

Constructive nonlinear dynamics in process systems engineering

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Abstract

To date, sensitivity, bifurcation and singularity analysis have been employed to identify and characterize the qualitative nonlinear behaviour of chemical process systems. The phenomena of interest include multiple steady states and periodic or even chaotic oscillations. The analyses have been aiming at proper understanding of the relation between the observed behaviour on the one and the process parameters as well as the underlying physical–chemical phenomena on the other hand. These methods have rarely been used to address synthesis problems, neither in process design nor in process control, where a desired process behaviour has to be realized according to given design specifications in a constructive manner. The present paper reviews the authors' recent work on constructive nonlinear dynamics that extends and applies ideas from nonlinear dynamics to address synthesis rather than analysis problems. The suggested method systematically accounts for process economics and process operability in an integrated framework. Further, model as well as process uncertainties can be addressed systematically. The suggested formalism is illustrated by means of examples from various areas of process systems engineering including process design, controller tuning and the integration of design and control under uncertainty. Additional opportunities for future research and application are pointed out.

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1. Introduction

The development and application of a variety of methods for the analysis of the nonlinear dynamics of process systems has a long tradition in chemical engineering research. Continuously improving software for numerical bifurcation analysis (Kuznetsov & Yu, 1999) by parameter continuation has made such analyses more and more attractive. The software package AUTO2000 (Doedel et al., 2001) and its predecessors have often been used by researchers in chemical engineering. Other software package also exist, but have not found such a widespread use, for example, CONTENT (Kuznetsov & Yu, 1998), which provides an easy-to-use interface to support a variety of analysis tasks, or DIVA (Mangold, Kienle, Gilles, & Mohl, 2000), which is particularly well-suited for the analysis of large-scale process models.

Early applications of numerical bifurcation analysis were aimed at deepening the understanding of the dynamics of chemical process systems in general. For this purpose, model problems have been chosen carefully to reflect the qualitative behaviour of an important class of process systems. The dynamic behaviour of these model processes can be represented by low-order nonlinear models, which can be treated with the analytical and numerical methods from nonlinear dynamics in a straightforward manner. The process studied most frequently is the continuous stirred tank reactor (CSTR) with various types of chemical reaction systems (for an overview see Razon & Schmitz, 1987). The seminal paper on the dynamics of CSTR with an exothermic irreversible first order reaction $A \rightarrow B$ by Uppal, Ray, and Poore (1974) is still up-to-date in that it demonstrates what type of information on the dynamics can be inferred from a bifurcation analysis by numerical parameter continuation. Most importantly, numerical bifurcation analysis is used to systematically detect and disclose stability boundaries due to saddle-node and Hopf bifurcations by one- and two-parameter continuation. While the first examples treated were restricted to small models ei-

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ther to illustrate the application of a mathematical technique or to get a fundamental understanding of a class of problems, this type of analysis has been applied more recently to industrially relevant process models of significant complexity including single chemical or biochemical reactors (e.g., Bildea & Dimian, 1998; Harold, Ostermaier, Drew, Lerou, & Luss, 1996; Khinast, Luss, Harold, Ostermaier, & McGill, 1998; Lei, Olsson, & Jørgensen, 2003; Morud & Skogestad, 1998; Ray & Villa, 2000) and distillation columns (Bekiaris, Meski, Radu, & Morari, 1993; Bekiaris, Meski, & Morari, 1996; Dorn & Morari, 2002) and distillation sequences (e.g., Esbjerg, Andersen, Müller, Marquardt, & Jørgensen, 1998; Güttinger & Morari, 1996), multi-functional processes such as reactive distillation columns (e.g., Kienle & Marquardt, 2002) as well as simple process plants (e.g., Kiss, Bildea, Dimian, & Iedema, 2002, 2003; Pushpavanam & Kienle, 2001; Zeyer, Pushpavanam, & Kienle, 2003).

While bifurcation analysis by continuation is an established method, there has been no systematic attempt so far to employ the rich theory of nonlinear dynamics to address synthesis problems in a rigorous manner. Rather, an iterative application of nonlinear analysis techniques embedded into a manual and time-consuming search in the parameter space has been employed. Typically, the designing engineer starts with an initial design with fixed process structure and parameters. He or she then employs nonlinear analysis methods to understand the behaviour and performance of this design in parameter space in the vicinity of the nominal design. From the results of such an analysis the designing engineer heuristically derives design modifications to better meet the design specifications. The process understanding accumulated during previous analysis phases can be effectively used to guide process design (for an example see Bildea & Dimian, 1998).

All these methods are focusing on analysis and are not directly addressing the synthesis problem. Synthesis has to be accomplished by the design engineer applying the analysis methods during a time consuming iterative search process. To overcome this limitation, a new set of nonlinear dynamics methods has been suggested by the authors in recent years (Mönnigmann, 2003; Mönnigmann & Marquardt, 2000, 2002, 2003). These nonlinear dynamics methods systematically address the synthesis rather than the analysis problem. The next section introduces the basic ideas without mathematical rigor first. Section 3 summarizes the technical issues to be tackled in order render the ideas operational. Section 4 introduces a number of problem formulations and

reviews selected case studies to illustrate the capability of the method. Section 5 puts this new approach into perspective with alternative problem formulations and solution techniques. We conclude with a summary and with an outline of future research issues.

2. Conceptual problem formulation

2.1. Preliminaries

In the present paper we assume that process models can be stated as a system of ordinary differential equations (ODE):

$$\dot{x} = f(x, u, \theta), \quad (1)$$

where x , u , and θ denote n_x -, n_u -, and n_θ -dimensional vectors of state variables, inputs, and parameters of the model, respectively. The vector-valued function f is assumed to be smooth with respect to x , u , and θ . The parameters θ comprise model parameters (such as a heat of reaction), equipment design parameters (such as a vessel volume), and operational parameters that are not manipulated by a controller or an operator (such as a feed temperature). For convenience, the notation $\eta^T = (u^T, \theta^T)$ is introduced, equation (1) is rewritten as:

$$\dot{x} = f(x, \eta), \quad (2)$$

and the domain of η is referred to as the parameter space. The problem class can easily be extended to differential–algebraic systems of index one (Mönnigmann, 2003). It is important to note that (2) can represent both open- or closed-loop processes. The parameter vector η may approximate time-varying quantities, if their dynamics is much slower than that of the process. For a more thorough discussion of the problem class, the reader is referred to other publications (Mönnigmann, 2003; Mönnigmann & Marquardt, 2003).

