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A single particle method for the stochastic simulation of coagulation processes

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Abstract

A Monte-Carlo stochastic simulation algorithm based on a single-particle method is suggested to describe steady state particle coagulation processes. The method does not require any information on the nearby particles, instead a fictitious coalescence partner with a given size is generated. The main drawback that limited applicability of this method in the past is that for each control volume the particle size distribution function has to be sampled and stored. In the present study we applied a discrete representation of the distribution function that requires only small memory resources and allows fast updating.

A Single Particle Method

Coagulation and breakage of particles, droplets and bubbles that are suspended in complex recirculating flows are typical processes in numerous industrial and environmental systems. Due to the complexity and multidimensional nature of the processes involved, Monte Carlo statistical simulations become one of the most efficient and maybe the only accessible numerical technique. The analogy between particle collision in suspensions and molecular collisions enables the application of methods previously developed for rarefied gas dynamics [7, 2, 6].

The direct simulation Monte Carlo (DSMC) method can be formulated as follows. The flow volume is divided into cells. The particle ensemble is represented by computational particles such that a group of identical particles in the physical system is substituted by one computational particle. Provided that the particle sizes, positions and other necessary parameters are known at time t, the particle distribution at time $t + \Delta t$ is calculated by an operator-splitting technique which comprises free flow and a collision step.

In the free flow phase the particles move, without any collisions occurring, during the time interval Δt. Their positions, velocities, sizes, temperatures, etc., are determined from the equations of motion, heat and mass transfer. In the second splitting step a new particle ensemble is calculated by simulating spatially homogeneous coagulation in each cell, when binary collisions between particles are sampled randomly. At this step, a particle can collide only with those particles that are in the same cell irrespective of the relative positions of the particles within the cell. The overall solution is thus accurate to first order in Δt. This can be improved by higher order splitting schemes, such as the Strang splitting scheme [13].

An accurate spatial discretisation ranges from $10^3 - 10^4$ cells in the computational domain in the two-dimensional case, while a typical three dimensional flow is usually resolved with $10^5 - 10^6$ cells. For a reasonable representation of a polydispersed particle ensemble one needs $10 - 100$ computational particles in each spatial cell. Thus modelling a spatially inhomogeneous polydispersed system requires simultaneous tracking of $10^4 - 10^8$ particles. Given that the particle number density is nonuniform over the flow region, it is either necessary to increase the total number of computational particles, or use a weighted particle method with splitting and termination of particle trajectories [10] to resolve low-density regions. Both approaches are time-consuming.

However, a significant simplification can be achieved for steady flows. A single particle (or alternatively, test particle) method (SPM) can be applied. The spatially homogeneous coagulation step does not require any information about neighbouring particles, instead a particle (which is referred to as test particle) coagulates with a fictitious collision partner that is generated according to the local particle distribution [12], thus, the particle always has a collision partner even in a low-density region. As soon as the particle leaves the system, the new particle distribution function is recalculated. Since a particle visits many cells (especially if the flow

has recirculation zones) as it cross the flow region, a relatively small number of the particles is sufficient in order to update the new distribution function. In our work [14] the SPM has been used to calculate droplets coagulation and fragmentation in an axisymmetric rotating disc contactor. Less then 5000 particles were sufficient to reach a steady state solution.

This procedure is iterative until convergence is achieved. Note, that if the coagulation submodel is integrated in a computational fluid dynamics (CFD) code, the iterative nature of the above described method should not be considered as a drawback. Most CFD methods use iterations to calculate a steady solution, i.e., an intermediate velocity field is used to calculate an approximated temperature distribution, etc. Thus the coagulation SPM step naturally fits the general iterative strategy.
Note that SPM for the Boltzmann equation has a history longer than the DSMC method [5]. Recently this method has been applied for spatially homogeneous coagulation-fragmentation problem [9, 8]. The main obstacle that limited wide application of SPM in the past was the necessity to store and update the particle distribution function in each cell of the computation domain. This difficulty has been resolved by Vlasov [15]. According to this approach only the number density and parameters of few (maybe even one) particles are stored in each cell. These particles are referred to as field (or target) particles. When a test particle crosses a cell, one of the field particles is replaced by the test particle with a probability p that is proportional to the residence time t_{res} of the test particle in the cell. The number density of the target particles is also updated according to t_{res}. In the present study we investigate the applicability of the SPM to coagulation processes and discuss associated numerical issues.

