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Two methods for sensitivity analysis of population balance equations by a Monte Carlo method

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Abstract

We consider two stochastic simulation algorithms for the calculation of parametric derivatives of solutions of a population balance equation, namely, forward and adjoint sensitivity methods. The dispersed system is approximated by an N-particle stochastic weighted ensemble. The infinitesimal deviations of the solution are accounted for through infinitesimal deviation of the statistical weights that are recalculated at each coagulation. In the forward method these deviations of the statistical weights immediately give parametric derivatives of the solution. In the second method the deviations of the statistical weights are used to calculate a finite-mode approximation of the linearized version of the population balance equation. The linearized equation allows for the calculation of the eigenmodes and eigenvalues of the process, while the parametric derivatives of the solution are given by a Lagrange formalism.

Contents

1 Introduction

Equations of population balances give the general mathematical framework for the description and modelling of particulate systems [1]. Starting from the famous Boltzmann equation [2], equations of population balances span over a wide range of physical, technological and environmental applications such as mixing [3, 4], liquid/liquid dispersion [5, 6], soot formation [7], dynamics of atmospheric aerosols [8], breakage and agglomeration of powders [9, 10, 11], growth of microbial cell population [12], polymerization [13] and crystallization [14]. The general area of applicability implies that the equations of population balances account for the most basic physical principles (mass, momentum and energy conservation), i.e., they provide only a framework, which has to "be filled in" by the physical information specific to the system under consideration.

The general form of the equation of population balance reads:

$$\frac{\partial m(t,x;\lambda)}{\partial t} = B(m(t,x;\lambda);\lambda) - D(m(t,x;\lambda);\lambda) \equiv L(m(t,x;\lambda);\lambda), \qquad (1)$$

where $m(t,x;\lambda)$ is mass density of the particles with mass x (in the case of multicomponent particles x can by vector containing masses of the components), $B(m(t,x;\lambda);\lambda)$ and $D(m(t,x;\lambda);\lambda)$ are birth and death rates of the particles due to collision and breakage, respectively, and λ is a parameter. λ can be a scalar, a vector or, even a function, which contains the empirical information about the system. Thus, a researcher who starts modelling a particulate system has to specify λ which is unknown a priori, i.e., the following questions have to be addressed.

1. In many cases the dimension of the model is not obvious i.e., we do not know a priori how many parameters are necessary to describe the shape of the particles, or how much the internal parameters such as porosity, chemical composition, humidity content or age of a microbial cell are important for the problem under consideration, etc.

2. Since many physical processes and mechanisms are simultaneously involved in the particulate systems, the most important of them have to be identified in order to keep the model tractable. For example, in a granulation system we do not know whether particle's breakage is important and if so, which type of breakage (attrition, breakage due to collisions with walls or with other particles, etc.) it is.

3. When the qualitative physical picture of the system is set up, the parameters of the model such as collision and breakage rate or number of fragments during a breakage have to be specified.

Obviously, these problems cannot be solved on the basis of theoretical considerations only, requiring intensive use of experimental data. The difficulty specific to dispersed systems is that while the mathematical models of population balances are based

on consideration of particle to particle interaction, the available experimental data characterizes behaviour of the particulate ensemble as whole and, therefore, most of the physical characteristics of the system are not accessible for direct measurement. Thus there is a need for numerical methods which can close the gap between an experiment and a model and assist in the determination of the physical structure of the particulate systems. These methods can be classified as follows.

Sensitivity analysis, i.e., calculation of the parametric derivatives

$$\frac{\partial m(t,x;\lambda)}{\partial \lambda} \tag{2}$$

in order to characterize the effect of a parameter on the solution of Eq. (1). Thus the parameters with high values of $\partial m/\partial\lambda$ have to be determined with a high precision, while those with lower values can be approximately estimated from some general considerations or, alternatively, another experimental conditions under which the parameter λ has more distinct effect

Method of perturbations can be considered as an extension of the sensitivity analysis (although requires a different mathematical technique). In order to isolate the effect of a single physical mechanism or process (attrition, coagulation, etc.) on the dynamics of the system this process is treated as a perturbation of a "background" solution:

$$\frac{\partial m(t,x;\lambda)}{\partial t} = L_0(m(t,x;\lambda);\lambda) + \epsilon L_1(m(t,x;\lambda);\lambda), \tag{3}$$

where ϵ is a small parameter, L_0 and L_1 are operators of the unperturbed problem and the perturbation, respectively. The solution of Eq. (1) is represented as $m = m_0 + \epsilon m_1 + ...$, where m_1 characterizes (up to $O(\epsilon)$) the effect of the considered physical mechanism.

