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# Monte Carlo simulation for simultaneous particle coagulation and deposition

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**Abstract** The process of dynamic evolution in dispersed systems due to simultaneous particle coagulation and deposition is described mathematically by general dynamic equation (GDE). Monte Carlo (MC) method is an important approach of numerical solutions of GDE. However, constant-volume MC method exhibits the contradictory of low computation cost and high computation precision owing to the fluctuation of the number of simulation particles; constant-number MC method can hardly be applied to engineering application and general scientific quantitative analysis due to the continual contraction or expansion of computation domain. In addition, the two MC methods depend closely on the "subsystem" hypothesis, which constraints their expansibility and the scope of application. A new multi-Monte Carlo (MMC) method is promoted to take account of GDE for simultaneous particle coagulation and deposition. MMC method introduces the concept of "weighted fictitious particle" and is based on the "time-driven" technique. Furthermore MMC method maintains synchronously the computational domain and the total number of fictitious particles, which results in the latent expansibility of simulation for boundary condition, the space evolution of particle size distribution and even particle dynamics. The simulation results of MMC method for two special cases in which analytical solutions exist agree with analytical solutions well, which proves that MMC method has high and stable computational precision and low computation cost because of the constant and limited number of fictitious particles. Lastly the source of numerical error and the relative error of MMC method are analyzed, respectively.

**Keywords: multi-Monte Carlo method, general dynamic equation, coagulation, deposition, particle size distribution, dispersed systems.**

The phenomena of simultaneous particle coagulation and deposition are of great ubiquity and of key importance in nature and various engineering applications, including the precipitation and downfall of rain, ice, snowflakes, fog and hail, the scavenging of fly ash

in electrostatic precipitator of power plant, the transplantation of fume in smoke flue of pulverized coal-fired plant, nuclear reactor analysis modeling aerosol's diffusion and deposition when blasting, the suspension and sedimentation of emulsoids and floccules in chemical engineering, the migration of indoor air pollutant, and the production of nanoparticle agglomerates, etc. Both particle coagulation, in which two particles collide together and form a bigger particle, and deposition, where one particle settles and does not participate in the dynamic evolution of particle population any longer, result in the time evolution of particle size distribution (PSD). Because many physico-chemical properties of particle population, such as light scattering, electrostatic charging, toxicity, radioactivity of suspended particle, sediment and capturing strategy, depend heavily on their size distribution, the time evolution of size distribution is of fundamental interest and a key issue. The familiar general dynamic equation (GDE) describes the time evolution of PSD for simultaneous coagulation and deposition as following[1]:

$$\left\{\frac{\mathrm{d}n_{\mathrm{p}}(v,t)}{\mathrm{d}t}\right\} = \left\{-R(v)\,n_{\mathrm{p}}(v,t)\right\}_{\text{deposition}} + \left\{\frac{1}{2}\int_0^v \beta(v-u,u)n_{\mathrm{p}}(v-u,t)n_{\mathrm{p}}(u,t)\mathrm{d}u - n_{\mathrm{p}}(v,t)\int_0^\infty \beta(v,u)n_{\mathrm{p}}(u,t)\mathrm{d}u\right\}_{\text{coagulation}}. \quad (1)$$

GDE, in which only binary coagulation is considered, is based on the assumption of dilute particles and molecule chaos. In eq. (1), $n_{\mathrm{p}}(v,t)$ is the function of particle size distribution at time $t$, so that $n_{\mathrm{p}}(v,t)\mathrm{d}v$ is the number concentration of particles whose sizes range between $v$ and $v+\mathrm{d}v$ per volume unit at time $t$; the dimension of $n_{\mathrm{p}}(v,t)$ is $\mathrm{m}^{-3}\cdot\mathrm{m}^{-3}$; the term on the left-hand side of eq. (1) describes the change in the number concentration of particles of volume $v$ with time; $R(v)$ is the deposition kernel or deposition coefficient for a particle with volume $v$, $\mathrm{s}^{-1}$, describing the probability of deposition event of particle with volume $v$ per time unit; $R(v)$ encapsulates all kinds of deposition mechanism such as gravitational sedimentation, Brownian diffusion, turbulent diffusion, thermophoresis, electrostatic forces, and wet removal, etc. The "deposition" term on the right-hand side describes the loss in number concentration due to the deposition of particle with volume $v$; $\beta(v,u)$ is the coagulation kernel for two particles with volume $v$ and $u$, $\mathrm{m}^3\cdot\mathrm{s}^{-1}$; the physics of the problem, such as Brownian diffusion, turbulent transport effect, accumulation effect and gravitational settling, is encapsulated in the coagulation kernel; the "coagulation" term describe the gain and loss in number concentration due to coagulation, where the first part accounts for the formation of particle with volume $v$ and the second part shows the disappearance of particle with volume $v$ due to coagulation with any particle. It is noticed that the coagulation kernel and deposition kernel of particles may change along with time.

GDE is a typical nonlinear partial integro-differential equation. Generally speaking, there are not analytical solutions, and more unfortunately the traditional numerical methods (such as finite volume method and finite difference method) are difficult to take GDE into account when polydispersed particle population as well as nonlinear models of coagulation and deposition kernel. The popular numerical solutions of GDE include moments of method[2], sectional method[3], discrete method[4], discrete-sectional

