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A Monte Carlo method for identification and sensitivity analysis of coagulation processes

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Abstract

A stochastic simulation algorithm is presented to calculate parametric derivatives of solutions of a population balance equation. The dispersed system is approximated by an N -particle stochastic weighted ensemble. The derivatives are accounted for through infinitesimal deviation of the statistical weights that are recalculated at each coagulation. Thus, all the parametric derivatives can be calculated along one trajectory of the process, given N sufficiently large. We use an operator-splitting technique to account for surface growth of the particles. The obtained solution is in good agreement with the available analytical solutions. As soon as the parametric derivatives are known the gradient-based methods can be applied to the control and identification of the coagulation process. The extension of the proposed technique to a multi-dimensional case is straightforward.

1 Introduction

Sensitivity analysis and inverse problems in population balances have a wide range of applicability in different technological processes such as grinding systems [1], emulsion polymerization [2], and liquid/liquid dispersions [3] to mention just a few (for a comprehensive review see Ref. [4], Chap. VI).

The main object of the present investigation is the space-homogeneous Smoluchowski's coagulation equation

$$\frac{\partial n(t, x; \lambda)}{\partial t} = \frac{1}{2} \int_0^x K(x - x', x'; \lambda) n(t, x - x'; \lambda) n(t, x'; \lambda) dx' - \int_0^\infty K(x, x'; \lambda) n(t, x; \lambda) n(t, x'; \lambda) dx', \quad (1)$$

where $n(t, x; \lambda)$ is the number density of particles that have mass x at the time t . 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 λ is an unknown (generally speaking, vector) parameter. At the initial moment $t = 0$ the particles are distributed according to the initial condition:

$$n(0, x) = n_0(x). \quad (2)$$

The presence of the undefined parameter λ in the kernel of Eq. (1) gives rise to two extremal problems, namely, optimization and identification. Consider a functional $H[n(t, x, \lambda)]$, so λ can be viewed as a control parameter that minimizes H :

$$\min_{\lambda} (H[n(t, x; \lambda)]). \quad (3)$$

A problem which is similar to the above is parameter identification or the *inverse* problem when the parameter λ has to be extracted from experimental observations. We denote our experimentally obtained functional H as H^{exp} . The solution of the inverse problem is the parameter λ^* that minimizes the difference between the calculated values of H and those observed experimentally, i.e.,

$$\lambda^* = \min_{\lambda} \|H[n(t, x; \lambda)] - H^{exp}\|. \quad (4)$$

The solution of the above problems is a nontrivial task that is often aided by the self-similar behaviour of many practically important dispersed systems [4]. In the present study we solve problem (4) by a more general gradient-search algorithm, where a Monte Carlo method is used for the solution of Eqs. (1)-(2) and the calculation of the gradient $\partial H/\partial \lambda$. This approach has two important advantages: (i) a Monte Carlo method easily can be extended to a multidimensional case, and (ii) gradient search is faster than other methods of mathematical programming. A common method for the calculation of parametric derivatives is to run the simulation for several values of the parameter and then to apply a finite difference method. The method is highly sensitive to numerical noise that is absolutely unavoidable in

the case of a Monte Carlo simulation. In the present investigation we formulate 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.

The paper is organized as follows. In §2 we represent coagulation as a Markov process for an N -particle weighted stochastic system. §3 contains a discussion of the applicability of weighted Monte Carlo methods for the estimation of parametric derivatives and possible ways to reduce dispersion of the estimation. In §4 we apply the obtained method to several model examples and discuss the numerical results. One of the possible extensions of the proposed method, namely, coagulation/condensation problem is treated by an operator-splitting technique.

2 Weighted N -particle system and the corresponding Markov process

In order to proceed further let us reformulate Eq. (1) in terms of mass density. Advantages of this formulation are discussed in [5], note also, that particle distributions according to their mass are encountered in technological applications more frequently than number distributions. The mass density of the particles that have mass x at a time t is $m(t, x) = xn(t, x)$. The total mass of the system is $M = \int m dx$. In order to rewrite the collision equation (1) in terms of mass density, we express $n(t, x)$ as $m(t, x)/x$, substitute it into (1) and multiply the equation by x . Note, that if $K(x, x') = 0$ for $x \leq 0$ or $x' \leq 0$, the limits of integration in (1) can be extended from $-\infty$ to ∞ . After some algebra we obtain [4], [5]:

$$\begin{aligned} \frac{\partial m(t, x; \lambda)}{\partial t} = & \int \frac{K(x - x', x'; \lambda)}{x'} m(t, x - x'; \lambda) m(t, x'; \lambda) dx' - \\ & \int \frac{K(x, x'; \lambda)}{x'} m(t, x; \lambda) m(t, x'; \lambda) dx'. \end{aligned} \quad (5)$$

