

Chapter 1

Parameter estimation for stochastic differential equations: algorithm and application to polymer melt rheology

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A parameter estimation algorithm for stochastic differential equation (SDE) systems is proposed. The problem is formulated using the maximum likelihood (MLE) objective function, and a modified Levenberg-Marquardt (LM) algorithm is developed for its solution. Stochastic sensitivity equations are derived and used in order to obtain reliable parameter estimates. Computational efficiency is addressed by varying the simulation size according to the proximity of the current iterate to the optimal solution, as indicated by the magnitude of the trust-region radius. To evaluate the confidence intervals of the parameters, a global uncertainty analysis is proposed, which is based on sampling and accounts for experimental uncertainty and model noise. The algorithm is applied to a stochastic model of polymer rheology.

1. Introduction

SDEs have gained popularity in recent years, for their ability to model systems that are subjected to fluctuations. The general form of an SDE is:

$$dX_t = \mu(t, X_t; \theta)dt + \sigma(t, X_t; \theta)dW_t \quad (1)$$

where t is time, X_t is the state variable of interest, μ and σ are the drift and diffusion term respectively, θ is a vector of model parameters and W_t is a Gaussian $N(0, \Delta t^{1/2})$ noise term (a stochastic process). Applications of SDEs include material modelling (e.g. polymer rheology), environmental pollution

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models, reactor systems, and finance. [1-4] Due to the existence of the noise term it is difficult to obtain closed-form solutions for most SDEs, and numerical methods such as the Euler and the Taylor schemes are required to calculate discrete-time trajectories of the state variables. [5] The accuracy and cost of integrating an SDE system depends on the number of stochastic processes generated (size of simulation). A key issue in using SDEs for practical applications is the estimation of model parameters. This is hindered by the stochastic nature of the model, which makes the computation of gradients unreliable, and by the high computational cost of integrating SDEs by numerical methods. The objective of this work is to develop a gradient-based parameter estimation algorithm for SDE models, that provides reliable values of the parameters and their confidence intervals at reasonable computational cost.

In section 2, the parameter estimation algorithm is outlined. The application of the algorithm to a model of polymer rheology is demonstrated in section 3, and in section 4, a global uncertainty analysis method for evaluating confidence intervals is described.

2. Methodology and algorithm

The algorithm is a modification of the LM algorithm [6], which takes into account the stochastic nature of the problem by careful consideration of the reliability of the gradients, and by using a variable simulation size.

The parameter-estimation problem is formulated using the MLE objective function:

$$\Phi = \frac{N}{2} \ln(2\pi) + \frac{1}{2} \min \left(\sum_{i=1}^{NE} \sum_{j=1}^{NM_i} \left[\ln(\sigma_{ij}^2) + \frac{(X_{ij}^{\text{exp}}(t) - X_{ij}^{\text{model}}(t; \theta))^2}{\sigma_{ij}^2} \right] \right) \quad (2)$$

where NE is the number of experiments performed and NM_i is the number of measurements in the i^{th} experiment; the LM algorithm is specifically designed to solve least-square problems.

The algorithm requires reliable gradients to successfully identify optimal parameters. The issue of gradient calculation is addressed by deriving the sensitivity equations for the SDE model. The original SDEs are differentiated with respect to the model parameters, and the augmented SDE system is then integrated to obtain the sensitivities of the state variables. The importance of this is illustrated in the following example, using the stochastic model of polymer-melt rheology (described in the next section) as a representative stochastic process. A comparison of analytical gradients, derived using the sensitivity equations, with numerical gradients, derived using the central finite-difference method, for different step sizes h (Figure 1) reveals that the numerical gradients are noisier. The numerical gradient with $h = 0.1$ appears unstable as the small step size amplifies the noise of the model predictions, and

results in a gradient with the wrong sign for part of the trajectory. For the larger step size $h = 0.5$, even though the trajectories follow the same shape as the analytical gradient, the same accuracy cannot be achieved. Moreover, the analytical gradient calculations, which involve one integration of the augmented SDE, require about 40% less computational time than the numerical gradient calculations which involve two integrations of the SDE model. As a result, the analytical gradients are more reliable, less noisy and faster to compute.

