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Robust multi-objective optimal control of uncertain (bio)chemical processes

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Abstract

Dynamic optimization or optimal control problems are omnipresent in the (bio)chemical industry. In addition, these problems often involve multiple and conflicting objectives, leading to a so-called set of Pareto optimal solutions, instead of one single optimum. Alternatively, robustness of the obtained solutions with respect to model uncertainties, e.g., guaranteeing that critical constraints are not violated, is of the highest importance in process industry. Moreover, robustness can also be interpreted as an additional and conflicting objective, since more robust solutions typically induce a performance decrease. The current manuscript exploits advanced deterministic techniques to efficiently and accurately generate Pareto sets in the presence of model uncertainty. The developed procedures allow the presentation of robust Pareto sets, i.e., Pareto sets in which robustness is an additional objective. Based on these Pareto sets all trade-offs can clearly be assessed by the decision maker. Two illustrative case studies are presented for the optimal design and operation of a jacketed tubular reactor with conflicting conversion and energy objectives and a fed-batch bioreactor with conflicting productivity and yield aims.

Keywords: bioreactors, chemical reactors, dynamic optimization, optimal control, multi-objective optimization, robust optimization, uncertainty

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1. Introduction

Due to market saturation and global competition process industry continuously strives for performance increases. To achieve this aim in a systematic and efficient way, mathematical models and optimization techniques are invaluable tools. When optimal trajectories have to be determined for processes described by differential equations, this leads to so-called *optimal control* or *dynamic optimization* problems (see, e.g., Sargent (2000) for a historical review). However, in practice additional questions rise.

1.1. When one is not enough . . . : the challenge of multiple objectives

Often multiple and conflicting objectives are present in (bio)chemical engineering problems. Such *multi-objective optimization* problems have in general not only one single point as a solution, but give rise to a set of mathematically equivalent compromise solutions (i.e., the Pareto set), where no objective can be improved without worsening at least one of the others (Miettinen, 1999). Consequently, it is important to present the set of Pareto optimal solutions to the so-called *decision maker* such that he/she can pick one solution according to his/her preferences.

Illustrative example. As an intuitive illustration of the conflicting nature of traditional objectives in dynamic (bio)chemical processes the following classic example is given (Bhaskar et al., 2000). In an isothermal batch reactor the following series of first-order reactions takes place: $A \xrightarrow{k_1} B \xrightarrow{k_2} C$. Initially, only reactant A is present. The final batch time t_f has to be chosen such that the yield of B and the selectivity of B are maximized. Here, yield is defined as the final amount of B over the initial amount of A . Maximization of the yield is important as it leads to higher amounts of B . Selectivity is defined as the final amount of B over the total amount of products formed. Maximization of the selectivity is desirable as it reduces the downstream separation costs. When adopting $k_1 = 0.1$, $k_2/k_1 = 0.1$ and batch times $t_f \leq 50$, the same results as described by Bhaskar et al. (2000) are obtained (see Figure 1). Here, the solid line between points Q and R indicates the Pareto set, as it is not possible to improve one of the objectives without worsening the other. Hence, it is clear that an intrinsic trade-off is present. The systematic representation of the Pareto set allows the process engineer to analyze

the trade-offs involved and to select one solution according to his/her own preferences for implementation in practice.

Figure 1: Isothermal batch reactor with consecutive first-order kinetics: Pareto set.

An overview of over 50 articles on multi-objective optimization in chemical engineering applications was given in Bhaskar et al. (2000). Additional results on multi-objective dynamic optimization of chemical processes were reported for instance in the following studies. Wang and Shieh (1997) studied the batch polymerization of styrene as well as the sugar crystallization in a vacuum pan crystallizer. For the first case, the aims were to achieve the desired number average molecular chain length and monomer conversion in minimum time, while for the second the objectives aimed at producing a desired crystal size in minimum time. Chen et al. (2003) tackled the heating of a nylon 6 batch polymerization in view of minimizing the concentrations of unreacted monomer and undesirable cyclic compounds, while minimizing the reaction time. A semi-batch epoxy polymerization process was analyzed in Deb et al. (2004); Mitra et al. (2004) with respect to maximizing the number average molecular weight with a specified value of the polydispersity index and number average molecular weight in the shortest possible time. Sarkar et al. (2006) dealt with conflicting objectives in seeded batch crystallization processes. Agrawal et al. (2007) investigated a Low-Density PolyEthylene (LDPE) tubular reactor in view of maximizing the monomer conversion while minimizing the undesirable side products.

58 With respect to multi-objective optimization of biochemical and fermen-
59 tation processes a number of results have been reported. Wang et al. (1998)
60 evaluated the fed-batch bioethanol production in view of maximizing the pro-
61 duction, while minimizing the batch time as well as the use of the substrates
62 glucose and xylose. Chen et al. (2003) studied a Penicillin G fed-batch fer-
63 mentation process for maximizing the production in minimum time. Yield,
64 i.e., the amount of product over the amount of substrate fed, was added as
65 a competing objective by Sarkar and Modak (2005) for the fed-batch pro-
66 duction of lysine. Sendin et al. (2006) dealt with the optimal operation of
67 an aerobic fermentor for producing *Saccharomyces cerevisiae*. The objec-
68 tives considered were product profit and capital investment. Similarly, these
69 authors (Sendin et al., 2010) also investigated the thermal sterilization of
70 canned food with respect to the retention of a nutrient versus the surface
71 retention of the luminosity quality factor.

72 1.2. Optimality in an uncertain world: the search for robustness

73 Besides the presence of multiple and conflicting objectives another dif-
74 ficulty is often encountered in industrial (bio)chemical processes. Namely,
75 model uncertainty as well as external process disturbances are inevitably
76 present when modeling, controlling and optimizing practical processes (see,
77 e.g., Srinivasan et al. (2003); Srinivasan and Bonvin (2007); Diehl et al.
78 (2008)). Despite this intrinsic uncertainty, *robust optimization* (Ben-Tal
79 and Nemirovski, 2001) can guarantee that critical constraints are met safely,
80 which is required for many engineering applications. Here, the nature of the
81 uncertainty can be modeled in various ways. Sometimes stochastic infor-
82 mation about the occurrence of uncertainties is available such that chance
83 constraints can be formulated (Wendt et al., 2002; Mitra, 2009). Here, the
84 stochastic information can, for example, be obtained from a parameter iden-
85 tification where typically confidence regions are generated along with the
86 estimates for the model parameters (Bock, 1983; Tjoa and Biegler, 1991;
87 Lohmann et al., 1992; Franceschini and Macchietto, 2008). In a different
88 set-up the uncertainty is known to be in a given set, which can for example
89 be a box or an ellipsoid. In this case, it is desirable to guarantee that all
90 constraints are satisfied in all possible worst case situations and/or what is
91 the worst possible performance to be expected.

92
93 As including more robustness typically gives rise to a lower performance
94 (Pinto, 1998), robustness can be seen as an additional conflicting objective

(Datskov et al., 2006). For instance, the scaling of the confidence region may be an important tuning parameter in a decision making process.

97

Illustrative example. A scalar example allows illustrating the underlying concepts. Consider the optimization of a parabola in which the linear term is affected by uncertainty:

$$\min_x f(x, w) \quad \text{with} \quad f(x, w) := x^2 + (1 + w)x, \quad (1)$$

where w is an uncertain parameter satisfying $|w| \leq \gamma$. Looking for a robust solution in which the worst possible case is considered, yields a typical min – max formulation:

$$\min_x \max_{w, |w| \leq \gamma} f(x, w) = \min_x x^2 + x + \gamma|x| \quad (2)$$

$$= -\frac{1}{4}(1 - \gamma)^2 \quad (3)$$

where we assume $0 \leq \gamma \leq 1$. Note that the optimal value $-\frac{1}{4}(1 - \gamma)^2$ can be interpreted as a function of γ . As expected, increasing γ , which corresponds to incorporating more robustness, is in conflict with the performance as this induces higher objective function values. The Pareto front is in this case a parabola as depicted in Figure 2. So in summary, the selection of the confidence region based on γ can be seen as an additional conflicting objective.

