Direct Simulation Monte Carlo Method for Particle Coagulation and Aggregation

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A Monte Carlo simulation technique developed describes dispersed-phase systems with emphasis on coagulation and aggregation. The method does not use particle trajectories, but is based on the transformation of known collision frequencies into collision probabilities of particle pairs. The particle evolution was computed as a stochastic game, computing the time step after each collision. The simulations were validated by comparing with exact mathematical solutions for aggregation of solid particles and with numerical solutions based on sectional methods for coagulation of droplets. The direct simulation Monte Carlo (DSMC) method is advantageous, because the simulation of complex, multidimensional systems results in very elaborate models when using sectional models and is implemented very easily. Two examples of industrial importance are chemical reaction in coagulating droplets and coating of particles with small solid particles.

Introduction

The time evolution of systems consisting of colliding particles plays an important role in both nature and engineering. These systems include the atmospheric aerosol and interstellar dust, where describing the particle-dynamical processes helps us understand meteorological or astrophysical phenomena, and industrial systems, where understanding the behavior of crystallizers, liquid–liquid reactors, and aerosol reactors can bring economical advantages.

The classic way of dealing with coagulation (describing colliding droplets) and aggregation (describing colliding solid particles) is the Smoluchowski equation (Smoluchowski, 1917). An analytical solution to this equation is only possible in the case of simple collision kernels, which are not relevant for the systems of interest just discussed. In the case of complex coagulation kernels, however, the problem has to be solved by numerical means.

This population-balance problem is traditionally solved by discrete population balances, moment methods, or sectional methods. Solving the discrete particle (or "cluster") -dynamic equation based on the Smoluchowski equation for the whole submicron-size spectrum implies that more than 10^9 differential equations have to be solved simultaneously in order to describe the particle-size distribution $n(v, t)$ between 1 nm and 1 μm. This is computationally too expensive at the moment. It has been done, however, for only a small part of the particle-size spectrum, for example, up to 20 basic units in a particle, whereas the larger particles are described by the sectional representation (Landgrebe and Pratsinis, 1990). Another method of numerical simulation assumes some form of the particle size distribution, for example, a log-normal one, that results in a few relatively easy to solve equations for the moments of the distribution (Pratsinis, 1988; Otto and Fissan, 1999). Finally, the particle-size distribution can be divided into sections for which differential equations can be derived (Litster et al., 1995). This method provides the solution accurately when the number of sections is high enough at a reasonable computational time.

All these methods have the following drawbacks:

1. There is no information about the history of each particle. The particles are described by means of their volume. Which particles collided to form a bigger particle cannot be determined, so the information about the internal structure is lost. This is a problem, especially in case of aggregates, as sometimes the size distribution of the primary particles com-

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posing an aggregate has to be known. Such cases are, for example, the mixing of solid particles with different composition and the coating of large particles with smaller ones.

2. When multidimensional systems have to be dealt with, the sectional representation results in very complex algorithms. Examples are additional information about the agglomerate form (Xiong and Pratsinis, 1993) using a particle-size distribution $n(v, a, t)$ to describe the particle volume and area or about the charge distribution (Vemury et al., 1997), which requires the solution of $n(v, q, t)$ to describe volume and charge level.

A classic method for overcoming such drawbacks is to use Monte Carlo simulation methods. A well-known example of this method is the simulation of the Brownian movement by stochastic processes. When a sufficient number of particles is taken, collisions will result from the random Brownian motions and a change in the particle-size distribution can be recorded. With the help of such trajectory methods based on the Langevin equation, Brownian coagulation of droplets (Pearson et al., 1984) and agglomerate formation by Brownian coagulation (Gutsch et al., 1995) have been described. Such methods can deliver *a priori* coagulation rates, but are time-consuming and are not suited to simulate large numbers of particles. Here, the number of simulated particles is usually between 500 and 1000.

When the collision rates are known, a much simpler method can be used. This method is based on choosing collision partners by a stochastic game and is known as the direct simulation Monte Carlo method (DSMC; Bird, 1976). It was originally developed for describing gas dynamics and is essentially the simulation of molecular collisions without regarding their spatial positions. Since the number of particles in real systems is too large for the present computer memories, the simulation volume is chosen such that a reasonable number (10^4–10^5) of simulation particles results. One advantage is that information about the history and internal structure of the particles is available, and that multidimensional systems, such as charged and multicomponent aerosols, requiring the solution of $n(v, q, c, t)$ describing particle volume, charge level, and composition, can be simulated. Monte Carlo (MC) simulations are computationally expensive; however, computer power has reached a level where simulations with up to 10^5 particles are now possible on fast PCs. Furthermore, more care has to be taken to ascertain that the numerical solutions, which vary due to the use of random numbers, are representative ones. The MC algorithms for solving particle dynamics are easily programmed, however, and it can be said that in comparison to sectional models, the time spent on solving a problem has shifted from the programmer to the computer.

