear program, which is in general of order $\mathcal{O}^3(n_o)$ (3). Thus the effort for solving (DP_δ) grows in general in the sixth order in the number of uncertain variables. Clearly, for large systems this might be inhibitive if (DP_δ) does not exhibit a particular structure, that can be exploited for speeding up the solution process, e.g. sparsity in (bio-)chemical reaction networks.

Additionally, the cardinality of $\mathcal{I}^\delta$ influences directly the necessary computational effort, as $(DP)_\delta$ has to be solved for all $\delta \in \mathcal{I}^\delta$ independently. In case that $\text{card}(\mathcal{I}^\delta) \gg 1$, checking all the distinct combinations of decision variables can become very costly. One possibility to reduce the problem size is to divide $\mathcal{I}^\delta$ into a smaller number of subsets $\mathcal{D}_d \subseteq \mathcal{I}^\delta$, such that

$$\mathcal{I}^\delta = \bigcup_{d\in\mathcal{I}^d} \mathcal{D}^d. \tag{17}$$

Here the nodes $\delta \in \mathcal{D}^d$ are merged to one common node, which contains $\text{card}(\mathcal{D}^d)$ nodes of the original system. For notational simplicity and to avoid extreme conservatism, we assume here that the state variables, parameters, inputs and constraints have the same physical meaning for all $\delta \in \mathcal{D}^d$. In this case, the feasibility problem (P) can then be restated as

$$(R): \begin{cases} \text{find} & d \in \mathcal{I}^d, x \in \mathbb{R}^{n_d^x}, u \in \mathbb{R}^{n_d^u}, p \in \mathbb{R}^{n_d^p} \\ \text{subject to} & \sum\limits_{\delta\in\mathcal{D}^d} c_\delta f_\delta(x,u,p) = 0 \\ & \sum\limits_{\delta\in\mathcal{D}^d} c_\delta k_\delta(x,u,p) \geq 0 \\ & \sum\limits_{\delta\in\mathcal{D}^d} c_\delta = 1 \\ & x \in \mathcal{X}_d, u \in \mathcal{U}_d, p \in \mathcal{P}_d, c_\delta \in [0,1] \forall \delta \in \mathcal{D}^d, \end{cases}$$

in which $\mathcal{P}_d = \cup_{\delta\in\mathcal{D}_d} P_\delta, \mathcal{U}_d = \cup_{\delta\in\mathcal{D}_d} U_\delta$.

The feasibility problem (R) is a relaxed version of (P), since the solution set of (R) is a superset of the solution set of (P). This holds, as a solution $\{\delta^s, x^s, u^s, p^s\}$ of (P) with $f_{\delta^s}(x^s,u^s,p^s) = 0$ and $k_{\delta^s}(x^s,u^s,p^s) \geq 0$ is also a solution of (R), for $c_\delta = 0\ \forall \delta \neq \delta^s$ and $c_{\delta^s} = 1$. Thus, the solution set of (R) contains the solution set of (P), but for c_δ chosen differently, additional solutions may exist.

For this relaxed infeasibility (R) again the dual problem can be derived using the same tools and performing the same relaxation as done before. The main differences are that the vector ξ contains now also the new variables c_i and additional equality and inequality constraints have to be considered.

The advantage of (R) is that a lower number of dual problems has to be considered in order to show the infeasibility of (P). If this is combined with a hierarchical refinement of the subdivision dependent on the feasibility of (R), the computational effort to check the infeasibility of a set $\mathcal{X}$ can be reduced dramatically.

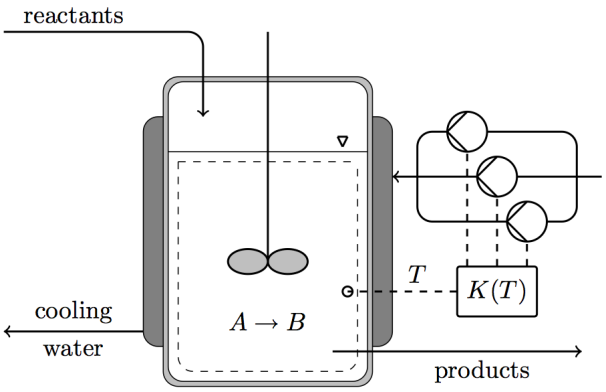

Fig. 2 Schematic of the considered simple CSTR.