method[5], and Monte Carlo (MC) method[6−9]. Monte Carlo method has the same physical foundation and physical assumption with GDE[10]. The discrete nature of MC method adopts itself naturally to discrete dynamic events in dispersed systems. The advantages of MC method lie in that[6]: it can gain information about history, trajectory crossing and internal structure of particles; it can take easily account of polydisperse and multi-component population; it can address restructuring, coating, chemical reaction, fractal aggregation and even rapping re-entrainment; its algorithms for solving GDE are easily programmed. So MC method is fully developed and applied. MC method is however time-consuming comparatively. With regard to ordinary "constant-volume" method[7], the total number of real particles decreases continuously when simultaneous coagulation and deposition, so does the total number of simulation particles. Since statistical precision of the MC is inversely proportional to the square root of the total number of simulation particles[7], "constant-volume" method must increase the initial number of simulation particles in order to protect against statistical fatigue, which burdens worse computation cost of the MC method. So "constant-volume" MC method exhibits the contradiction of low computation cost and high computation precision. With respect to "constant-number" method[8], the total number of simulation particles is constant and the simulation volume is continuously adjusted so as to contain the same number of simulation particles. "Constant-number" method can maintain constant statistical accuracy and can simulate growth over arbitrarily long time with a finite number of simulation particles. However, it can hardly be applied in engineering and scientific quantificational analysis, and can hardly take account of boundary conditions, space diffusion of size function and flow-particle dynamics, owing to the expansion or contraction of the simulated domain along with dynamic evolution in order to maintain constant number of simulation particles in the domain.

The paper promoted a new multi-Monte Carlo (MMC) method to solve GDE for simultaneous coagulation and deposition. MMC method has the characteristics of both constant number and constant volume method. It assorts with computation cost and computation precision, and has the latent expansibility of simulation for the space evolution of PSD, boundary conditions and even particle dynamics.

## 1 Description for multi-Monte Carlo method

### *1.1 The introduction of weighted fictitious particle*

A reasonably sized dispersed system contains approximately $10^{10}$ or more particles, however, Monte Carlo code can only examine $10^3 — 10^7$ particles at a time on fast PCs because of the limitation of CPU speed and memory capacity. So in any known Monte Carlo method[6−9] a "subsystem" of the total system is considered. "Subsystem" contains $10^3 — 10^7$ simulation particles, where each of simulation particles represents some real particles, says, each of simulation particles has a value of number-weight. Ones assume the whole system is fully-stirred and spatially isotropic, and the subsystem satisfies the constraint of periodic boundary conditions, i.e., as some particles move out from one

boundary of the subsystem, some identical particles move in from the symmetrical boundary of the subsystem. By those hypotheses the behavior of the subsystem duplicates the system as a whole. The number-weight in constant-volume MC is same for all particles and constant during simulation. As far as constant-number MC is considered, in the cases of breakage and nucleation that result in net generation of real particles, the volume of "subsystem" is contracted in order to maintain the constant number of simulation particles; with mechanisms such as coagulation and deposition which make for net depletion of real particles, this amounts to the expansion of the simulated "subsystem". Factually the number-weight in constant-number MC is the same for all particles but varies with time.

Different from the above approaches, MMC method introduces the concept of "weighted fictitious particle" in order to maintain the volume of computational domain and the number of fictitious particles. Real particles which have same or similar volume can be considered to have the same properties and, hence, the same behaviors. Those real particles can be represented by one or several weighted fictitious particles, where fictitious particles are an indicator of those real particles. So the time evolution of fictitious particles duplicates that of real particles.

Fig. 1 shows a pictorial representation of weighted fictitious particle. As shown in Fig. 1, every class of real particle population is represented by some fictitious particles, and the volume of fictitious particle is just the volume of those represented real particles. Those fictitious particles of the same class have the same value of transform-weight "$w$", however, the different value for the different classes. The value of transform-weight "$w$" of one fictitious particle is equal to the number concentration of those real particles represented by the fictitious particle. Generally speaking, MMC method still maintains high computation precision even though the value of "$w$" reaches to the magnitude of $O(10^3)$ $-O(10^4)$.

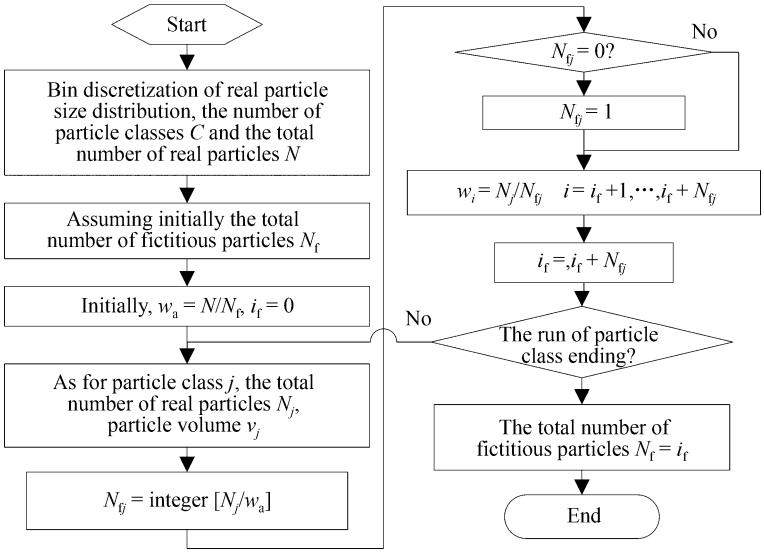


Fig. 1. The steps of numerical selection of transform-weight "$w$" of fictitious particle.

According to the different consequence of the dynamic events, the transform-weight "$w$" and volume of those related fictitious particles are adjusted following each time step; instead, both the computational domain and the number of fictitious particles are maintained during the time evolution of particle population, which will be described detailedly in the following text. In fact, only those fictitious particles which participate in coagulation and deposition event change their "$w$" and volume with time in MMC method.

Although the concept of "weighted fictitious particle" is introduced, the total number of fictitious particles is still a large magnitude as for the whole system with a large number of real particles. Under the condition, MMC method must introduce the concept of "subsystem", alike other MC methods. If the total number of real particles in a whole system is small, or, one fictitious particle is allowed to represent more real particles at the cost of low computation precision, the total number of fictitious particles might be within the scope of computer ability. Under the condition, MMC method can discard the hypothesis of "subsystem" and, then, has the expansibility of simulation for the space evolution of size distribution, boundary conditions and even gas-particle dynamics coupling with two-phase turbulent model[11].

