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MULTI-MONTE-CARLO METHOD FOR GENERAL DYNAMIC EQUATION CONSIDERING PARTICLE COAGULATION *

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Abstract: Monte-Carlo (MC) method is widely adopted to take into account general dynamic equation (GDE) for particle coagulation, however popular MC method has high computation cost and statistical fatigue. A new Multi-Monte-Carlo (MMC) method, which has characteristics of time-driven MC method, constant number method and constant volume method, was promoted to solve GDE for coagulation. Firstly MMC method was described in details, including the introduction of weighted fictitious particle, the scheme of MMC method, the setting of time step, the judgment of the occurrence of coagulation event, the choice of coagulation partner and the consequential treatment of coagulation event. Secondly MMC method was validated by five special coagulation cases in which analytical solutions exist. The good agreement between the simulation results of MMC method and analytical solutions shows MMC method conserves high computation precision and has low computation cost. Lastly the different influence of different kinds of coagulation kernel on the process of coagulation was analyzed: constant coagulation kernel and Brownian coagulation kernel in continuum regime affect small particles much more than linear and quadratic coagulation kernel, whereas affect big particles much less than linear and quadratic coagulation kernel.

Key words: numerical solution; population balance equation; fictitious particle; particle size distribution; constant kernel; computation cost; computation precision

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Introduction

Particulate Matter (PM10, aerodynamic diameter less than 10 μm) has been widely investigated. Some kinds of combustion including coal combustion, gasoline/diesel oil combustion of vehicle, municipal solid waste combustion, *etc*, are one of the main sources

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of PM10[1]. Particle coagulation and breakage during combustion are the main mechanisms of the formation of PM. Furthermore the measures, which boost PM's coagulation during combustion, flight, emission, are the main PM's control strategies. In addition, solid particle (or droplet) coagulation (or aggregation) is also an important mechanism in both nature and engineering fields, including the formation and evolvement of air aerosols and emulsion droplets, the manufacture of nanoparticle agglomerates. Particle size distribution (PSD) due to particle coagulation is of fundamental interest and a key issue during the process of particle dynamic evolution[2]. PSD along with time is described by general dynamic equation (GDE), which is also called Population Balance Equation (PBE)[3]. It is focused on the algorithm solving GDE for particle coagulation. GDE for coagulation is as follows:

$$\frac{\mathrm{d}n_{\mathrm{p}}(v,t)}{\mathrm{d}t} = \frac{1}{2}\int_0^v \beta(v-u,u)n_{\mathrm{p}}(v-u,t)n_{\mathrm{p}}(u,t)\,\mathrm{d}u - n_{\mathrm{p}}(v,t)\int_0^\infty \beta(v,u)n_{\mathrm{p}}(u,t)\,\mathrm{d}u, \tag{1}$$

where $n_{\mathrm{p}}(v,t)$ is the number concentration of particles of volume v at time t ($(\mathrm{N/m^3})/\mathrm{m^3}$, where N denotes the number of particles); $\beta(u,v)$ is coagulation kernel for two particles of volume v and u ($\mathrm{m^3}/N$)/s, representing the probability of a binary coagulation event per unit volume per unit time. In the paper, particle collision leads to particle coagulation directly and absolutely, and collision and coagulation are used indiscriminately. Any coagulation is binary coagulation. The term on the left-hand side of Eq. (1) describes the change in number concentration of particle of volume v with time; and the first term on the right-hand side is growth term, accounting for the formation of particle of volume v due to coagulation between particle of volume $(v-u)$ and particle of volume u, and the second term is death term, showing the disappearance of particle of volume v due to coagulation with any particle.

Nowadays the most popular numerical methods of GDE are moments of method, Monte-Carlo (MC) method[3-8], sectional method, discrete model, discrete-sectional method, finite element method, *etc.* Those methods have both advantages and disadvantages. MC method can be divided into two classes according to the approach of time-step setting: one refers to "time-driven" MC method [3,4], which calculates time-step before the dynamic event; The other MC method is called "event-driven" MC method[5-8], in which the time between events is set on the basis of the known event probability. On the other hand, MC method can also be classified into two general classes according to whether or not the number of simulation particles and computational domain are changed along with the evolvement of time. The first approach refers to "constant-volume method"[3-5], which tracks a constant computational domain and thus grows or shrinks the number of simulation particle, N, in direct proportion to the number concentration of the physical system; since computation precision of MC is inversely proportional to the square root of N, constant-volume method can not maintain constant statistical accuracy. The second class is "constant-number MC"[6-8], which is based on event-driven MC, and in which the simulation volume is continuously contracted or expanded so as to contain the same number of simulation particles; constant-number method maintains constant statistical accuracy, however the

contraction or expansion of computational domain results in bad applicability in engineering computation.