5 Analysis of a CSTR

In order to illustrate the proposed approach the set of feasible steady states of a CSTR is studied. The reactor considered consists of a simple vessel filled with fluid stirred by an impeller, an inflow and an outflow, as depicted in Figure 2. To avoid over-temperature active water-cooling, which is provided by three pumps, is considered. The objective is now to classify the set of steady states assuming uncertain parameters and possible pump failures.

5.1 System description

We assume in the following constant reaction volume and adiabatic conditions. The reaction in the reactor is given by a first-order, exothermal liquid-phase reaction

$$A \xrightarrow{k} B.$$

The conversion rate is of the form $R = k(T)c_A$, in which the reaction rate constant is modeled using Arrhenius' equation,

$$k(T) = k_\infty e^{-\frac{E}{RT}}. \tag{18}$$

Simple mass and energy balances lead to the following set of ordinary differential equations:

$$\begin{aligned} \frac{dc_A}{dt} &= \frac{1}{\theta}(c_{Af} - c_A) - k(T)c_A \\ \frac{dT}{dt} &= \frac{1}{\theta}(T_f - T) - \frac{\Delta H_R}{C_p\rho}k(T)c_A - \frac{k_w A_w}{V_R C_p \rho}(T_w - T)K(T, m_p), \end{aligned} \tag{19}$$

capturing the dynamics of the CSTR (19). The state variables are the concentration c_A of reactant A, and the reactor temperature T. The parameters are the mean residence time $\theta = V_R/Q$, the reactor volume V_R, the flowrate Q, the concentration of A in the feed stream c_{Af}, the feed stream temperature T_f, the reaction enthalpy ΔH_R, the heat capacity of the fluid C_p, the fluid density ρ, the cooling water temperature T_c, the effective heat exchange coefficient $\frac{k_w A_w}{V_R}$, and the number of operating cooling pumps m_p.

The over-temperature controller is denoted by $K(T, M_p)$. This controller regulates the active cooling water flow to keep the temperature below the critical reactor temperature T_{crit}. This controller is inactive as long as the temperature stays below a threshold temperature T_{th}. Above this threshold, the cooling water flowrate increases linearly with the temperature until the maximal flow rate is reached. This over-temperature control is a proportional control with input saturation and threshold, implemented via the control law

$$K(T, m_p) = \begin{cases} \max\{k_c(T - T_{th}), m_p\} & \text{for } T > T_{th} \\ 0 & \text{otherwise,} \end{cases} \tag{20}$$

Table 1 Parameter values.

Parameter	Value	Units	Uncertainty
T_f	298	K	±2K
c_{Af}	2.0	kmol/m^3	±2%
k_∞	5.0×10^8	1/min	±2%
E/R	8.0×10^3	K	±2%
ΔH_R	-3.0×10^5	kJ/kmol	±2%
C_p	4.0	KJ/(kg K)	–
ρ	10^3	kg/m^3	–
θ	10	min	–
$\frac{k_w A_w}{V_R}$	40	kJ/(min K m^3)	–
T_w	298	K	±2K
m_p	3.0	–	–
k_c	0.1	1/K	–
T_{th}	390	K	–

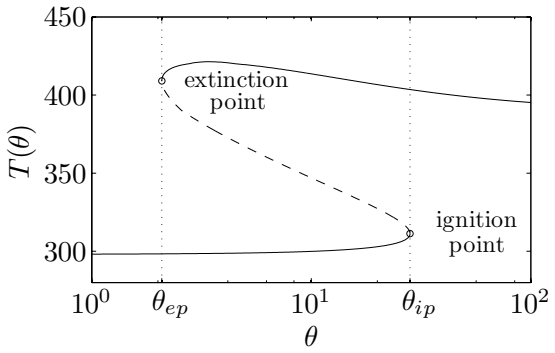

Fig. 3 Bifurcation diagram of CSTR without parameter uncertainties.

in which k_c is the controller gain. The numerical values of the nominal parameters are provided in Table 1.

