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Optimal experimental design for ill-posed problems

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Abstract

Modern high-resolution measurement techniques offer the possibility to determine unknown functional dependencies directly from the data. The underlying inverse problems, however, are much more demanding than standard parameter estimation. Still, systematic strategies for experimental design of such ill-posed problems are missing. A new approach is proposed here that in particular achieves the sound integration of the bias-variance trade-off critical to the solution of ill-posed problems. The new design approach is based on the minimization of the expected total error (ETE) between true and estimated function. The ETE design approach is exemplified for the classical example of determination of reaction rates from measured data.

Keywords: experimental design, inverse problem, parameter estimation, reaction kinetics, numerical differentiation.

1. Introduction

In model-based experimentation, the goal is often to extract an unknown functional relationship from the data. Standard examples are e.g. reaction rates or phase equilibria as function of the state variables. The usual approach is to reduce the problem complexity: first, a model structure (or several candidates) is specified; then the unknown parameters contained are determined from experiments [1].

However, it would often be desirable to avoid the separation of the problem in two parts and to determine the unknown function directly. With the advent of high-resolution measurement techniques, modern process information management systems and advanced mathematical methods (e.g. data mining) this direct route is now becoming increasingly feasible [2].

Still, the identification of unknown functions represents an infinitely dimensional inverse problem. In addition, these problems are generally ill-posed, i.e. the solution is not unique or does not depend continuously on the data [3]. The solution of ill-posed problems for function estimation therefore poses much higher requirements on the data than standard parameter estimation problems where a finite number of parameters are determined in a known model structure.

Despite the increased complexity, the systematic generation of optimal experimental conditions for ill-posed problems has received only little attention. Model-based optimal design theory for parameter estimation, pioneered by Box & Lucas [4], is now well established. The approaches available for ill-posed problems are generally direct extensions of these classical design methods [5,6,7]. Since they are set in the maximum likelihood framework they assume unbiased estimates. However, in the solution of ill-posed problems, bias is systematically introduced to stabilize the problem. The trade-off between variance and bias is then the key element [3]. A sound approach to optimal

experimental design for ill-posed problems therefore has to incorporate this trade-off. However, none of the approaches currently available includes the bias effect.
A new design criterion for ill-posed problems is therefore introduced in this work. The criterion minimizes the statistically expected total error (ETE) between the true and the estimated function. It thus incldues both error contributions: bias and variance. The new criterion is derived next. Estimation of reaction rates from experimental data is then considered as an example application. A discussion of the new approach concludes this paper.

2. Design criterion for ill-posed problems

In order to limit the discussion to the essence of the method only linear problems are considered. Nonlinear problems can be treated using proper linearization as in standard design theory [4,5]. Linear ill-posed problems are often obtained from integral equations [3]

$$g(t) = \int_T K(t,s;d) f(s) ds \,, \tag{1}$$

where $f(t)$ is the unknown function to be identified from the measured data $g(t_i)$. Data is usually available only at discrete points t_i and corrupted by measurement errors (assumed here to be Gaussian with zero mean and variance σ^2). The kernel function $K(t,s;d)$ is generally known from theory and contains also the design parameters d that can be chosen by the experimenter. It is the goal of experimental design to find the optimal settings for these parameters.
For the solution of the inverse problem, direct inversion of Eq. (1) would lead to unstable solutions. Therefore, regularization methods have to be employed. The most common approach is Tikhonov regularization where the estimate for the unknown function f is determined as [3]

$$\hat{f} = \arg\min \sum_{i=1}^{n} \frac{1}{\sigma_i^2} \left(g(t_i) - \int_T K(t_i,s;d) f(s) ds \right)^2 + \lambda \left\| Lf \right\|_{L_2}^2 \,. \tag{2}$$

Here, the first term is the data error. The second term represents a penalty ensuring smoothness. For the operator L, the identity or the second derivative are frequently used. The regularization parameter λ gives the relative weight to both contributions of the objective.

The goal of a successful experiment should be that the estimate $\hat{f}$ is as close as possible to the true solution f. The expected value for the total error (ETE) between the Tikhonov estimate and the true function can be computed as [8]

$$E\left(\left\| f - \hat{f} \right\|_{L_2}^2 \right) = \left\| f - K^{-1}(\lambda) K f \right\|_{L_2}^2 + \sigma^2 trace\left(K^{-1}(\lambda) \left(K^{-1}(\lambda) \right)^T \right), \tag{3}$$
$$\text{where} \quad K^{-1}(\lambda) = \left(K^T K + \lambda L^T L \right)^{-1} K^T .$$

Assuming that an initial guess of the true solution and the measurement error is available it is therefore proposed here to obtain the optimal experimental design from minimizing the expected total error with respect to the design variables *d*. Thus, the optimal design *d** is determined from ETE criterion

$$\min_{d,\lambda} E\left(\left\|f-\hat{f}\right\|_{L_2}^2\right). \tag{4}$$

The first term of the ETE criterion in Eq. (3) reflects the bias introduced by the penalty term whereas the second term summarizes the variance in the estimate. Thus, the bias-variance trade-off is properly incorporated into the new ETE design criterion.

