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An Integration Based Optimization Approach for Parameter Estimation in Dynamic Models

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Abstract
A common problem in model verification is to determine the values of model parameters that provide the best fit to measured data, based on some type of least squares or maximum likelihood criterion. In the most general case, this requires the solution of a nonlinear and frequently nonconvex optimization problem. Some of the available software lack in generality, while others do not provide ease of use. As the need for a user-interactive parameter estimation software, especially for identifying kinetic parameters, was needed; in this work we developed an integration based optimization approach to provide a solution to such problems. For easy implementation of the technique, a parameter estimation software (PARES) has been developed in MATLAB environment. When tested with extensive example problems from literature, the suggested approach is proven to provide good agreement between predicted and observed data within relatively less computing time and iterations.

Keywords: parameter estimation, dynamic simulation

1. Introduction

Parameter estimation is a common problem in many areas of process modeling, both in ‘on-line’ applications such as real time optimization and in ‘off-line’ applications such as the modeling of reaction kinetics and phase equilibrium. The goal is to determine the values of model parameters that provide the best fit to measured data, generally based on some type of least squares or maximum likelihood criterion. The estimation of parameters in kinetic expressions from time series data is essential for the design, optimization, and control of many chemical systems. The models that describe the kinetics take the form of a set of differential algebraic equations. The statistics and formulation of this parameter estimation problem are well studied (Bard, 1974). In the most general case, this requires the solution of a nonlinear and frequently nonconvex optimization problem, which can be approached by dynamic programming (with substantial programming effort and computer time), Pontryagin’s maximum principle (that requires solution of adjoint vector whose initial values creating extra problems) or nonlinear programming techniques. Nonlinear programming may be pursued through discretization of all variables by finite difference approximations or orthogonal

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collocation. However, inclusion of the collocation variables immensely increases the size of the optimization domain and the degree of polynomial used has a significant impact on the error. The other option is to use the integration approach without transforming the differential equations into a fully algebraic NLP. Pontryagin's two theorems provide continuity and differentiability of the input-output (parameters - state variables) map with respect to the parameters. This method requires calculation of the sensitivities by integrating an extra set of differential equations at the expense of computation time. Esposito & Floudas (2000) provided an underestimating formulation for this method. Vassiliadis *et al.*'s (1994) control vector parameterization method in the same direction, although carries out the optimization in the space of decision variables, uses Lagrange polynomials for expressing control variables and again converts the problem into a finite dimensional NLP with added complexity in the algorithm.

From the perspective of existing software for dynamic model calibration, there are a number of programs available commercially, some of them being accessible via internet. However, most of them cannot fit some needs of the studied system satisfactorily even though they excel in other fields. For example the OPTKIN software supports large scale sets of equations with many parameters, but little customizability of these equations is allowed (Huybrechts & Van Assche, 1998) Therefore it is quite an efficient tool for the treatment of large sets of first order reactions (e.g. various radical mechanisms) but it does not allow application on some more complicated models. There are also limited possibilities for more detailed analysis of reliability and significance of parameters. The only generally applicable types of programs are the academic and semi-academic ones (Stewart *et. al*., 1992) but they are supplied as source code and provide poor user interface and require programming skills. Although ERA software package developed by Zamostny and Belohlav (1999) is a useful regression analysis tool, its input data matrix is limited to 20 independent variables and 20 responses, with up to 256 experimental points in each response and the number of model parameters is restricted to 15. Therefore, the floor seems to be open for further developments.

In this work, we have developed an integration based efficient integration algorithm, and a software for implementation, for parameter estimation in dynamic models without requiring calculation of sensitivity equations. The results obtained for some problems from the literature are compared, and its effectiveness is demonstrated.

2. Problem Formulation and Optimization Algorithm

The mathematical formulation of chemical reaction mechanisms, taken as exemplary systems to demonstrate the optimization algorithm, is given by a coupled system of stiff nonlinear differential equations

$$\frac{dy}{dt} = f(t, y; p), \quad y(t^0) = y^0, \quad t^0 \le t \le t^f \tag{1}$$

where y is the state vector of the system, p is the model parameters.

We used the control vector parameterization approach for integration such that the search space is discretized in time, i.e. in the space of control variables only. By assuming a piecewise constant control, we have been able to integrate the model equations so that the objective function can be evaluated. Running a numerical constrained/unconstrained optimization technique on top of this space allowed us to obtain the values of decision variables, i.e. parameters in the kinetic models, after a reasonable set of iterations to satisfy a stopping criterion. Since no approximation polynomials or sensitivity functions were required, the calculations were substantially simplified.

Figure 1 shows the logic flow diagram for the parameter estimation algorithm developed, which has been extensively tested before with a number of different problems (Agun 2002). The algorithm offers the possibility of employing different numerical optimization routines with ease to estimate the updated p in order to satisfy the particular needs of the model employed.