Monte-Carlo code can only examine $10^3 - 10^7$ particles at a time because of the limit of computation capacity of PCs, however a reasonably sized system of gas contains approximately 10^{10} or even more particles. So in some known Monte Carlo method the concept of a "subsystem" of the total system is introduced and it is assumed the whole system is fully-stirred and spatially isotropic. The behavior of the subsystem, which satisfies the constraint of periodic boundary conditions and contains $10^3 - 10^7$ simulation particles, duplicates the system as a whole. "Subsystem" hypothesis makes it difficult for those MC methods to simulate the dynamic evolution of the whole system and to consider further space dispersion of PSD.

In order to overcome those drawbacks of those known MC methods (especially such as the contradiction of computation cost and computation precision, and the constraint of the introduction of the concept of "subsystem"), it is tried to perform a new Multi-Monte-Carlo method (MMC) to consider GDE for coagulation.

1 Description for Multi-Monte-Carlo Method

1.1 Introduction of concept of "weighted fictitious particle"

The MC method discards the concept of "subsystem" and does not describe directly the evolution of real particles. Instead the concept of "weighted fictitious particle" is introduced. It is believed that those real particles, who have the same or similar volume, have the same properties and hence the same behaviors. Those real particles are represented by one or several fictitious particles, depending on the number of those real particles. It is considered the fictitious particles is an "indicator" of those real particles, says, the evolution process of fictitious particles denotes that of real particles within computational domain. The approaches of transforming real particles into fictitious particles are as follows: real particles within computational domain are classified; and then according to local particle size distribution, some proper fictitious particles are assigned to represent real particles of any particle class; One fictitious particle, of which serial number is i, is endowed with a transform-weight "w_i", the physical meaning of which is the number of local real particles which are represented by the fictitious particle i. Since the number of fictitious particles is far less than the number of real particles, the MC method, which tracks those fictitious particles, will have much less computation cost than those ordinary MC methods which track those real particles, especially when the number of real particles is large within computational domain. Furthermore the introduction of the concept of "weighted fictitious particle" makes it possible for the MC method to simulate the space evolution of particle size distribution of the whole system.

1.2 Scheme of MMC method

"Time-driven" MC technique is developed to consider possible binary coagulation event within time step Δt, which is set real-timely. Although the total number of real particles cuts down continuously along with the occurrence of coagulation event, the total number fictitious particles is constant by the means of adapting the transform-weight and volume of

the related fictitious particle. In addition, the volume of computational domain is conserved. The MC method is named with multi-Monte-Carlo (MMC) method since the MC method has the characteristics of time-driven MC method, constant number method and constant volume method at the same time. The scheme of MMC method for particle coagulation is showed in Fig. 1.