The DSMC method has been used before to solve complex problems that involved particle collisions (such as Shah et al., 1977; Bapat et al., 1983; Smith and Matsoukas, 1998). There are, however, few studies in which different DSMC methods for solving the coagulation and aggregation problem are investigated and tested more in detail. In this work, these methods are briefly described and then we propose a related method that is computationally optimized. We test the accuracy of the solutions by comparing them with (1) analytical solutions, and (2) numerical results from a sectional model. Two examples of industrial relevance are given to show the suitability of the DSMC method for describing complex multidimensional particle dynamics.

Direct-Simulation Monte Carlo Methods

It is assumed that the N colliding particles with an initial distribution $n_0(v)$ are contained in a fluid of volume V. This volume is chosen such that N is a reasonable number, with the present personal computer power not more than 10^5. We further assume that all collisions are binary, that is, we are not dealing with concentrated dispersions or slurries, and that the sticking coefficient is unity. The collision rate β is a known function that depends on the properties of the medium and on the properties of the colliding particles.

The particulate system considered here is usually described by the Smoluchowski equation:

$$\frac{\partial n_k}{\partial t} = \frac{1}{2}\sum_{i=1}^{k-1} \beta_{i,k-i} n_i n_{k-i} - n_k \sum_{i=1}^{\infty} \beta_{i,k} n_i, \qquad (1)$$

where t is the time, n_k is the number concentration of the k-fraction, and $\beta_{i,k}$ is the collision rate of the particle pair (i,k) that defines the kinetics of the process. In the DSMC method, particle pairs are selected by stochastic methods based on the transformation of collisions rates into collision probabilities.

Garcia et al. (1987) describes two stochastic methods based on the simulation procedure for a Markov process. The methods were not tested by these authors, however. The mean time between two collisions events is τ, which can be calculated by

$$\tau = \frac{1}{\sum_{i=1}^{N}\sum_{j=i+1}^{N} \beta_{i,j}^{V}}, \qquad (2)$$

in which β^V is the collision rate in a volume V. It can be proved that τ is an exponentially distributed random variable. This time, τ, is therefore selected by means of a random number generator. The coagulation pair (i, j) is chosen on the basis of the probability $Pr_{i,j}$, which is calculated by:

$$Pr_{i,j} = \frac{\beta_{i,j}^{V}}{\sum_{k=1}^{N}\sum_{l=k+1}^{N} \beta_{k,l}^{V}}. \qquad (3)$$

Because it is necessary in this method to calculate the collision rates of all possible particle pairs (inverse method), which is time-consuming, Garcia et al. (1987) proposed a second method based on the acceptance–rejection procedure (Bird, 1976). Here, a particle pair (i, j) is selected entirely at random independent of the collision probability. Then this pair is accepted as a collision pair if

$$R < \frac{\beta_{i,j}^{V}}{\max\left\{\beta_{k,l}^{V}\right\}}, \qquad (4)$$

where R is a random number uniformly distributed between 0 and 1. When the pair is rejected, a new random number is selected, and so on, until the condition is fulfilled. This method is only practical when the maximum of the collision rate can easily be determined and might introduce an error according to Garcia et al. (1987). In order to investigate this error, we compared simulations based on this method with the analytical solution for the simple collision kernel $\beta = A(i + j)$ (described further in this article). The results are shown in Figure 1. Even with 10^4 simulation particles, the error in the total particle number is rather large in comparison with the results that are shown later in this article (see Figure 3b). Furthermore, when the range of particle sizes increases the ratio of the randomly generated collision rate and the maximum collision rate becomes very small, the large number of rejections slows down the program considerably. This problem was discussed by Smith and Matsoukas (1998), who also used the acceptance–rejection method. As an example, in an aerosol system with a wide range of particle classes, undergoing Brownian coagulation $\beta(d_1 = 1 \text{ nm}, d_2 = 1 \text{ nm})/\beta(d_1 = 1 \text{ nm}, d_2 = 100 \text{ nm})$ at standard conditions is on the order of 10^{-3}.

A third method is proposed by Liffman (1992), who uses a two-step method. The mean time that particle i experiences between two collision events is

$$\tau_i = \frac{1}{\sum_{j=1}^{N} \beta_{i,j}^V}. \tag{5}$$

This time is an exponentially distributed function. A time step is chosen that should be much smaller than the smallest value of all possible τ_i. Then, the particle i is accepted if

$$R < 1 - e^{[(-\Delta t/2\tau_i)]}. \tag{6}$$

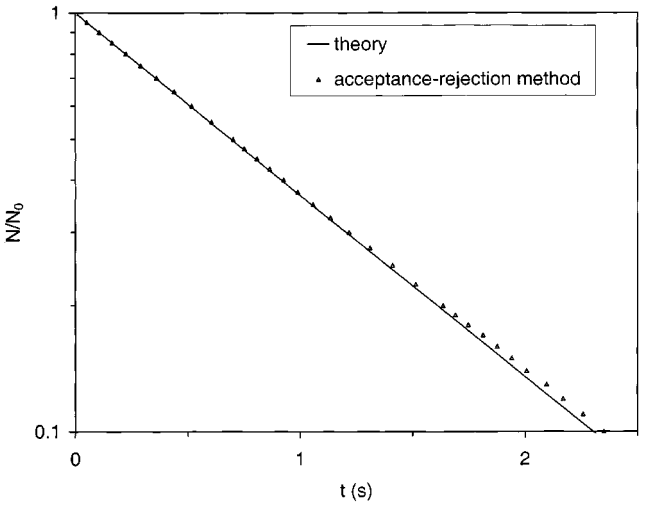

Figure 1. Decrease in relative total number concentration N/N_0.