In the space of the parameters η , regions with qualitatively different process behaviour can be distinguished. These regions are separated by nonlinear boundaries, the so-called critical manifolds (for a sketch see Fig. 1). The critical manifolds are not apparent from the process model, but must be identified by often-tedious calculations. A typical case for a critical manifold is a stability boundary that separates a region of the parameter space in which a single stable steady state exists from a region with sustained oscillations around an un-

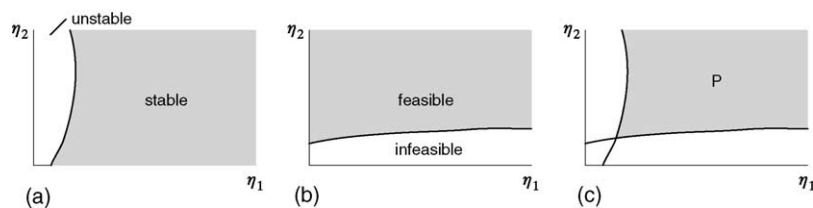


Fig. 1. Critical manifolds separate regions with qualitatively different process behaviour from one another: (a) stability boundary, (b) feasibility boundary, and (c) the intersection of the regions in (a) and (b).

stable steady-state (Fig. 1a). In this case, the critical manifold is a manifold of Hopf bifurcations, which can be found with, for example, a numerical bifurcation analysis (Kuznetsov & Yu, 1998). It is noted that simple inequality constraints on state variables, such as an upper bound on the process temperature, or on functions of state variables, give rise to critical manifolds too (Fig. 1b). In addition, other constraints on the process dynamics than stability boundaries can be described by critical manifolds. Mönnigmann and Marquardt (2000) show, for example, how information on the location of critical manifolds of cusp singularities can be used to avoid multiple steady states. Similarly, Gerhard, Mönnigmann, and Marquardt (2004) solve optimization problems with constraints on the location of nontransversal Hopf bifurcations. These constraints ensure that no stability loss can occur in a finite, user-specified, range of a bifurcation parameter. The concept of a critical manifold in fact provides a unified description of constraints on both process operation and dynamics in the parameter space (Mönnigmann, 2003; Mönnigmann & Marquardt, 2003).

2.2. Steady state process design by optimization

Any steady state $0=f(x, \eta)$ of the model (2) corresponds to a stationary operating point of a continuous process. Since a particular value $\eta=\eta^*$ fixes design and operational parameters of the process (for the set of chosen model parameters), the point η^* represents a certain design in the parameter space. Finding an appropriate value η^* therefore amounts to designing the process. The selection of the desired point in the parameter space can be interpreted as a simple synthesis problem if we assume a fixed process and model structure. The restriction to a fixed process and model structure has been guiding our research in the past. However, we expect that this restriction can be overcome by an appropriate extension of our method in the future.

In a typical design scenario, the design objectives are cast into an economic objective function ϕ . In a first attempt, an economically optimal steady state can therefore be determined by solving a problem of the form:

$$\min \phi(x, \eta) \text{ subject to } 0 = f(x, \eta). \quad (3)$$

Clearly this problem statement does not take information on the critical manifolds of the particular model into account.

In a next step, we therefore have to force the design into a particular region of the parameter space constrained by critical manifolds. For example, we want to make sure that any design results in a stable steady-state operating point rather than in an unstable point with an oscillatory regime. Similarly, critical manifolds due to feasibility constraints have to be taken into account in (3). If we introduce the region P , which is the intersection of those regions with a certain desired behaviour reflecting the design objectives (cf. Fig. 1c), the problem (3) can be replaced by:

$$\min \phi(x, \eta) \text{ subject to } 0 = f(x, \eta) \text{ and } \eta \in P. \quad (4)$$

The boundaries of P are given by parts of the critical manifolds separating regions in the parameter space with different qualitative process properties. If none of the critical manifolds bounding P gives rise to an active constraint, the problems (3) and (4) will result in the same optimal design. In the sequel, however, we assume that at least one critical manifold imposes a nontrivial restriction, cf. Fig. 2a and b. In this case, the optimal design is not in the desired region P in the parameter space for problem formulation (3) but is forced onto one of the boundaries of P for problem formulation (4). Denoting the values of the objective ϕ resulting from problem formulations (3) and (4) by $\phi^{(3)}$ and $\phi^{(4)}$, respectively, this implies $\Delta\phi = \phi^{(4)} - \phi^{(3)} \geq 0$. Hence, there is a loss $\Delta\phi$ in the objective due to the restrictions imposed by confining the design to a particular region P , which, for example, guarantees a certain qualitative dynamic behaviour. This loss is a quantitative measure for the cost of enforcing such qualitative dynamic behaviour or another constraint.