Note that the considered system can be modeled as a hybrid differential system. The decision variable is hereby directly connected to the temperature. At T_{th} and $T = T_{th} + \frac{m_p}{k_c}$ a change in the systems dynamics occurs. Additionally, the number of operating pumps is discrete, which causes additional switching if pump failures are occur.

5.2 Analysis of the nominal CSTR

In case that all parameters are known, one can exactly predict how the reactor behaves under different operating conditions. Since the mean residence time θ is the easiest parameter to manipulate, the operating condition is often defined in terms of θ. Using continuation methods it is possible to numerically compute the steady state curve (bifuraction diagramm) for varying residence times (6), as shown in Figure 3. θ_{ep} and θ_{ip} denote the mean residence time at the extinction and the ignition point respectively. The computed bifurcation diagram can be used to determine precisely the set of feasible operating points if no uncertainties are present.

5.3 Analysis of CSTR with parameter uncertainties

Unfortunately, in practice, most parameters are subject to uncertainties. In this case, calculating the set of feasible steady states is significantly more challenging. Typically, sampling based techniques such as Monte-Carlo like methods are used. These allow to approximate the union of all feasible equilibrium points $\mathcal{X}_s^*$. However, as for all Monte-Carlo like methods no outer bounds can be provided.

Our approach overcomes this problem and enables us to compute an guaranteed outer approximation of the set of feasible equilibrium points of the uncertain system.

Initial outer bounding set $\mathcal{X}_0$

In order to apply the proposed method, a set $\mathcal{X}_0 \in \mathbb{R}^n$ which contains all feasible steady states of the uncertain system has to be determined. This set $\mathcal{X}_0$ can be arbitrary large but has to be bounded. For system (19) such a conservative set is given by

$$\mathcal{X}_0 = \left\{ [c_A, T]^T \in \mathbb{R}^n | 0 \leq c_A \leq c_{Af,max}, T_{f,min} \leq T \leq T_{f,max} - \frac{\Delta H_{R,max}}{C_{p,min}\rho} \right\},$$

where p_{min} and p_{max} are minimal and maximal values of parameter p under the given uncertainties. This outer bounding set $\mathcal{X}_0$ can be determined using the physical properties of the system and the state equations (19).

Note that these outer bounds are extremely conservative and are strongly refined by in the following by the proposed algorithm.

Bounding the rate constant $k(T)$

Besides the set $\mathcal{X}_0$, the presented method also requires a polynomial systems. Hence, the Arrhenius-like rate constant has to be bounded from below and from above using polynomial functions, as outlined in Section 3. In this paper $k(T)$ is bounded via three linear functions,

$$\max(g_1(T), g_2(T)) \leq k(T) \leq g_3(T), \quad \forall T \in [T_{min}, T_{max}], \tag{21}$$

as depicted in Figure 4, in which

$$g_1(T) = k_{min}(T_{min}) \tag{22}$$

$$g_2(T) = \frac{E k_{min}(T_{max})}{R T_{max}^2}(T - T_{max}) + k_{min}(T_{max}) \tag{23}$$

$$g_3(T) = \frac{k_{max}(T_{max}) - k_{max}(T_{min})}{T_{max} - T_{min}}(T - T_{min}) + k_{max}(T_{min}), \tag{24}$$

and

$$k_{min}(T) = k_{\infty,min} e^{-\left(\frac{E}{R}\right)_{max} \frac{1}{T}}, \tag{25}$$

$$k_{max}(T) = k_{\infty,max} e^{-\left(\frac{E}{R}\right)_{min} \frac{1}{T}}. \tag{26}$$

This approach to outer bound $k(T)$ is simple and has the disadvantage that the bounding of $k(T)$ is less precise if the difference of T_{min} and T_{max} becomes large. Therefore, we do not use a static approximation but rather select g_1, g_2 and g_3 in each iteration of the bisection algorithm dependent on the box $\mathcal{X}$ in state space currently under consideration. This adaption allows to keep the overestimation of the set of feasible steady states small.