1.3. Motivation for the paper

To enhance the decision making in the (bio)chemical industry, systematic mathematical techniques and software tools are required to present different alternatives (i.e., Pareto optimal solutions) to the decision maker in an efficient way. The decision maker can for example be (i) an engineer who selects a final Pareto optimal solution based on the actual situation without modeling his/her personal preferences mathematically or (ii) an independent higher-level post-processing tool whose final decision criterion is confidential or for any other reason not available at the time when the optimal control problem has to be solved. As outlined above, the presentation of alternatives is required to include robustness as well, allowing the assessment of the performance loss due to conservatism. Hence, from a conceptual point of view it is obvious to cast robust dynamic optimization within a multi-objective frame. However, in practice this is mostly a non-trivial question as

Figure 2: Pareto curve for trade-off between performance and robustness.

multi-objective dynamic optimization as well as robust optimal control are often on their own already challenging and computationally expensive problems. This is especially the case if nonlinear dynamic systems are involved, which typically lead to non-convex optimal control problems. Finally, to the best of the authors' knowledge, robust Pareto sets, i.e., Pareto sets in which robustness is an additional objective, for dynamic chemical processes were only computed by Mitra (2009) exploiting evolutionary strategies. Hence, the current contribution can be regarded as the first to include uncertainty in multi-objective dynamic optimization using efficient deterministic optimization approaches.

1.4. Aim of the paper

The main contribution of this paper is to establish a computationally efficient frame for robust multi-objective optimal control yielding robust Pareto sets for dynamic (bio)chemical processes. Based on the presentation of these Pareto optimal alternatives, the decision maker can select one according to his/her preferences. The proposed frame is illustrated on two conceptual benchmark case studies which are relevant for both (bio)chemical engineers as well as developers of nonlinear optimal control algorithms. First, a jacketed tubular reactor is considered with conflicting conversion and energy costs. However, the reactor temperature must not exceed a given upper bound de-

spite uncertain fouling at the reactor wall which affects the heat transfer. Second, a fed-batch fermenter for the production of lysine with conflicting yield and productivity objectives is studied. Despite uncertainties in the feeding a minimum amount of substrate has to be added which influences, e.g., the objective values. The examples exhibit a variety of challenging features which may arise in general multi-objective and optimal control problems, e.g., highly nonlinear dynamics, state and control constraints, uncertainty on model inputs and parameters, fixed and free-end time, singular and non-singular nature, and convex and non-convex Pareto sets. All results have been obtained by using the Automatic Control And Dynamic Optimization toolkit ACADO (Houska et al., in press) and its Multi-Objective extension (Logist et al., 2010), which are both freely available at www.acadotoolkit.org.

1.5. Structure of the paper

This paper is divided into two major parts. The first part (Section 2) reviews the formulation and methods for robust as well as for multi-objective optimal control. Besides introducing the basic syntax and main algorithmic ideas it outlines the connections between these two fields and introduces a generic solution frame. The second part (Section 3) is dedicated to two extensive case studies: a jacketed tubular reactor and a fed-batch fermenter, which exhibit a variety of typical optimal control problem features. Section 4 finally summarizes the conclusions.

2. Methodology for robust multi-objective optimal control

The methodological part starts in Section 2.1 with a general overview on nonlinear optimal control, but specializes in Section 2.2 and Section 2.3 to robust optimal control and multi-objective optimal control, respectively. The proposed framework is summarized in Section 2.4.

2.1. Nonlinear optimal control

This section starts with a short review of existing methods in standard nonlinear optimal control introducing the necessary syntax which is used throughout the paper. Optimal control problems are a special class of optimization problems which focus on the optimization of possibly nonlinear dynamic systems. A fairly general formulation of a such a problem reads as follows:

$$\begin{aligned} \min_{x(\cdot), u(\cdot), p, \xi_f} \quad & m(p, \xi_f) \\ \text{s.t.} \quad & \begin{cases} x(0) = x_0 \\ \frac{d}{d\xi}x(\xi) = f(\xi, u(\xi), p, x(\xi), w(\xi)) \\ 0 \geq h_i(\xi, u(\xi), p, x(\xi)) \end{cases} \quad , \end{aligned} \quad (4)$$

for all $\xi \in \Xi_i(\xi_f)$.

In this context, $x : \mathbb{R} \rightarrow \mathbb{R}^{n_x}$ denotes the state vector, $u : \mathbb{R} \rightarrow \mathbb{R}^{n_u}$ the control function which may vary along the interval $[0, \xi_f]$, and $p \in \mathbb{R}^{n_p}$ the constant parameters. Note that the end time ξ_f can in a general setting be optimized too. Furthermore, there are three types of functions assumed to be given. First, the function f represents the dynamic system equations. Second, the functions h_i (with $i \in \{1, \dots, n_h\}$) indicate inequality path- or point constraints depending on the definition of the sets $\Xi_i(\xi_f) \subseteq [0, \xi_f]$. And third, the function m denotes a Mayer objective term which is in the above notation assumed to be independent of $x(\xi_f)$ as such a dependence can always be eliminated by introducing an additional *slack parameter*. In such a situation, the Mayer objective term $M(x(\xi_f), p, \xi_f)$ in the minimization is replaced by the slack parameter p_s , while an additional constraint is added $0 \geq M(x(\xi_f), p, \xi_f) - p_s$. Hence, the parameter p_s is forced to decrease until the additional constraint becomes an equality and p_s takes the value of the Mayer objective $M(x(\xi_f), p, \xi_f)$. Similarly, additional Lagrange terms $\int_0^{\xi_f} L(\xi, u(\xi), p, x(\xi), w(\xi)) d\xi$ can always be reformulated by the inclusion of an additional cost state $dx_c/d\xi = L(\xi, u(\xi), p, x(\xi), w(\xi))$ with $x_c(0) = 0$. Besides these model functions f , h , m and the optimization variables x, u, p, ξ_f there are two remaining variables: (i) the vector x_0 denoting the initial state of the dynamic system and (ii) the possibly time varying function w which may influence the right-hand side of the differential equation. Here, a main difficulty is that the initial state x_0 as well as the external input w might not be known at the time when the process is optimized, i.e., these two inputs can be uncertain. In the next Section 2.2 it shall be discussed how this uncertainty can be taken into account.

Assuming for a moment that x_0 and w are known, i.e., in the case that no uncertainty is present, the above optimal control problem can be solved with standard software. Numerical methods for solving optimal control problems are generally classified into two categories, *direct* and *indirect* approaches.

Methods of the latter class try to find solutions according to Pontryagin's Minimum Principle (Pontryagin et al., 1962), i.e., the first-order necessary conditions for optimality, while techniques of the former class convert the original infinite dimensional optimal control problem into a finite dimensional Non-Linear Programming problem (NLP) via discretization. Most optimal control problems are nowadays solved by direct approaches.

In *sequential direct methods* as *Single Shooting* (Sargent and Sullivan, 1978; Vassiliadis et al., 1994a,b) only the controls u are discretized, most often using piecewise polynomials. For each parameterization the differential equations are solved using a standard integration algorithm, and the objective function is evaluated. The parameter values are then updated employing a standard optimization algorithm. Hereto, most often a deterministic, sequential quadratic programming routine is employed. Consequently, the solution of the differential equations and the minimization are decoupled, and proceed sequentially within one iteration step of the optimizer. As these methods provide a correct solution to the differential equations during all iterations, they are also known as *feasible path* methods. The most important advantage of the sequential approach is its straightforward implementation resulting in rather small-scale NLPs. However, the sequential approach may be slow, especially in the presence of path constraints on the states, since these constraints cannot be enforced directly.