Inverse problems, when the unknown parameter λ has to be extracted from experimental observations. The most common formulation of an inverse problem, namely, an extremal formulation reads as follows. Consider a functional $H[m(t,x,\lambda)]$ and let us measure the values of H and denote them as H^{exp}. The solution of the inverse problem is the parameter λ^* that minimizes the difference between the calculated values of H and these observed in an experiment, i.e.,

$$\min_{\lambda} \|H[m(t,x;\lambda)] - H^{exp}\|. \tag{4}$$

Since (4) is a non-linear minimization problem, its efficient solution requires knowledge about the local behaviour of $H[m(t,x,\lambda)]$ as a function of λ. Therefore, sensitivity analysis and methods of perturbations are an important part of any parameter identification procedure.

Certainly there are other problems where sensitivity analysis of Eq. (1) can be very helpful. Many industrial reactors such as bioreactors and crystallizers exhibit an

oscillatory behaviour, i.e., the steady-state solution of the corresponding population balance equation is unstable. Investigation of the unstable regimes consists of the following steps: (i) detection of the steady-state solution, i.e., calculation of roots of the right hand side of Eq. (1); (ii) linearization of Eq. (1) around this unstable solution and calculation of the eigenvalues and eigenmodes of the linearized equation; (iii) design of a controller in order to stabilize the steady-state solution; (iv) detection of the bifurcation points in the parameter space where a stable solution becomes unstable and vice versa. All these tasks require an efficient procedure for the linearization and sensitivity analysis of Eq. (1).

One of the difficulties associated with the sensitivity analysis of population balance equations arises from the nature of Eq. (1). Equations of population balances are nonlinear integro-differential equations, which makes a numerical treatment of Eq. (1) very time-consuming. If the dimension of x is higher then 2, Monte Carlo statistical simulation becomes the only available numerical technique. Note that many researches believe that since a Monte Carlo solution suffers from high statistical fluctuations, finite differences and finite elements methods have better performance, at least for one-dimensional problems. Recent results show [15] that the convergence rate of Monte Carlo method can be significantly improved, which together with the obvious logarithmical simplicity and flexibility makes it an attractive method even for one-dimensional problems. A common method for sensitivity analysis is to run the simulation for several values of the parameter and then to apply a finite difference method. However, the method is highly sensitive to numerical noise which is absolutely unavoidable in Monte Carlo simulations. In our recent investigations [16] we formulated the equations for parametric derivatives of the solution and solve these equations together with Eq. (1). This method gives the solution of the equation and its parametric derivatives simultaneously. In [17] this method has been used for parametric identification of a population balance model of a liquid/liquid dispersion process.

The present work focuses on the further development of Monte Carlo methods for the sensitivity analysis of the equations of population balances, namely, we consider two approaches to the problem: forward sensitivity analysis and adjoint sensitivity analysis of the linearized version of Eq. (1).

2 Sensitivity analysis and Monte Carlo methods

Before we start our considerations let us briefly mention the two methods of sensitivity analysis, namely forward and adjoint methods. The equations for forward sensitivity can be obtained by formally differentiating Eq. (1) with respect to λ. Denote the forward sensitivity $\partial m/\partial\lambda$ by $m'(t,x;\lambda)$, the equation for $m'(t,x;\lambda)$ and the initial conditions are

$$\frac{\partial m'(t,x;\lambda)}{\partial t} = L'm'(t,x;\lambda) + \frac{\partial L(m;\lambda)}{\partial \lambda},\ m'|_{t=0} = 0, \tag{5}$$

where L' is the linearization of the operator $L(m(t,x;\lambda);\lambda)$. Then the parametric derivative of a functional $H[m(t,x,\lambda)] = \int\int m(t,x,\lambda)h(x)dtdx$, where $h(x)$ is an arbitrary function, reads

$$\frac{\partial H[m(t,x;\lambda)]}{\partial \lambda} = \int\int m'(t,x;\lambda)h(x)dtdx. \tag{6}$$

The adjoint sensitivity analysis is based on the Lagrange formalism. Let us define an adjoint variable ψ and rewrite the functional H as follows

$$H[m(t,x,\lambda)] = \int\int_0^T \left\{ m(t,x;\lambda)h(x) - \psi\left(\frac{\partial m(t,x;\lambda)}{\partial t} - L(m;\lambda)\right)\right\} dtdx. \tag{7}$$

Since $m(t,x;\lambda)$ satisfies Eq. (1), the last term in the above equation is 0.