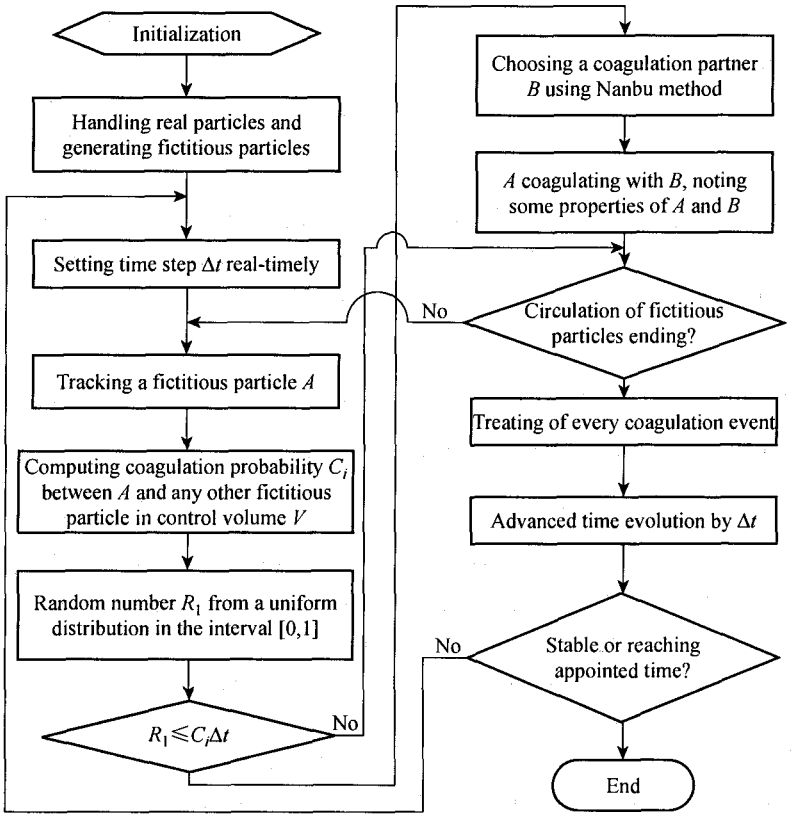

Fig. 1 Schematic diagram of multi-Monte-Carlo method

It is noticeable that coagulation event does not influence immediately the properties and the behaviors of the tracked fictitious particles and the related fictitious particles within current time step. The influence will occur in the next time step. So treating of particle coagulation should be delayed until the end of the current time step.

1.3 Setting of time step

MMC method is based on time-driven technique, and the rule of the setting of time step is: within time step Δt which is adjusted real-timely, the number of coagulation event of any fictitious particle must be less than or equal to one. The rule can assure that every coagulation event can be counted. Within computational domain V, one assumes the number of real particles is N and the number of fictitious particles N_f. Then C_i, the total coagulation probability per unit time of fictitious particle i with any fictitious particle in V, is as follows:

$$C_i = \sum_{j=1, j\neq i}^{N_f} [\beta_{ij} \times w_j] \approx \sum_{j=1}^{N_f} [\beta_{ij} \times w_j]. \tag{2}$$

So the mean coagulation time scale for fictitious particle i interacting with any fictitious particle is: $t_{i,c} = 1/C_i$. Then, time step Δt should be less than or equal to the minimum value of the coagulation time scale of any fictitious particle, *i. e.*, $\Delta t \leqslant \min(t_{i,c}) = 1/\max(C_i)$. Generally, in order to increase the number of Monte-Carlo loop, time step is usually defined as $\Delta t = \alpha/\max(C_i), \alpha \leqslant 0.01$.

1.4 Judgment of occurrence of coagulation event

The Nanbu method[9] is used to judge the occurrence of coagulation event and to search coagulation partner. A random number R_1 from a uniform distribution in the interval $[0,1]$ is generated, and then a coagulation event is calculated when the random number R_1 becomes smaller than the coagulation probability within Δt, *i. e.*, $R_1 \leqslant C_i \Delta t$.

1.5 Choice of coagulation partner

Once coagulation event occurs, the next issue is choosing coagulation partner of the tracked fictitious particle i. Firstly, the probability of fictitious particle i coagulating with any fictitious particles j is expressed as $P_{ij} = w_j \times \beta_{ij} \times \Delta t$; Secondly the random number R_1 is still used for the determination of true coagulation partner. If the relation

$$\sum_{k=1}^{j-1} P_{ik} \leqslant R_1 \leqslant \sum_{k=1}^{j} P_{ik}, \qquad j \in [1, N_f] \tag{3}$$

is satisfied, it is decided that the tracked particle i coagulate with fictitious particle j.

1.6 Consequential treatment of coagulation event

If fictitious particle i coagulates with its coagulation partner j, MMC method adjusts their transform-weight (w_i and w_j, respectively) and volume (v_i and v_j, respectively) to realize the consequence of coagulation event and to conserve the total number of fictitious particles and computational domain. Nanbu[10] considered the number of coagulation event between two weighted particles is the minimum between transform-weight w_i and w_j, that is, $\min(w_i, w_j)$, and Ref. [11] treated the consequence of coagulation event of weighted fictitious particles according to the above theory. However there exists some numerical bias when the evolution time excesses greatly the characteristics coagulation time. In fact, because fictitious particle is an indicator of some real particles, one can neglect the factual progress of the coagulation event. It is considered the consequence of the coagulation event is the appearance of real particle with the number of $(w_i + w_j)/2$ and the volume of $(v_i + v_j)$. Because during each time step every fictitious particle needs to judge the occurrence of coagulation event and then choose its partner, a coagulation event of particle pairs is double counted. If the tracked fictitious particle is i, only some properties of i is changed to satisfy the consequence of the coagulation event and no changes in its partner j:

$$w_{i,\text{new}} = w_i/2; \quad v_{i,\text{new}} = v_i + v_j. \tag{4}$$

When the tracked fictitious particle is assigned to j in turn, the same coagulation event will be checked and its coagulation partner will be i in theory. According to Eq. (4), only some properties of i are changed to satisfy the consequence of the coagulation event. The measures not only can conserve the number of fictitious particle, but also accord with the reality of coagulation event, no matter how many coagulation events there are and how long evolution time is.

2 Computational Cases

The paper uses some special cases in which analytical solutions exist to validate MMC method.

2.1 Case 1, constant coagulation kernel of monodisperse particles, $\beta(u,v) = A$

Here A is a constant. Constant coagulation kernel indicates that the rate of coagulation is independent of particle volume, *i. e.*, particle size distribution. The constant kernel reproduces the integral value behavior in the Brownian coagulation. Analytical solution of the case can refer to Ref. [5].

2.2 Case 2, linear coagulation kernel of monodisperse particles, $\beta(u,v) = A(u + v)$

Here u and v is respectively the volume of two coagulation particles. The linear coagulation kernel is frequently used to approximate turbulent gravitational coagulation, as the kernels have about the same degree of growth with increasing particle size. Analytical solution of the case can refer to Ref. [5].

2.3 Case 3, quadratic constant kernel of monodisperse particles, $\beta(u,v) = Auv$

The quadratic coagulation kernel leads to critical phenomena ("gelation") during coagulation. In this case there exists only an analytical solution for $t < 1/(N_0 A)$ [5], where N_0 is the initial number of real particles.

2.4 Case 4, Brownian coagulation kernel of monodisperse particles in the continuum regime

When particle diameter is so small that particle enters continuum regime, the coagulation kernel for Brownian diffusion are given[12]:

$$\beta(u,v) = \frac{2k_B T}{3\mu}\left[2 + \left(\frac{u}{v}\right)^{1/3} + \left(\frac{v}{u}\right)^{1/3}\right], \tag{5}$$

where k_B is Boltzmann's constant, T is the temperature and μ is the viscosity of the medium. Analytical solution of the case can refer to Ref. [6].

2.5 Case 5, Brownian coagulation kernel of log-normal polydisperse particles in the continuum regime

The analytical solution[13] of the case with initial lognormal PSD and Brownian coagulation kernel in the continuum regime is as follows:

$$n_p(v,t) = \frac{1}{3v}\frac{N(t)}{\sqrt{2\pi}\ln\sigma(t)}\exp\left\{-\frac{\ln^2[v/v_g(t)]}{18\ln^2\sigma(t)}\right\}, \tag{6}$$

where $v_g(t)$ is the geometric number mean particle volume, $\sigma(t)$ is the geometric standard deviation based on particle radius. Coagulation kernel in the case is the same with that of Case 4.

Coagulation kernel is generally bounded as follows: $A \leqslant \beta(u,v) \leqslant A(u + v)$ or Auv.

3 Numerical Simulation

In the paper, for all cases, the initial total particle number is $N_0 = 10^8$, and $A = 10^{-9}$; for Case 4 and Case 5 of Brownian coagulation, the temperature T is 2 500 K and is always constant, and Boltzmann's constant k_B is $1.380\,54 \times 10^{-23}$ J · K, and the medium viscosity μ

is 1.83×10^{-5}kg/(m·s).

The initial number of fictitious particles in MMC method is set as 3 000 for Case 1, and 2 000 for Case 2, 1 000 for both Case 3 and Case 4, 10 000 for Case 5, respectively. The initial particle size distribution for Case 1 – Case 4 is monodisperse, however polydisperse for Case 5. In order to initialize particle population and collect statistics properties of PSD at some appointed time-point, polydisperse particles are divided into 200 classes between the largest and smallest particle volumes in the simulation.