2.3. Design optimization under uncertainty

Depending on the nature of the objective function and the critical manifolds, the optimal design can either lie in the interior of the region P or on its boundary. In fact, the applications treated in Section 4 suggest that the latter case is more likely. If the solution of the optimization problem (4) results in a design on the boundary of P , this result is not robust, since even a slight change in η may cause the design to leave the desired parameter space region P . Thus, the design may cross a stability boundary, or an infeasibility may occur in the real process due to the parametric uncertainties in either model or operational parameters. In order to make use of the

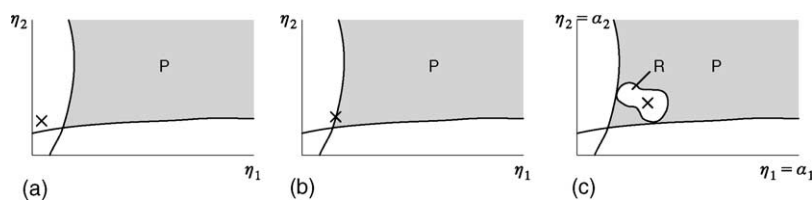


Fig. 2. The parameters of the optimal design are marked with a cross. (a) Problem statement (2) fails to take critical manifolds into account. (b) Problem statement (3) is likely to result in a steady state on one or more critical manifolds. (c) Problem statement (5) forces design to back off from the critical manifold into region P .

information on the location of critical manifolds in a manner that is meaningful for practical applications, parametric uncertainty has to be taken into account. To do so, the parameter vector η is split into two parts, a subset of n_p parameters p that are known precisely, and a subset of $n_\alpha = n_\eta - n_p$ parameters α that are uncertain. The parameter space is consequently split into two subspaces, which correspond to uncertain and certain parameters, respectively.

While a nominal process design corresponds to a single point in parameter space, taking the parametric uncertainty into account now unfolds this point into a region denoted by R in the (p, α) -space (for a sketch of a situation where only uncertain parameters exist, see Fig. 2c). With this uncertainty description, we now require the resulting design to lie in the desired region of the parameter space P despite the given uncertainties. Geometrically, the uncertainty region R that surrounds the nominal design has to be in the interior of the region P . The optimization problem to be solved therefore is:

$$\min \phi(x, \eta) \text{ subject to } 0 = f(x, \eta) \text{ and } R \setminus P = \emptyset. \quad (5)$$

The solution of the optimization problem is sketched in Fig. 2c. Obviously, it is not only determined by the design constraints but also by the shape of the uncertainty region R .

2.4. Leveraging the design loss by structural modifications

Assuming that the constraint $R \setminus P = \emptyset$ is not trivially met, the objective function value $\phi^{(5)}$ resulting from (5) will be larger than or equal to $\phi^{(4)}$, i.e., $\Delta\phi' := \phi^{(5)} - \phi^{(4)} \geq 0$. This profit loss is larger or equal to the loss $\Delta\phi$ that does not account for parametric uncertainty. The successive introduction of design specifications and parametric uncertainty will result in different desirable regions P and robustness regions R and ultimately to different losses after the solution of the associated optimization problem. This way, a systematic evaluation of the cost of a certain design specification or the uncertainty in a specific model or design parameter becomes possible. If the critical manifold is a stability boundary, for example, the loss measures the cost of requesting a stable operating point for a given uncertainty in selected model or process parameters. If the loss $\Delta\phi'$ is not acceptable, the designer might try to reduce the level of uncertainty in one or more of the parameters α . Geometrically, this uncertainty

reduction leads to a smaller uncertainty region R , which facilitates a design closer to the boundaries of P . If a reduction of the level of parametric uncertainty is not sufficient, a structural modification of the process can be envisioned. This structural modification may lead to a process with the desired qualitative behaviour, for example, stability, but with a smaller profit loss even in case of parametric uncertainty. Typically, if the nominal steady-state process is open-loop unstable, such a structural change is implemented by some type of feedback control or—less frequently—by some modification of the process or equipment itself.

3. Mathematical problem formulation and solution

This section presents some of the mathematical background necessary to implement the concept sketched in the previous section. The style is kept informal. References to more detailed literature are given.

3.1. Critical manifolds

In order to understand the concept of a critical manifold, it is instructive to consider a simple feasibility constraint first. Assume that a feasibility constraint has to be enforced for steady states of the process model (1), i.e., we are interested in steady states that obey $0 = f(x, \alpha, p)$ and further satisfy:

$$0 \leq g(x, \alpha, p) \quad (6)$$

where g is scalar and real-valued. In this simple case, the set of points at which the inequality is active defines the critical manifold M^c of interest

$$M^c = \{(x, \alpha, p) : 0 = f(x, \alpha, p) \text{ and } 0 = g(x, \alpha, p)\}. \quad (7)$$

As sketched in Fig. 3a, the projection of this critical manifold separates the space of the uncertain parameters α into the region in which (6) holds on the one hand, and the region in which (6) is violated on the other hand. Fig. 3b shows the projection of M^c into the space of the uncertain parameters α along with a robustness region R to be discussed below. The nominal values of the uncertain parameters are denoted by $(\alpha_1^{(0)}, \alpha_2^{(0)})^T$ in Fig. 3.

A larger class of critical manifolds can be described if the single equation $0 = g(x, \alpha, p)$ in (6) is replaced by a set of

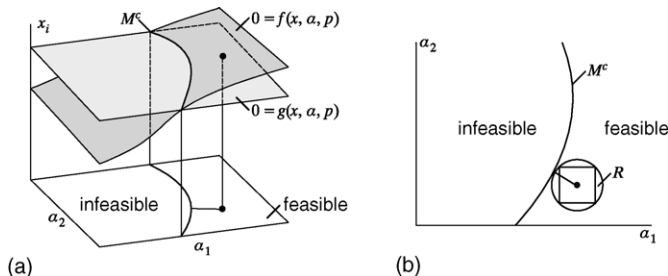


Fig. 3. (a) Illustration of a critical manifold M^c . (b) Closest distance along normal direction to the manifold. Nominal design at $(\alpha_1^{(0)}, \alpha_2^{(0)})^T$.

equations, i.e.,

$$M^c = \{(x, \tilde{x}, \alpha, p) : 0 = f(x, \alpha, p) \text{ and } 0 = \tilde{g}(x, \tilde{x}, \alpha, p)\} \quad (8)$$

In equation (8), \tilde{x} denotes a $n_{\tilde{x}}$ -dimensional vector of auxiliary variables that are necessary to state the defining equations \tilde{g} of the particular critical manifold. The function \tilde{g} has a range of dimension $n_{\tilde{x}} + 1$ and hence implicitly constrains a single state variable (Mönnigmann, 2003; Mönnigmann & Marquardt, 2003). For critical manifolds of the process model (1), f and \tilde{g} form the so-called augmented system for the critical phenomenon of interest. Often, these critical phenomena are bifurcations. Most importantly, saddle-node and Hopf bifurcations give rise to stability boundaries. Higher order bifurcations and singularities such as cusp or nontransversal Hopf points can also be related to engineering applications as demonstrated with an example in Section 4. A thorough discussion of the theoretical background is beyond the scope of the present paper. In the sequel, we will only make use of the fact that these systems can be stated in the form (8). The reader is referred to Kuznetsov and Yu (1999) for an introduction to applied bifurcation theory and to Golubitsky and Schaeffer (1985) for singularities of higher codimension.