We require that the adjoint variable ψ satisfies the following problem:

$$-\frac{\partial \psi(t,x;\lambda)}{\partial t} = L^*\psi(t,x;\lambda) + h(x),\ \ \psi|_{t=T} = 0, \tag{8}$$

where L^* is an operator adjoint to L'. Differentiating Eq. (7) with respect to λ, integrating by parts with respect to time and using Eq. (8) we obtain after some algebra the following expression for the parametric derivative of the functional H:

$$\frac{\partial H[m(t,x,\lambda)]}{\partial \lambda} = \int\int_0^T \psi(t,x;\lambda)\frac{\partial L(m;\lambda)}{\partial \lambda}dtdx. \tag{9}$$

Both forward and adjoint approaches have advantages and disadvantages. As one can see, Eq. (5) contains the derivative $\partial L/\partial\lambda$. This means that the equation for the forward sensitivity m' has to be solved for the each of parameters λ involved in the problem. On the other hand, for the known forward sensitivity m' the parametric derivative $\partial H/\partial\lambda$ can be calculated for *any* functional H. The equation for adjoint sensitivity (8) does not contain a derivative with respect to λ, instead it contains the function h. As soon as the adjoint sensitivity ψ is known, the parametric derivatives of the *given* functional H with respect to *any* number of parameters is given by Eq. (9). In conclusion, if we need the sensitivities of a large number of functionals with respect to a small number of parameters, the forward sensitivity approach is more efficient. While the adjoint sensitivity method is better if we need sensitivities of a small number of functionals with respect to a large (maybe infinite) number of parameters. The last case is rather common in population balances, because in an experiment, only the first few moments of the particles' distribution (mass of the particles, mean diameter, mean surface area, etc.) are accessible. Therefore, in the present article we discuss Monte Carlo algorithms for the forward sensitivity analysis, linearization and adjoint sensitivity analysis of the equations of population balances.

2.1 Weighted particles method and forward sensitivity analysis

The main idea of the Monte Carlo method is the representation of $m(x)$ and $H[m(t,x,\lambda)]$ as

$$m(t,x;\lambda) \approx \sum_{i=1}^{N} w_i \delta(x - x_i),\ H[m(t,x,\lambda)] \approx \sum_{i=1}^{N} w_i h(x_i), \tag{10}$$

respectively, i.e., the ensemble of a large number of physical particles is replaced by an ensemble of N computational particles. Each computational particle represents w_i identical real particles. If all statistical weights w_i are equal, this is called the direct simulation Monte Carlo (DSMC) method, while the more general case is called the stochastic weighted particle method (SWPM). Let us consider the following formulae:

$$m(t,x;\lambda+\Delta\lambda) \approx \sum_{i=1}^{N} w_i(1+\Delta\lambda W_i)\delta(x - x_i),$$

$$H[m(t,x,\lambda+\Delta\lambda)] \approx \sum_{i=1}^{N} w_i(1+\Delta\lambda W_i)h(x_i).$$

Comparison with Eq. (10) reveals that

$$\frac{\partial m(t,x;\lambda)}{\partial\lambda} \approx \sum_{i=1}^{N} W_i \delta(x - x_i),\ \frac{\partial H[m(t,x,\lambda)]}{\partial\lambda} \approx \sum_{i=1}^{N} W_i h(x_i). \tag{11}$$

We refer to the system with statistical weights $w_i(1+\Delta\lambda W_i)$ and operator $L(n;\lambda+\Delta\lambda)$ as the "disturbed" system, while the original system is referred to as an "undisturbed" one. We then simulate the undisturbed system and after each event (coagulation, breakage, etc.) the factors W_i which account for the infinitesimal deviation of the solution due to an infinitesimal deviation of the parameter are recalculated as follows [16].

