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Bounded-error optimal experimental design via global solution of constrained min–max program

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ABSTRACT

We present an improvement of existing methods for globally solving optimal experimental design (OED) for bounded-error estimation based on a bilevel formulation from Mukkala et al. (2017). The proposed solution method for the min–max program is based on our method for generalized semi-infinite programs (via restriction of the right-hand side). The algorithm employed has the advantage that it guarantees a global solution for the OED assuming the global solution of two subproblems. To obtain a feasible solution only the lower-level problem has to be solved globally. In case of a local solution of the upper-level problem, the solution is still feasible though it is an upper bound of the global solution. The min–max method for OED is illustrated with four examples: two simple chemical reactions, BET-adsorption and a reformulated predator-prey system. The benefits of global methods are shown along with the limitations of state-of-the-art global solvers.

1. Introduction

Models are ubiquitously used throughout all fields of science and engineering. For example in process systems engineering, in model-based process and product synthesis and design, models are used to find the optimal process configuration or the optimal working fluid leading to minimal investment and/or operating expenses (Mechleri et al., 2017; Schilling et al., 2017). Similarly, in process planning, scheduling, and control, models are employed in order to find the process input leading to the optimal production schedule or to the desired process operation with respect to a certain objective (Grüne and Pannek, 2011; Pattison et al., 2017; Rawlings and Mayne, 2009). Due to the complexity of real-life systems, obtaining reliable models is a difficult task to be addressed during model development and validation processes (Franceschini and Macchietto, 2008; Marquardt, 2005), which consists of three major steps: experimental data collection, specification of various model candidates and data-model comparison. In order to select the most promising model and to obtain precise parameter values, model discrimination and parameter estimation methods are applied. A final validated model is furnished at the end of the model building process.

Models usually include parameters that need to be estimated before they can be used for further purposes. This is done by fitting the model predictions to experimental data, i.e., via parameter estimation. Two different categories of parameter estimation methods can be distinguished: statistical parameter estimation (Aster et al., 2013), often based on maximum likelihood methods, or bounded-error parameter estimation methods (Belforte et al., 1990; Milanese and Vicino, 1991), also termed set-membership or guaranteed parameter estimation (Kieffer and Walter, 2011; Paulen et al., 2015). In the former approach a point in the parameter space is estimated by selecting the most probable parameter value based on the experimental statistical error distribution. A confidence region shows how the measurement error distribution propagates into the parameter space. In contrast, in the bounded-error approach, the experimental error is given by attributing upper and lower bounds to the errors for every measurement point without making further assumptions on the statistical distribution. In other words, the only assumption made on the measurement errors is that they are bounded. In the context of bounded-error estimation, a parameter set is estimated that is consistent with the measurements and their error bounds, i.e., all model predictions from the estimated parameter set lie in a set defined by the measurements and their error bounds. The set of all consistent parameter values is called the feasible parameter set.

It is clear that independent of the parameter estimation method, the estimated parameters are known only within a given uncertainty, due to the experimental measurement errors. Uncertainties resulting from model structure inaccuracies will not be

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considered in this article. The parameter uncertainties can be influenced by the experimental conditions, e.g., the initial concentration of a reactant in a chemical reaction and measurement errors. In order to ensure the model reliability, these uncertainties have to be as small as possible. Thus, the question arises how to perform an experiment such that the uncertainties of the parameters are minimal, i.e., the parameter precision is maximal. This question can be answered by the methods termed optimal experimental design (OED) for parameter precision (Atkinson, 2011; Pronzato, 2008; Pronzato and Walter, 1990).

Following the parameter estimation classifications above, two respective approaches for OED exist: statistical OED (Atkinson, 2011; Franceschini and Macchietto, 2008; Pronzato, 2008) and bounded-error OED (Pronzato and Walter, 1990). The former approach corresponds to the statistical parameter estimation methods, where the size of the confidence region is a measure for the uncertainty of the parameters. Hence, the goal of statistical OED is to find the experimental conditions under which the confidence region is as small as possible. In OED for bounded-error estimation, the uncertainty in the measurements is considered only by the error bounds as is done in bounded-error parameter estimation. The size of the feasible parameter set is used as a measure for the parameter uncertainty. It is particularly advantageous in cases where no statistical assumption can be made on the measurement errors, e.g., if experimental measurement data is rare, as in biological systems, so that collecting sufficient data for proving the statistical assumptions on the measurement data is not possible (Milanese and Vicino, 1991; Walter and Piet-Lahanier, 1990).

Little literature is available on bounded-error OED despite the fact that it offers some advantages over the statistical OED. Norton (1987), Belforte et al. (1984, 1987), and Pronzato and Walter (1990) give an overview of OED for guaranteed parameter estimation. Borchers and Findeisen (2011) presented an OED for guaranteed parameter estimation for differential systems that are linear in the parameters. Hasenauer et al. (2010) and Marvel and Williams (2012) chose the optimal conditions from a given discrete set of conditions. Recently, Gottu Mukkula and Paulen (2016a) gave a rigorous and general method of OED for guaranteed parameter estimation neither restricted to linear models nor to discrete sets of conditions. It is based on bilevel optimization techniques, which determine the optimal experimental conditions for general nonlinear models. However, in their work, an optimization method was used which can not generally guarantee finding a globally optimal solution. To our best knowledge, there is no work that is generally applicable in the context of OED for bounded-error parameter estimation and can guarantee a globally optimal solution. For better understanding of the proposed method, a short review on bilevel and semi-infinite programing will be presented later on.