Since the coagulation kernel is symmetric, i.e., $K(x, x') = K(x', x)$, the factor of $1/2$ before the first integral in Eq. (5) disappears. From the physical point of view it means that coagulation reduces the number of particles but does not affect their mass. The initial conditions become

$$m(0, x) = m_0(x) = xn_0(x). \quad (6)$$

Consider a stochastic particle system

$$x_1(t), \dots, x_N(t),$$

which approximates the mass density function $m(t, x)$ as

$$m(t, x) \approx \sum_{n=1}^N w_n \delta(x - x_n(t)), \quad (7)$$

i.e., each particle in the above N -particle system represents a group of identical physical particles with size x_n . The total mass of the n^{th} group is w_n and the number of particles in the group is w_n/x_n . Since the probability that during a small time interval dt the k^{th} particle will coagulate with one particle from the l^{th} group is $K(x_k, x_l; \lambda)dt$, probability that the k^{th} particle will coagulate with any of the l^{th} particles is

$$\pi_{kl}(\lambda)dt = \frac{K(x_k, x_l; \lambda)w_l}{x_l}dt.$$

Thus, the coagulation rate of the k^{th} particle is given by summation of the above formula over l . The formula for the total collision rate reads:

$$\sum_{\alpha=1, \beta=1}^N \pi_{\alpha\beta}(\lambda), \quad (8)$$

and the collision pair is chosen with the relative probability

$$\frac{\pi_{kl}(\lambda)}{\sum_{\alpha=1, \beta=1}^N \pi_{\alpha\beta}(\lambda)}. \quad (9)$$

In the present investigation we use the acceptance-rejection technique similar to that used in [5]. Let us consider a majorant kernel and majorant weights satisfying

$$K(x_k, x_l; \lambda) \leq \hat{K}(x_k, x_l), \quad w_l \leq \hat{w}_l, \quad \pi_{kl}(\lambda) \leq \hat{\pi}_{kl} = \hat{K}(x_k, x_l)\hat{w}_l.$$

The corresponding stochastic particle algorithm reads:

1. Generate an exponentially distributed time increment τ with parameter

$$\sum_{\alpha=1, \beta=1}^N \hat{\pi}_{\alpha\beta}; \quad (10)$$

2. Choose a pair (k, l) to collide according to the distribution

$$\frac{\hat{\pi}_{kl}}{\sum_{\alpha=1, \beta=1}^N \hat{\pi}_{\alpha\beta}}; \quad (11)$$

3. Accept the coagulation with probability

$$\frac{\pi_{kl}(\lambda)}{\hat{\pi}_{kl}}; \quad (12)$$

i.e., x_k is replaced by $x_k + x_l$;

4. Or reject the coagulation and perform a *fictitious* jump that does not change the size of the colliding particles with probability

$$1 - \frac{\pi_{kl}(\lambda)}{\hat{\pi}_{kl}}. \quad (13)$$

Notably, the number of particles in this algorithm does not change during the calculations. The acceptance-rejection method described above is equivalent to the well-known maximum section method [6], [7]. The particle ensemble at time t is an approximation of the mass density function $m(t, x)$.

Now, consider a functional of the solution of Eqs. (5) - (6):

$$H(t, m; \lambda) = \int m(t, x; \lambda) h(x) dx,$$

where $h(x)$ is an integrable function of x . Substitution of Eq. (7) into the above equation gives a Monte Carlo estimate of the functional H that is averaged over the N -particle ensemble:

$$H(t, m; \lambda) \approx \sum_{n=1}^N w_n h(x_n(t)). \quad (14)$$

3 Calculation of parametric derivatives of the solution of the coagulation equation

Let us consider the following formula:

$$H(t, m; \lambda + \Delta\lambda) \approx \sum_{n=1}^N w_n (1 + \Delta\lambda W_n) h(x_n(t)). \quad (15)$$

Comparison with Eq. (14) reveals that

$$\frac{\partial m(t, x; \lambda)}{\partial \lambda} \approx \sum_{n=1}^N w_n W_n \delta(x - x_n(t)) \quad (16)$$

which can be interpreted as a parametric derivative (in weak sense) of the solution of Eq. (5), and $W_n = (\partial_\lambda w_n)/w_n = \partial_\lambda \ln w_n$.