The probability that two particles with masses x and x' respectively, coalesce during a small time interval dt is $K(x,x';\lambda)dt$, where K is a coagulation kernel. In the present investigation we use an acceptance-rejection technique similar to that used in [18]. The probability that the k^{th} particle collides with the l^{th} particle is $\pi_{kl}(\lambda)dt = K(x_k,x_l;\lambda)x_l^{-1}w_l dt$. Consider a majorant $\hat{\pi}_{kl}$ satisfying $\pi_{kl}(\lambda) \leqslant \hat{\pi}_{kl}$. A pair of particles is chosen based on the distribution $\hat{\pi}_{kl}/\sum_{\alpha\beta=1}^{N}\hat{\pi}_{\alpha\beta}$. The coagulation is then either accepted with the probability $\pi(\lambda)_{kl}/\hat{\pi}_{kl}$ and x_k and W_k are recalculated as

$$x_k = x_k + x_l,\ W_k = W_k + \partial_\lambda \ln(\pi(\lambda)_{kl}), \tag{12}$$

or the coagulation is rejected and W_k is recalculated as

$$W_k = W_k - \frac{\pi(\lambda)_{kl}}{\hat{\pi}_{kl} - \pi(\lambda)_{kl}}\partial_\lambda \ln(\pi(\lambda)_{kl}). \tag{13}$$

Thus, we obtain the solution to Eq. (1) and its parametric derivatives simultaneously.

As the calculations proceed, dispersion of the estimation (11) increases further and the procedure becomes very time-consuming. The nature of this error and methods of its reduction are discussied in the Appendix.

2.2 Linearization of Eq. (1) and adjoint sensitivity analysis

Consider a solution of Eq. (1) $m_0(t,x)$ and a perturbation of the solution $m'(t,x)$, which we represent in the following form:

$$m'(t,x;\lambda) = m_0(t,x;\lambda)\sum_{j=1}^{J} a_j(t)\gamma_j(x). \tag{14}$$

In other words the deviations of the statistical weights W_k read

$$W_k = \sum_{j=1}^{J} W_{k,j} = \sum_{j=1}^{J} a_j(t)\gamma_j(x_k). \tag{15}$$

In the above equation $\gamma_j(x)$ are known polynomial while $a_j(t)$ are unknown functions of time. Substitution of $m(t,x) = m_0(t,x) + m'(t,x)$ into Eq. (1) and substraction of the undisturbed equation from the disturbed one yields the following linearized version of the equation of population balance

$$\frac{\partial m'(t,x;\lambda)}{\partial t} = L'm'(t,x;\lambda). \tag{16}$$

Multiplication of (16) by $\gamma_i(x)$ and integration by x yields the finite-mode approximation of the linearized equation:

$$\frac{d}{dt}A_{ij}a_j(t) = B_{ij}a_j(t), \tag{17}$$

where

$$A_{ij} = \int m_0(x)\gamma_i(x)\gamma_j(x)dx,\ B_{ij} = \int \gamma_i(x)L'\{m_0(x)\gamma_j(x)\}dx. \tag{18}$$

The finite-mode approximation of (8) and (9) then read

$$-A_{ij}\frac{d\psi_i(t)}{dt} = B_{ij}\psi_i(t) + \int m_0(x)\gamma_i(x)h(x)dx,\ \psi(T) = 0, \tag{19}$$

$$\frac{\partial H[m(x,\lambda)]}{\partial\lambda} = \sum_i \psi_i \int \gamma_i(x)\frac{\partial L(m_0;\lambda)}{\partial\lambda}dx. \tag{20}$$

While the calculation of A_{ij} by Monte Carlo methods is straightforward, the calculation of B_{ij} is more complicated. Combining of Eqs. (12, 13, 18) yields the following