### *1.2 The scheme of MMC method considering simultaneous coagulation and deposition*

MMC method is based on the "time-driven" technique[9], saying, each of fictitious particles is tracked and any possible events are considered within one time step $\Delta t$. It is considered for the "time-driven" technique that any coagulation events and any deposition events are decoupled within a sufficiently small time step $\Delta t$, that is, the evolution is decomposed into two distinct processes: coagulation and deposition. The scheme of MMC method for simultaneous coagulation and deposition is shown in Fig. 2. It is noticeable that coagulation and deposition event does not change immediately the properties and the behaviors of the tracked fictitious particles and the involved fictitious particles within current time step. The changes will occur in the next time step. So the treatment of coagulation and deposition event should be delayed until the end of current time step, which refers to Fig. 2.

The "time-driven" MC method, which needs explicit time discretization, has generally more computation cost comparing with the opposite "event-driven" MC method[7].

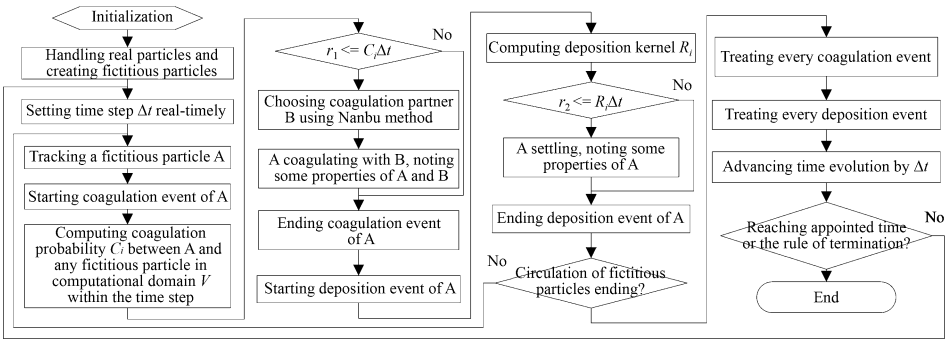


Fig. 2 The scheme of MMC method for simultaneous coagulation and deposition.

However, because numerical solutions of two-phase turbulent model is mostly based on the "time-driven" technique, the "time-driven" MC method can coupled with those numerical solutions more easily and then has stronger expansibility than the "event-driven" MC method.

### *1.3 The setting of time step*

The setting of time step is a key of MMC method. As for the "time-driven" MC technique, in order to track any fictitious particles and any events, the number of coagulation or deposition events of one fictitious particle must be less than or equal to one within time step $\Delta t$.

The total coagulation probability of fictitious particle $i$ is firstly investigated. Ones assume the total number of real particles is $N$ and fictitious particles $N_{\mathrm{f}}$ within computational domain $V$, and one fictitious particle is considered as one group of real particles. Thus the number of coagulation event occurring among $i$th-group (fictitious particle $i$, number concentrations per unit volume $w_i$) and $j$th-group particles (fictitious particle $j$, number concentrations per unit volume $w_j$) per unit time per unit volume is given by

$$NC_{ij} = \beta_{ij} \times w_i \times w_j, \tag{2}$$

where $\beta_{ij}$ is the coagulation kernel for the two particles $i$ and $j$.

For like particles, the number of coagulation event per unit time per unit volume is taken as[12]:

$$NC_{ii} = \beta_{ii} \times w_i \times \left(w_i - 1\right)/2. \tag{3}$$

So $NC_i$, the total number of coagulation event of fictitious particle $i$ per unit time per unit volume, is computed by

$$NC_i = NC_{ii} + \sum_{j=1, j\neq i}^{N_{\mathrm{f}}} NC_{ij} = \beta_{ii} \times w_i \times \frac{\left(w_i - 1\right)}{2} + \sum_{j=1, j\neq i}^{N_{\mathrm{f}}} \left(\beta_{ij} \times w_i \times w_j\right). \tag{4}$$

Because fictitious particle $i$ represents $w_i$ real particles, the average number of coagulation events of each real particle with any real particle per unit time is followed as

$$C_i = \frac{NC_i}{w_i} = \frac{\beta_{ii} \times \left(w_i - 1\right)}{2} + \sum_{j=1, i\neq j}^{N_{\mathrm{f}}} \left(\beta_{ij} \times w_j\right). \tag{5}$$

Since fictitious particle $i$ is an indicator of those represented real particles, the average number of coagulation events of fictitious particle $i$ (including coagulation event within unlike particle and within like particle) per unit time is equal to $C_i$, which is also called as the total coagulation probability of fictitious particle $i$.

Then, the coagulation time scale of fictitious particle $i$, within which only one coagulation event occurs, is expressed by

$$t_{i,\mathrm{coag}} = 1/C_i. \tag{6}$$

According to the definition of deposition kernel, the deposition time scale, within which only one deposition event occurs, is as follows:

$$t_{i,\mathrm{depo}} = 1/R_i, \tag{7}$$

where $R_i$ is the deposition kernel of fictitious particle $i$.

In MMC method, time step $\Delta t$ should be less than or equal to the minimum coagulation time scale ($\min(t_{i,\mathrm{coag}})$), and also be less than or equal to the minimum deposition time scale ($\min(t_{i,\mathrm{depo}})$), that is:

$$\Delta t \leqslant \min\left\{1\Big/\max_{i=1,\ldots,N_\mathrm{f}}(C_i), 1\Big/\max_{i=1,\ldots,N_\mathrm{f}}(R_i)\right\}. \tag{8}$$

In order to increase the number of Monte Carlo loop, time step is usually defined as

$$\Delta t = \alpha \times \min\left\{1\Big/\max_{i=1,\ldots,N_\mathrm{f}}(C_i), 1\Big/\max_{i=1,\ldots,N_\mathrm{f}}(R_i)\right\}, \tag{9}$$

where the multiplicative constant, $\alpha$, has the value of 0.01 or less.