As a natural extension to the stability boundary, critical manifolds can be defined to be steady states at which the real part of the leading eigenvalue attains a user specified value $\sigma_0 < 0$. Such a critical manifold is interesting from a technical point of view because it separates those steady states which have a decay rate of σ_0 or faster to linear order from steady states for which disturbances are rejected more slowly. Formally, the resulting critical manifolds are a simple extension of the augmented system of the Hopf bifurcation (Mönnigmann & Marquardt, 2002). A simple example is given in Section 4.3.

3.2. Distance to a critical manifold

Based on the concept of a critical manifold, the robustness of a candidate nominal design $\eta^{(0)T} = (\alpha^{(0)T}, p^{(0)T})$ can be quantified. The distance r of $\alpha^{(0)}$ to the critical manifolds in the subspace of the uncertain parameters α is used as a robustness measure. The locally closest distance between $\alpha^{(0)}$ and the projection of the critical manifold onto the α -space

occurs along the direction that is normal to the critical manifold as shown in Fig. 3b. In this figure, the uncertainty box $\alpha_i \in [\alpha_i^{(0)} - \Delta\alpha_i, \alpha_i^{(0)} + \Delta\alpha_i], i = 1, \dots, n_\alpha$, is overestimated by a ball. By enforcing the distance $|r|$ between $\alpha^{(0)}$ and the critical manifold along the normal direction r to be larger than the radius of the ball, the critical manifold is guaranteed not to be crossed, regardless of the actual values of the uncertain parameters in the robustness box.

Mönnigmann and Marquardt (2000) show that the normal vector r can be calculated from the defining equations $0 = \tilde{g}(x, \tilde{x}, \alpha, p), 0 = f(x, \alpha, p)$ in equation (8). Here, we do not digress to discussing the construction of sets of equations for the calculation of normal vectors, but only cite the result. According to Mönnigmann and Marquardt (2000) the normal vector can be calculated from equations of the form:

$$0 = G^{(c,i)}(x^{(c,i)}, \tilde{x}^{(c,i)}, \alpha^{(c,i)}, p^{(c,i)}, r^{(c,i)}), \quad (9)$$

where the upper index (c, i) denotes the quantities that belong to the critical manifold number i , r refers to the desired normal vector, and $G^{(c,i)}$ comprises $n_x + n_{\tilde{x}} + n_\alpha + n_p + n_r$ equations which have full rank at solutions (Mönnigmann, 2003; Mönnigmann & Marquardt, 2000). The structure of these equations depends on the type of critical manifold such as one stemming from saddle-node or Hopf bifurcations or from a feasibility constraint.

As pointed out in the previous section, the approach presented here is not restricted to describing parametric uncertainty by boxes $\alpha_i \in [\alpha_i^{(0)} - \Delta\alpha_i, \alpha_i^{(0)} + \Delta\alpha_i], i = 1, \dots, n_\alpha$. Fig. 4a sketches a general robustness region around a candidate nominal value $\alpha^{(0)}$ for the uncertain parameters. Parametric robustness can be enforced in such a case by requiring the locally closest connections between the robustness manifold M^r and the critical manifold $M^{(c,i)}$ to be larger than or equal to zero. The locally closest connections between M^r and $M^{(c,i)}$ occur along directions that are normal to both, the critical manifold and the robustness manifold. Mönnigmann and Marquardt (2003) show that a large class of robustness regions M^r can be described by considering the boundary of M^r to be a manifold of the same form (8) as the critical manifolds. In order to distinguish the robustness manifold normal vector system from (8), all quantities for the robustness manifold normal vector system are labeled with an upper index (r, i) instead of (c, i) .

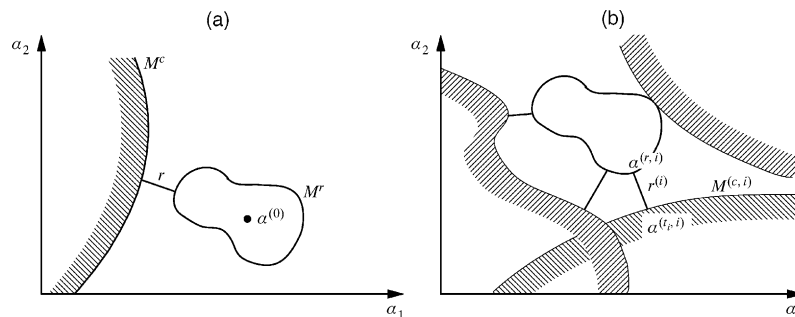


Fig. 4. (a) Uncertainty need not be described by a box. (b) Multiple closest connections exist due to multiple critical manifolds and non-convexity.