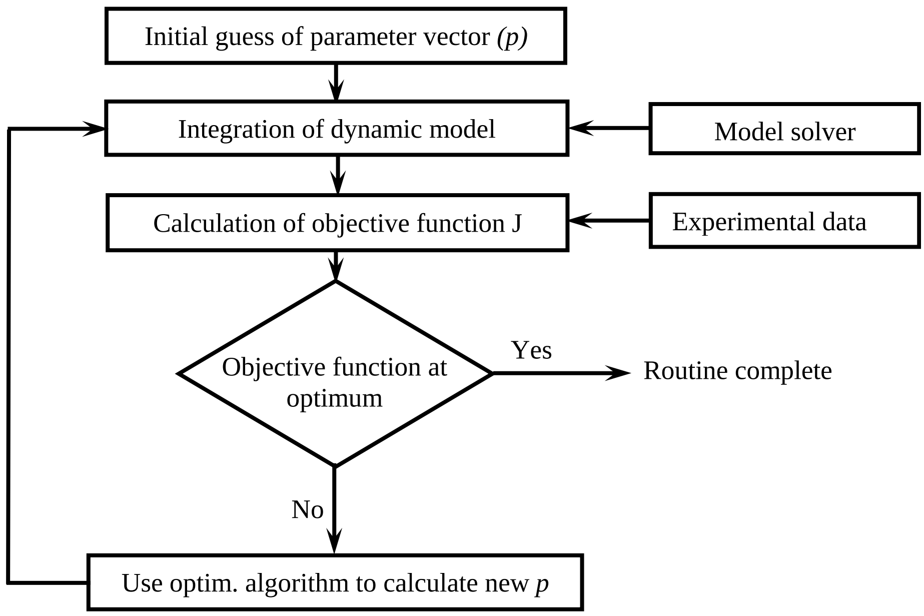


Figure 1. Basic iteration routine for parameter estimation

Model parameters were estimated by Quasi-Newton (QN), Nelder-Mead Simplex (NMS), Gauss-Newton (GN), Levenberg-Marquardt (LM), Sequential Quadratic Programming (SQP) algorithms by minimizing the objective function, which is the sum of squares of errors between the predicted and measured values for all of the state variables for a dynamic run as follows;

$$J = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(x_{ij} - x_{o,ij} \right)^2 \tag{2}$$

where x: computed value, x_o : observed value, n: total number of state variables and m: total number of observations. All computation was done in MATLAB environment.

3. PARES Software

A software PARES (PARameter EStimation), coded in MATLAB™ 6.5 has been developed to implement the suggested parameter estimation technique. PARES is an interactive software system to identify parameters in differential algebraic equation system models. The program has ability to make parameter estimation with different optimization methods. For unconstraint optimization problems, Gauss-Newton, Nelder-Mead Simplex, Levenberg-Marquardt or Quasi-Newton methods can be optionally used whereas sequential quadratic programming needs to be used for constraint optimization problems.

The program requires the input data in six main steps described as follows:

1. Model equations (user can enter any model, on the window provided as an M-file)
2. Number of model parameters
3. Initial, lower and upper bounds of the parameters
4. Starting, final and sampling times for experimental data
5. Number of state variables
6. Experimental measurements of state variables

The software, whose graphical user interface of the software is shown in Figure 2, has also the ability to choose different objective functions (i.e. least square error, absolute error or standardized absolute error). It shows optimum model parameters, CPU time, objective function and the fit between experimental and predicted values of each state variable graphically. Contrary to the ERA software package, there is no limit on the number of parameters and state variables.

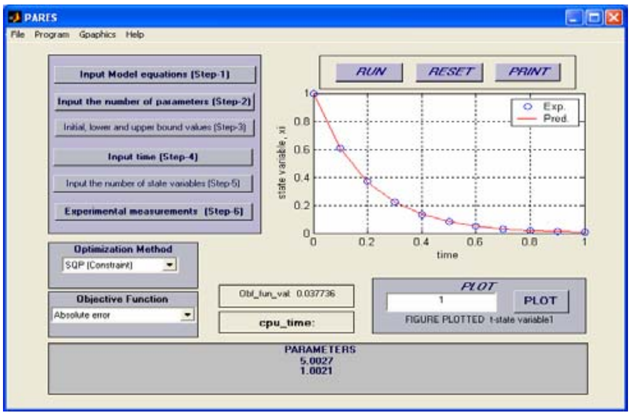


Figure 2. Opening menu of PARES software

4. Example Problems, Results and Conclusion

Although many linear and nonlinear models with varying degree of kinetic complexity have been tested, only one example is included here briefly for the sake of space.