We will refer to a system with weights $w_n(1 + \Delta\lambda W_n)$, kernel $K(x_k, x_l; \lambda + \Delta\lambda)$ and

$$\pi_{kl}(\lambda + \Delta\lambda) = \frac{K(x_k, x_l; \lambda + \Delta\lambda) w_n (1 + \Delta\lambda W_n)}{x_l}$$

as the “*disturbed*” system, while the original system is referred as an “*undisturbed*” one. The time evolution of the disturbed system is as follows. Since Eqs. (10) - (11) do not depend on λ , the only difference between the disturbed and undisturbed systems is at the acceptance-rejection step. The coagulation is accepted with probability

$$\frac{\pi_{kl}(\lambda + \Delta\lambda)}{\hat{\pi}_{kl}}, \quad (17)$$

or rejected with probability

$$1 - \frac{\pi_{kl}(\lambda + \Delta\lambda)}{\hat{\pi}_{kl}}. \quad (18)$$

After this step the average contribution of the k^{th} particle to the functional (15) reads:

$$h(x_k + x_l)w_k(1 + \Delta\lambda W_k) \times \frac{\pi_{kl}(\lambda + \Delta\lambda)}{\hat{\pi}_{kl}} + \\ h(x_k)w_k(1 + \Delta\lambda W_k) \times \left(1 - \frac{\pi_{kl}(\lambda + \Delta\lambda)}{\hat{\pi}_{kl}}\right).$$

Expanding the above formula with respect to $\Delta\lambda$ and keeping the terms up to $O(\Delta\lambda)$ one obtain the average contribution of the k^{th} particle to the functional H :

$$h(x_k + x_l)w_k(1 + \Delta\lambda(W_k + \partial_\lambda \ln \pi)) \times \frac{\pi_{kl}(\lambda)}{\hat{\pi}_{kl}} + \\ h(x_k)w_k(1 + \Delta\lambda(W_k - \pi \frac{\partial_\lambda \ln \pi}{\hat{\pi} - \pi})) \times \left(1 - \frac{\pi_{kl}(\lambda)}{\hat{\pi}_{kl}}\right).$$

Comparison of the above formula with Eqs. (12)-(13) shows that the probabilities of acceptance and rejection in the disturbed system can be the same as in the undisturbed one, i.e., initially all factors $W_k = 0$, the system evolves along the trajectory of the undisturbed system, while at each step (fictitious and non-fictitious) the factors W_k have to be recalculated as

$$W_k = W_k + \partial_\lambda \ln(\pi_{kl}) = W_k + \partial_\lambda \ln(K) + W_l, \quad (19)$$

if the coagulation is accepted, or as

$$W_k = W_k - \pi_{kl} \frac{\partial_\lambda \ln \pi_{kl}}{\hat{\pi}_{kl} - \pi_{kl}} = W_k - w_l K \frac{\partial_\lambda \ln(K) + W_l}{\hat{w}_l \hat{K} - w_l K}, \quad (20)$$

if the coagulation is rejected. The same procedure can be applied for the calculation of second and higher order derivatives.

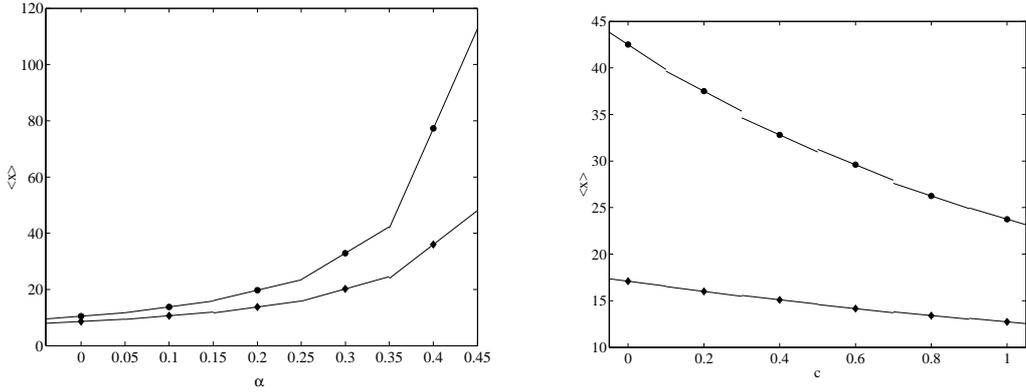
As an illustrative example let us consider a coagulation problem with the following kernel:

$$K(x, x', \alpha, c) = (xx')^\alpha \exp\left(-c \frac{|x^{1/3} - x'^{1/3}|}{(x + x')^{1/3}}\right). \quad (21)$$