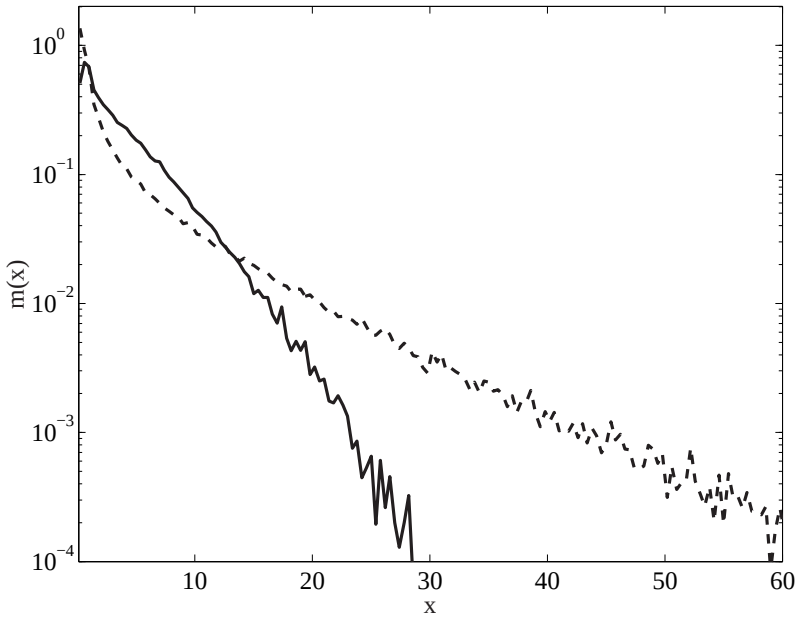


Figure 1: *Mass densities for $\alpha = 0$ (solid line) and $\alpha = 0.5$ (dashed line), $Q_{in} = 1$.*

expression for B_{ij} averaged over a short time interval: $B_{ij} = \sum_n b_{ij}^n$, where the contribution of n^{th} collision reads:

$$b_{ij}^n = \frac{w_k}{\tau_n} \left[\gamma_i(x_k + x_l)\{\gamma_j(x_k) + \gamma_j(x_l)\} - \gamma_i(x_k)\gamma_j(x_k)\right], \tag{21}$$

if the coagulation between the k^{th} and l^{th} particles is accepted, or

$$b_{ij}^n = -\frac{w_k}{\tau_n}\gamma_i(x_k)\gamma_j(x_l), \tag{22}$$

if the coagulation is rejected. In the above equations τ_n is the time interval between the $(n-1)^{th}$ and n^{th} collisions.

3 Results and discussion

We simulated the coagulation of particles in a stirred reactor. Particles enter the reactor at a mass flux Q_{in} and are uniformly distributed in mass ranging from 0 to 1. The collision kernel is $K(x, y) = (xy)^\alpha$, and the probability that a particle with mass x leaves the reactor during a small time interval dt is $0.1xdt$. After some transient period the system reaches a steady state. Mass density functions are shown in Fig. 1. As the exponent α increases the total mass of the system and the mean size of the particles decrease, while a long tail of large particles is formed which slowly decreases with x. An accurate resolution of the tail is very important

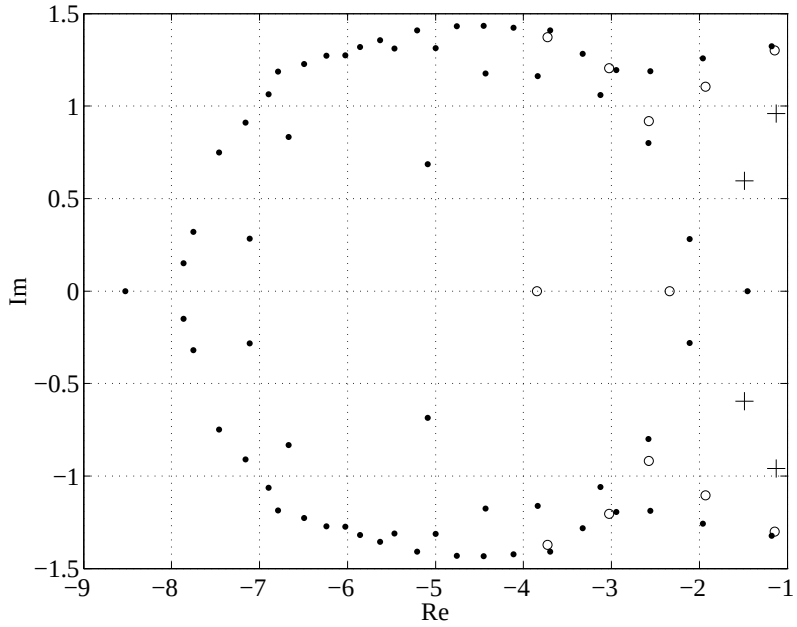
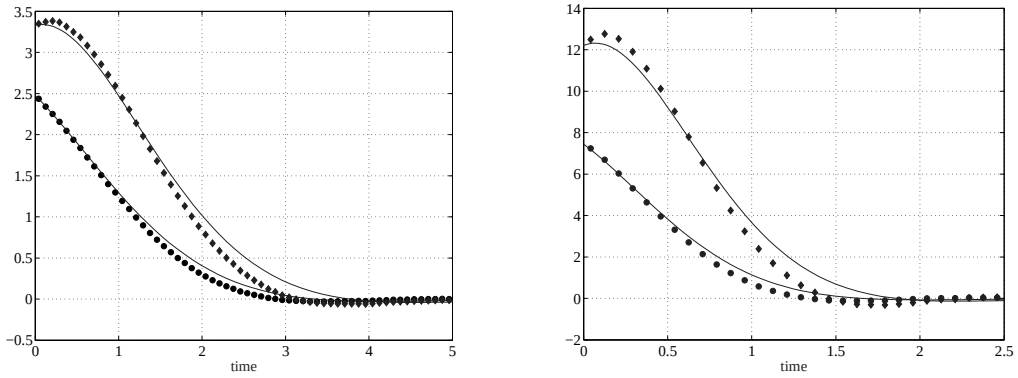