Particle size distribution evolves with the occurrence of dynamic events, which will change the minimum coagulation and deposition time scale. So time step must be adjusted real-time-wise, not just as a fixed value, which refers to Fig. 2.

### *1.4 The treatment of particle coagulation event*

The process includes three steps: the judgment of the occurrence of coagulation event, the choice of coagulation partner, and the consequently treatment of coagulation event.

(i) The judgment of the occurrence of coagulation event. It is deemed that the process of dynamic evolution due to particle coagulation is a Markov process[7]. As far as a standard procedure for a Markov process is considered, the probability of fictitious particle $i$ interacting with any fictitious particle within $\Delta t$, $Pr_{\mathrm{coag},i}(\Delta t)$, is represented by an exponential function[9]: $Pr_{\mathrm{coag},i}(\Delta t)=1-\exp(-C_i\Delta t) \approx C_i\Delta t$. In the paper the Nanbu method[13] is used to judge the occurrence of coagulation event and to search coagulation partner, which is shown in Fig. 3. Firstly a random number $r_1$ from a uniform distribution in the interval [0,1] is generated. Then a coagulation event is calculated when the random number $r_1$ becomes smaller than the coagulation probability within time step $\Delta t$, i.e. if $r_1 \leqslant C_i\Delta t$ fictitious particle $i$ will be ascertained to coagulate with its partner.

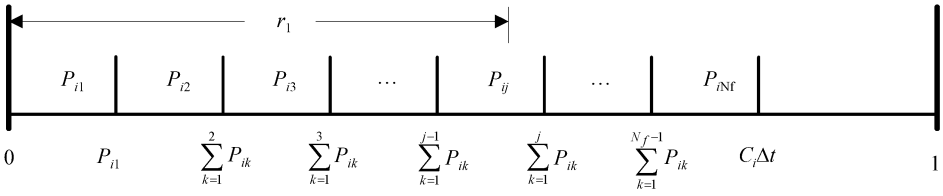


Fig. 3. The schematic diagram of the Nanbu method.

(ii) The choice of coagulation partner. If fictitious particle $i$ will be related to one coagulation event, the Nanbu method[13] is still used to search its coagulation partner. Firstly, the probability of fictitious particle $i$ coagulating with any other fictitious particle $j$ is expressed as $P_{ij}=w_j\times\beta_{ij}\times\Delta t$, and the coagulation probability among the same fictitious particle $i$ is expressed as $P_{ii}=[(w_i-1)/2]\times\beta_{ii}\times\Delta t$. Obviously,

$$C_i\Delta t = \left[\sum_{j=1,i\neq j}^{N_\mathrm{f}}\left(\beta_{ij}\times w_j\right)+\frac{\beta_{ii}\times\left(w_i-1\right)}{2}\right]\Delta t = \sum_{k=1}^{N_\mathrm{f}} P_{ik}. \tag{10}$$

Secondly the random number $r_1$ is still used for the determination of coagulation partner. If the following relation is satisfied, it is considered that the tracked particle $i$ coagulate with fictitious particle $j$:

$$\sum_{k=1}^{j-1} P_{ik} \leqslant r_1 \leqslant \sum_{k=1}^{j} P_{ik}, \qquad j \in [1, N_f]. \tag{11}$$

(iii) The consequently treatment of coagulation event. One of important points of MMC method is to maintain the total number of fictitious particles during the process of coagulation in order to protect against statistical fatigue. The consequently treatment of coagulation event concentrates the idea of constant number of fictitious particles and constant computational domain in MMC method. MMC method adjusts the transform-weight "$w$" of the "coagulated" fictitious particle to realize both "constant-number" MC and "constant-volume" MC.

When the tracked fictitious particle $i$ (its transform-weight $w_i$, its volume $v_i$) coagulates with coagulation partner $j$ (its transform-weight $w_j$, its volume $v_j$), it means, some real particles represented by $i$ coagulate with those real particles represented by $j$. Coagulation between real particle A with volume $v_A$ and real particle B with volume $v_B$ results in a new real particle with volume ($v_A$+$v_B$). Regardless of the factual progress of the coagulation event between the two fictitious particles $i$ and $j$, as a result, those "old" real particles represented by $i$ and $j$ are replaced by the "new" real particles, and the total number of those "new" real particles is the half of the total number of those "old" real particle, saying, ($w_i$+$w_j$)/2. In order to maintain the total number of fictitious particles, both the tracked fictitious particle and its partner are conserved; instead, their transform-weight and their volume are adjusted to externalize the consequence of coagulation event. It is considered that the transform-weights of both the tracked fictitious particle $i$ and its partner $j$ are halved respectively, and the volumes of both $i$ and $j$ are changed as "$v_i$+$v_j$". Because every fictitious particle will be judged the occurrence of coagulation event, a coagulation event of particle pairs is double-counted within each time step $\Delta t$. If the tracked fictitious particle is $i$, only some properties of the current tracked fictitious particle are changed and there are no any changes in its partner, saying,

$$\begin{aligned} &(w_i)_{new} = (w_i)_{old}/2; \quad (v_i)_{new} = (v_i)_{old} + (v_j)_{old}; \\ &(w_j)_{new} = (w_j)_{old}; \qquad (v_j)_{new} = (v_j)_{old}. \end{aligned} \tag{12}$$

When the tracked particle is assigned to fictitious particle $j$ in turn, its coagulation partner will be fictitious particle $i$ in theory and the same measures are taken:

$$\begin{aligned} &(w_j)_{new} = (w_j)_{old}/2; \quad (v_j)_{new} = (v_i)_{old} + (v_j)_{old}; \\ &(w_i)_{new} = (w_i)_{old}; \qquad (v_i)_{new} = (v_i)_{old}. \end{aligned} \tag{13}$$

The measures not only conserve the number of fictitious particles, but also maintain the computation domain.