The first part of the kernel (21) is homogeneous with respect to x and x' with exponent $\gamma = 2\alpha$. Kernels with $\gamma > 1$ are expected to give rise to gelation, i.e., formation of a particle with mass equal to the mass of the system within a finite time. In the present study we consider only non-gelling kernels. The exponential part of the kernel prevents coagulation of particles with too different diameters, for example, due to hydrodynamic interaction. The majorant kernel is

$$\hat{K}(x, x') = r(xx')^\alpha, \quad (22)$$

where r is a constant that is greater than 1 (we have used $r = 1.5 - 2$) in order to avoid the singularity in Eq. (20). Majorant weights are $\hat{w}_n = w_n$. Note that in a general case, the summation in Eqs. (10) - (11) is a time consuming



(a) $\partial \langle x \rangle / \partial \alpha$, $c = 0$ (circles) and $c = 1$ (diamonds). (b) $\partial \langle x \rangle / \partial c$, $\alpha = 1/3$ (circles) and $\alpha = 1/6$ (diamonds).

Figure 1: Derivative of mean size of the particles with respect to the parameters of coagulation kernel at $t = 10$.

procedure that requires $O(N^2)$ operations. Due to the special structure of the majorant kernel (22) the summation over the indices can be done independently, i.e., $\sum (x_k^\alpha x_l^{\alpha-1}) = (\sum x_k^\alpha)(\sum x_l^{\alpha-1})$. The generalization of the above procedure to other majorant kernels, like sums of products, is straightforward.

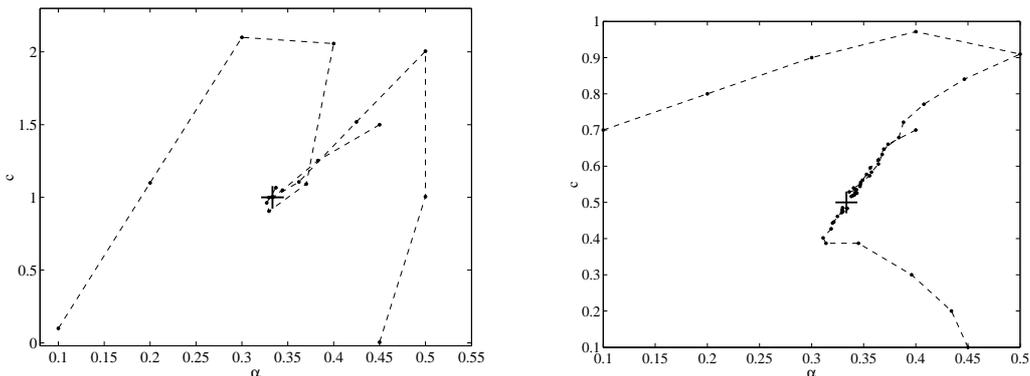
The derivatives of mass-mean size $\langle x \rangle$ of the particles with respect to the parameters of the coagulation kernel are presented in Fig. 1. The bold symbols are the values of $\langle x \rangle$, and the slopes of the lines crossing the corresponding points are calculated according to Eqs. (19)-(20). As one can see, the proposed procedure provides a good estimation for the parametric derivatives of the solution.

Since the total mass of the system does not depend on λ , the mean value of W_k is 0. According to Eqs. (19)-(20), $\sum w_k W_k = 0$ only in a statistical sense, while at each moment it deviates significantly from 0. As a result, the estimation of parametric derivatives based on Eqs. (19)-(20) has a high dispersion. While for calculation of mass-mean size $\langle x \rangle$ in the above mentioned example one needs about 100 repetitions of the algorithm (10)-(13) for $N = 100$, reliable calculation of parametric derivatives requires up to 10^4 repetitions.