Comparison between theory (Case 2, $\beta_{i,j} = A(i + j)$) and the acceptance–rejection Monte Carlo simulation procedure of Garcia et al. (1987). Number of simulation particles: 10,000.

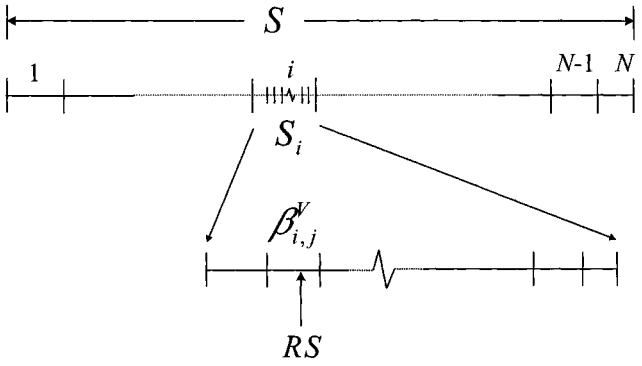

Figure 2. Illustration of the MC technique used to choose pairs of colliding particles (i, j) by means of a random number R.

In this procedure the collision rates of all possible particle pairs are weighted.

When i has been found, the collision partner j is found by first computing all collision probabilities between the chosen particle i and any other particle j:

$$Pr_j = \frac{\beta_{i,j}^V}{\sum_{k=1}^{N} \beta_{i,k}^V}, \tag{7}$$

and then selecting j by means of

$$\sum_{k=1}^{j-1} Pr_k \le R \le \sum_{k=1}^{j} Pr_k. \tag{8}$$

As in the acceptance–rejection method of Garcia et al. (1987), selecting one particle pair might take a large number of random numbers, which is computationally expensive. A further problem is that it is not a clear criterion for selecting the time step. Nevertheless, the simulation results were extensively compared with analytical solutions and found to be correct.

A Fast DSMC Method

We propose a new method that is related to the inverse method. As the collision kernel represents the probability of a binary collision in unit time, the sum over all collision kernels will be the probability that one collision takes place in the particle ensemble in unit time. So the mean time $\langle \tau \rangle$ needed for one collision taking place in the particle ensemble is the inverse of the sum of all collision kernels:

$$\langle \tau \rangle = \frac{2}{\sum_{i=1}^{N} \sum_{j=1, i \ne j}^{N} \beta_{i,j}^V} \tag{9}$$

The factor 2 is added in order to account for the double counting of the particle pairs. We choose a collision pair by making a list of all pairs and accumulate the collision rate until the sum surpasses a random number. If a random number R is uniformly distributed in (0,1], $R\sum\sum\beta^V_{i,j}$ is uniformly distributed in $(0, \sum\sum\beta^V_{i,j}]$. We can write the selection criterion as follows:

$$\sum_{k=1}^{i}\sum_{l=1}^{j}\beta^V_{k,l} \le R\sum_{k=1}^{N}\sum_{l=1}^{N}\beta^V_{k,l} \le \sum_{k=1}^{i}\sum_{l=1}^{j+1}\beta^V_{k,l} \quad \text{with } k \neq l, \tag{10}$$

which causes the particle pair (i, j) to form a new particle with volume $v_i + v_j$. Only one random number is used for the selection of a particle pair and no acceptance–rejection procedure takes place. If m steps are completed, the total time can be simply calculated from:

$$t = \sum_{i=1}^{m}\langle\tau\rangle_i. \tag{11}$$

When an initial size distribution has to be modeled, it is generated by means of a random generation method (Rubinstein, 1981). The DSMC method can be applied in an aggregation mode, in which each aggregate is assigned an array indicating which primary particles compose the aggregate, or in a coagulation mode, in which after each collision the new (droplet) properties such as volume and density are stored.

During the simulation the number of simulation particles N drops, which means that the error increases, as the sample error is on the order of $\sigma/\sqrt{N}$. In order to counter this, the simulation volume V and the number of particles N are doubled when the concentration has dropped by half. The properties of each "old" particle are of course copied to the "new" particles in the added volume in order to conserve the statistical properties. This procedure is discussed by Liffman (1996).