The regularization parameter λ integrates naturally as an additional free variable of the design optimization problem (4) and is determined along with the experimental settings. The ETE criterion thus provides a consistent rule to determine λ. Previous approaches had to rely on *a priori* knowledge [5,6].

Discretization of Eq. (3) is not critical. A simple trapezoidal scheme is usually sufficient since the discretization error is typically much smaller than regularization and data error [8].

3. Example: Identification of reaction rates – Numerical differentiation

The specific merits of the new approach are discussed in the light of an example. For this purpose, the determination of reaction rates as function of time from measured concentration data is considered [9]. Such model-based reaction rate measurements typically form the starting point for the identification of constitutive equations [2]. The core of the underlying mathematical problem is the differentiation of experimental data. This by itself is a standard problem in chemical engineering beyond the area of reaction kinetics since often not the measured quantity itself but its derivative is of interest.

In practice, the finite difference scheme is often employed to determine the unknown derivative $f=dg/dt$ from the measurements $g(t_i)$ [9]. Equidistant measurements with sampling interval $dt=t_i-t_{i-1}=const.$ are assumed here. The discretized kernel *K* is then a lower triangular matrix with all entries identical to *dt* [6].

In an experiment, the sampling interval *dt* can be chosen by the experimenter himself. It thus serves as design parameter. It is well known that if the sampling is too coarse the approximation will be poor. However, in the inverse problem, too fine sampling can also lead to an amplification of the error since the measurement noise will corrupt the result and the variance increases [3].

The ETE design criterion (3) is now applied to determine the optimal sampling interval for finite differences. No additional regularization parameter λ is required as the sampling interval itself has a regularizing effect. For the sound incorporation of the bias effect, the first term in the objective (3) is therefore computed using interpolation of the estimated solution on a finer grid.

In the example, a first-order reaction, leading to an exponential decay, serves as true function, i.e. $f=exp(-10t)$. Measurement standard deviation is $\sigma=0.01$.

The ETE design objective (3) is shown as function of the sampling interval *dt* in Fig. 1. The new criterion shows the expected behavior for the ill-posed problem. The optimal sampling time is found as the trade-off point between bias and variance contribution. Variance dominates the error for small time steps while bias increases for large time steps.

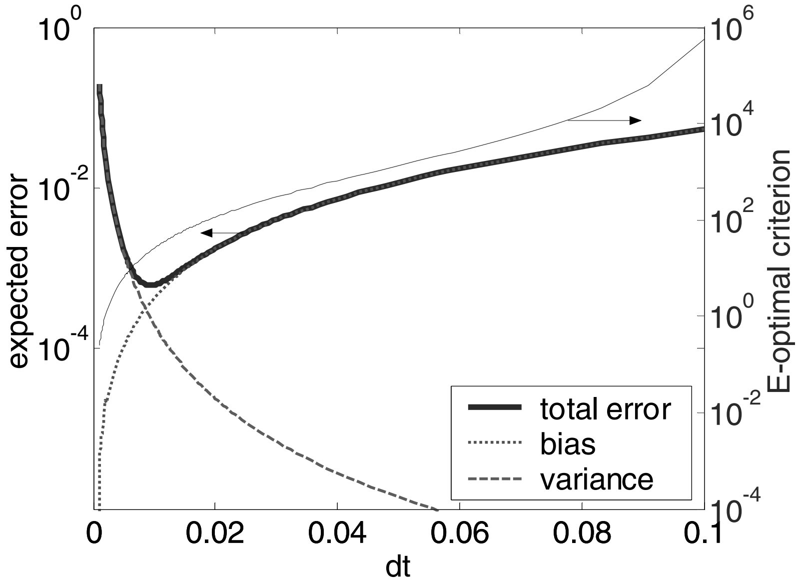

Fig. 1: ETE design objective as function of sampling interval dt. Bias (dotted) and variance (dashed) contributions to the objective are also shown (left axis). The E-optimal design criterion (thin full line) is shown on the right axis.

Criteria proposed previously for the design of ill-posed problems [5,6,7] solely focus on the variance contribution. In these methods, the so-called Fisher information matrix is usually introduced as a variance measure. The Fisher matrix corresponds here to the term inside the trace in Eq. (3). As an example for these design criteria, the E-optimal experimental design criterion [5,6] is plotted on the right axis in Fig. 1. In E-optimal design, the smallest eigenvalue of the Fisher information matrix is maximized [10]. It can be seen that the classical design criteria suggest the use of the maximum sampling time. Thus, these criteria are not able to reflect the specific nature of ill-posed problems.
In order to assess the quantitative accuracy of the ETE criterion a simulation study was performed. Simulated measurement data was corrupted with random noise and the finite difference scheme was applied to this data. The average deviation from the true signal was then evaluated and averaged over 10,000 replications. The average error is shown in Fig. 2. It can be seen that the ETE criterion truly captures the behavior found in the actual experiment. The predicted optimal sampling time is slightly larger than the value found in the simulation study which adds to the robustness of the estimate. In summary, it can be concluded that the ETE criterion is able to find the best sampling time with good accuracy.
The example of numerical differentiation studied here is well suited to show the specific properties of the new approach. However, it is also special since the design variable, the sampling time, serves at the same time as implicit regularization parameter. The success of the approach therefore shows at the same time that the new method is also able to initialize a regularization parameter. This step was missing in previous approaches [5,6].