Alternatively, *simultaneous direct approaches* discretize both the control and the states. Consequently, the simulation and optimization are performed simultaneously in the space of both the discretized controls and states, yielding a large-scale NLP which requires tailored numerical methods. Since in this case the differential equations are only satisfied at the solution of the optimization problem, these methods are also called *infeasible path* methods. Two different approaches exist: *Multiple Shooting* (Bock and Plitt, 1984; Leineweber et al., 2003a,b) and *Orthogonal Collocation* (Biegler, 1984; Logsdon and Biegler, 1989; Biegler, 2007). In the former case, the total integration range is split into a finite number of intervals on which integration of the states is continuous. The value of the control and the initial value of the states in each interval are chosen by the NLP-solver in each iteration while trying to ensure the continuity of the states between the different intervals. In the latter case, also the states are fully discretized based on (orthogonal) polynomials. Hence, the minimal objective value has to be found while

satisfying the discretized differential equations. Clearly, Multiple Shooting and Orthogonal Collocation allow a more direct enforcement of the state constraints. However, the size of the NLPs significantly increases, requiring tailored optimization algorithms that exploit the problem's structure and sparsity. Obviously, the largest but also sparsest NLPs are encountered with Orthogonal Collocation. Consequently, most often Interior Point (Wächter, 2002) and partially reduced SQP methods (Leineweber et al., 2003a) are employed for Orthogonal Collocation and Multiple Shooting, respectively.

2.2. Robust optimal control

This section returns to the discussion of the case that the inputs x_0 and w in the optimal control problem (4) are uncertain. Here, the knowledge about the uncertainty is typically assumed to be of the form $(x_0, w) \in \mathcal{W}$, where $\mathcal{W}$ is a given bounded uncertainty set. The aim is now to take this knowledge about the uncertainty into account by formulating the associated robust counterpart problem. Note that the idea of robust counterpart, *worst case* or min-max formulations has, for example, been established for finite dimensional static optimization problems in Ben-Tal and Nemirovski (2001). These developments transfer conceptionally also to optimal control problems.

In order to outline this aspect, the solution of the differential equation is assumed to uniquely exist. Thus, the state $x(t)$ can at any time t be interpreted as a function of the inputs x_0 , u , p , and w writing:

$$\forall \xi \in [0, \xi_f] : \quad x(\xi) = \zeta[\xi, x_0, u(\cdot), p, w(\cdot)] .$$

Here, the functional ζ can numerically be evaluated by integrating the differential equation on the interval $[0, \xi]$ using the corresponding arguments x_0 , u , p , and w as initial value, control input, parameter and disturbance input, respectively. With this notation, the robust counterpart problem of the optimal control problem (4) can be written as:

$$\begin{aligned} \min_{x(\cdot), u(\cdot), p, \xi_f} \quad & m(p, \xi_f) \\ \text{s.t.} \quad & \max_{(w(\cdot), x_0) \in \mathcal{W}} h_i(\xi, u(\xi), p, \zeta[\xi, x_0, u(\cdot), p, w(\cdot)]) \leq 0 , \end{aligned} \quad (5)$$

where the constraints have to be satisfied for all times $\xi \in [0, \xi_f]$ and all components $i \in \{1, \dots, n_h\}$ of the constraint function h .

272 The main problem with the above min-max optimal control formulation
273 is that there are no efficient algorithms known which can solve the nonlin-
274 ear min-max problem in full generality and with a high numerical accuracy.
275 Thus, in order to obtain an efficient computational framework, a trade-off
276 between computational tractability and numerical accuracy has to be made.
277 There are basically two options. First, *heuristics* can be applied which can,
278 for example, be based on *linearization strategies* or even *higher-order Taylor*
279 *expansions* of the model functions with respect to the uncertainty as orig-
280 inally proposed in Ma and Braatz (2001); Nagy and Braatz (2004); Diehl
281 et al. (2006); Houska and Diehl (2009). And second, the above min-max op-
282 timal control problem can be *approximated conservatively* which does require
283 a more involved analysis and consequently more expensive algorithms as dis-
284 cussed in Stein (2003); Floudas and Stein (2007); Houska and Diehl (2010)
285 where mathematical guarantees on the conservatism of the corresponding
286 approaches are proven. However, in this paper the focus is on the heuristic
287 approaches based on *linearization*, which can be justified if the uncertainty is
288 small with respect to the curvature of the model functions such that higher-
289 order terms can either be neglected or overestimated if conservatism is re-
290 quired.

291

In order to outline the main idea of the linear approximation approach,
a reference trajectory x_{ref} and a deviation δx are introduced such that $x =$
 $x_{\text{ref}} + \delta x$. For small uncertainties w the original dynamic equation:

$$\frac{d}{d\xi}x(\xi) = f(\xi, u(\xi), p, x(\xi), w(\xi))$$

292 can be split into the nominal dynamics for the reference trajectory and the
293 approximate linearization based dynamics of the uncertainty affected devia-
294 tion which satisfy the differential equations:

$$\begin{aligned} \frac{d}{d\xi}x_{\text{ref}}(\xi) &= f(\xi, u(\xi), p, x_{\text{ref}}(\xi), w_{\text{ref}}) \quad \text{with} \quad x_{\text{ref}}(0) = x_0^{\text{ref}} \\ \text{and} \quad \frac{d}{d\xi}\delta x(\xi) &= A(\xi)\delta x(\xi) + B(\xi)\delta w(\xi) \quad \text{with} \quad \delta x(0) = B_0\delta w_0, \end{aligned}$$

295 respectively. Here, the following short-hands are used:

$$\begin{aligned} A(\xi) &:= \frac{\partial f(\xi, u(\xi), p, x_{\text{ref}}(\xi), w_{\text{ref}})}{\partial x}, \\ B(\xi) &:= \frac{\partial f(\xi, u(\xi), p, x_{\text{ref}}(\xi), w_{\text{ref}})}{\partial w}, \\ C_i(\xi) &:= \frac{\partial h_i(\xi, u(\xi), p, x_{\text{ref}}(\xi), w_{\text{ref}})}{\partial x}, \\ \text{and } d_i(\xi) &:= h_i(\xi, u(\xi), p, x_{\text{ref}}(\xi)), \end{aligned}$$

where $i \in \{1, \dots, n_h\}$. The plan is now to replace the exact robust counterpart functions of the form:

$$V_i[\xi, u(\cdot), p, \xi_f] = \max_{(w(\cdot), x_0) \in \mathcal{W}} h_i(\xi, u(\xi), p, \zeta[\xi, x_0, u(\cdot), p, w(\cdot)])$$

296 with an approximate robust counterpart based on the linearization:

$$\begin{aligned} \tilde{V}_i[\xi, u(\cdot), p, \xi_f] &:= \max_{\delta x(\cdot), \delta w(\cdot)} d_i(\xi) + c^T \delta x(\xi) \\ \text{s.t. } &\begin{cases} \frac{d}{d\xi} \delta x(\xi) = A(\xi) \delta x(\xi) + B(\xi) \delta w(\xi) \\ (x_0^{\text{ref}} + \delta x(0), w_{\text{ref}} + \delta w) \in \mathcal{W}. \end{cases} \quad (6) \end{aligned}$$

297 In order to analyze the influence of the uncertainty in the linearized dynamic
298 systems, i.e., evaluating the maximization problems of the form (6) Lyapunov
299 equations (Kalman, 1960, 1963; Bittanti et al., 1984) have turned out to be a
300 useful tool. This has, e.g., been discussed in Houska and Diehl (2009), where
301 also the following technical result is proven:

302
303 **Lemma:** If the uncertainty set $\mathcal{W}$ is modeled as a *ball* of size $\Gamma \in \mathbb{R}^+$,
304 i.e., if $\mathcal{W}$ contains the L_2 bounded uncertainty functions:

$$\mathcal{W} := \left\{ (x_0^{\text{ref}} + B_0 \delta w_0, w_{\text{ref}} + \delta w) \left| \delta w_0^T \delta w_0 + \int_0^{\xi_f} \delta w(\tau)^T \delta w(\tau) d\tau \leq \Gamma^2 \right. \right\}, \quad (7)$$

305 then we have:

$$\tilde{V}_i[\xi, u(\cdot), p, \xi_f] = d_i(\xi) + \Gamma \sqrt{C_i(\xi) P(\xi) C_i(\xi)^T}, \quad (8)$$

where the matrix valued function P satisfies a Lyapunov differential equation of the form:

$$\frac{d}{d\xi}P(\xi') = A(\xi')P(\xi') + P(\xi')A(\xi')^T + B(\xi')B(\xi')^T \text{ with } P(0) = B_0B_0^T \quad (9)$$

for all $\xi' \in [0, \xi_f]$.