Strictly speaking, MMC method conserve perfectly the total volume of particle population before and after one coagulation event only when $w_i$=$w_j$ or $v_i$=$v_j$, saying,

$$
\begin{aligned}
&(v_i)_{\text{new}} \times (w_i)_{\text{new}} + (v_j)_{\text{new}} \times (w_j)_{\text{new}} \\
&= \left[(v_i)_{\text{old}} + (v_j)_{\text{old}}\right] \times (w_i)_{\text{old}} \Big/ 2 + \left[(v_i)_{\text{old}} + (v_j)_{\text{old}}\right] \times (w_j)_{\text{old}} \Big/ 2 \\
&= \left[(v_i)_{\text{old}} + (v_j)_{\text{old}}\right] \times \left[(w_i)_{\text{old}} + (w_j)_{\text{old}}\right] \Big/ 2 \\
&= \begin{cases} (v_i)_{\text{old}} \times \left[(w_i)_{\text{old}} + (w_j)_{\text{old}}\right] = (v_i)_{\text{old}} \times (w_i)_{\text{old}} + (v_j)_{\text{old}} \times (w_j)_{\text{old}} & \text{if } (v_i)_{\text{old}} = (v_j)_{\text{old}}, \\ \left[(v_i)_{\text{old}} + (v_j)_{\text{old}}\right] \times (w_i)_{\text{old}} = (v_i)_{\text{old}} \times (w_i)_{\text{old}} + (v_j)_{\text{old}} \times (w_j)_{\text{old}} & \text{if } (w_i)_{\text{old}} = (w_j)_{\text{old}}. \end{cases}
\end{aligned}
$$

Obviously, the transform-weight and volume of the tracked fictitious particle may be not always equal to those of the partner during the process of dynamic evolution, so MMC method for coagulation event exhibits some numerical error, which is the inevitable cost for the purpose of the forcibly maintenance of constant number and constant volume. If the difference between transform-weight and volume of the tracked fictitious particle and those of the partner is small, the propagation and delivery of numerical error can be restrained by means of the repetitious MC loops and the averaging operation.

### *1.5 The treatment of particle deposition event*

The process involves the two-step procedure: the judgment of the occurrence of deposition event, and the consequently treatment of deposition event.

(i) The judgment of the occurrence of deposition event. Similarly, the occurrence of deposition event is considered as a Markov process, and $Pr_{\text{depo},i}(\Delta t)=1-\exp(-R_i\Delta t)\approx R_i\Delta t$, where $Pr_{\text{depo},i}(\Delta t)$ is the deposition probability of fictitious particle $i$ within $\Delta t$. So, as for a standard Monte Carlo simulation, if

$$r_2 \leqslant R_i \Delta t \tag{14}$$

is satisfied, deposition event of fictitious particle $i$ occurs; otherwise fictitious particle $i$ does not deposit, and will be tracked within next time step. Here $r_2$ is the random number from a uniform distribution in the interval [0,1].

(ii) The consequently treatment of deposition event. The direct consequence of deposition event is net depletion of real particles. If any measures are not taken, the total number of fictitious particles will continue to decrease. If the tracked fictitious particle A is ascertained to deposit, the following measures are taken. Firstly a random number $r_3$ from a uniform distribution in the interval [0,1] is used to select fictitious particle B from fictitious particle array, the serial number of which is integer[$r_3\times N_f$], where integer[] indicates the integer part of its argument. The progress of random choice will be repeated until the selected fictitious particle B is not the tracked fictitious particle A. Secondly the randomly selected fictitious particle B (transform-weight $w_B$, volume $v_B$) is divided equally into two fictitious particles——C (transform-weight $w_C$, volume $v_C$) and D (transform-weight $w_D$, volume $v_D$), where $w_B=2w_C=2w_D$, and $v_B=v_C=v_D$. Fictitious particle C is placed in the position of the tracked fictitious particle A, and the randomly selected fictitious particle B is replaced by fictitious particle D. Factually those measures mean that the leaving null of the deposited fictitious particle A is filled with the half of the randomly selected fictitious particle B. Those measures not only conserve both the number

of fictitious particles and computation domain but also accord with the reality of deposition event.

## 2 Validation for multi-Monte Carlo method

As for some special coagulation kernel, deposition kernel and initial particle size distribution, analytical solutions of the time evolution of PSD can be obtained. Those analytical solutions are used to validate MMC method for simultaneous coagulation and deposition.

The initial particle size distribution is represented by an exponential function, i.e.

$$n_{\mathrm{p}}(v,0)=\left(N_0/v_{\mathrm{g0}}\right)e^{-v/v_{\mathrm{g0}}}, \tag{15}$$

where $N_0$ is the initial total number concentration of real particles, and $v_{\mathrm{g0}}$ is the initial mean volume. In the paper, we choose $N_0=10^6$ cm$^{-3}$ and $v_{\mathrm{g0}}=0.029$ μm$^3$. Two special cases in which analytical solutions exist are as follows:

Case 1: constant coagulation kernel and constant deposition kernel, $\beta(u,v)=K_{\mathrm{C}}$, $R(v)=K_{\mathrm{D}}$;

Case 2: constant coagulation kernel and gravitational deposition kernel, $\beta(u,v)=K_{\mathrm{C}}$, $R(v)=K_{\mathrm{D}}v^{2/3}$;

where both $K_{\mathrm{C}}$ and $K_{\mathrm{D}}$ are constant number. Constant coagulation kernel reproduces the integral value behavior in the Brownian coagulation. Constant deposition kernel describes leakage through a hole or crack. Gravitational deposition kernel accounts for sediment due to gravitation. Computational conditions of two special cases are listed in Table 1. Analytical solutions of two cases refer to ref. [14].