It should be pointed out that this method necessitates the calculation of the collision rates of all possible particle pairs. If N is large, the storage of N^2 collision rates becomes prohibitive; 10^5 simulation particles would mean the storage of 10^{10} collision rates. In order to overcome this, only the sum of possible collision rates for each particle $S_i = \sum_{l=1, l\neq i}^{N}\beta^V_{i,l}$

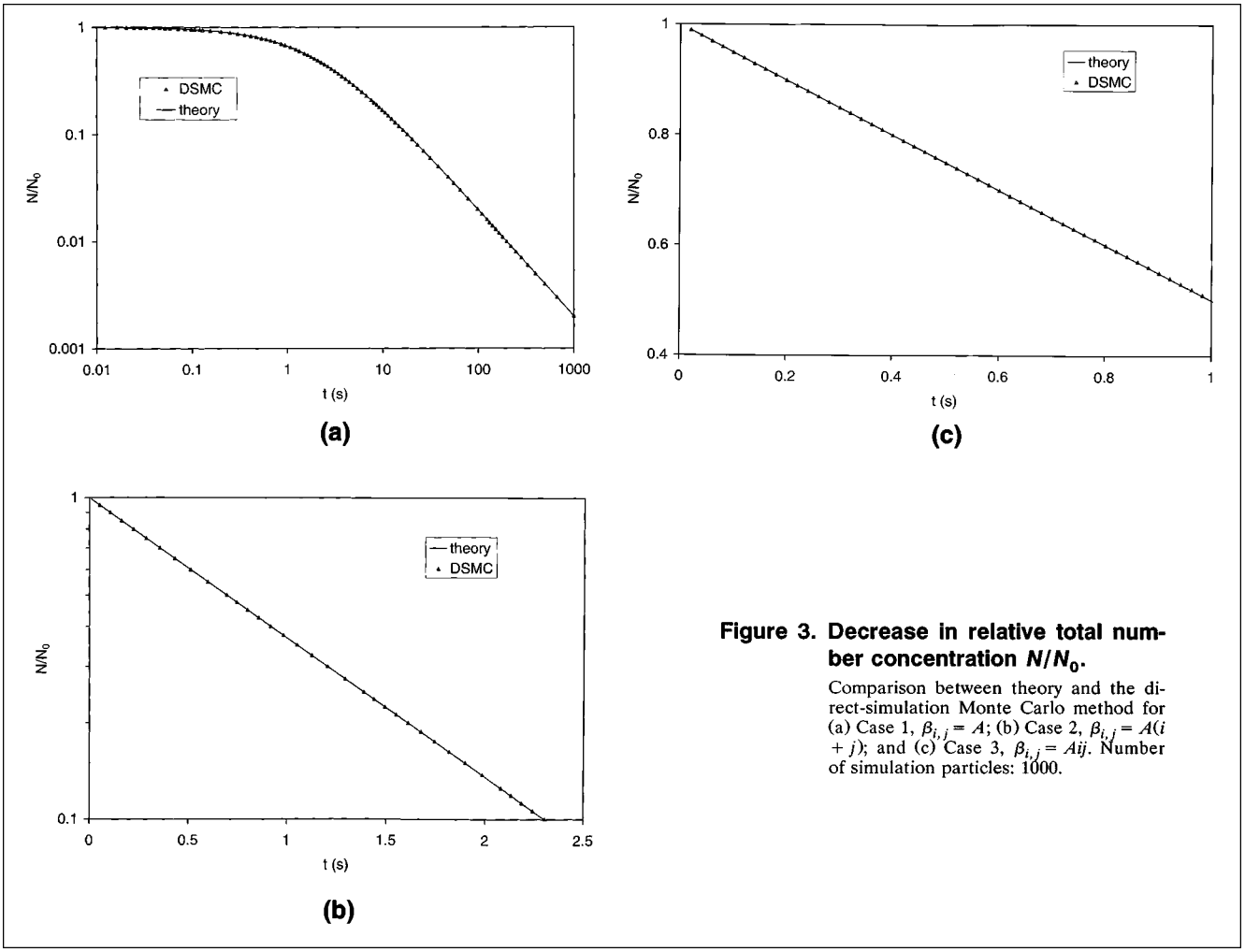

Figure 3. Decrease in relative total number concentration N/N_0.

Comparison between theory and the direct-simulation Monte Carlo method for (a) Case 1, $\beta_{i,j} = A$; (b) Case 2, $\beta_{i,j} = A(i + j)$; and (c) Case 3, $\beta_{i,j} = Aij$. Number of simulation particles: 1000.

is stored. The condition (Eq. 10) can now be rewritten as a sequence of two steps. First, i is selected by means of

$$S_{i-1} \le RS \le S_i, \tag{12a}$$

in which $S = \sum_{i=1}^{N} S_i$. The second step yields j by means of:

$$S_{i-1} + \sum_{l=1,\, l\neq i}^{j-1} \beta_{i,l}^{V} \le RS \le S_{i-1} + \sum_{l=1,\, l\neq i}^{j} \beta_{i,l}^{V}. \tag{12b}$$

This is shown in Figure 2. This method implies the storage of only N values. After each pair selection, a simple correction can be made for all S_k values to account for the change in particle properties and, consequently, in the collision rates. Our evaluation of $\langle \tau \rangle$ according to Eq. 9 implies a double counting of coagulation coefficients, which could be avoided by using Eq. 2. This is inherent to our algorithm mainly due to solution chosen for the storage problem (Eq. 12). This double counting takes place only in the initial calculation of all possible interactions, and even could be avoided at the cost of increasing the complexity of the algorithm. Each aggregation or coagulation step leads only to correction of the S_k values, and there is no further double counting during the simulation itself. The large number of calculations of the collision rate, which are independent of each other, makes the simulation very suitable for parallel computing.