Figure 2: *Eigenvalues of Eq. (17) for* 4 *modes* (+), 12 *modes* (∘) *and* 64 *modes* (·). $\alpha = 1/3$, $Q_{in} = 1$.



(a) $Q_{in} = 1$.

(b) $Q_{in} = 6$.

Figure 3: *Relaxation of* μ_0 (•) *and* μ_1 (⋄), *lines show the corresponding results by the direct method,* $\alpha = 1/3$. *The initial conditions are* $a_1(0) = 1$, $a_{j\neq 1}(0) = 0$.

because large particles have higher probability of leaving the reactor and the tail controls the total mass of the particles in the reactor.

When the system reaches a steady state we simulate its infinitesimal disturbances both by the forward and the adjoint methods. The eigenvalues of Eq. (17) are plotted in Fig. 2. In the present study we used two forms of the polynomial $\gamma_j(x)$:

$$\gamma_j(x) = \left(\frac{6x}{\pi}\right)^{(j-1)/3}, \quad \gamma_j(x) = \cos\left(\frac{2\pi}{\chi}x(j-1)\right), \tag{23}$$

where χ is a characteristic size. While for a small number of modes both approximations work well, large J the trigonometric functions are better. As one can see, even a small number of modes is enough to capture the eigenvalues with highest real parts. Additional modes have shorter relaxation times, and we can suppose that truncation error of the finite-mode approximation is small.

Since the forward sensitivity method has been extensively tested in our previous investigations [16], we refer to the results obtained by this method as "exact". In order to compare the results obtained by Eqs. (12), (13) and Eq. (17) we define the following moments of the particles size distribution:

$$\mu_n = \int \left(\frac{6x}{\pi}\right)^{n/3} m(x)dx. \tag{24}$$

Fig. 3 shows the relaxation of these moments toward a steady state calculated by the both methods. Given that only 4 modes have been used in Eq. (17) in these calculations, the accordance between the results is satisfactory. Parametric derivatives $\partial\mu_0/\partial\alpha$ and $\partial\mu_1/\partial\alpha$ by the forward and the adjoint methods are given in Table 1. When α approaches 0.5 the particles size distribution has a long tail, then up to 12 modes are necessary to make the adjoint method insensitive to the number of modes. As one can see, the discrepancy between the two methods ε is about 10%, which is a good result since both methods are subjected to numerical noise.

4 Conclusion

In this work we considered two methods for sensitivity analysis of the equations of population balance. These methods are based on the weighted particles Monte Carlo method, where the sensitivity is accounted for by infinitesimal deviations of the statistical weights. The forward sensitivity method directly uses these deviations of the statistical weights to calculate parametric derivatives of the solution. Another method uses a similar procedure to calculate a finite-mode approximation of the linearized version of the equation. Then, the Lagrange formalism can be used to calculate parametric derivatives of linear functionals (statistical moments, etc.) of the solution. Both methods give similar results when they are applied to the same problems, while each method has its advantages and disadvantages.