To proceed further consider a control volume V. The number of particles with size x that enter V is $n_{in}(x)$, and the size-dependent residence time of a particle is $\theta(x)$. The Smoluchowski coagulation equation reads:

$$\frac{\partial n(t,x)}{\partial t} = \frac{1}{2}\int_0^x K(x-x',x')n(t,x')n(t,x-x')dx' + \frac{n_{in}(x)}{V} - \int_0^\infty K(x,x')n(t,x')n(t,x)dx' - \frac{n(t,x)}{\theta(x)}, \quad (1)$$

where $n(t,x)$ is the number density of the particles that have mass x at time t. The probability that two particles with masses x and x', respectively, coalesce during a small time interval dt is $K(x,x')dt$.

Let us reformulate Eq. (1) in terms of mass density. Advantages of this formulation are discussed in [9, 1, 3, 4], note also, that the description of particle distributions according to their mass is 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 density that V contains is $M = \int m(t,x)dx$, and the mass of particles with size x that enter V is $m_{in}(x) = xn_{in}(x)$. The total flow rate of particles through V is $Q_{in} = \int m_{in}(x)dx$. In order to reformulate 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 $x \leqslant 0$ or $x' \leqslant 0$, and so the limits of integration in (1) can be extended from $-\infty$ to ∞. After some algebra we obtain [8, 3]:

$$\frac{\partial m(t,x)}{\partial t} = \int \frac{K(x-x',x')}{x'} m(t,x')m(t,x-x')dx' + \frac{m_{in}(x)}{V} - \int \frac{K(x,x')}{x'} m(t,x')m(t,x)dx' - \frac{m(t,x)}{\theta(x)}. \quad (2)$$

The factor 1/2 before the first integral in Eq. (2) disappears because coagulation reduces the number of particles but does not affect their mass. Eq. (2) can be solved by the mass flow algorithm (MFA) [1, 3] which simulates evolution of N-particles until convergence to a steady state. Below we will use the MFA to validate the results obtained by SPM.

In order to formulate the single particle process, let us rewrite Eq. (2) as

$$\frac{\partial m(t,x)}{\partial t} = \int \frac{K(x-x',x')\phi(t,x')}{x'}\, m(t,x-x')dx' + \frac{m_{in}(x)}{V} - \int \frac{K(x,x')\phi(t,x')}{x'}\, m(t,x)dx' - \frac{m(t,x)}{\theta(x)}, \quad (3)$$

where $\phi(t,x)$ is the mass density of field particles which converges to the mass density of test particles $m(t,x)$ as $t \to \infty$. Thus, formally, Eq. (3) is a linear transport equation with respect to $m(t,x)$ with transition probability depending on $\phi(t,x)$. The Monte Carlo algorithm for single particle process is as follows. The test particle enters the control volume and initially has size x that is generated according to the distribution $m_{in}(x)$. We denote the mass density of field particles at the n^{th} iteration as $\Phi^n = \int \phi^n dx$, and represent the field particles ensemble by N particle groups with sizes $y^n = (y_1^n, ..., y_i^n, ..., y_N^n)$. The total mass of the i^{th} group is Φ^n/N and number of particles in the group is $\Phi^n/(Ny_i^n)$. Since the probability that during a small time interval dt the test particle coagulates with a field particle from the i^{th} group is $K(x,y_i^n)dt$, the probability that the test particle coagulate with any of the i^{th} field particle is $K(x,y_i^n)(\Phi^n/y_i^n)dt$. Thus, the coagulation rate $\rho(x)$ of the test particle is given by summation of the above formula over i:

$$\rho(x) = \frac{\Phi^n}{N} \sum_{i=1}^{N} \frac{K(x,y_i^n)}{y_i^n}. \quad (4)$$

The test particle leaves V with probability $dt/\theta(x)$. Then the simulation algorithm reads:

1. Set $\Phi^{n+1}=0$, $y^{n+1}=y^n$.
2. Generate a test particle according to the distribution $m_{in}(x)$.
3. Generate an exponentially distributed time increment τ with parameter $\rho(x)+1/\theta(x)$.