The method described here is similar to the inverse method of Garcia et al. (1987). However, we do not use exponentially distributed time steps between collisions, which would necessitate additional generation of a random number, but instead use a calculated deterministic mean time. This is possible due to the large number of simulated particles, and it simplifies the simulation procedure.

Comparison with Theoretical Solutions for Aggregation

Obviously, the fast DSMC must be compared to known theoretical solutions to determine if it is a valid method of analysis and to get an idea of its accuracy. There are only a few forms of the collision kernel for which an analytical solution of Smoluchowski equation exists (Spouge, 1983): $\beta_{i,j} = A$, $\beta_{i,j} = A(i + j)$, $\beta_{i,j} = Aij$. Here A is a constant, and i,j are the section numbers. We examine the fast DSMC method using these collision kernels. The initial particle-size distribution is monodisperse.

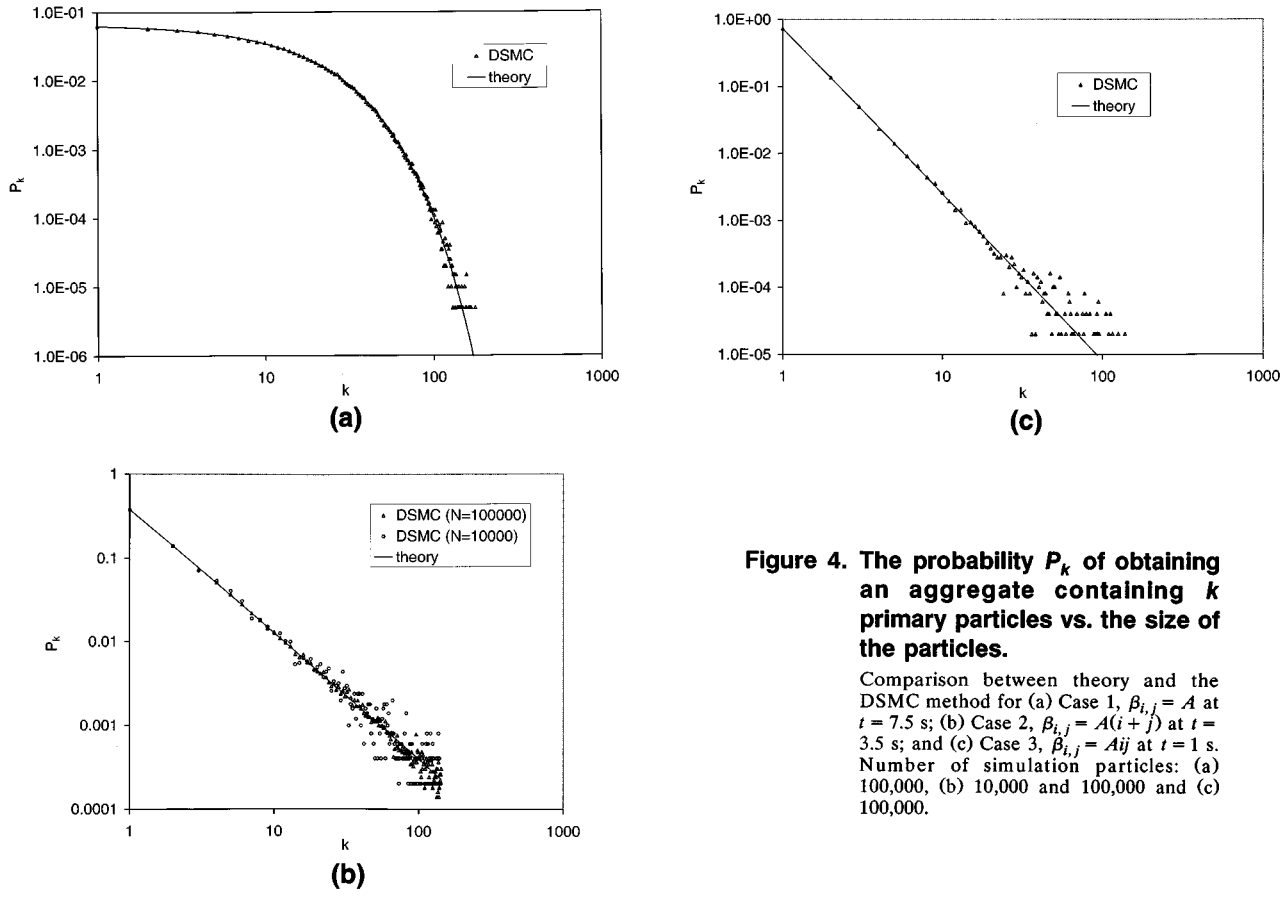

Figure 4. The probability P_k of obtaining an aggregate containing k primary particles vs. the size of the particles.