Fig. 4b illustrates that generally more than one critical manifold exists. Assuming that i_{\max} locally closest connections exist, the optimization problem with constraints for robustness reads:

$$\min_{x^{(0)}, \alpha^{(0)}, p^{(0)}} \phi(x^{(0)}, \alpha^{(0)}, p^{(0)}) \quad (10a)$$

$$\text{s.t. } 0 = f(x^{(0)}, \alpha^{(0)}, p^{(0)}) \quad (10b)$$

$$0 = G^{(r,i)}(x^{(r,i)}, \tilde{x}^{(r,i)}, \alpha^{(r,i)}, p^{(r,i)}, r^{(i)}) \quad (10c)$$

$$0 = G^{(c,i)}(x^{(c,i)}, \tilde{x}^{(c,i)}, \alpha^{(c,i)}, p^{(c,i)}, r^{(i)}) \quad (10d)$$

$$0 = l^{(i)} r^{(i)} - (\alpha^{(c,i)} - \alpha^{(r,i)}) \quad (10e)$$

$$0 \leq l^{(i)} \quad (10f)$$

$$i = 1, \dots, i_{\max}. \quad (10g)$$

Equation (10b) ensures that the optimal design $(x^{(0)}, \alpha^{(0)}, p^{(0)})$ is a steady state of process model (1). Equations (10c) and (10d) ensure that the critical manifold $M^{(c,i)}$ and the robustness manifold $M^{(r)}$ are connected by a common normal direction $r^{(i)}$, cf. Fig. 4b. Constraints (10e) and (10f) guarantee that a distance larger than or equal to zero exists along this direction. For a more detailed discussion the reader is referred to Mönnigmann (2003) or Mönnigmann and Marquardt (2003).

3.3. Numerical solution

In order to describe the robustness of a candidate design $(x^{(0)}, p^{(0)}, \alpha^{(0)})$ by its distance to the critical manifolds, the location of these critical manifolds must be known. An analysis of the critical manifolds is often tedious, however. Since existing methods for the analysis of critical manifolds for process dynamics strongly rely on visualizations, a thorough analysis of these manifolds is only practical for process models with a few uncertain parameters. Clearly, an optimization method for parametric robustness must not rely on an a priori analysis of the critical manifolds, but it must take the critical manifolds into account automatically.

Rather than analyzing the critical manifolds a priori, they can be detected as the optimization proceeds. From research in applied bifurcation analysis, real-valued test functions are known which signal the crossing of a critical manifold by a sign change (Kuznetsov & Yu, 1999). With these test functions, an optimal robust design can be found by solving the optimization problem (10a)–(10g) repeatedly while iteratively building up information on the critical manifolds. Assuming that a feasible solution and some critical manifolds $i = 1, \dots, j_{\max}$ are known (possibly none to start with), the optimization can be started with constraints on the distance to these j_{\max} known critical manifolds. Loosely speaking, the optimizer will push the robustness region through the search space, and previously unknown critical manifolds are signalled by sign changes in the test functions. Constraints on the distance to these previously unknown critical manifolds are then

added to (10a)–(10g), and the process is repeated until no new critical manifolds must be taken into account. Mönnigmann and Marquardt (2005) successfully demonstrated that this approach can be used for the optimization of examples with a few hundred model equations without a priori knowledge on the existence and location of critical manifolds.

The algorithm described so far does not guarantee a global solution to the problem. In some cases, the algorithm may miss a critical manifold, which cuts the robustness region R after convergence. In this case, the critical distance would not be maintained to this manifold. Such cases can only be avoided, if rigorous search methods are employed. Mönnigmann et al. (2004) present such a rigorous search method based on interval arithmetics, which is however limited to problems of moderate complexity.

3.4. Software implementation

Several technical issues need to be resolved for an implementation of the method sketched here. Most importantly, equation (9) and the defining equations $0 = \tilde{g}(x, \tilde{x}, \alpha, p)$ in equation (8) contain higher order derivatives of the process model equations. These derivatives are currently calculated with symbolic and automatic differentiation by MAPLE (Monagan et al., 2000) and ADIFOR (Bischof, Carle, Hovland, Khademi, & Mauer, 1998), respectively.

Furthermore, it must be pointed out that the test functions are only meaningful at steady states of the process model. The optimization algorithm used to solve (10a)–(10g) therefore must be of the feasible path type (e.g., FSQP, Lawrence & Tits, 2001), if the test functions are to be evaluated simultaneously. The restriction of having to use a feasible path optimizer can be relaxed, however, by evaluating the test functions along a linear connection between the starting and end points of the optimization. For details, the reader is referred to Mönnigmann (2003).

4. Illustrating applications

The previous sections introduced the concept of a critical manifold and the idea of stating constraints in terms of distance between candidate points of operation and critical manifolds in the space of the uncertain parameters. Due to the generality of these concepts, the sketched approach is applicable to a variety of problems. This section demonstrates the application to process design, robust controller tuning and integration of design and control. The examples given here are simple and the discussions are brief due to limitations in space. References to more detailed discussions and larger examples are given, however.

4.1. Process design

In this application, a simple model for a fermentation in a well-mixed tank is optimized. The fermenter model is not stated here for brevity, but the reader is referred to

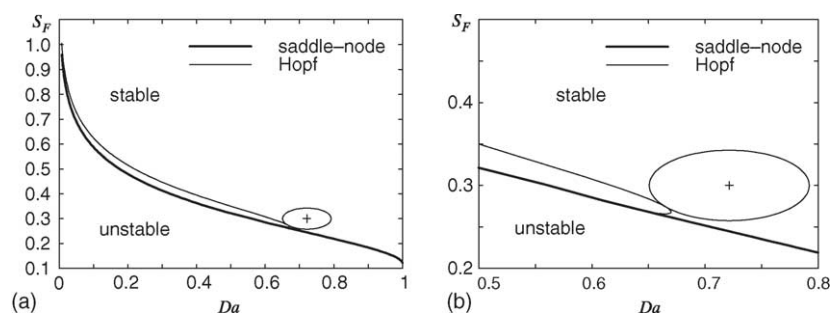


Fig. 5. (a) Result of the optimization of the fermenter with constraints for robust stability. (b) Enlargement of the robustness ellipse at the critical manifold due to saddle-node and Hopf bifurcations.