4. Recalculate the future field particles mass as $\Phi^{n+1} := \Phi^{n+1} + \tau Q_{in}/V$.

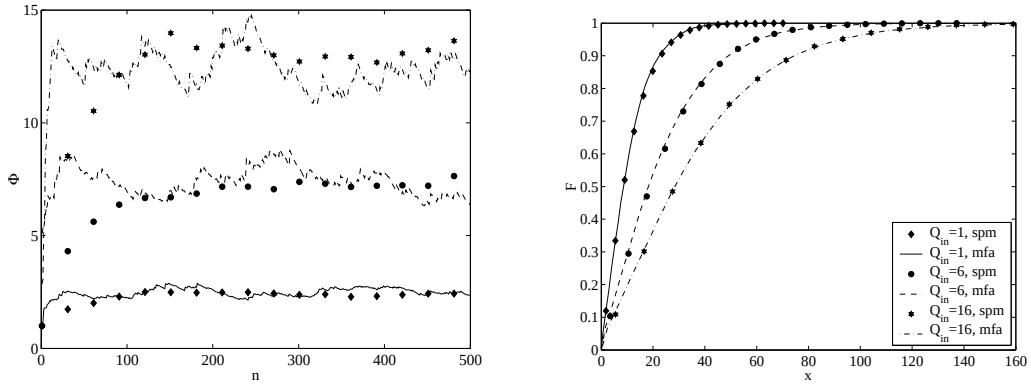
5. With probability
$$\alpha_1 \frac{\tau Q_{in}}{\Phi^n V} \tag{5}$$
replace an uniformly chosen y_i^{n+1} by x, where α_1 is a constant.

6. With probability $\rho(x)/(\rho(x) + 1/\theta(x))$ choose a coagulation step, overwise go to step 8.

7. Choose a collision partner according to the distribution
$$\frac{K(x, y_i^n)\Phi^n}{N\rho(x)y_i^n}, \tag{6}$$
and replace x by $x + y_i^n$. Go to step 3.

8. The test particle leaves the system. Underrelax the field particles mass density as
$$\Phi^{n+1} := \alpha_2 \Phi^{n+1} + (1 - \alpha_2)\Phi^n, \tag{7}$$
where α_2 is a constant, $n := n + 1$. Go to step 1.

In order to underline the connection between the above described algorithm and coagulation equation (2) note that steps 4, 5, and 8 are equivalent to the following equations with respect to Φ and ϕ:
$$\frac{\partial \Phi}{\partial t} = \frac{1}{\tau_1}(M - \Phi), \ \frac{\partial}{\partial t}\frac{\phi}{\Phi} = \frac{1}{\tau_2}(\frac{m}{M} - \frac{\phi}{\Phi}), \tag{8}$$
where $\tau_{1,2}$ are relaxation times. Eqs. (8) together with Eq. (3) provides solution of (2) in a steady state limit.
In order to complete the description of the algorithm one needs to specify the appropriate values of the constants, namely, N and $\alpha_{1,2}$. The probability that a test particle will leave the control volume without being registered in the field particles array is given by the product of Eq. (5) over all time steps:
$$\prod_j (1 - \alpha_1 \frac{\tau_j Q_{in}}{\Phi^n V}) \approx 1 - \alpha_1 \frac{Q_{in}}{\Phi^n V} \sum_j \tau_j = 1 - \alpha_1 \frac{Q_{in}}{\Phi^n V} t_{res}. \tag{9}$$
In the above formula we used the assumption that $\tau \ll t_{res}$. Since $\Phi \sim t_{res} Q_{in}/V$, in order to keep Eq. (9) positive one needs $\alpha_1 < 1$. In our calculations we used $\alpha_1 = 0.1 - 0.5$. The specification of the other parameters is as follows. Our numerical experiments reveal that in order to represent ϕ adequately and avoid correlation between consecutive collisions one needs the number N of field particles to be approximately equal to the number of collisions that a test particle undergoes before it leaves the control volume. Thus, for CFD applications where the size of the control volumes is chosen in such a way that the probability of more than one collision in a