In this work, a bounded-error OED method is presented based on the work of Gottu Mukkula and Paulen (2016a, 2017). The bilevel optimization problem as proposed in these articles is recast as a min–max program and solved globally via an adaptation of an algorithm for generalized semi-infinite programs. The method is used for designing experiments for four test problems: a simple second-order reaction, a consecutive first-order reaction, BET-adsorption and a simplified prey-predator model.

2. Optimal experimental design for bounded-error estimation

Process systems are often described using dynamic models, such as ordinary differential equations (ODE) or differential-algebraic equations (DAE), or using static (steady state) models. In this work, the focus is on systems that can be written analytically as an input-output relationship:

$$\boldsymbol{y} = \boldsymbol{g}(\boldsymbol{v}, \boldsymbol{p}), \tag{1}$$

where $\boldsymbol{y}$ are the system outputs of dimension n_y, $\boldsymbol{p}$ the system parameters of dimension n_p, and $\boldsymbol{v}$ are the manipulated variables of dimension n_v consisting of the system's discretized control inputs $\boldsymbol{u}$ and/or initial values of the systems states $\boldsymbol{x_0}$. If the system is described using an ODE or DAE model, the system outputs $\boldsymbol{y}$ can be obtained via the numerical solution of the given dynamic system (Gear and Petzold, 1984), Ascher and Petzold (1998). In this case, global optimization dynamic solvers have to be used (Chachuat et al., 2006; Mitsos et al., 2009), which require substantial more effort. Global solution methods of dynamic systems are still limited to small systems; thus herein global solvers for algebraic systems are used and, therefore, only dynamic models with an explicit solution are considered in the following case studies.

As seen from Eq. (1), to accurately describe the behavior of the system and predict its outputs, the system parameters $\boldsymbol{p}$ need to be specified. Assuming that the model Eq. (1) correctly describes the system, i.e., the model is structurally correct, then the model accuracy depends on the parameter uncertainties. Increasing the model accuracy via parameter precision with the least experimental effort is the domain of optimal experimental design. In the following no assumptions on the parameter and measurement error distributions are made except that the parameters are bounded and measurement errors are bounded between $\boldsymbol{\eta}^L$ (lower error bound) and $\boldsymbol{\eta}^U$ (upper error bound), i.e., the measurement error $\boldsymbol{\eta}$ lies between $[\boldsymbol{\eta}^L, \boldsymbol{\eta}^U]$.

The following OED formulation is part of an overall model validation cycle (Franceschini and Macchietto, 2008; Marquardt, 2005). In general the starting point of the cycle is (i) initial experiments, followed by (ii) obtaining measurement data for the relevant system states, (iii) conducting parameter estimation and model refinement if need be, (iv) doing optimal experimental design and finally (v) carrying out new experiments, which closes the model validation cycle.

2.1. Problem formulation

The OED problem for bounded-error estimation utilizes a worst-case formulation, i.e., the largest possible parameter set that is still consistent with the model inequality constraints is minimized. In this formulation it is assumed that all possible manipulated variables $\boldsymbol{v}$ can be used for determining the optimal experimental design. The following formulation, with different notation, is taken from Gottu Mukkula and Paulen (2016a, 2016b) and Gottu Mukkula and Paulen (2017):

$$\Phi^* = \Phi(\boldsymbol{v}^*, \boldsymbol{p}^*) = \min_{\boldsymbol{v}\in V} \max_{\substack{\boldsymbol{p}^{L,i}\in P \\ \boldsymbol{p}^{U,i}\in P}} \sum_{i}^{n_p} (p_i^{U,i} - p_i^{L,i}) \tag{2}$$

$s.t.$

$$\left.\begin{array}{l} 2\boldsymbol{\eta}^{\boldsymbol{L}} \le \boldsymbol{g}(\boldsymbol{v}, \hat{\boldsymbol{p}}) - \boldsymbol{g}(\boldsymbol{v}, \boldsymbol{p}^{j,i}) \\ 2\boldsymbol{\eta}^{\boldsymbol{U}} \ge \boldsymbol{g}(\boldsymbol{v}, \hat{\boldsymbol{p}}) - \boldsymbol{g}(\boldsymbol{v}, \boldsymbol{p}^{j,i}) \end{array}\right\} \forall j \in \{U, L\}, i \in \{1, \ldots, n_p\} \tag{3}$$

with $\boldsymbol{p}^{L,i} = (p_1^{L,i}, \ldots, p_{n_p}^{L,i})^T$ and $\boldsymbol{p}^{U,i} = (p_1^{U,i}, \ldots, p_{n_p}^{U,i})^T$ with $i = 1 \ldots n_p$. P and V are a subset of real numbers of dimension n_p and n_v, respectively. The functions $\boldsymbol{g}$ correspond to the system outputs (see Eq. (1)), whereas, $\boldsymbol{g}(\boldsymbol{v}, \hat{\boldsymbol{p}})$ are calculated using nominal parameters $\hat{\boldsymbol{p}}$, which are known (or estimated) *a priori*. The total number of optimization variables is $2n_p^2 + n_v$ and total number of inequality constraints is $2(2n_y n_p)$. An optimal solution is denoted by $\boldsymbol{v}^*$ and $\boldsymbol{p}^*$. A trivial solution that always satisfies the above inequality constraints is $\boldsymbol{p}^{j,i} = \hat{\boldsymbol{p}}$.

The idea behind this OED formulation is to find the experimental conditions, $\boldsymbol{v}$, that would result in the smallest feasible parameter set that is consistent with the measurement errors, $\boldsymbol{\eta}^L$