307

In the following context, the optimization variables are given by $y := (x_{\text{ref}}(\cdot), P(\cdot), u(\cdot), p, \xi_f)$. Thus, under the assumption that the uncertainty set is a *ball* as in the above Lemma, then the original exact robust counterpart problem (5) can in a linear approximation be written as:

$$\min_y m(p, \xi_f) \quad \text{s.t.} \quad y \in S.$$

Here, the set S is comprising the set of all variables $y := (x_{\text{ref}}(\cdot), P(\cdot), u(\cdot), p, \xi_f)$ which satisfy:

$$\frac{d}{d\xi}x_{\text{ref}}(\xi) = f(\xi, x_{\text{ref}}(\xi), u(\xi), p, 0) \quad \forall \xi \in [0, \xi_f] \quad (10)$$

$$\frac{d}{d\xi}P(\xi) = A(\xi)P(\xi) + P(\xi)A(\xi)^T + B(\xi)B(\xi)^T \quad (11)$$

$$x_{\text{ref}}(0) = x_0 \quad (12)$$

$$P(0) = B_0B_0^T \quad (13)$$

$$0 \geq d_i(\xi) + \Gamma \sqrt{C_i(\xi)P(\xi)C_i(\xi)^T} \quad \forall \xi \in \Xi_i(\xi_f). \quad (14)$$

The second term in the right-hand side of (14) can be interpreted as a *back-off* value. For a constraint, this term can be regarded as an additional *safety margin* required for guaranteeing the meeting of the constraint. For cost functions, which have been reformulated using a slack variable p_s , this term indicates the direct *performance loss*.

315

Finally, it has to be noted that the above formulation has also a stochastic interpretation (Houska and Diehl, 2009; Kalman, 1960, 1963; Bittanti et al., 1984). If the disturbance w entering the dynamic system is a Gaussian white noise process with:

319

$$\begin{aligned} \mathbb{E}\{w(\xi)\} &= 0 \quad \text{and} \\ \mathbb{E}\{w_i(\xi_1)w_j(\xi_2)^T\} &= \Sigma(\xi_1) \delta(\xi_2 - \xi_1) \delta_{i,j} \end{aligned}$$

for all $\xi, \xi_1, \xi_2 \in \mathbb{R}$ and for all $i, j \in \{1, \dots, n_w\}$ while the uncertain value w_0 is Gaussian distributed with variance-covariance matrix $\mathbb{I} \in \mathbb{R}^{n_x \times n_x}$ then $P(\xi)$ is a linear approximation of the variance-covariance matrix of the state $x(\xi)$. (Note that $\Sigma(\xi_1)$ denotes here the variance-covariance matrix of the white noise at ξ_1 , while $\delta_{i,j}$ is Kronecker's delta.) Consequently, the variance-covariance matrix associated with constraint function h is in a linear approximation given by $C(\xi)P(\xi)C(\xi)^T$ such that the *robustified constraints* can also be interpreted as *single chance constraints*.

On the one hand, the above approximation strategy leads to a tractable optimal control problem which includes robustness aspects. However, on the other hand, this formulation requires not only an evaluation of the right-hand side function f but also of the Jacobian matrices A and B associated with the right-hand side. Moreover, robust optimal control problems are in general more challenging to solve compared to their nominal variant. For example, if the nominal optimal control problem is feasible, the robust version might not be feasible as the constraints are required to be satisfied for all possible disturbances.

2.3. Multi-objective optimal control

In this section the general formulation of robust multi-objective optimal control problems is introduced:

$$\min_{y, \Gamma} \{J_1(y, \Gamma), \dots, J_m(y, \Gamma)\} \quad \text{s.t.} \quad y \in S \quad (15)$$

Here, the set S has already been defined in the previous section. Recall that this set contains the differential equations for the reference function x_{ref} , the Lyapunov differential equation for the additional states P which describe the influence of uncertainty in a linear approximation, the corresponding initial values, as well as the robustified constraints which depend on the confidence level Γ . The main difference to the previous considerations is that more than one objective $J_1, \dots, J_m$ is present, i.e., a multi-objective optimal control problem is considered. Note that the objectives are allowed to depend on Γ too, as robustness itself can be an additional objective which can conflict with other aims.

Contrary to single objective optimization, typically no single solution exists in multi-objective optimization. Hence, to determine the set of optimal

solutions a certain optimality criterion is needed, i.e., *Pareto optimality*:

Definition: A point $(y^*, \Gamma^*) \in \mathcal{S}$, is Pareto optimal iff there does not exist another point with $\Gamma \geq 0$ and $y \in \mathcal{S}$, such that $J_i(y, \Gamma) \leq J_i(y^*, \Gamma^*)$ for all i and $J_j(y, \Gamma) < J_j(y^*, \Gamma^*)$ for at least one objective function j .

Alternatively, it could be stated that a point is Pareto optimal if there exists no other point that improves at least one objective function without worsening another.

Multi-objective methods for determining Pareto optimal solutions are in literature most often divided in two categories (see, e.g., Marler and Arora (2004) for a review): (i) *scalarization methods* (e.g., Miettinen (1999)) and (ii) *vectorization methods* (e.g., Deb (2001)). Approaches from the former class exploit additional scalar variables to convert the multi-objective optimization problem into a (series of parametric) single-objective optimization problem(s), while techniques from the latter class tackle directly the multi-objective optimization problem by employing a population of candidate solutions that gradually evolves to the Pareto front.

These vectorization approaches consisting of evolutionary or stochastic optimization strategies (see, e.g., Bhaskar et al. (2000) for an overview) have often been used successfully in various applications. However, as they require the repeated solution of the model equations, the computation time can easily grow, especially for large-scale models. Also the incorporation of other constraints than simple bounds and the tuning of algorithmic parameters like the population size may be non-trivial. Moreover, when employing these methods in a context where model uncertainty is present, the computation times may increase even stronger because sampling over the uncertain parameters may be needed and each sample gives rise to an additional simulation run.

On the other hand *scalarization* techniques can and have to take advantage of fast and efficient deterministic optimization routines to find a (local) optimum for the series of possibly large-scale single-objective problems.

- **Weighted Sum (WS).** Although the convex Weighted Sum is still

most often used in practice, it exhibits as drawbacks that (i) the returned solutions strongly depend on the scale and scaling of the objectives, that (ii) a uniform variation of the weights does not necessarily result in nice spread on the Pareto set and (iii) that points in non-convex regions of the Pareto cannot be obtained (Das and Dennis, 1997).

- **ϵ -constraint.** The ϵ -constraint method (Haimes et al., 1971) minimizes a selected single most important objective function, while the $m - 1$ other objective functions are added as (in)equality constraints of the form that have to be equal to $J_i \leq \epsilon_i$. However, the selection of the ϵ_i values is not always trivial and a uniform approximation of the Pareto set is not ensured.

- **Normal Boundary Intersection (NBI).** NBI (Das and Dennis, 1998) has been developed based on geometrically intuitive arguments in order to overcome the deficiencies of the WS. The multi-objective optimization problem is reformulated as to maximize the distance from a point on the convex hull of individual minima along the (quasi-)normal toward the utopia point. Hence, this formulation gives rise to additional equality constraints. As a result, a nice spread along the Pareto set is obtained, and possible Pareto points in non-convex regions can be detected. However, in adverse cases also non-Pareto optimal solutions can be returned.