Comparison between theory and the DSMC method for (a) Case 1, $\beta_{i,j} = A$ at $t = 7.5$ s; (b) Case 2, $\beta_{i,j} = A(i + j)$ at $t = 3.5$ s; and (c) Case 3, $\beta_{i,j} = Aij$ at $t = 1$ s. Number of simulation particles: (a) 100,000, (b) 10,000 and 100,000 and (c) 100,000.

Case 1: $\beta_{i,j} = A$. If we define N_0 as the initial number of primary particles at $t = 0$, the total number of particles N at time t is

$$N = \frac{N_0}{1 + N_0 At/2}. \tag{13}$$

The probability of obtaining a particle containing k primary particles is

$$P_k = \frac{(N_0 At/2)^{k-1}}{(1 + N_0 At/2)^k}. \tag{14}$$

Case 2: $\beta_{i,j} = A(i + j)$. In this case, the decrease of particle number can be expressed as

$$N = N_0 e^{-N_0 At} \tag{15}$$

with the probability of obtaining a k-aggregate

$$P_k = \frac{k^{k-1}}{k!}(1 - e^{-N_0 At})^{k-1} e^{-k(1-e-N_0 At)}. \tag{16}$$

Case 3: $\beta_{i,j} = Aij$. In this case there exists only an analytical solution for times shorter than a finite time $t = 1/N_0 A$,

$$N = N_0(1 - N_0 At/2), \qquad 0 \le t < \frac{1}{N_0 A} \tag{17}$$

$$P_k = \frac{k^{k-2}}{k!} \frac{(N_0 At)^{k-1}}{(1 - N_0 At/2)} e^{-k N_0 At}, \quad 0 \le t < \frac{1}{N_0 A}. \tag{18}$$

The results of the simulations for $N_0 = 10^{12}$ m^{-3} and $A = 1/N_0$ are shown in Figures 3 and 4.

Figure 3 demonstrates the high degree of accuracy of the DSMC method in simulating the decrease in relative total number concentration. Here, using just 1000 simulation particles results in very accurate solutions for the total number concentration. The doubling procedure is a necessity, as can be seen in Figure 3a, where the total number concentration drops by a factor of 10^3. This results in some eight doubling procedures, clearly resulting in an accurate description. Figures 3b and 3c show the two other test cases.

The probability distribution of aggregates with k primary particles for the just mentioned test cases is, however, a more sensitive function of the number of simulation particles. The statistical nature of Monte Carlo simulations become evident when there are only a few aggregates in the simulation volume. This can be seen in Figures 4a and 4c, where the scatter increases at large k-numbers. Figure 4b clearly shows the decrease in statistical scattering when increasing the number of simulation particles from 10,000 to 100,000.

Comparison with Numerical Solutions for Coagulation

The form of the particle size distribution is an important issue in particle production units such as aerosol reactors or crystallizers. In order to check the validity of the DSMC method in a more realistic system than cluster aggregation, a coagulating aerosol is simulated. Figure 5 shows the development of the geometric standard deviation σ_g in an initially polydisperse aerosol. A sectional model (Landgrebe and Partsinis, 1990) is used as a reference, with a section spacing of 1.08. It can be seen that two simulations with the same number of simulation particles can give different results due to the stochastic nature of the modeling. It is seen that when a sufficient number of particles is taken, the deviations to the sectional model are several percent. Since both methods are numerical, it is difficult to say which one is the more correct. The stochastic effect can be reduced by repeating the simulation with different random numbers. This is shown in Figure 5 for the case of 40,000 simulation particles, where, after taking the mean of three different simulations, the simulation shows a smooth curve.

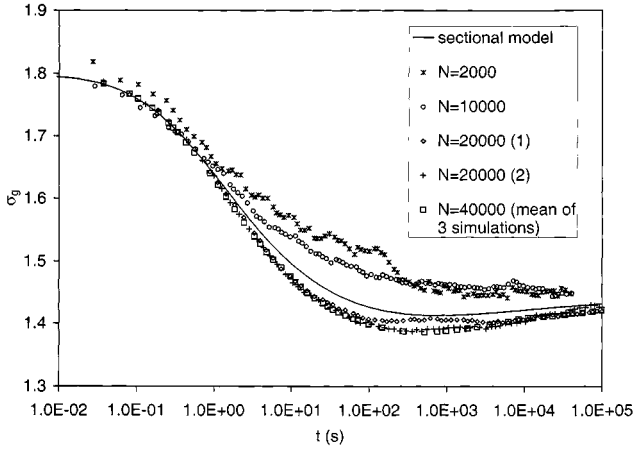

Figure 5. The geometric standard deviation σ_g of a coagulating aerosol system (initially $d_g = 53.5$ nm, $N = 10^9$ cm^{-3}, and $\sigma_g = 1.8$) as a function of time t.