Table 1 Computational condition of two special cases

| Case | $K_{\mathrm{C}}$ (cm$^3$·s$^{-1}$) | $K_{\mathrm{B}}$ | $\tau_{\mathrm{coag}}$ (s) | $\tau_{\mathrm{depo}}$ (s) | $\tau_{\mathrm{coag}}$ (s) | $\tau_{\mathrm{depo}}$ (s) |
|---|---|---|---|---|---|---|
| Case 1 | $6.405\times10^{-10}$ | $3.2025\times10^{-4}$ s$^{-1}$ | $2/(K_0N_0)$ | $1/K_{\mathrm{B}}$ | ≈3122.6 | ≈3122.6 |
| Case 2 | $6.405\times10^{-10}$ | $3.3928\times10^{-3}$ s$^{-1}$·μm$^{-2}$ | $2/(K_0N_0)$ | $1/(K_{\mathrm{B}}v_{\mathrm{g0}}^{2/3})$ | ≈3122.6 | ≈3122.6 |

In Table 1, $\tau_{\mathrm{coag}}$ is the characteristic coagulation time scale, and $\tau_{\mathrm{depo}}$ is the characteristic deposition time scale, and Ω ($=\tau_{\mathrm{coag}}/\tau_{\mathrm{depo}}$) is the ratio between the two characteristic time scales. In order to have a real coupling between two processes, Ω is chosen to be approximatively equal to 1 in the two cases.

The initial total number of fictitious particles is set as 3000 for the two cases, and the real number of fictitious particles is 3081, that is, one fictitious particle represents about 325 real particles on the average. The length of time evolution is $\tau_{\mathrm{coag}}/2$, i.e., 1561.3 s. In order to initialize PSD and collect statistical properties of PSD at some appointed time-point, polydisperse particle population is divided into 200 classes between the largest and smallest particle volume in the simulation. Nevertheless, MMC method needs no information about bin discretization during simulating, which avoids numerical bias and numerical diffusion. In addition, the first-order moment of PSD is defined as $M_1(t)=\int n_{\mathrm{p}}(v,t)v\mathrm{d}v$, which denotes the sum of particle volume.

In Fig. 4 and Fig. 5, we present a comparison between MMC solutions and analytical

solutions for Case 1 and Case 2, respectively. Here Fig. 4(a) and Fig. 5(a) show the time evolution of total number concentration, geometric mean volume and the first-order moment for Case 1 and Case 2; the information about PSD at $t$=750 and 1500 s for Case 1 and Case 2 is exposed in Fig. 4(b) and Fig. 5(b), respectively. As for Case 1, the total number of real particles at $t$=750 and 1500 s is only 65.43% and 44.57% of initial total number, respectively. As for Case 2, the total number of real particles at $t$=750 and 1500 s is only 66.62% and 47.10% of initial total number, respectively. Although the total number of real particles continues to decrease, the agreement between MMC solution and analytical solution is good since the total number of fictitious particles is always kept 3081. Those simulation results show MMC method has very high and stable computation precision.

It is remarkable that computation cost of MMC method is very low. As for Case 1 and Case 2, CPU time is 1564.03 and 350.55 s, respectively. Computational environment is as follows: Athlon Xp2500+, 512M. Low computation cost comes mainly from that the number of fictitious particles is far less than that of real particles. The most expensive part of MMC method is the computation of time step, so even the very complicated coagulation kernel and deposition kernel in engineering and nature will not contribute

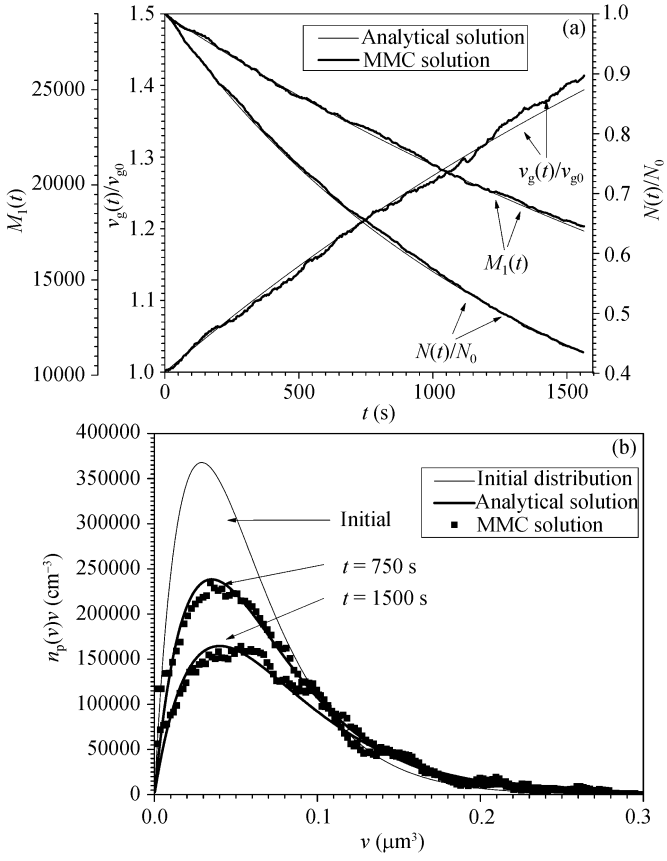


Fig. 4. Time evolution of PSD with constant coagulation kernel and constant deposition kernel.