Agrawal, Lee, Lim, & Ramkrishna (1982) for details. The cost function ϕ in (10a) is the cost of the substrate diminished by the profit from produced cells in this example. The constraints (10b)–(10f) comprise the fermenter model and constraints on the distance to critical manifolds for stability. Since the process model has been analyzed before (Agrawal et al., 1982), we know a priori that two critical manifolds due to saddle-node and Hopf bifurcations exist. The constraints (10c)–(10f) have to be stated for saddle-node and for Hopf bifurcations, or, in other words, $i_{\max} = 2$ in (10g). The Damköhler number Da and the substrate feed concentration S_F are assumed to be uncertain parameters α with uncertainties $\Delta\alpha_1 = \Delta Da = 0.05$ and $\Delta\alpha_2 = \Delta S_F = 0.03 \text{ kmol m}^{-3}$. The constraints (10c)–(10f) ensure that the resulting optimal point of operation is stable despite this parametric uncertainty.

The model is first optimized *without* the constraints (10c)–(10g) for reference. The result is an optimal but unstable point of operation. The optimization is then repeated with the robustness constraints. This optimization results in an optimal stable point of operation which is robust in the sense that it remains stable despite the uncertainty in Da and S_F . This result is visualized in Fig. 5. The loss for guaranteeing robust stability is about 66% of the profit in the nominal case. Such a loss calls for a stabilizing controller (see Section 4.2) or a process design modification. For details on this example, the reader is referred to Mönnigmann (2003).

A similar but more involved application to a continuous polymerization process is given by Mönnigmann and Marquardt (2003). The polymerization is optimized with respect to an economic profit function. In order to guarantee parametric robustness with respect to stability, critical manifolds due to Hopf and saddle-node bifurcations have to be taken into account. In addition, an upper bound on the process temperature gives rise to a critical manifold of the feasibility constraint type. The example demonstrates that the approach presented here can be used to treat feasibility constraints and constraints on the dynamics in a unified manner.

4.2. Robust controller tuning

The previous examples addressed the robustness of a single optimal point of operation. Robust stability often has to be

guaranteed over wide ranges of operating conditions rather than for a single point of operation, however. For example, if various grades are to be produced for a range of production capacities, the process must be stable despite the demanded flexibility. In this section, we discuss a simple example, where the robustness constraints (10c)–(10f) are used to guarantee parametrically robust stability for a large range of operating conditions. The example considered is a cooled CSTR with an exothermic first order reaction $A \rightarrow B$. Unmodeled dynamics are represented by an overdamped second order process. A feedback linearizing controller is used to control the temperature in the vessel. We are interested in a controller tuning which guarantees robust stability in a large region of operating temperatures. A bifurcation analysis of the model reveals that a lower bound on the controller time constant exists below which the region of process instability vanishes (Hahn, Mönnigmann, & Marquardt, 2003).

This study has been extended to output feedback control systems (Hahn, Mönnigmann, & Marquardt, 2004), where an observer is implemented to estimate the full state of the CSTR from available measurements. It is found that the plant–model mismatch has a much more profound impact on the tuning of the observer than it has on the controller tuning. Further, this study reveals that an observer design, which makes use of additional knowledge about the system, will not necessarily result in better stability properties as the level of uncertainty in the model increases.

A manifold of a particular type of bifurcation, a so-called nontransversal Hopf bifurcations, splits the closed-loop process parameter space into two regions with qualitatively different behaviour. While in one region unstable behaviour can occur depending on the value of the temperature controller set-point T_{sp} , process stability can be guaranteed for the entire range of T_{sp} in the other region. By backing off the critical manifold of nontransversal Hopf bifurcations at a user-specified distance, process stability can be guaranteed for the entire range of T_{sp} despite parametric uncertainty.

In this application, the cost function ϕ in (10a) is the yield of product B. Equations (10b)–(10f) are the CSTR process model and the robustness constraints for the critical manifold of nontransversal Hopf bifurcations of the form (8). For details on the defining relations of the critical manifold the reader is referred to Gerhard et al. (2004).

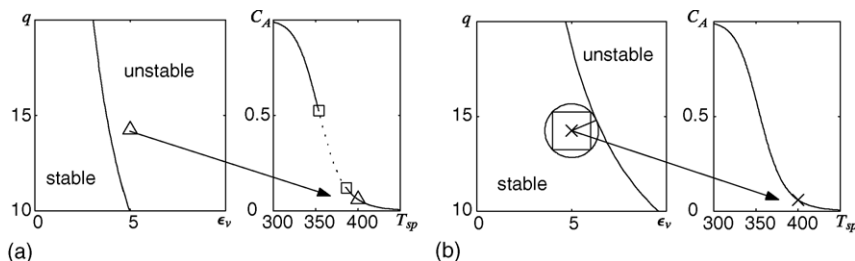


Fig. 6. (a) Optimization without constraints (10c)–(10f). (b) Optimization with constraints (10c)–(10f).

The feed rate q to the reactor and the time constant ε_v of the unmodelled dynamics are considered to be uncertain parameters α . The robustness ball in Fig. 6 overestimates the uncertainties $\Delta\alpha_1 = \Delta q = 10 \text{ mol min}^{-1}$ and $\Delta\alpha_2 = \Delta\varepsilon_v = 0.01 \text{ min}$. Fig. 6 illustrates the result. Fig. 6a shows the result of the optimization *without* constraints (10c)–(10f). This optimization has been carried out for reference only. For the resulting point of operation some values of the set-point T_{sp} are not admissible, since the process may become unstable (dotted line) due to Hopf bifurcations (\square). With robustness constraints, the process is stable for the entire range of T_{sp} (solid line) as illustrated in Fig. 6b

4.3. Integration of design and control

The example in Section 4.1 addressed the design of an open-loop fermentation process. This section presents a simple application to a closed-loop model. The fermenter model of Section 4.1 is augmented by a simple P-controller to demonstrate that both model and controller parameters can be determined by solving the optimization problem (10a)–(10g). It is stressed that this amounts to simultaneously tuning the controller, and designing the process for optimal operation with respect to an economic cost function.