Example 1 represents a First-Order Reversible Chain Reaction Series, and was presented by Esposito and Floudas (2000). The reaction kinetics is expressed as,

$$A \underset{k_2}{\overset{k_1}{\Leftrightarrow}} B \underset{k_4}{\overset{k_3}{\Leftrightarrow}} C \tag{3}$$

All of the components are supposed to be measured, and therefore their concentrations are included in the model used. The dynamic model is written as

$$\frac{dx_1}{dt} = -\beta_1.x_1 + \beta_2.x_2, \quad \frac{dx_2}{dt} = \beta_1.x_1 - (\beta_2 + \beta_3).x_2 + \beta_4.x_3,$$
$$\frac{dx_3}{dt} = \beta_3.x_2 - \beta_4.x_3, \qquad x_0 = [1,0,0] \qquad t \in [0,1] \tag{4}$$

where the state vector, x, is defined as [A, B, C], and parameter vector, β, is defined as [k_1, k_2, k_3, k_4]. The data used in the study was generated with values for the parameters of β=[4, 2, 40, 20] with no added error. The results obtained were summarized in Table 1.

Table 1. Optimization results for Example 1.

Method	Po	Iter.	Obj. Func.	CPU(s)	Parameters			
					P1	P2	P3	P4
LM	P^{01}	135	2.1125e-4	249.4	4.004	2.003	38.431	19.205
	P^{02}	554	8.9698e-4	1321	4.003	1.997	38.571	19.331
NM	P^{01}	310	1.8897e-7	116.8	4.000	2.000	40.012	20.006
	P^{02}	469	1.8897e-7	203.7	4.000	2.000	40.012	20.006
QN	P^{01}	13	1.9003e-7	22.2	4.000	2.000	40.000	20.000
	P^{02}	27	0.0037	55.2	3.8947	1.877	194.521	100.18
SQP	P^{01}	21	1.9012e-7	22.2	4.000	2.000	40.020	20.010
	P^{02}	71	1.8893e-7	97.9	4.000	2.000	40.007	20.003
Esposito&Floudas[1a] (2000)		338	3.367e-7	549.07	4.001	2.001	39.80	19.90
Esposito&Floudas[1b] (2000)		280	1.890e-7	1546.05	4.000	2.000	40.01	20.01
Optimum					4	2	40	20
Lower Bounds					0	0	10	10
Upper Bounds					10	10	50	50

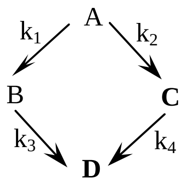
[1a] Collocation approximation

[1b] Integration approximation

Initial parameter set-1:	P^{01} =	10	10	30	30
Initial parameter set-2:	P^{02} =	50	50	150	150

Example 2 tackles with catalytic hydrogenation of cinnamaldehyde, which was previously investigated by Zamostny and Belohlav (1999). Reactions occurring in the system are indicated in the scheme as follows. The experiment was carried out in the

isothermal, isobaric, stirred semi-batch reactor, the mass balance of which is given by the following equations.



$$r_1 = k_1\theta_A \quad r_2 = k_2\theta_A \quad r_3 = k_3\theta_B \quad r_4 = k_4\theta_C$$
$$\frac{dC_A}{dt} = -r_1 - r_2, \quad \frac{dC_B}{dt} = r_1 - r_3, \quad \frac{dC_C}{dt} = r_2 - r_4, \quad \frac{dC_D}{dt} = r_3 + r_4 \tag{5}$$

where r : reaction rate, k : rate constant, t : time, and θ_i : catalyst surface coverage for i-th compound. The parameter estimation problem was solved using different optimization routines. Figure 3 shows that the predicted data fits the experimental measurements very well.

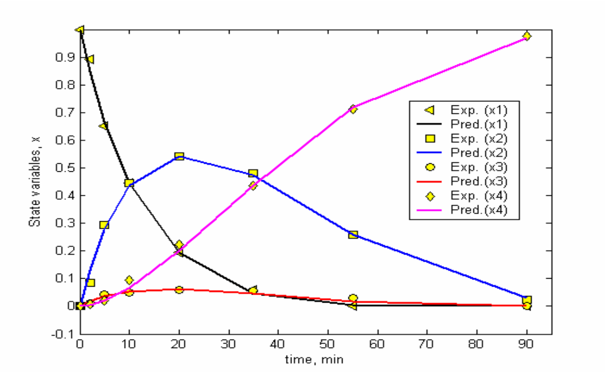


Figure 3. Comparison of experimental and observed data using SQP for Example 2

As reflected by the results, solution to such difficult dynamic optimization problems for parameter estimation seems to be effectively achieved by suggested approach. Particularly, Table 1 reveals that PARES software, which is available for download at http://chemeng.ankara.edu.tr/berber is capable of accomplishing the task with much less iterations and CPU time than previously published techniques, and furthermore the SQP method appears to be favourable to other numerical optimization routines.

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