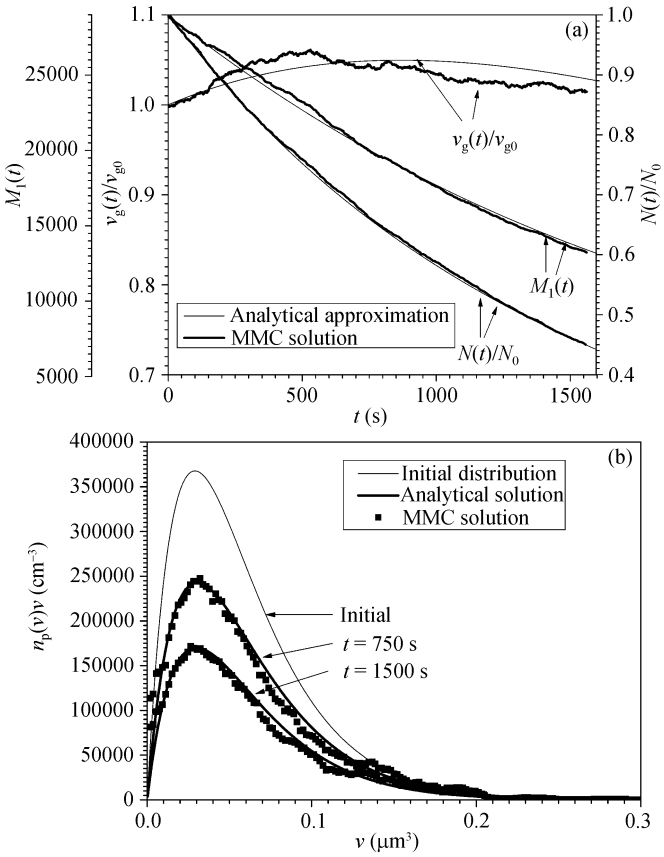


Fig. 5. Time evolution of PSD with constant coagulation kernel and gravitational deposition kernel.

markedly to computation cost.

## 3 Discussion

### *3.1 The analysis of numerical errors of MMC method for simultaneous coagulation and deposition*

Since it is impossible that every "sub-process" of MMC method is separated and realized numerically, it is difficult to analyze quantitatively and accurately the influence of every "sub-process" on computation precision of MMC method. However it is possible to analyze qualitatively the source of numerical errors of MMC method for simultaneous coagulation and deposition. MMC method is based on time-driven technique, which assumes dynamic events in dispersed systems are decoupled each other within a sufficiently small time step $\Delta t$. In fact, within the time-step $\Delta t$, there are several coagulation and/or deposition events; the same particle may participate in one coagulation event and one deposition event within the same interval; the same particle may be involved with several coagulation events, acting as the main particle or coagulation partner. Obviously those dynamic events may depend on each other and are NOT uncoupled absolutely each

other, so the "time-driven" MC method shows so-called "uncoupling error". The introduction of multiplicative constant $\alpha$ is precisely for the purpose of reducing the "uncoupling error", seeing eq. (9). The smaller multiplicative constant, the fewer coagulation events or/and deposition events take place within one time-step and, then, the smaller "uncoupling error" but on the expense of computation time. The other source of numerical errors come form the consequently treatment of dynamic events. When MMC method takes account of particle coagulation, the transform-weight of two "new" fictitious particles is halved and the volume of two "new" fictitious particles is changed as the sum of the two "old" fictitious particles, respectively. Those measures don't accord perfectly with the reality of coagulation event; however maintain both computation domain and the number of fictitious particles. We might as well call the error as "constant volume and number error". The repetitious MC loops and the averaging operation will restrain greatly the propagation and delivery of numerical error. As far as deposition event is considered, the disturbance of transform-weight of fictitious particles due to the forcible maintenance of constant volume and constant number contributes to "constant volume and number error". The third numerical error originates from random process. MMC method adopts many random numbers and random processes, for example, when judging the occurrence of coagulation event and deposition event. Those random procedures exhibit some inevitable error, which is called as random error. Lastly, the statistical error is inevitably demonstrated by all MC methods. The statistical error is inverse proportional to the square root of the sample size. If the number of simulation particles reaches the magnitude of $O(10^3)$ or more, statistical error shows a weak effect on numerical results of the macro-variables or the whole variables such as the total number concentration, geometric mean volume and the total volume concentration; however the total number of simulation particles plays an important role in the instantaneous variables or the detailed variables such as PSD at the appointed time-point when polydisperse population with a wide range. In fact, only when the number of simulation particles in every particle class reaches to the magnitude of $O(10^2)$—$O(10^3)$ does MC method protect well against the statistical fatigue of properties of PSD. To increase the total number of simulation particles will decrease the statistical error.

Case 1 is illustrated the relative error of MMC method for simultaneous coagulation and deposition. The radio of initial number concentration and real-time number concentration during evolution, $N_0/N(t)$, is chosen as the reference variable. The relative error of $N_0/N(t)$ is computed via the relation:

$$\delta = \left| \frac{\left[N_0/N(t)\right]_{\mathrm{MMC}} - \left[N_0/N(t)\right]_{\mathrm{theory}}}{\left[N_0/N(t)\right]_{\mathrm{theory}}} \right| = \left| \frac{\left[N(t)\right]_{\mathrm{theory}}}{\left[N(t)\right]_{\mathrm{MMC}}} - 1 \right|. \tag{16}$$

As for direct simulation Monte Carlo promoted by Liffman (noted by Liffman's DSMC), Liffman[9] analyzed the relative error of $N_0/N(t)$, which satisfies approximatively the condition $\delta < 2/\sqrt{N_{\mathrm{c}}(t)}$, where $N_{\mathrm{c}}(t)$ is the real-time total number concentration of simulation particles in "subsystem". Because Liffman's DSMC adopts the procedure of stepwise "doubling" subsystem, $N_{\mathrm{c}}(t)$ always varies the value between $N_{\mathrm{c}0}/2$ and