One could of course choose other methods to bound $k(T)$, for instance based on high order polynomials and the Taylor series expansion. However, the computational effort to solve the semidefinite program once will increase significantly and the presented simplistic approach will be computationally more efficient for the considered example system.

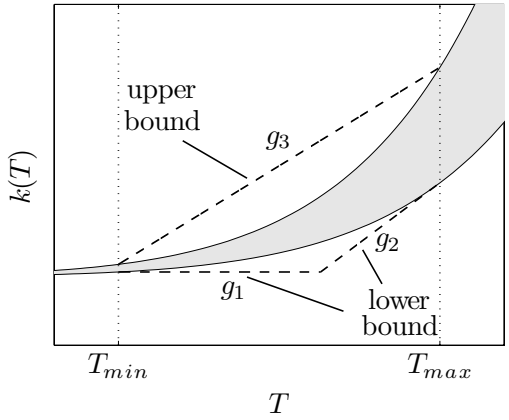

Fig. 4 Bounding of uncertain Arrhenius term (light gray area) with three linear functions (– –).

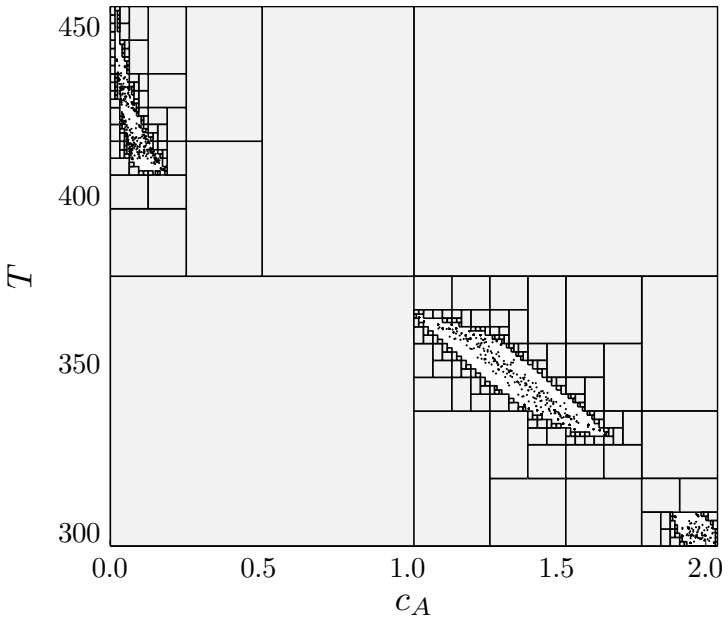

Fig. 5 Region in state space which cannot contain steady states for given parameter uncertainties (light gray rectangles) versus steady states (·) computed using Monte-Carlo sampling.

Set of feasible steady states

Using the bounding of $k(T)$ and $\mathcal{X}_0$ an outer bounding set of feasible steady states of the CSTR can be computed employing the methods presented in Section 4. Here, uncertainties in several parameters are allowed. The amounts of uncertainty with respect to the nominal values are provided in Table 1. The decision variables δ is hereby a set of seven possible system configurations, as up to the hybrid dynamics introduced by the over-temperature protection also failures of two of the three pumps are considered, $m_p \in \{1, 2, 3\}$.

The algorithm outlined in Section 4.1 is in the following used to estimate the set of all feasible equilibrium points of (19) for the given parameter uncertainties and $m_p \in \{1, 2, 3\}$. The results are shown in Figure 5, where the part of the state space which is certified infeasible is marked light gray. To compare our results with classical approaches, one thousand equally distributed Monte-Carlo samples for the accessible parameter set were taken and the steady states were determined.