- **(Enhanced) Normalized Normal Constraint ((E)NNC).** NNC (Messac et al., 2003) is based on similar geometric ideas as NBI but combines them with an ϵ -constraint approach. Hence, a selected (normalized) objective has to be minimized, while $m-1$ half planes that are orthogonal to the plane connecting all (normalized) individual minima, are added as inequality constraints. Although NNC can avoid returning part of the non-Pareto optimal points generated by NBI, non-Pareto optimal solutions may still be provided and, hence, a Pareto filter algorithm to remove these candidate solutions may be required. ENNC (Sanchis et al., 2008) is an extension of NNC, which uses adapted normalization strategies that also in cases with more than two objectives lead to a unique and uniform coverage of the Pareto set.

Several of the above mentioned scalarization techniques (e.g., NBI and

NNC) have been shown to enable an efficient and accurate solution of multi-objective optimal control problems (Logist et al., 2009b). In addition, a multi-objective extension **ACADO Multi-Objective** (Logist et al., 2010) to the freely available Automatic Control and Dynamic Optimization toolkit **ACADO** Houska et al. (in press) exists.

2.4. A generic robust multi-objective framework

The proposed generic frame for robust multi-objective optimal control can be summarized as follows. It consists of an integration of the linear approximation techniques for robust optimal control with novel scalarization based approaches for multi-objective optimization (e.g., NBI and (E)NNC). The former techniques enable the reformulation of the hard to solve min-max problem into more tractable minimization problems. To this end, the Lyapunov differential variables have to be added as additional state variables with appropriate initial conditions and the uncertainty parameter Γ has to be included in the constraints and/or objective functions. The latter methods ensure the generation of accurate solutions with a nice spread along the Pareto set. The underlying force driving the accurate and efficient solution is that the combination of approaches still allows the use of fast deterministic single-objective optimization routines.

Note that the above formulation also includes the case that the robustness itself is an additional objective as, e.g., an additional parameter γ can be used to scale w such that this parameter can be interpreted as a *confidence level* to be maximized: $\Gamma = \gamma w$. Enlarging the uncertainty set allows the incorporation of more severe disturbances and parameter uncertainties. This results in more robust solutions, but also in a worse performance. As the uncertainty parameter γ for the robustness objective typically takes values in between 0 and 100%, several values can easily be selected a priori. This procedure boils down to an ϵ -constraint approach.

As a computational platform **ACADO** toolkit is used. An ϵ -constraint approach is adopted for the robustness objective, while NBI and NNC are exploited for the original objectives. In each of the resulting parametric single-objective optimization subproblems the approximate robust counterpart formulation for the robust optimal control problem is tackled using Multiple Shooting. Consequently, robust Pareto fronts, i.e., Pareto fronts for different levels of uncertainty, can be obtained.

3. (Bio)chemical case studies

To illustrate the approach two (bio)chemical cases are studied: a jacketed tubular reactor and a fed-batch bioreactor. These have been selected as exemplary processes because (i) they exhibit a variety of typical features of practical optimal control problems, (ii) they have been well-studied in the literature and (iii) they allow an easy (bio)chemical interpretation of the results.

The cases are first introduced in Sections 3.1 and 3.2. To ensure a clear explanation, the subsequent sections gradually add complexity to the problem statement, going from the solution of nominal single-objective optimal control problems in Sections 3.3 and 3.4, over the approximate computation of robust optimal control policies in Sections 3.5 and 3.6, to the inclusion of multiple and conflicting objectives in Sections 3.7 and 3.8. Finally, Section 3.9 discusses some general remarks.

3.1. Jacketed tubular chemical reactor

The first benchmark problem involves a tubular chemical reactor operating under steady-state conditions. Inside the reactor an exothermic reaction takes place, while a surrounding jacket enables the heat removal. The reactor model adopted is based on the 1D plug flow model with an irreversible first-order reaction from Smets et al. (2002); Logist et al. (2008). It is based on the following assumptions: (i) no axial dispersion, (ii) steady-state conditions, (iii) perfect radial mixing, (iv) a constant density and heat capacity of the fluid, (v) a negligible heat resistance of the reactor wall, and (vi) an Arrhenius law dependence of the reaction rate on the temperature. Using the spatial coordinate $\xi := z \in [0, L]$ along the reactor with length L , the reactor can in steady state be described by a highly nonlinear model:

$$\frac{dC}{dz}(z) = -\frac{k_0}{v} C e^{\frac{-E}{RT}} \quad (16)$$

$$\frac{dT}{dz}(z) = -\frac{\Delta H}{v\rho C_p} k_0 C e^{\frac{-E}{RT}} + \frac{4h}{v\rho C_p d} (T_w - T) \quad (17)$$

$$\frac{dH}{dz}(z) = \frac{4h}{\rho C_p d L} (T_w - T) \quad (18)$$

where C [mole/L], T [K] and H [1/s] indicate the reactant concentration, the reactor temperature, and the scaled heat exchanged. T_w [K] denotes the

jacket fluid temperature. v [m/s] is the fluid velocity and ΔH [J/kmole] is the heat of reaction ($\Delta H < 0$ for an exothermic reaction). ρ [kg/m³], C_p [J/kg K], k_0 [1/s], E [J/mole], R [J/mole K], h [W/m² K], and d [m] are the fluid density, the specific heat, the kinetic constant, the activation energy, the ideal gas constant, the heat transfer coefficient and the reactor diameter.

Initial conditions are $C(0) = C_F$, $T(0) = T_F$ and $H(0) = 0$, where, T_F and C_F are the temperature and the reactant concentration of the feed stream, respectively. Bounds are imposed on the reactor and jacket temperatures for constructive reasons:

$$T_{\min} \leq T(z) \leq T_{\max}, \quad (19)$$

$$T_{w,\min} \leq T_w(z) \leq T_{w,\max}. \quad (20)$$

For convenience dimensionless variables x_1 , x_2 , x_3 and u are introduced:

$$x_1 = \frac{C_F - C}{C_F}, \quad x_2 = \frac{T - T_F}{T_F}, \quad x_3 = H \quad \text{and} \quad u = \frac{T_w - T_F}{T_F},$$

as well as the constants α , β , γ and δ :

$$\alpha = k_0 e^{\frac{-E}{RT_F}}, \quad \beta = \frac{4h}{\rho C_p d}, \quad \gamma = \frac{E}{RT_F} \quad \text{and} \quad \delta = -\frac{\Delta H C_F}{\rho C_p T_F}.$$

Parameter values can be found in Smets et al. (2002); Logist et al. (2008).

As a result the following model is obtained:

$$\frac{d}{dz} x_1(z) = \frac{\alpha}{v} (1 - x_1) e^{\frac{\gamma x_2}{1+x_2}} \quad (21)$$

$$\frac{d}{dz} x_2(z) = \frac{\alpha \delta}{v} (1 - x_1) e^{\frac{\gamma x_2}{1+x_2}} + \frac{\beta}{v} (u - x_2) \quad (22)$$

$$\frac{d}{dz} x_3(z) = \frac{\beta}{L} (u - x_2) \quad (23)$$

with initial conditions:

$$x(0) = (0, 0, 0)^T \quad (24)$$

and constraints:

$$x_{2,\min} \leq x_2(z) \leq x_{2,\max}, \quad (25)$$

$$u_{\min} \leq u(z) \leq u_{\max}. \quad (26)$$

504
505

The aim is to derive an optimal profile along the reactor for the jacket temperature profile $u(z)$. The two objectives considered are similar to the conflicting ones treated in Logist et al. (2009a): maximizing the *conversion*, which is related to minimizing the reactant concentration at the outlet [mole/L]:

$$J_1 = C(L) = C_F(1 - x_1(L)) \quad (27)$$

and maximizing the net *heat transfer* between the reactor and its jacket, where heat transferred from the reactor to the jacket is assumed to a profit. Note that the unit is [1/s] as the heat transferred is made relative to the heat present in a reactor which is at T_F :

$$J_2 = H(L) = x_3(L). \quad (28)$$

506 The uncertain aspect is introduced via the dimensionless heat transfer co-
507 efficient $w(z) = \beta(z)$ along the reactor. This variable may fluctuate due to
508 fouling at the wall. As a result, it has a direct effect on the reactor tem-
509 perature constraint (26). Hence, in the current case a robustification of this
510 constraint is focused on.