$N_{c0}$, where $N_{c0}$ is the initial number of simulation particles. So the upper limitation of $\delta$ satisfies the relation: $\delta < 2\sqrt{2}/\sqrt{N_{c0}}$ . With respect to constant number method, Smith *et al*.[15] formulated an analysis of the relative error and scaled approximatively the quantity as the relation: $\delta = \left\{\ln\left[N_0/N(t)\right]\right\}^{0.8}/\sqrt{2N_c}$, where $N_c$ is a constant in the constant number method. Fig. 6 shows the relative error of MMC method with the time evolution. Obviously, the simulation result of MMC method, the scattering of which is basically within 1.5%, shows the higher precision than that of Liffman's DSMC on the whole. The computation precision of MMC method shows the same magnitude with that of constant number method. Similar with the fitting of constant number method, the relative error of MMC method is represented approximatively by the relation:

$$\delta = \left\{\ln\left[N_0/N(t)\right]\right\}^{0.8}/\sqrt{2N_c}\,.$$

Generally speaking, the computation precision of MMC method will be worse and worse along with the advancement of dynamic evolution in dispersed systems. When the relative error of MMC method is greaten than 10%, it's considered that MMC method doesn't describe accurately the dynamic evolution in dispersed systems any longer. So the criterion of convergence or termination in MMC method is as follows:

$$\delta = \left\{\ln\left[N_0/N(t)\right]\right\}^{0.8}/\sqrt{2N_c} < 10\%.$$

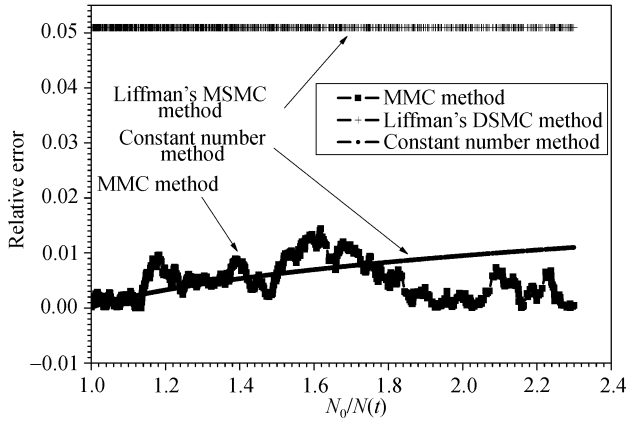


Fig. 6. The relative error of $N_0/N(t)$.

### *3.2 The expansibility of MMC method for GDE*

Those complicated mechanisms of coagulation and deposition progress are encapsulated in the coagulation kernel and the deposition kernel by virtue of general dynamic equation. As for any system where particle coagulation, deposition, breakage, nucleation, condensation/evaporation, and chemical reaction occur, GDE describes the time and space evolution of PSD. GDE has very solid physical foundation and very simple physical assumption, which is able to describe nanoparticle material or galaxies, aerosol or

particulate matter (PM), and so on. Once GDE is well solved numerically by MMC method, ones can focus one's mind on the complicated "kernel" model in various domains and conditions. GDE itself, as given in the paper, is the Eulerian style, however, MMC method is Langrangian approach. It should be noted that GDE and MMC method is integrated perfectly into one framework because they have the same physical foundation and physical assumption. Furthermore, In comparison with those Eulerian methods such as moments of method, sectional method, discrete method, or discrete-sectional method, MMC method has the wider scope of application and stronger expansibility since it obtains the information about trajectory crossing and history effect of particles.

Nowadays any numerical methods for GDE focus on the solution of GDE describing the time evolution of PSD, which factually describes the spatially zero-dimension system. The computational ability of MMC method described in the paper is also zero-dimension. The information about the time evolution of PSD is sufficient for some application in engineering and science. However, the information about the space evolution of PSD is very important and necessary for some cases. For example, when researching the principle of the particulate matter enrichment with heavy metals by coagulation, condensation and nucleation progress in pulverized-coal flames[16], the spatial information about PSD will help to design the reasonable capturing strategy and control measures. However, it is difficult for all of numerical methods for GDE to describe the space evolution of PSD. It's more disappointed that those MC methods don't even have the expansibility considering space diffusion of PSD because of the introduction of subsystem. On the contrary, MMC method, which introduces the concept of weighted fictitious particle, not only has low computation cost and stable computation precision, but also has stronger expansibility to simulate the time and space evolution of PSD within the whole system if the concept of subsystem is abandoned at some cases. MMC method for space diffusion will be developed in the next stage.

The strong expansibility of MMC method also includes the conveniently consideration of other dynamic events such as condensation/evaporation, nucleation and breakage etc. Even MMC method has ability to couple with the numerical solution for the model of two-phase turbulent flow, which will describe not only the time and space evolution of PSD but also the fields of particle phase and flow phase etc. MMC method can also be applied to consider GDE for multi-component, more-dimensional and polydisperse population. Those works will also be developed in the next stage.

## 4 Conclusion

Multi-Monte Carlo (MMC) method for general dynamic equation (GDE) introduces the concept of weighted fictitious particle and is based on the "time-driven" technique. MMC method maintains forcibly the total number of fictitious particles and computational domain by means of the adjustment of the transform-weight and volume of the involved fictitious particles. Similar with constant-volume method, MMC method adapts well to engineering application and general scientific quantitative analysis; similar with constant-number method, MMC method maintains high and stable statistical precision.

The introduction of weighted fictitious particle makes it possible to abandon the concept of "subsystem", where MMC method has stronger expansibility to consider the space evolution of PSD, boundary conditions and gas-particle dynamics.

The computation precision of MMC method is constrained by the "uncoupling error", the "constant volume and number error", the random error and the statistical error. The relative error of MMC method is approximatively formulated as $\delta = \left\{\ln\left[N_0/N(t)\right]\right\}^{0.8}\Big/\sqrt{2N_c}$, which shows that MMC method has similar precision with constant number method but higher precision than Liffman's DSMC.

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