In order to reduce the statistical error, we modified the steps (12)-(13) as follows. At each coagulation step we create two particles. One has size $x_k + x_l$, weight $w_k(\pi_{kl})/(\hat{\pi}_{kl})$ and factor W that is calculated according to Eq. (19). The second particle has size x_k , weight $w_k(1 - (\pi_{kl})/(\hat{\pi}_{kl}))$ and factor W that is calculated according to Eq. (20). The direct calculation shows that

$$w_k \frac{\pi}{\hat{\pi}} \partial_\lambda \ln \pi - w_k \left(1 - \frac{\pi}{\hat{\pi}}\right) \pi \frac{\partial_\lambda \ln \pi}{\hat{\pi} - \pi} = 0$$

and $\sum w_k W_k$ does not change. This procedure leads to an increase in the number of particles in the system. Thus, number of particles has to be reduced in order to keep



(a) Kernel (21), $\alpha = 1/3$, $c = 1$.

(b) Kernel (27), $\alpha = 1/3$, $c = 1/2$.

Figure 2: Trajectories of searching algorithm and corresponding target points.

the system computationally tractable. This problem has already been discussed in the context of the weighted particles method for the Boltzmann equation [8]. The method proposed in [8] is based on *clustering*. Consider a group of particles that has total weight $\bar{w} = \sum_{k=1}^{\bar{n}} w_k$ and a mean size $\bar{x} = \bar{w}^{-1} \sum_{k=1}^{\bar{n}} w_k x_k$. If the quantity

$$\rho = \sum_{k=1}^{\bar{n}} w_k (x_k - \bar{x})^2 \quad (23)$$

is small, the group of particles can be replaced by one particle with $w = \bar{w}$, $x = \bar{x}$, $W = \bar{w}^{-1} \sum_{k=1}^{\bar{n}} w_k W_k$. It was shown in [8] that the effect of this replacement on the accuracy is negligible given N sufficiently large. In the present investigation we performed this procedure after each step of the algorithm. The pair of particles that has a minimum or close to minimum value of ρ has been determined by a random search method, i.e., we randomly generated an a priori fixed number ($O(N)$) of pairs (k, l) , the pair with minimum ρ being replaced by one particle. Thus, $\sum w_k W_k = 0$ identically and this procedure decreases deviation of the estimation (16) by orders of magnitude.

4 Results and discussion

4.1 Inverse problems

As soon as the parametric derivatives are known, an efficient gradient search method can be applied to problems (3), (4). Our numerical test is as follows. We solve Eq. (1) with initial particles' masses uniformly distributed from on $[0, 1]$ for some $\vec{\lambda} = (\alpha, c)$ and calculate the mean diameter of the particles $\langle D_{exp} \rangle = \int m(x) D(x) dx$, and

its second and third moments $\langle D_{exp}^2 \rangle = \int m(x)D^2(x)dx$ and $\langle D_{exp}^3 \rangle = \int m(x)D^3(x)dx$, which are referred to as "experimental". Then we solve an identification procedure, i.e., look for the set of parameters $\vec{\lambda}^* = (\alpha^*, c^*)$ that minimizes the residual

$$\frac{1}{2} \sum_{i=1}^3 (D^i(\vec{\lambda}) - D_{exp}^i)^2. \quad (24)$$

Note that an inverse problem is ill-posed [9], a small experimental noise leads to large error in the extracted parameters. Since a Monte Carlo method by itself is a source of statistical noise, we use the overdetermined formulation when three measurements are used for identification of only two parameters.

We use the following minimization algorithm, at each iteration $\vec{\lambda}$ is represented as $\vec{\lambda} + \Delta\vec{\lambda}$, and then $D^i(\vec{\lambda}) = D^i(\vec{\lambda}) + \Delta\lambda_l \partial_{\lambda_l} D^i(\vec{\lambda})$. Substitution of this formula into (24) and differentiation with respect to $\vec{\lambda}$ yields the following system of linear algebraic equations with respect to the increment $\Delta\vec{\lambda}$:

$$\sum_{m=1}^3 a_{lm} \Delta\lambda_m = b_l, \quad (25)$$

where

$$a_{lm} = \sum_{i=1}^3 \left(\frac{\partial D^i}{\partial \lambda_l} \frac{\partial D^i}{\partial \lambda_m} \right) \text{ and } b_m = \sum_{i=1}^3 (D_{exp}^i - D^i(\vec{\lambda})) \frac{\partial D^i}{\partial \lambda_l}. \quad (26)$$

We terminate the calculations when three successive iterations do not improve the residual (24). The results of the calculations are presented in Fig. 2(a). We started the search from different initial points, the points on the plot represent successive values of the identified parameters, the target point is designated by the cross. As one can see, the first few iterations approach the target points and then the solution oscillates near this value, a situation frequently encountered in identification problems.