Table 1: *Parametric derivatives of μ_0 and μ_1 with respect to α by the direct and the adjoint methods.*

α	μ_0	μ_1	μ_0' (direct)	μ_0' (adjoint)	ε (%)	μ_1' (direct)	μ_1' (adjoint)	ε (%)
0.0	2.64	4.61	-0.840	-0.835	0.59	-1.50	-1.40	7.1
0.1	2.57	4.46	-0.776	-0.783	0.96	-1.23	-1.29	5.0
0.2	2.51	4.32	-0.557	-0.608	8.30	-1.46	-1.34	8.6
0.3	2.46	4.12	-0.372	-0.361	2.90	-1.30	-1.18	10.0
0.4	2.45	4.02	-0.088	-0.101	13.0	-1.15	-1.06	7.8
0.5	2.44	3.91	-0.053	-0.050	7.10	-0.79	-0.851	7.1

Although the adjoint method (unlike the forward one) requires intensive postprocessing, it can be very helpful in those situations where the applicability of the forward method is questionable, namely, stability analysis of particulate processes. Many industrial reactors involve two distinct kinetic processes: inception of particles (nucleation of crystals, birth of cells, etc.) and their subsequent growth. The rate equations of these kinetic processes are usually nonlinear, and this property can lead to complex dynamic behavior including self-sustained oscillations, multiple steady states, etc. Oscillations in particle size distribution have been reported for industrial crystallizers (see e.g., [19], [12] and references therein). These oscillations represent an important practical problem leading to off-specification products. The proposed sensitivity analysis methods have a high potential for bifurcation and stability analysis of such systems. These methods can be used to find unstable steady-state solution which cannot be detected by direct simulation. The equation can then be linearized around the obtained solution in order to calculate its eigenmodes and eigenvalues. The resulting linear model can be used to develop a control strategy that regulates the particles' number distribution by manipulating the feed and removal rate and other operating conditions.

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A Statistical error of Eq. (11) and methods of its reduction.

The forward method for sensitivity analysis of population balance equations has been tested in our previous work [16] and it was shown that the estimation of the parametric derivatives based on Eq. (11)) has a high dispersion. To understand the origin of this dispersion it is enough to note that two identical particles with factors W and $-W$, respectively, give zero contribution to the parametric derivative for any (high) value of W. The numerical experiments show that the factors W can grow infinitely which leads to unbounded dispersion of the calculated derivatives. In order to reduce the dispersion we used a weighted particles algorithm which creates one additional particle after each collision, then we used a clustering method [20] to reduce the number of particles.

In the present investigation we use an alternative method which does not require the time-consuming clustering procedure. Assume that we do not need to calculate the parametric derivative of the solution $\partial m(x)/\partial\lambda$ but only derivatives of the first few moments of the particle size distribution: total mass, mean diameter, etc. Defining a set of functions $h_0(x)$, $h_1(x)$,... $h_K(x)$ (for example, $h_k(x) = x^{k/3}$) and substituting these into Eq. (11) yields a system of linear equations with respect to W_n:

$$\sum_{n=1}^{N}\{h_k(x_n)w_n\}W_n = \frac{\partial H_k(m;\lambda)}{\partial\lambda}. \tag{A.1}$$

The solution of the problem

$$\min_{W_n}\frac{1}{2}\sum_{n=1}^{N}W_n^2 \tag{A.2}$$

which satisfies Eq. (A.1) does not change the parametric derivatives $\partial H_k/\partial\lambda$ of the first K moments and reduces the scatter of the factors W_n.

Finally, the algorithm for the calculation of the parametric derivatives is as follows. We simulate the coagulation process and recalculate W_n according to Eqs. (12), (13). When the scatter of the factors W_n becomes too high we solve the problem (A.1) – (A.2). Since we do not need an exact minimum of (A.2), $2-4$ iterations of a projective gradient method are enough to stabilize the computations. Note that this procedure is not time-consuming and the required CPU time scales almost linearly with the number of preserved moments K. Computational experiments with K spaning from 6 to 24 give very similar results. All the results presented in this work are obtained with $K = 12$.

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