In this example, the same cost function (10a) as in Section 4.1 is used. Equations (10b)–(10f) comprise the closed-loop model and the constraints for robustness with respect to a bound $\sigma_0 < 0$ on the real part of the leading eigenvalue as discussed in Section 3.1. The bound on the eigenvalues is chosen to be $\sigma_0 = -1/60$. By staying off this manifold, a decay rate of $1/(60 \text{ s})$ or faster is guaranteed for the closed-loop process to first order. Since only one critical manifold exists, $i_{\max} = 1$ in equation (10g).

The fermenter model is stated in dimensional variables for this application (Mönnigmann & Marquardt, 2003). The feed flowrate F is considered an input. A P-controller $F = F_0 + k_p(S - S_0)$ is added to the process, where S is the substrate concentration in the tank. The controller bias F_0 and the substrate feed concentration S_F are considered to be uncertain parameters α . The parametric uncertainties were assumed to be $\Delta\alpha_1 = \Delta F_0 = 0.7 \text{ m}^3 \text{ s}^{-1}$ and $\Delta\alpha_2 = \Delta S_F = 0.03 \text{ kmol m}^{-3}$. The result is illustrated in (Fig. 7). In the shaded area shown in (Fig. 7), the lead-

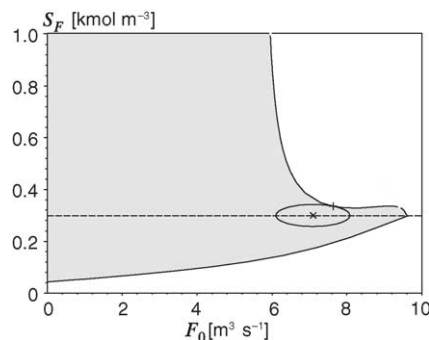


Fig. 7. Steady states of the closed-loop fermenter in the shaded area have a decay rate of $1/(60 \text{ s})$ or faster.

ing eigenvalue is smaller than $1/(60 \text{ s})$. The robustness ellipse touches the critical manifold thus guaranteeing a decay rate of $1/(60 \text{ s})$ despite the user-specified parametric uncertainty.

The same approach has successfully been used in the optimization of larger process models. Grosch, Mönnigmann, and Marquardt (2003) optimized a continuous crystallization process. The crystallization is modeled with a population balance, which is discretized by the methods of moments. Simple crystallization kinetics given by Volmer's law for nucleation and McCabe's law for crystal growth are used. The open-loop process turns out to have an optimal point of operation, which is unstable. In order to avoid sustained oscillations due to a Hopf bifurcation, the process is augmented by a PI-controller. The closed-loop model is then optimized with an upper bound $\sigma_0 < 0$ on the real part of the dominant eigenvalue, simultaneously tuning the controller and obtaining an optimal robust steady state of operation.

Similarly, Mönnigmann and Marquardt (2005) use the approach sketched in the present section to optimize the reaction section of Douglas' HDA process. An optimal point of operation is found in this example for which a user-specified decay rate can be guaranteed despite parametric uncertainty. The HDA model comprises several hundred equations and twelve uncertain parameters. This example therefore demonstrates that the proposed approach can be applied to large-scale models.

5. Discussion

5.1. Limitations and obvious extensions

Several extensions of the approach presented here are currently being investigated. We give a brief account of the major ideas. A more detailed description along with first examples to illustrate the potential of synthesis methods based on critical manifolds and robustness regions can be found in the thesis of Mönnigmann (2003).

Most importantly, the restrictive assumption on the dynamics of the quantities η has to be relaxed. These quantities have to be either constant, or they may vary on a time-scale that is much slower than the dominating process time. A suitable parameterization of time-varying inputs and performance indices can be used to address this issue. Bounds on performance indices can also be cast into a new type of a critical manifold. By means of an example Mönnigmann (2003) shows that a bound on performance indices such as the integral squared error (ISE) gives rise to critical manifolds of the same type as those of a stability boundary. Since the ISE increases, loosely speaking, both with larger frequencies of oscillation and smaller decay rates, the idea of bounding the ISE above is a natural extension of the critical manifolds defined by the bounds on the eigenvalues as briefly sketched in Section 4.3.

In an alternative extension of the existing method, bounds on trajectories of the dynamical system can be used to define critical manifolds for the response of a nonlinear system to time-varying disturbances. As opposed to the extension employing critical manifolds of performance indices and input parameterization, the critical boundaries for trajectories do not have to rely on the steady-state assumption.

On a different track, the stability boundaries known from applied bifurcation theory have to be generalized to critical manifolds that are more relevant from a practitioners point of view. While bifurcation theory focuses on the stability of solutions, a stable solution with a very small real part of the leading eigenvalue is of little interest from a practical point of view. Critical manifolds defined as the steady-states at which a user-specified bound on the leading real part is attained remedy this problem as demonstrated in Section 4.3. A natural extension to bounding the real part is to confine eigenvalues to a sector in the open right half of the complex plane.

All of the examples investigated so far in our research have been based on process models of the ODE type. An extension of the theory to DAE models of index one is straightforward. An implementation of such an extension is planned for the near future. A more interesting extension relates to the treatment of distributed parameter systems. A straightforward extension of our method is the approximation of the distributed parameter model by a lumped ODE or DAE model by means of the method of lines. This would be in line with research related to the analysis of the nonlinear dynamics of distributed parameter systems (e.g., Jensen & Ray, 1982;

Pathath & Kienle, 2002). However, it is well known that the spatial discretization may significantly impact the stability behaviour. Stability boundaries may just move quantitatively but they also may vanish completely (Liu & Jacobsen, 2004). Hence, the impact of the discretization on the critical manifolds must be investigated in the future.

Our method, currently, only addresses a very restricted class of synthesis problems as a fixed and given model structure must be assumed. Typically, not only the process and control parameters but also the structure of the process and its associated control system are of interest during design, requiring the formulation of mixed-integer or disjunctive programming problems (for a review, see Grossmann (2002)). Even though we did not address this problem yet in our research, we would expect that the method can be extended in the longer run to such problems replacing the dynamic process model (1) by a disjunctive dynamic model (Oldenburg Marquardt, Heinz, & Leineweber, 2003) that allows for structural design alternatives.