511

512 **Motivation for the example.** It has to be noted that the model -
513 despite its conceptual character- can be regarded as a highly simplified ver-
514 sion of one peak of a tubular low-density polyethylene reactor under steady-
515 state conditions (Van Erdeghem et al., 2010). In this reactor the irreversible
516 propagation reaction mainly generates heat, while surrounding jacket ele-
517 ments allow the supply and/or removal of heat. As confirmed by industrial
518 partners, there exists a large interest in industry to maximize conversion and
519 minimize energy consumption, while accounting for effects due to fouling at
520 the wall, which is hard to predict in practice. From a mathematical point
521 of view, the model is highly nonlinear and has control and state constraints,
522 which need to be robustified. For the nominal case, results by Logist et al.
523 (2009a) can serve as a starting point.

524 3.2. Fed-batch bioreactor

525 The second benchmark case study involves a bioreactor based on the
526 fed-batch lysine fermentation process investigated in Logist et al. (2009b).
527 The aim is to determine an optimal feeding profile and batch length with

528 respect to conflicting yield and productivity objectives. Assuming perfect
529 mixing and no product degradation yields the following ODE equations with
530 as independent variable ξ the time $t \in [0, t_f]$ (Ohno et al., 1976):

$$\frac{d}{dt}x_1(t) = +\mu x_1 \quad (29)$$

$$\frac{d}{dt}x_2(t) = -\sigma x_1 + uC_{s,F} \quad (30)$$

$$\frac{d}{dt}x_3(t) = +\pi x_1 \quad (31)$$

$$\frac{d}{dt}x_4(t) = +uC_{s,F} \quad (32)$$

$$\frac{d}{dt}x_5(t) = +u \quad (33)$$

with initial conditions:

$$x(0) = \left(0.1 \text{ g}, 14 \text{ g} + \sqrt{2}w_0, 0 \text{ g}, 0 \text{ g}, 5 \text{ L} \right)^T \quad (34)$$

531 and constraints:

$$5 \text{ L} \leq x_5(t) \leq 20 \text{ L} \quad (35)$$

$$0 \text{ L/h} \leq u(t) \leq 2 \text{ L/h} \quad (36)$$

$$20 \text{ h} \leq t_f \leq 40 \text{ h} \quad (37)$$

$$20 \text{ g} \leq x_4(t_f). \quad (38)$$

532 Here, the state variables are the biomass x_1 [g], the substrate x_2 [g], the
533 product (lysine) x_3 [g], the amount of substrate added x_4 [g] and the volume
534 x_5 [L]. The control u [L/h] is the volumetric rate of the feed stream, which
535 contains a limiting substrate concentration $C_{s,F}$ of 2.8 g/L. The specific rates
536 for growth μ [1/h], substrate consumption σ [g/gh], and production π [g/gh]
537 are given as:

$$\mu = 0.125C_s \quad (39)$$

$$\sigma = \mu/0.135 \quad (40)$$

$$\pi = -384\mu^2 + 134\mu \quad (41)$$

with C_s [g/L] the substrate concentration. For practical reasons constraints
are imposed on the volume, the feed rate, the operation time and the amount

of substrate added through feeding after time $t = 0$ s. As the process is operated in fed-batch mode with an unspecified duration, both the feed rate $u(t)$ and the final time t_f have to be determined. The aim is to derive a feeding strategy which maximizes the productivity, i.e., the ratio between the product formed and the process duration:

$$J_p = \frac{x_3(t_f)}{t_f} \quad (42)$$

while maximizing the yield, i.e., the ratio between the product formed and the substrate added during the process:

$$J_y = \frac{x_3(t_f)}{x_4(t_f)}. \quad (43)$$

538 These maximization problems are cast into a minimization framework by
539 defining as cost functions the negative productivity and yield: $J_1 = -J_p$
540 and $J_2 = -J_y$. The uncertain parameter considered in this case is the sub-
541 strate concentration in the feed $w(t) = C_{s,F}(t)$, which needs a careful dosing
542 along the batch duration. It should be noted that a similar uncertainty is
543 also present the initial amount of substrate. Consequently, the uncertainty
544 does not only affect the possible satisfaction of constraint (38), but also the
545 biomass growth and lysine production, and thus both performance measures.
546 Hence, in the current case a robustification of both the constraint and the
547 objectives is concentrated on.

548
549 **Motivation for the example.** Although specific formulations for the
550 growth, consumption and production rates are employed that were originally
551 derived for the production of lysine by an auxotrophic mutant (Ohno et al.,
552 1976), it has to be noted that the methods remain applicable. The example
553 exhibits typical (i) optimal control features, e.g., a free end time, singular
554 arcs, end point constraints, and (ii) multi-objective challenges, e.g., a non-
555 convex Pareto set. Nevertheless, for a broader view on diverse kinetics and
556 their influence on nominal fed-batch reactor optimization, the reader is re-
557 ferred to, e.g., (Parulekar et al., 1988; Parulekar, 1992; Banga et al., 2003)
558 and the references therein. Industrial partners also confirmed that trade-offs
559 between yield and productivity have to be made on a regular basis depend-
560 ing on the market demand and the availability of fermenters. However, up
561 till now these decisions are made based on heuristics and intuition. Finally,
562 results for the nominal case can be compared to the ones reported by Sarkar
563 and Modak (2005) and Logist et al. (2009b).

3.3. Jacketed tubular chemical reactor: Nominal solution for conversion optimization

As an illustration, the nominal solution (i.e., without uncertainty) for the case with only the conversion cost (27) is computed using Multiple Shooting with a control discretization of 20 intervals. The resulting optimal state and control trajectories are depicted in Figure 3. As can be seen, the reactor is first heated as fast as possible until the upper reactor temperature limit is hit. From this point on an intermediate jacket temperature is used to remove exactly the amount of the heat produced by the reaction and hence remain at the upper reactor temperature bound. This observation is easily explained as higher temperatures stimulate conversion in irreversible first-order reactions. Moreover, it can be analytically shown that all arcs in the optimal profile are determined by active state and control constraints (Logist et al., 2009a).

Figure 3: Nominal jacketed tubular reactor profiles: concentration (top), reactor temperature (middle) and jacket temperature (bottom).

Figure 4: Nominal fed-batch fermenter profiles: states (top) and feed rate (bottom).

3.4. Fed-batch bioreactor: Nominal solution for yield optimization

Also the nominal solution for the fed-batch fermenter is calculated for the case that solely focuses on yield (43). To this end, a Multiple Shooting approach on 50 uniform discretization intervals is employed. Figure 4 displays the resulting state and control trajectories. As the duration is unimportant, the batch duration clearly reaches its upper limit. In the first part no additional substrate is fed and the originally present substrate is exploited. However, when the major amount has been taken up, an additional 20 g of substrate are fed according to a singular control law analytically derived by Modak and Lim (1987), in order to maintain biomass growth and lysine production. The amount of 20 g is chosen because an additional batch phase follows until the end and the constraint has to be satisfied. It should be noted that the singular arc significantly complicates the numerical optimal control computation due to the intrinsic low sensitivities of the cost with respect to the control. In addition, the free-end time condition, adding the total batch duration as a decision variable introduces additional nonlinearities. Hence,

accurate derivatives with respect to all decision variables are of major importance and to this end ACADO's automatic differentiation features have been exploited.