Comparison between a sectional model and the DSMC method. The collision kernel used is from Dahneke (1983), and the sectional model used is by Landgrebe and Pratsinis (1990). The number of simulation particles N is indicated in the legend.

The statistic errors can be quantified by calculating the standard deviation from different simulations. This procedure is illustrated in Figure 6, which shows the so-called self-preserving particle-size distribution (Friedlander, 1977) for the collision kernel $\beta(v_i, v_j) = (v_i^{1/3} + v_j^{1/3})(v_i^{-1/3} + v_j^{-1/3})$. Ten DSMC simulations were made so that an estimation of the statistical errors could be obtained by the standard σ method (corresponding to a probability of 67%). The figure shows that the form of the distribution is correctly described by the DSMC method and that the standard σ method generally overestimates the error in the simulations. The statistical errors can be further reduced by increasing the number of simulations and/or the number of simulation particles. This was not pursued further. Curve-fitting can also be applied here. The most striking feature in the figure is that the left-hand side of the curve shows much larger statistical deviations than the righthand side does. This is due to the fact that the method is particle number-based; to the left of the

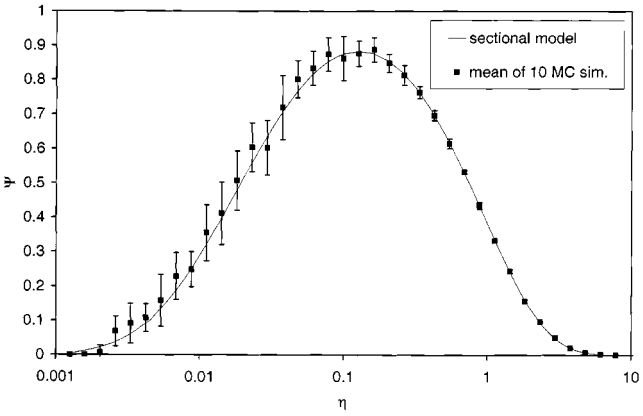

Figure 6. Self-preserving particle-size distribution for the collision kernel $\beta(v_i, v_j) = (v_i^{1/3} + v_j^{1/3})(v_i^{-1/3} + v_j^{-1/3})$, in which the dimensionless particle-density function Ψ (defined as $Vn(v,t)/N^2$, where V and N are the total particle volume and number) is a function of a dimensionless particle volume η (defined as Nv/V).

The DSMC results are compared with results from Vemury et al. (1994), who use a sectional model. A confidence range of $\pm\sigma$ is calculated from 10 simulations, with 60,000 simulation particles in each simulation.

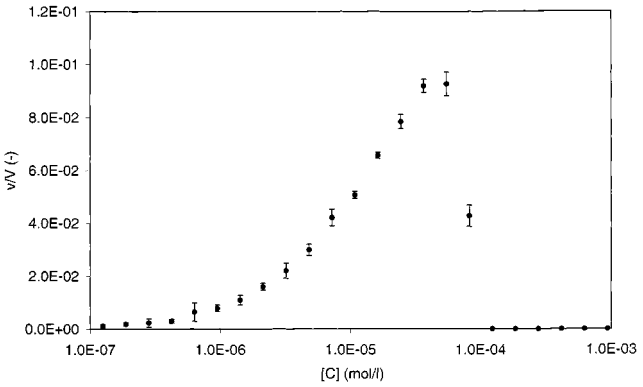

Figure 7. Distribution of product concentration $[C]$ resulting from the chemical reaction $A + B \rightarrow C$ with a reaction rate $k = 2.067\ 10^{-3}$ L/(mol s) between an A-containing spray (initially $[A] = 10^{-3}$ mol/L, $d_g = 1\ \mu$m, $N = 5\ 10^2$ cm^{-3}, and $\sigma_g = 1.5$) and a B-containing spray (initially $[B] = 10^{-3}$ mol/L, $d_g = 1\ \mu$m, $N = 5\ 10^2$ cm^{-3}, and $\sigma_g = 1.6$).

υ/V is the volume fraction of droplets having a concentration $[C]$. Not shown is the fraction unreacted droplets with $[C] = 0$ mol/L. The time elapsed is $4.37\ 10^5$ s. The initial number of simulation particles is 30,000 particles for each spray. The error bars ($\pm\sigma$) result from repeating the simulation four times.

maximum, fewer simulation particles are available for one data point due to the logarithmic axis.