One of the main advantages of the Monte Carlo method is its ability to treat multi-dimensional problems without significant complications. In order to demonstrate how the proposed method performs with two internal coordinates, namely, size and mixture fraction of an admixture, let us consider droplets that contain an additive. The mass fraction of the admixture in a droplet is denoted by y . We assume that the admixture decreases the probability of coagulation. The coagulation kernel reads:

$$K(x, x', y, y', \alpha, c) = (xx')^\alpha \left(1 - c \frac{xy + x'y'}{x + x'} \right),$$

where $0 < c < 1$. We use the same search procedure in order to extract the constants α and c from "experimental" data. Since we minimize the residual (24), the variable y is a "shadow" variable that is not accessible for direct observation. The results are presented in Fig. 2(b). Although the convergence rate is not as good as in the one dimensional case, less than 20 iterations are sufficient to determine the unknown parameters.

4.2 Coagulation/condensation problems

The problems of simultaneous growth and coagulation of droplets or particles are of high importance for many dispersed systems. Provided that the particles sizes and other necessary parameters are known at a time t , the particles distribution at time $t + \Delta t$ is calculated by operator-splitting technique [10]. In the first phase of the algorithm the particles growth, without any collisions occurring, during the time interval Δt . Sizes of the k^{th} particle and mass of the k^{th} group are determined from the equations of mass transfer:

$$\frac{dx_k}{dt} = F(x_k; \lambda), \quad \frac{dw_k}{dt} = \frac{w_k}{x_k} \frac{dx_k}{dt} = n_k F(x_k; \lambda), \quad (27)$$

where $n_k = w_k/x_k$ is number of particles in the k^{th} group. In the second splitting step coagulation equations (1) or (5) are solved during the same interval Δt .

In order to extend the sensitivity analysis on coagulation/condensation problems we use the operator-splitting method as follows. In the first stage we solve Eq. (27) together with the equation for the parametric derivative of x with respect to λ

$$\frac{d}{dt} \partial_\lambda x_k = \frac{\partial F(x_k; \lambda)}{\partial x_k} \partial_\lambda x_k + \frac{\partial F(x_k; \lambda)}{\partial \lambda}. \quad (28)$$

In order to describe the change of W_k as the result of mass transfer, we rewrite the second of Eqs. (27) as $d(\ln w_k)/dt = d(\ln x_k)/dt$. Differentiation of this equation with respect to λ yields

$$\frac{d}{dt} \partial_\lambda \ln w_k = \frac{dW_k}{dt} = \frac{d}{dt} \frac{\partial_\lambda x_k}{x_k}. \quad (29)$$

In the second stage we solve Eq. (5), while the factors W_k are recalculated according to Eqs. (19), (20). In this case the derivatives $\partial \ln \pi / \partial \lambda$ are calculated as

$$\frac{\partial \ln \pi_{kl}}{\partial \lambda} = \frac{\partial K(x_k, x_l; \lambda)}{\partial \lambda} + \frac{\partial K(x_k, x_l; \lambda)}{\partial x_k} \partial_\lambda x_k + \frac{\partial K(x_k, x_l; \lambda)}{\partial x_l} \partial_\lambda x_l - \frac{\partial_\lambda x_l}{x_l} + W_l. \quad (30)$$

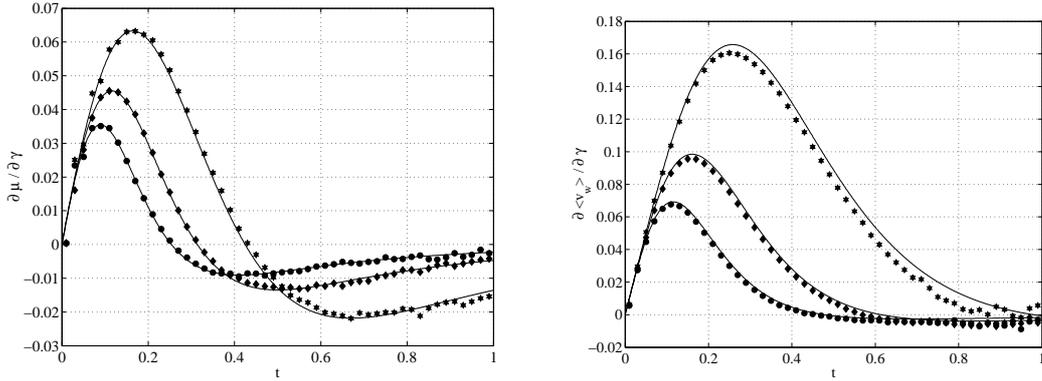
The formula for parametric derivative of a functional $H(t, m; \lambda)$ reads:

$$\frac{\partial H(t, m; \lambda)}{\partial \lambda} \approx \sum_{n=1}^N w_n \left\{ W_n h(x_n(t)) + \frac{h(x_n(t))}{\partial x} \partial_\lambda x_k \right\}.$$

In order to verify our method we compared the numerical predictions with the analytical results available in the literature [11]. We consider growth of an aerosol that contains both water drops and particles of ice. Coagulation kernel

$$K(x, x') = x + x'$$

does not depend on the type of the colliding particles. If an ice particle coagulate with a water droplet, the resulting particle is an ice particle. Coagulation of two ice



(a) Parametric derivative of mass μ of the liquid phase

(b) Parametric derivative of the number-mean volume $\langle v_w \rangle$ of the droplets.

Figure 3: Time evolution of parametric derivatives with respect to the condensation constant γ : $\gamma = 3$ (stars); $\gamma = 5$ (diamond); $\gamma = 7$ (circles). The lines correspond to the analytical solution [11].

particles or two droplets gives ice and droplet, respectively. Vapor condensation on the k^{th} particle is given by the equation

$$\frac{dx_k}{dt} = \gamma M_{ss} x_k, \quad (31)$$

where γ is a constant and M_{ss} is the vapor supersaturation. Thus, mass of k^{th} group and vapor concentration evolve as

$$\frac{dw_k}{dt} = \gamma M_{ss} w_k, \quad \frac{d}{dt} M_{ss} = - \sum_{k=1} \frac{dw_k}{dt}. \quad (32)$$

Differentiation of the above equation with respect to γ yields the equation for sensitivity derivative of the vapor concentration

$$\frac{d}{dt} \partial_\gamma M_{ss} = - \sum_{k=1} \frac{d}{dt} \partial_\gamma w_k = - \sum_{k=1} \frac{d}{dt} w_k W_k.$$

The time derivatives of $\partial_\gamma x_k$ and $\partial_\gamma W_k$ are calculated according to Eqs. (28), (29). Initially, the mass μ of the droplets is, $\mu = 0.7$, the mass m of the ice particles is, $m = 0.3$, $M_{ss} = 1$, both the droplets and the ice particles are distributed exponentially the number-mean volume $\langle v_w \rangle$ is, $\langle v_w \rangle = 1$. The results are shown on Fig. 3. Condensation rate affects the liquid phase through two mechanisms. Directly, it increases the mass of the droplets, but indirectly the growth of the particles increases the coagulation rate between the droplets and the ice particles and leads to the “freezing” of the water phase. Initially, the parametric derivative $\partial\mu/\partial\gamma$ is positive,

but at the late stages of the process higher growth rate means higher collision and freezing rate and $\partial\mu/\partial\gamma < 0$ as one can see on Fig. 3(a). As the supersaturation M_{ss} approaches 0, the spectrum of the droplet phase is stabilized by the freezing, as the theory predicts, and the number-mean volume $\langle v_w \rangle$ of the droplets becomes insensitive to γ . The proposed method predicts correctly the parametric sensitivity of the system from the beginning of the process to its final stage.

5 Conclusion

In conclusion, we have presented an weighted Monte Carlo algorithm for the calculation of the derivatives of solution of the Smoluchowski's coagulation equation with respect to the parameters of the kernel. According to this approach each particle in the simulation algorithm has a statistical weight, and the parametric derivatives are represented as infinitesimal deviations of the statistical weights. The solution of the original problem and all the derivatives are calculated simultaneously. This method can be used for sensitivity analysis and control and identification of dispersed systems. The numerical predictions are in good agreement with the analytical results available in the literature. The generalization of this method to calculation of higher order derivatives as well as to multidimensional cases, to growth, fragmentation and spatially nonhomogeneous processes is straightforward.

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