3.5. Jacketed tubular chemical reactor: robust solution for conversion optimization

In order to illustrate how the approach described above results in robust optimal solutions, the conversion is optimized for different levels of uncertainty on the heat transfer coefficient β between the reactor and its jacket. In practice, this heat transfer coefficient is often hard to measure/estimate and may vary along the reactor, e.g., due to local fouling at the reactor wall. As the heat transfer coefficient directly affects the reactor temperature and the upper reactor temperature limit in the constraint (25) is typically active when conversion is optimized, this constraint needs to be robustified. Since only the robustification of the upper reactor temperature constraint is asked, only the influence of uncertainty on the first two states is needed and the size of the Lyapunov matrix P is two by two. For a given percentage γ of uncertainty on the nominal heat transfer coefficient β , i.e., $\Gamma = \gamma\beta$, formulation (14) then leads to the following robustified path constraint:

$$x_1(z) + \gamma\beta\sqrt{(0 \quad 1) \begin{pmatrix} P_{1,1}(z) & P_{1,2}(z) \\ P_{2,1}(z) & P_{2,2}(z) \end{pmatrix} \begin{pmatrix} 0 \\ 1 \end{pmatrix}} \leq \frac{T_{\max} - T_F}{T_F}. \quad (44)$$

$$x_1(z) + \gamma\beta\sqrt{P_{2,2}(z)} \leq \frac{T_{\max} - T_F}{T_F}. \quad (45)$$

where the second term in the left-hand side is easily understood as a *back-off* to safeguard the constraint satisfaction.

Figure 5 displays the optimal state and control profiles for various γ levels. Hence, it is clearly seen that the higher the uncertainty is allowed to be (i.e., the higher γ is), the further the reactor temperature stays away from the upper bound of 400 K and the lower the conversion is at the end of the reactor. The slower initial rise in reactor temperature is because not the maximum transferable amount of heat is supplied to the reactor (i.e., the jacket fluid temperature is not at its upper limit) but the supply of heat is more carefully dosed in order to avoid a too fast reaction and heat production. Hence, most differences are seen in the region between 0.2 and 0.7, where the reaction and also the heat production is the strongest. As the reaction is

624 more or less completed toward the end, most of the differences decrease again.
625

Figure 5: Robustified jacketed tubular reactor: concentration (top), reactor temperature (middle), and jacket temperature (bottom).

626 This evolution is also visible in the top plot of Figure 6 which depicts the
627 evolution of the fourth element P_{22} in the Lyapunov matrix P . Obviously,
628 the uncertainty in the states is the largest in the region where most of the
629 reaction takes place. This region also slightly shifts towards the end of the
630 reactor when more back-off is taken into account and heat is supplied more
631 carefully. The bottom plot of Figure 6 presents the *uncertainty ellipsoids* for
632 the different uncertainty levels measured at a certain point in the reaction
633 zone (i.e., at $z = 0.4$). The uncertainty ellipsoids are computed as the Γ^2
634 level lines of the inverse of $C(\xi)P(\xi)C(\xi)^T$ and are part of an *uncertainty*
635 *tube* in the state space. This tube contains all possible state trajectories in a
636 first-order approximation. Hence, instead of forcing the nominal trajectory
637 to satisfy the constraints, the tube is now forced to meet the constraints
638 inducing a back-off and a performance loss. Obviously, for the nominal case,

i.e., $\gamma = 0$, the tube is equal to a line, whereas for increasing γ values the tube enlarges gradually. It should also be noted that the axes of the ellipsoids are tilted, indicating that the uncertainty on the heat transfer affects both the concentration and the reactor temperature. Also the axes turn slightly when comparing the ellipsoids for the different γ values. As an illustration, Figure 7 depicts this uncertainty tube in the state space for a γ value equal to 0.2. These ellipses are drawn based on the matrix P .

Figure 6: Robustified jacketed tubular reactor: Lyapunov state P_{22} (top) and confidence ellipsoids at $z = 0.4$ (bottom).

Figure 7: Robustified jacketed tubular reactor: uncertainty tube in the state space for $\gamma = 0.2$.

3.6. Fed-batch bioreactor: robust solution for yield optimization

Here, the robust optimization of the yield for the bioreactor is tackled, despite uncertainty in the substrate dosing $w(t) = C_{s,F}(t)$ and the initial amount of substrate present. Figure 8 depicts the optimal state trajectories for the first four states and the optimal controls. The fifth state x_5 is not depicted as no uncertainty is induced on the volume. As a result, the Lyapunov matrix has only to be four by four. Clearly, the addition of substrate starts earlier when a higher confidence level is required. Also the total amount of substrate added increases, since more and more back-off is required with respect to the constraint on the minimum amount to be added:

$$\sqrt{(0 \ 0 \ 0 \ 1) \begin{pmatrix} P_{1,1}(t_f) & P_{1,2}(t_f) & P_{1,3}(t_f) & P_{1,4}(t_f) \\ P_{2,1}(t_f) & P_{2,2}(t_f) & P_{2,3}(t_f) & P_{2,4}(t_f) \\ P_{3,1}(t_f) & P_{3,2}(t_f) & P_{3,3}(t_f) & P_{3,4}(t_f) \\ P_{4,1}(t_f) & P_{4,2}(t_f) & P_{4,3}(t_f) & P_{4,4}(t_f) \end{pmatrix} \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix}} \geq 2 \quad (46)$$

$$x_4(t_f) - \gamma C_{s,F} \sqrt{t_f} \sqrt{P_{4,4}(t_f)} \geq 2 \quad (47)$$

Note that the batch duration t_f enters the robustness margins explicitly as the final time is a degree of freedom which is assumed to influence the model

of the uncertainty set. On a longer time horizon, we expect the L_2 -norm over the uncertainties to be larger. Clearly, this higher amount of substrate that is inserted results in a higher amount of biomass and lysine being produced. However, this increase is dominated by the increase in the amount of substrate fed such that the yield (still) decreases. Moreover, also a back-off in the objective is assumed to be taken into account:

$$\begin{aligned}
 J_2 &= -\frac{x_3(t_f)}{x_4(t_f)} + \gamma C_{s,F} \sqrt{t_f} \\
 &\cdot \sqrt{\left(0 \ 0 \ \frac{1}{x_4(t_f)} \ \frac{-x_3(t_f)}{x_4^2(t_f)}\right) \begin{pmatrix} P_{1,1}(t_f) & P_{1,2}(t_f) & P_{1,3}(t_f) & P_{1,4}(t_f) \\ P_{2,1}(t_f) & P_{2,2}(t_f) & P_{2,3}(t_f) & P_{2,4}(t_f) \\ P_{3,1}(t_f) & P_{3,2}(t_f) & P_{3,3}(t_f) & P_{3,4}(t_f) \\ P_{4,1}(t_f) & P_{4,2}(t_f) & P_{4,3}(t_f) & P_{4,4}(t_f) \end{pmatrix} \begin{pmatrix} 0 \\ 0 \\ 1 \\ \frac{x_4(t_f)}{-x_3(t_f)} \\ \frac{-x_3(t_f)}{x_4^2(t_f)} \end{pmatrix}\right)} \quad (48) \\
 &= -\frac{x_3(t_f)}{x_4(t_f)} + \gamma C_{s,F} \sqrt{t_f} \\
 &\cdot \sqrt{\frac{1}{x_4^2(t_f)} P_{3,3}(t_f) - \frac{1}{x_4(t_f)} \frac{x_3(t_f)}{x_4^2(t_f)} P_{3,4}(t_f) - \frac{1}{x_4(t_f)} \frac{x_3(t_f)}{x_4^2(t_f)} P_{4,3}(t_f) + \frac{x_3^2(t_f)}{x_4^4(t_f)} P_{4,4}(t_f)} \quad (49)
 \end{aligned}$$

Figure 8: Robustified fed-batch fermenter: states (top) and feed rate (bottom).

664

665 The diagonal elements of the Lyapunov matrix are depicted in Figure 9.
666 All Lyapunov states start at 0 except $P_{2,2}(0) = 2$ because uncertainty is also
667 present on the initial substrate concentration. A similar increasing behavior
668 is seen for $P_{1,1}$, $P_{3,3}$, $P_{4,4}$. Moreover the increase is stronger for higher γ
669 values. Alternatively, a decrease is observed for $P_{2,2}$, which is only coun-
670 teracted when additional substrate is added. Also here the effects become
671 (slightly) larger when γ is increased. In the current case, the visualization
672 of the uncertainty ellipsoids is not possible due to the high dimensionality of
673 the regular and Lyapunov states.