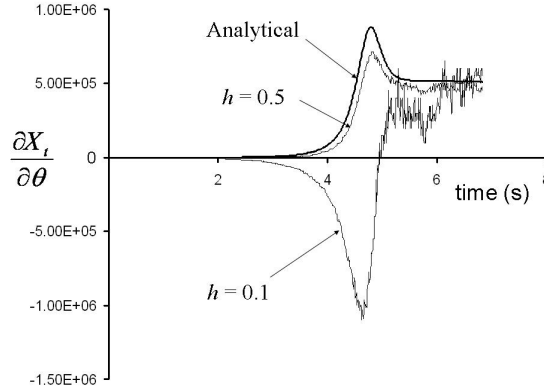


Figure 1: Numerical gradients ($h = 0.1, 0.5$) compared with analytical gradients from sensitivity equations

The stochastic nature of the problem reduces computational efficiency; a large simulation size is required to obtain reliable model predictions. This issue is addressed by varying the simulation size from iteration to iteration. In the LM algorithm, the size of the step to the next iterate is determined by the trust-region radius Δ . The magnitude of Δ is kept constant or increased after successful iterations, while it is reduced after unsuccessful iterations. In this work, upon each successful iteration the simulation size is kept constant or decreased; when the contrary happens, the simulation size is increased to improve the accuracy of the predictions of the state variables and the reliability of the gradients so as to increase the probability of identifying the optimal solution. As a result, at each iteration the simulation size is computed as an inverse function of Δ , increasing computational efficiency. The function used in this work is $size = 5000 \|\mathcal{D} \theta\| \Delta^{-1}$, where $\|\cdot\|$ denotes the Euclidean norm, and $\mathcal{D}$ is a diagonal scaling matrix (for details, see page 111 of Reference 6).

3. Application of algorithm to a stochastic model of polymer rheology

The parameter estimation algorithm has been applied to a stochastic model of polymer melt rheology [1]. In this model, the role of the SDEs is to mimic the random orientation of polymer chains under flow conditions, known as

reptation. The model is used to predict transient viscosity under different shear and extensional flow conditions. The key stochastic variable of the model is the random orientation vector $\mathbf{u}$; its stochastic process takes the form:

$$d\mathbf{u}_t = \left[\left(1 - \frac{\mathbf{u}_t \mathbf{u}_t}{|\mathbf{u}_t|^2} \right) \cdot \boldsymbol{\kappa} \cdot \mathbf{u}_t - 2D\mathbf{u}_t \right] dt + \sqrt{2D} \left(1 - \frac{\mathbf{u}_t \mathbf{u}_t}{|\mathbf{u}_t|^2} \right) \cdot d\mathbf{W}_t \quad (3)$$

where $\boldsymbol{\kappa}$ is the transpose of the velocity gradient tensor and D is the orientation diffusion coefficient associated with the reptation motion. The stress tensor, $\boldsymbol{\tau}$, is then obtained as a function of the expectation of the dyadic product $\mathbf{u}\mathbf{u}$ at time t . The transient shear viscosity η^+ and extensional viscosity μ^+ are then given by simple functions of the appropriate components of the stress tensor and the shear rate or strain rate (respectively). The maximum likelihood objective function is then computed as a function of the least-square of the difference between model-predicted viscosity and experimental data. There are three model parameters which are related to the dynamic properties as well as the architecture of polymer chains. They are:

- the plateau modulus, G_N^0 : this is the plateau value of the shear relaxation modulus, and it characterises the transition of the dynamics of polymer chain motion from vibration at short time scales to reptation at long time scales.
- the reptation time, τ_d : this is a characteristic relaxation time for polymer chains to diffuse away from an imaginary confined tube imposed by surrounding polymer chains.
- the maximum stretching ratio, λ_{max} : this is the ratio of the contour length of a fully stretched polymer chain to the length when it is in its equilibrium state.

The ability of the algorithm to identify known parameter values is tested by considering a “model” polymer. Pseudo-experimental data are generated from simulations using known parameter values at three different extensional rates, and noise, representing experimental error, is added to the data. Starting from parameter values some distance away from the true values, the algorithm estimates parameter values that are close to the true ones, both for fixed and variable-size simulations (see Table 1). The quality of fits to the data (Figure 2) is very high. However, the computational expense is 50% smaller when using a variable *size*, compared to the case of fixed *size*.

Table 1: Parameters used to generate pseudo-experimental data, starting point of the algorithm, parameters estimated by the algorithm and the computational expense (runs were performed on a hyperthreaded Pentium 4 3.4GHz computer running on RHEL 3 system)..