Case study 1: Chemical reaction in coagulating droplets (microreactors)

The standard numerical solution schemes, for example, sectional models, for solving coagulating particle systems are more computer time-efficient than the DSMC simulation. They are not, however very well suited to solving more complex systems, such as reacting droplets. In order to show the applicability of the DSMC simulations, we take the reactive mixing of two sprays containing two different solutions. A chemical reaction takes place when droplets of solution A coagulate with droplets containing solution B, producing a new droplet acting as a "microreactor" in which product C is formed. This process is used to mix two liquid reactants very fast. The chemical kinetics is given by

$$\frac{d[C]}{dt} = k[A][B], \tag{19}$$

where $[A]$ denotes the concentration of A and k is a reaction constant. The problem is analogous to that of a chemical reaction in the liquid–liquid dispersion described by Shah et al. (1977). However, we assume polydisperse initial-size distributions. The chemical reaction is closely related to the droplet dynamics, as a collision event changes the concentrations of the reactants and product, depending on the relative volume of the colliding droplets. The coagulation version of the DSMC code is very easily adapted for calculating the change in concentrations due to the chemical reaction between two collisions by adding only a few additional program lines.

The results of the simulation are plotted in Figure 7. As collision kernel the collision frequency for the continuum regime (Friedlander, 1977) is taken. The results for the volume fraction of droplets υ/V having the indicated product concentration $[C]$ are shown in a histogram fashion, with the values of the concentration located at the midpoint of a section. Here several simulations also were made to get an idea of the accuracy of the results by using the standard σ method. It is seen that only a few simulations are necessary to get a relatively accurate result. The DSMC is an easily programmed method for calculating complicated multidimensional-size distributions, in this case $n(\upsilon,[A],[B],[C],t)$.

Case study 2: Coating particles with smaller particles by aggregation

A second example of a problem that is very difficult to solve with standard particle dynamic models is the coating of solid particles with nanoparticles, with both particle fractions having an initially polydisperse size distribution. Particle coating is used in the industry to apply a functional, protective, or wear-resistive layer, and is often carried out in a fluidized-bed reactor (Li and Hua, 1997). The problem is to determine how much of the surface of the larger particles (1 μm) is covered with the smaller particles (100 nm). This is expressed in the surface coverage, which is defined as the ratio of the sum of the area of all nanoparticles deposited on a large particle and the surface area of the large particle. Due to the polydisperse size distribution and the random nature of the collision process, the surface coverage of the large solid particles will be a distributed variable.

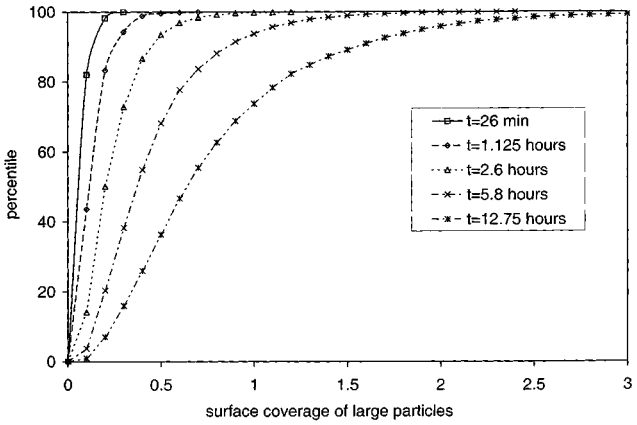

Figure 8. Cumulative distribution of the surface coverage as a result of coating large particles ($d_g = 1\mu$m, $N = 10^3$ cm^{-3}, and $\sigma_g = 1.2$) with nanoparticles ($d_g = 100$ nm, $N = 10^6$ cm^{-3}, and $\sigma_g = 1.5$) for different coating times indicated in the legend.

The initial number of simulation particles is 20,000 for the nanoparticles, the simulation was repeated five times.

For the DSMC simulation, it means that the aggregation mode has to be used where information about the primary particles composing an aggregate is stored. As an approximation, the collision kernel for the continuum regime (Friedlander, 1977) is used. The results are shown in Figure 8 as a cumulative distribution of the surface coverage for the different coating times indicated in the legend. As can be seen from the slopes of the curve, there is a rather broad distribution of surface coverage of the large particles. The results of the DSMC method are in fact a solution to the multidimensional size distribution $n(v_1, v_2, \ldots, v_{n_p}, t)$ in which n_p is the number of particles in the aggregate.

Conclusions

In this article, we have demonstrated the suitability of a direct simulation Monte Carlo (DSMC) method for simulating complex particle dynamics. Contrary to Langevin-based methods, where particle trajectories are used, collision frequencies are the basis of the stochastic game that chooses collision partners. The DSMC method developed here allows a sufficient number of simulation particles to be taken in order to get accurate results and, in order to retain this accuracy, includes a doubling procedure when the particle number has dropped by half. The method is validated by comparing with analytical results for particle aggregation and with numerical results for droplet coagulation. Care has to be taken that a sufficient number of simulation particles is used in order to minimize statistical errors. These errors can easily be estimated by repeating the simulation several times with other random numbers, because a Monte Carlo simulation can be seen as a numerical experiment. The method is especially suited for solving multidimensional problems of distributed variables that would normally need simplification or very complicated models, usually limited to two dimensions. Two examples of industrial relevance containing multidimensional particle dynamics are shown: coagulation and chemical reaction in droplets (microreactors), and the coating of solid particles with nanoparticles.

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