Figure 9: Robustified fed-batch fermenter: diagonal Lyapunov states.

674 3.7. Jacketed tubular chemical reactor: Robust multi-objective results

675 Figure 10 depicts the multi-objective results for the jacketed tubular re-
676 actor. In this case, NBI has been used with 41 approximation points for
677 the Pareto sets. As can be seen, there is a trade-off between conversion and
678 energy recuperation. These results for the nominal case are in correspon-
679 dence with the ones reported in Logist et al. (2009a). When looking to the
680 robust results, it is clear that the Pareto front shifts to higher values for both
681 objectives when more robustness is demanded (i.e., γ increases). Although
682 the expected conflict and trade-off between performance and robustness are
683 encountered for all parts of the Pareto set, it is more pronounced at low en-
684 ergy recuperation values, i.e., when focusing on the reactant conversion. The

685 explanation is that in this case, the temperature has to be as high as possible
686 requiring to be over a larger section close to the upper reactor temperature
687 bound, which has to be robustified. Hence, more back-off is induced to guar-
688 antee the constraint satisfaction, leading to a larger performance decrease.
689 On the other hand, when energy recuperation is focused on, the upper bound
690 is only active in a small part of the reactor. Hence, any robustification of
691 this constraint will not have a large impact on the performance. It has to be
692 noted that no post-processing or additional filtering is needed as the returned
693 Pareto fronts exhibit in all cases a nice convex shape.

Figure 10: Jacketed tubular reactor: robust Pareto set.

694 3.8. Fed-batch bioreactor: robust multi-objective results

695 In Figure 11 the resulting robust Pareto sets for the fed-batch bioreactor
696 are displayed using each time NNC and 41 Pareto optimal points. It has to
697 be mentioned that J_1 and J_2 have been depicted, which are the negative val-
698 ues of the productivity and yield. Hence, using J_1 and J_2 in a minimization
699 frame as introduced here, will result in a maximization of the the yield and
700 productivity. Also here a clear trade-off is visible, i.e., a focus on productivity
701 typically leads to short batch times and large substrate feedings, while con-
702 centrating on yield results in longer batches with a more economical use of

the substrate resource. Also for this case, the nominal results are the same as previous results reported in Sarkar and Modak (2005); Logist et al. (2009b). When the level of uncertainty γ is increased, a shift to worse objective values is observed despite the insensitivity that singular optimal control problems often exhibit. One explanation is that not only the constraint but also both objective functions have been robustified. The robustified productivity can be found as:

$$J_1 = -\frac{x_3(t_f)}{t_f} + \frac{1}{t_f}\gamma C_{s,F}\sqrt{t_f P_{3,3}} = -\frac{x_3(t_f)}{t_f} + \gamma C_{s,F}\sqrt{\frac{P_{3,3}}{t_f}} \quad (50)$$

while the expression for J_2 is given in (49).

In contrast to the previous case, the shift is now more or less uniform in all regions of the Pareto set. It has to be noted that the Pareto sets exhibit a non-convex region due to constraint (38). However, an accurate distribution of the candidate solutions is obtained and no post-processing with a Pareto filter is required as all candidates solutions are Pareto optimal. This non-convex part is apparently intrinsic to the solution and does not change with the robustification (47), as it is not too much affected by an increasing level of uncertainty.

3.9. Final remarks

The two conceptual benchmark case studies include a variety of different optimal control problem aspects, e.g., nonlinear dynamics, singular and non-singular controls, fixed and free interval ends, Based on the observed results, it is concluded that accurate and efficient approximations of the robust Pareto sets can be obtained by the deterministic strategies presented in this paper.

Concerning the algorithmic details, a Multiple Shooting algorithm has been used which is combined with an exact Hessian SQP method exploiting the band-structure by a condensing routine as proposed in Leineweber et al. (2003a,b). When looking into one SQP iteration for the robust fed batch bioreactor set-up, it is observed that most of the time is spent in the integration and sensitivity generation which takes about 2.2 s per SQP iteration. This computation time is mainly due to the fact that exact Hessian matrices are computed. Here, it should be noted that the algorithm needs already first-order derivatives for the approximate robustification and construction

Figure 11: Fed-batch bioreactor: robust Pareto set. Note that J_1 and J_2 are depicted which are the negative values of the productivity and the yield (i.e., productivity loss and yield loss). Using J_1 and J_2 in the minimization frame described above results in the desired maximization of the productivity and yield.

of the Lyapunov differential equations. Thus, a computation of the exact Hessian matrix within the SQP algorithm requires third-order derivatives of the differential states which can in the current context only be computed with sufficient accuracy as efficient automatic differentiation techniques are exploited. The remaining time of one SQP iteration is spent in the QP solver which takes about 35 ms only and, hence, is negligible in comparison to the expensive integration and sensitivity generation. Finally, the multi-objective algorithm based on normalized normal constraints (NNC) needs in hot-start mode typically between 4 and 10 SQP iterations. Here, it should be noted that the Pareto front is computed with a very high numerical accuracy, i.e., with a KKT-tolerance of less than 10^{-8} . This gives a worst case computation time of around 22 s per Pareto point. This computation time can certainly still be improved by exploiting the particular structure of the Lyapunov equations inside the algorithm which will be part of future research. For larger problems the computation of the Lyapunov matrices may become expensive as the number of elements scales quadratically with the number of states

affected by the uncertainty and linearly with the number of uncertainties accounted for. For the robust plug flow reactor example the computation times are similar: 0.94 s per SQP step for integration and sensitivity generation and 2.4 ms for the QP solution.

The robust optimization framework is flexible to deal with (i) different kinds of uncertainty, e.g., on the initial values, along the interval, for fixed and free-end times, and (ii) different kinds of variables to be robustified, e.g., path and end point constraints as well as objective functions. Finally, it has been illustrated how robustness can be regarded as an additional conflicting objective in a multi-objective optimal control problem, yielding robust Pareto sets. Concerning to the scalability of the approach, $(n+m-1)!/(m!(n-1)!)$ single-objective optimization problems have to be solved when using equally spaced, convex weights (i.e., $w_i \geq 0$ and $\sum_i^m w_i = 1$) for WS, NBI, and (E)NNC with n the number of points between two individual minima and m the number of objectives considered by these methods. When exploiting, the ϵ -constraint method for the robustness objective, this number has to be multiplied by the number of uncertainty values selected. Hence, for the tubular reactor and bioreactor case, this leads to 41 points for the trade-off between (i) energy and conversion, and between (ii) productivity and yield, respectively, times 5 for the selected ϵ -constraint values, resulting in 205 parametric single-objective optimization problems to be solved. However, this increase for increasing numbers of objective functions is similar to other other multi-objective methods that try to approximate the entire Pareto set. Hence, when a high number of objectives is present, interactive multi-objective methods may be more appropriate also because visualization of the entire front becomes difficult (Miettinen, 1999).

4. Conclusion

This paper has discussed robust multi-objective optimal control formulations together with a solution framework employing the software ACADO Toolkit. Here, two approaches have been integrated: (i) deterministic scalarization based multi-objective approaches as ϵ -constraint, Normal Boundary Intersection, and Normalized Normal Constraint as well as (ii) robust optimization techniques for dynamic systems based on Lyapunov differential equations. As a result, robust Pareto sets, i.e., Pareto sets in which robustness is an additional objective, can be efficiently provided to the decision

789 maker allowing him/her (*i*) to clearly assess the different trade-offs involved
 790 and (*ii*) to pick one solution according to his/her preferences.

791
 792 Moreover, the paper has established two benchmark problems for which
 793 the applicability of the presented robust multi-objective optimal control tech-
 794 niques has been illustrated. The first problem was concerned about the max-
 795 imization of conversion and heat recovery in a jacketed tubular reactor with
 796 uncertain heat transfer between reactor and jacket, while the second problem
 797 was focussed on the maximization of yield and productivity in a fed-batch
 798 fermenter with an uncertain substrate concentration in the feed and an un-
 799 certain amount of substrate initially present.

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