Parameter	G_N^0 (Pa)	τ_d (s)	λ_{max}	CPU (s)
“True” parameter	9876	54.3	2.1	-----
Starting point	1000	100	10	-----
Fixed <i>size</i> (= 100,000)	9894	53.44	2.130	49104
Variable <i>size</i> (min 10, max 100,000)	9774	55.00	2.090	99754

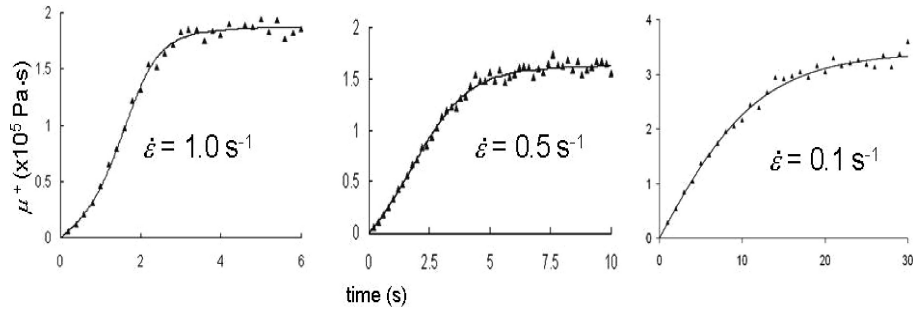


Figure 2: Model-predicted viscosity (lines) and pseudo-experimental data (symbols) for a model polymer at three different extensional rates. The fits for both fixed and variable *size* are shown, but they are so close that they are indistinguishable.

The algorithm is then applied to polystyrene data [7] at four different shear rates. Figure 3 shows the parameters estimated and the fits to the experimental shear viscosity. The model-predicted and experimental trajectories are in qualitative agreement while, quantitatively, the fits are better at shear rate $= 1.0 \text{ s}^{-1}$ than at higher rates, as is expected for this model.

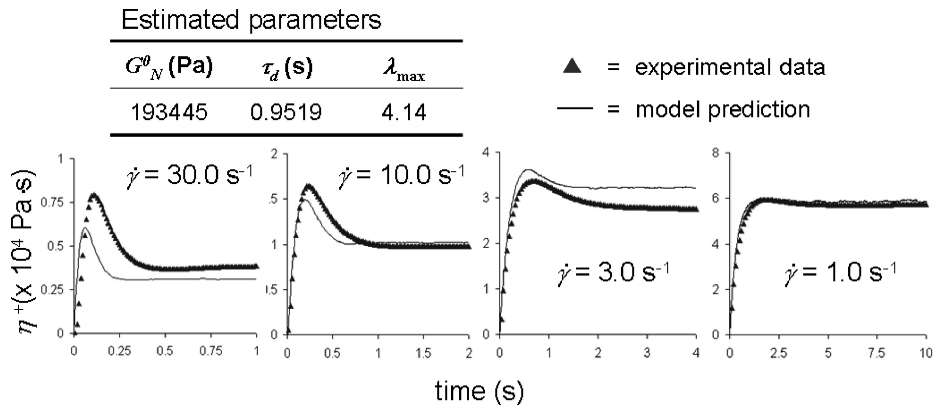


Figure 3: Parameter estimates for a polystyrene sample and the fits to experimental data

4. Evaluating the confidence intervals of the parameters

Uncertainty in the parameters estimated arises from experimental error and from the stochastic nature of the model. To obtain confidence intervals, a global uncertainty analysis is proposed in this work. A number of normally distributed experimental trajectories are first sampled, and the cumulative probability that a given set of parameters is optimal is then estimated, based on knowledge of the simulation noise ($\pm 0.5\%$ for *size* = 100,000). This is repeated for a set of uniformly distributed parameters values, and a distribution of the probability

that any given parameter value is optimal is obtained. This then gives an estimate of the expected parameters values and their confidence intervals.

Table 3 shows the results of the global uncertainty analysis for a sample of polyacrylate at extensional rate = 1.0 s^{-1} . The optimal parameters predicted by the algorithm are close to the expected value of the parameters, and the 95% confidence intervals are small, ranging between 2 to 7% of the parameter values.

Table 3: Results of global uncertainty analysis for polyacrylate at extensional rate = 1.0 s^{-1}

Parameter	Optimal value	Expected value	95% confidence interval
G_N^0 (Pa)	24413	24677	[23208, 26475]
τ_d (s)	69.71	69.48	[68.27, 71.13]
λ_{max}	15.70	15.62	[15.06, 16.87]

5. Conclusions

SDEs have found many applications in the modelling of complex systems subjected to randomness, but pose problems for parameter estimation due to their stochastic nature. In this work, a reliable parameter estimation algorithm for SDE models has been developed and implemented. This is based on a modified Levenberg-Marquardt algorithm, in which the simulation size is varied to improve computational performance. The gradients required for the successful identification of the parameters are derived from stochastic sensitivity equations. To quantify the uncertainty in the parameters due to experimental error and the stochastic nature of the model, a global uncertainty analysis is proposed. The application of this algorithm to a stochastic model of polymer rheology has been successfully demonstrated.

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