The system size that can be tackled with the current implementation of the method is limited by the use of the dense derivatives matrices generated by ADIFOR (Bischof et al., 1998). The tractable system size can be expected to increase considerably if the sparse option of ADIFOR is used in the future.

5.2. Relation to other work

Due to the general applicability of the concept of a critical manifold, the proposed approach cannot only be applied to design for a certain qualitative dynamic process behaviour but also to design for process feasibility.

The application to feasibility constraints relates the presented approach to research on design under uncertainty. Numerous articles have addressed this problem over the last two decades (for a brief summary see Mönnigmann & Marquardt, 2003). Many articles on design under uncertainty are based on feasibility and flexibility measures for nonlinear process models that were introduced by Grossmann and co-workers (Halemane & Grossmann, 1985; Swaney & Grossmann, 1985). These measures are based on assessing the constraint violation. The idea of constraint violation is to rate designs (x, α, p) by the value of the function g in (6). Clearly $g(x, \alpha, p) \geq 0$ and $g(x, \alpha, p) \leq 0$ indicate feasibility and infeasibility, respectively. In addition, however, the particular value of $g(x, \alpha, p)$ is used to compare designs. Among several infeasible designs $(x^{(i)}, \alpha^{(i)}, p^{(i)})$, the one that yields the smallest constraint violation $g(x^{(i)}, \alpha^{(i)}, p^{(i)})$ is, loosely speaking, considered to be the best one. While this seems to be obvious for simple feasibility constraints (such as an upper bound on the temperature in a unit, for example), it is not clear which assumptions must hold for the function g in (6) in general. Assume, for example, that we know a feasible steady-state $(x^{(1)}, \alpha^{(1)}, p^{(1)})$ for which a constraint $g(x, \alpha, p) \geq 0$ is active, i.e., $g(x^{(1)}, \alpha^{(1)}, p^{(1)}) = 0$. Further assume that we know that increasing $p^{(1)}$ by a small number $\varepsilon > 0$ renders

the feasible steady state infeasible, i.e., $g(x^{(1)}, \alpha^{(1)}, p^{(1)}) = 0$, $g(x^{(2)}, \alpha^{(1)}, p^{(2)}) < 0$ for $p^{(2)} = p^{(1)} + \varepsilon$. One would like to infer that for a third steady state $0 = f(x^{(3)}, \alpha^{(1)}, p^{(3)})$ with $g(x^{(3)}, \alpha^{(1)}, p^{(3)}) < g(x^{(2)}, \alpha^{(1)}, p^{(2)})$ that $p^{(3)} < p^{(2)} < p^{(1)}$. Unfortunately, this cannot be inferred for general constraints g in (6).

In contrast, the measure used here is not based on evaluating a measure in the range of the constraint functions, but the distance between the candidate point of operation and the critical manifolds in the space of the uncertain parameters. Note that this is a measure that is directly defined in the space of the uncertain parameters. While this detail seems to be technical at first sight, it is the key to an approach that covers both feasibility and dynamical constraints. For constraints on the dynamics, an inequality of the type (6) can in general not be stated. The concept of constraint violation can therefore not be extended from feasibility constraints of the form (6) to constraints on the dynamics. A meaningful definition of the critical manifold (8) can, however, be stated based on the so-called augmented systems for bifurcation points known from applied bifurcation theory (Kuznetsov & Yu, 1999). Since both feasibility constraints and constraints on the dynamics, such as stability boundaries, can be described by critical manifolds a unified approach to robust stability and feasibility is possible. Previous approaches to design under uncertainty made use of matrix measures (Kokossis & Floudas, 1994; Mohideen, Perkins, & Pistikopoulos, 1997). While matrix measures are amenable to implementation, they are known to be conservative. Unfortunately, this conservativeness may result in an overestimation of the stability boundary and thus to suboptimal process designs only. Furthermore, the approach suggested seems to be a viable approach to systematically studying the interaction between design and control for nonlinear systems. Only very few papers have been treating this subject (see, for example, Brengel & Seider, 1992; Lewin & Bogle, 1996).

6. Summary

Methods for the analysis of the dynamics of nonlinear process models are well established in the chemical engineering community. While these methods are very mature and powerful, they rely on visualizing numerical data and on subsequently interpreting diagrams. Unfortunately, an approach which depends on manual visualization and experience-based interpretation cannot be used systematically in process design. Further, nonlinear analysis becomes tedious or even impossible if the dimension of the space of relevant parameters is large. The present paper summarizes the ideas behind a new approach to taking dynamics into account at the design stage. To the authors' knowledge this is the first instance of a nonlinear dynamics method that is constructive in the sense that it does not rely on analysis, visualization and interpretation. The new approach is based on the concept of a critical manifold that separates regions of the design space with de-

sired process behaviour from those with undesired process behaviour. As the concept of a critical manifold can be applied to both dynamical properties and feasibility constraints, the proposed critical manifold-based constraints permit a unifying approach to robust stability and feasibility. Because the method relies on the distance to a critical manifold in parameter space, the curse of dimensionality limiting the applicability of analysis methods is not faced, because the normal to a manifold is always a one-dimensional object regardless the dimension of the parameter space. A number of examples have been briefly summarized to demonstrate the versatility of the approach.

To the authors' knowledge, the sketched critical manifold-based approach is the first systematic approach to considering stability at the process design stage which does not involve approximations such as matrix measures and which accounts for uncertainty. As the new approach allows to optimize a process model with respect to a profit function and to simultaneously take constraints on the dynamics into account, it is ideally suited for the integration of design and control. Our research will focus in the near future on a more detailed comparison to existing approaches to the design of robust controllers for nonlinear systems as well as on tailoring of the method to the integration of process and control system design.

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