(a) Mass density of the particles in the control volume as a function of the number of iteration

(b) Cumulative mass fraction of the particles that leave the reactor

Figure 1: *Comparison of SPM (symbols) and MFA (lines).*

cell is small, even one field particle per cell is sufficient. The parameter α_2 gives the interval which is used to calculate Φ. According to Eq. (7), the mass density at the n^{th} step of the algorithm is averaged over approximately $1/\alpha_2$ previous iterations. Usually, a choice of $\alpha_2 = 0.01 - 0.1$ prevents large oscillations of the particles mass density in the control volume.

In order to check the performance of the proposed method, we compared the results obtained by SPM with the results by constant particle number version of MFA [11], [3]. The version of MFA used in the present investigation is as follows. The particle ensemble is represented by N groups with sizes $x = (x_1, ..., x_i, ..., x_N)$. The total mass of the i^{th} group is M/N and number of particles in the group is $M/(Nx_i)$. A particle leaves V with probability $dt/\theta(x)$. A new particle enters V with probability $dt(Q_{in}N)/M$, the size of this particle is distributed according to $m_{in}(x)$ and this particle represents a group of identical particles with total mass M/N. The coagulation rate ρ, exit rate ϑ and influx rate μ are given by the formula:

$$\rho = \frac{M}{N}\sum_{i,j=1}^{N}\frac{K(x_i,x_j)}{x_j},\ \vartheta = \sum_{i=1}^{N}\frac{1}{\theta(x_i)},\ \mu = \frac{NQ_{in}}{M}.$$

Then the simulation algorithm reads:

1. Generate an exponentially distributed time increment τ with parameter $\rho + \vartheta + \mu$.

2. With probability $\rho/(\rho+\vartheta+\mu)$ choose coagulation step, overwise go to step 4.

3. Choose collision pair according to the distribution

$$\frac{M}{N}\frac{K(x_i, x_j)}{\rho(x)x_j},$$

and replace x_i by $x_i + x_j$. Go to step 1.

4. With probability $\mu/(\vartheta+\mu)$ a new particle enters the system. Increase the total mass density of the system M by the factor $(N+1)/N$. Uniformly choose an i^{th} particle. Generate a new particle according to distribution m_{in} and replace the i^{th} particle by this newly generated particle. Go to step 1.

5. A particle leaves the system. Decrease the total mass density of the system M by the factor $(N-1)/N$. Choose the particle according to the distribution $\theta(x_i)/\vartheta$. Uniformly choose an j^{th} particle $(i \neq j)$, and replace the i^{th} particle by the duplicate of the j^{th} particle. Go to step 1.

We simulated coagulation of particles in a stirred reactor. The particles that enter the reactor are uniformly distributed on $[0, 1]$. The collision kernel is $K(x, y) = (xy)^{1/3}$, and the probability that a particle with mass x leaves the reactor during small time interval dt is $0.1 \times dt$. The calculations have been performed with 100 field particles for SPM and MFA. Other constants are $\alpha_1 = 0.5$ and $\alpha_2 = 0.01$. The results of the calculations for different mass flowrates Q_{in} are presented on Fig. (1). The initial conditions are $\Phi^1 = 1$, $y_i^1 = 1$. After a short transient period of $n = 1/\alpha_2$ iterations, the mass density of the particles reaches a steady state limit. Then, every time when a particle leaves the system we register its size and mass density of the particles in the reactor. The numerical experiments show a good agreement between the two methods. The computational times for both methods are the same, provided the same number of particles is used in the simulations.

In conclusion, we have introduced a single particle method for a steady state coagulation process. Due to the discrete representation of the distribution function of the particles, the method is computationally and algorithmically simple and gives the same results as direct simulation. Generalization of this method for multidimensional population balance and spatially inhomogeneous problems is straightforward.

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