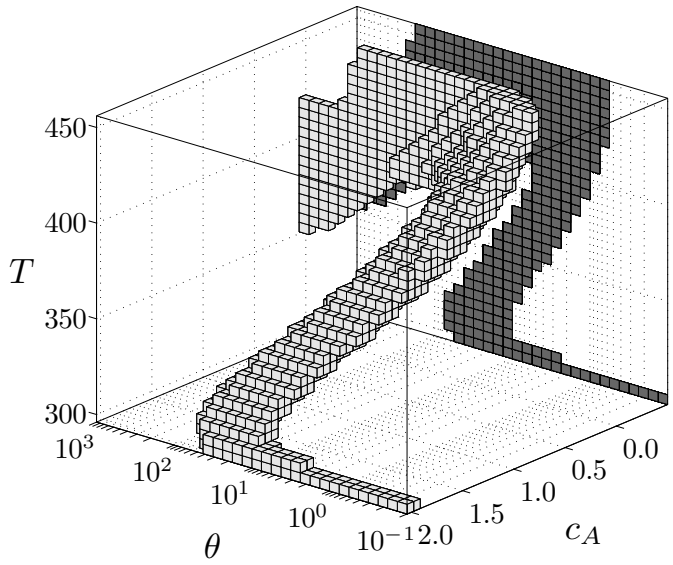

Fig. 6 Outer approximation of the envelope of the bifurcation diagram of an uncertain system and its projection of the θ-T-plane. Note that in contrast to Figure 5 here the region in space which may contain equilibrium points is marked with light gray cubes.

As one can see in Figure 6, points from the Monte Carlo samples are always contained in the outer bounded sets. However, the Monte-Carlo sampling provides only an analysis of a finite set of points and a conclusion about the region between these points can not be drawn directly. The set of feasible equilibrium points is always underestimated, even for exhaustive Monte-Carlo sampling. This can lead to problems if the mapping from parameters to equilibrium points is highly nonlinear (10).

The proposed set-based method on the other hand guarantees that all equilibrium points are contained in the determined set. Hence, it can also be used to verify process safety. In this work the method has for instance be used to compute certificates that a critical temperature $T_{crit} = 440$ K is not exceeded even if one pump fails.

Envelope of the bifurcation diagram

Besides the set of feasible steady states, this approach can also be used to determine an outer approximation of the envelope of the bifurcation diagram of the uncertain hybrid system. This is depicted in Figure 6. Knowledge of the envelope of the uncertain bifurcation diagram allows a better understanding of the system. One can for instance study the multiplicity of steady states or the stability of whole uncertain branches of the bifurcation diagram. For the later, set-based stability analysis of uncertain nonlinear systems has to be used, compare (23).

In this work, the envelope of the bifurcation diagram is used to determine lower and upper bounds on the extinction and the ignition point of (19),

$$\begin{aligned} 5.6 \times 10^{-1} \text{ min} \leq \theta_{ep} \leq 5.7 \times 10^{0} \text{ min} \\ 1.0 \times 10^{1} \quad \text{min} \leq \theta_{ip} \leq 7.6 \times 10^{1} \text{ min}. \end{aligned}$$

Bounds on θ_{ip} and θ_{ep} are of practical relevance as they can be used for operating the reactor in a region where small disturbances cannot lead to extinction or ignition of the reaction. Note that θ_{ip} and θ_{ep} can vary one order of magnitude despite very small uncertainty in parameters.

6 Conclusion

In this work we studied the problem of guaranteed outer bounding the region in state space containing all equilibrium points of uncertain hybrid differential algebraic systems. The proposed method is based

on the formulation as a feasibility problem and a relaxation to a SDP. It is shown that guaranteed outer bounds of the feasible set of equilibrium points can be determined.

The advantage of the proposed methodology in comparison to Monte-Carlo based approaches is explained and shown considering a simple CSTR process. In particular, the developed method does not rely on sampling and can deal with strongly nonlinear and non-unique mappings from parameters to steady states.

The computed set is guaranteed to contain all feasible steady states, thus worst case scenarios can be analyzed. This is of certain interest to evaluate controller performance in case of failures.

Finally, it is shown that the proposed method can also be used to determine the envelope of the bifurcation diagram of uncertain systems. This allows to obtain a better understanding of the possible complex process dynamics.

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