4. Discussion and conclusions

It could be shown that the new ETE criterion is suitable for the experimental design of ill-posed problems whereas other approaches fail. Still, the new approach requires some

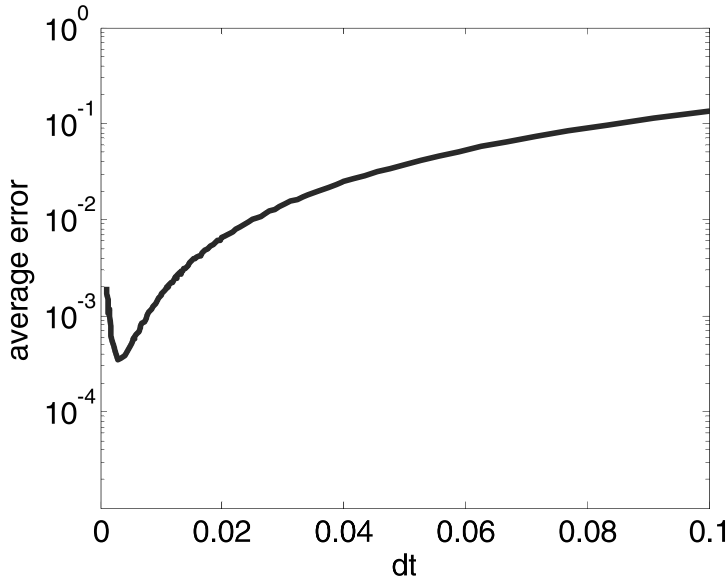

Fig. 2: Error as function of sampling interval dt computed using true solution and simulated measurement data averaged over 10,000 cases.

discussion. In particular, it relies on the assumption that an initial guess for the true solution *f* – which is actually sought – is available *a priori*. One may thus wonder about the practicality of the approach. However, it should be noted that this is the standard dilemma in experimental design theory. For nonlinear problems, more relevant in chemical engineering, it already cannot be avoided even in the simpler case of design for parameter estimation [4]. An iterative experiment cycle is thus usually required to find the desired solution [1]. This strategy may also be applied to the ETE approach. Still, even in the initial stage of an analysis, the ETE criterion can be adapted to the level of *a priori* knowledge available as briefly sketched in the following discussion.

Often, the experimenter has at least some qualitative knowledge about the general class of functions the solution should belong to. This is even true for more complex cases than presented in Section 3 (e.g. exponential decay for reaction rates, peak shaped functions for spectra, polynomials for transport coefficients). The criterion may then be used to study the influence of the design variables for the expected function class. This may already give important insight into the proper design. Robust design formulations (e.g. average, min-max design) could then be applied to obtain quantitative design rules [10]. These robust formulations could be of even more importance for nonlinear problems in order to capture the effect of local linearization.

In a case when there is really no reasonable assumption available the first term of the ETE criterion (3) may simply be neglected (*f=0*). The criterion then corresponds to a direct extension of the well-known *A*-optimal design criterion [10] to ill-posed problems. Such a design is therefore expected to provide at least a reasonable initial experiment.

In general, it is an important feature of the formulation that it identifies the individual contributions for bias and variance. Note that the assumed measurement error variance enters the formulation only as relative weight of these two terms (cf. Eq. (3)). A deeper

problem understanding can therefore be gained by a separate analysis of the dependence of bias and variance on the design variables.
In this context, it should be noted that the impact of the design variables on the bias could be approximately analyzed even without assuming any *a priori* knowledge on functional form for *f*. After discretization, the bias contribution is given by (cf. Eq. (3))

$$\left\| \left(I - K(\lambda)^{-1} K \right) f \right\| \le \left\| I - K(\lambda)^{-1} K \right\| \left\| f \right\|, \tag{5}$$

where I is the identity matrix. The right hand side follows from the submultiplicative property of the matrix norm. Assuming the true solution to be bounded and of order 1 (always possible by proper scaling) an analysis of the bias term could be based on the first matrix norm of the right hand side. Thereby, an upper bound for the bias would be studied. This would thus correspond to standard design theory where a lower bound for the variance from the Cramer-Rao theorem is used [10].
In summary, the expected total error (ETE) design criterion introduced in this work seems to provide the first sound framework for the experimental design of ill-posed problems. As discussed above, the method even yields design guidelines with minimal *a priori* knowledge. This property underlines the practical utility of the new approach.

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