The initial particle diameter for monodisperse particle cases is 1.24 (non-dimension). For Case 5 of polydisperse particles, in the paper we choose $\sigma_0 = 1.5$, $v_{g0} = 1.0$ μm^3. The volume range of lognormal distribution of Case 5 in the paper is considered as $\ln v_{g0} - 12\ln\sigma_0 \leqslant \ln v \leqslant \ln v_{g0} + 12\ln\sigma_0$. Now the minimum volume and diameter of particles is 7.701×10^{-3} μm^3 and 0.245 μm, respectively; the maximum volume and diameter is 127.746 μm^3 and 6.281 μm, respectively.

Figure 2 shows the comparison of the time evolution of some key parameters for Case 1 between analytical and MMC solution, including the curve of relative total number concentration $(N(t)/N_0)$ along with time t, relative geometric mean particle mass $(\tilde{M}(t)/\tilde{M}_0)$ along with t and geometric standard deviation (σ) along with t. Although time evolution is continued to a long time and the total number of particles decreases sharply, computation precision is conserved well because of the constant number of fictitious particles. The agreement between MMC solution and analytical solution is exact.

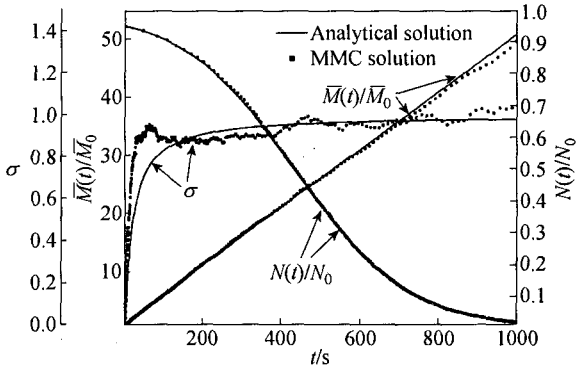

Fig. 2 The time evolution of parameters for Case 1

The results of numerical simulation for Case 2, Case 3, and Case 4 are exposed synthetically in Fig. 3, including the curve of relative total number concentration $(N(t)/N_0)$ along with time t for Case 2 and Case 3, and the curve of geometric mean particle mass $(\tilde{M}(t)/\tilde{M}_0)$ along with t for Case 4. As we can see, the simulation result of MMC method for GDE agrees with analytical solution well, even the difference between MMC solution and analytical solution is

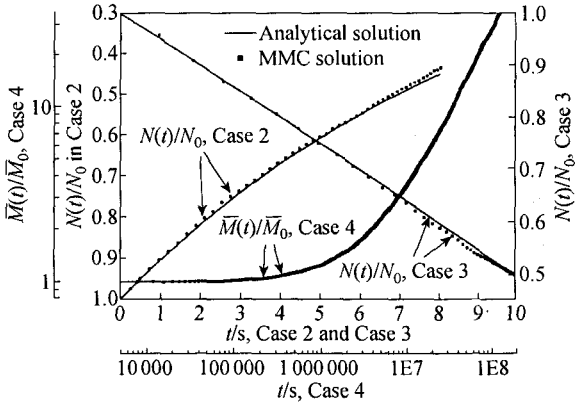

Fig. 3 The time evolution of parameters for Case 2, Case 3 and Case 4

less than the thickness of the line.

As far as Case 5 is considered, Fig. 4(a) is a plot of relative total number concentration ($N(t)/N_0$), relative geometric mean particle volume ($v_g(t)/v_{g0}$), and geometric standard deviation σ as a function of time. For parameters $N(t)$ and $v_g(t)$, the MMC solution agree well with analytical solution. The result of σ has some difference with analytical solution along with the evolution of time. Fig. 4(b) illustrates the evolution of the size distribution for Brownian coagulation of polydisperse particles. The agreement between MMC solutions and analytical solutions is mostly good. Along with the evolution of time, the peak value of size distribution curve is descending, which means particle number decreases continuously; and the location of peak is moving to the right side (the side of big particles), which means the particle volume is bigger and bigger. During the evolution, the particle size distribution remains basically lognormal distribution, *i. e.*, the "self-preserving" distribution[12]. However, along with advancing of time evolution, the agreement for size distribution between MMC solution and analytical solution becomes worse and worse, which inherits the striking difference of σ in Fig. 4(a) between MMC solution and analytical solution. The inapplicable measure of particle bin discretization and the insufficient number of fictitious particles may contribute partly to it, and certain additional approximations and simplifications of analytical solution in Ref. [13] may also contribute partly to those bias. Those need validate and probe farther.

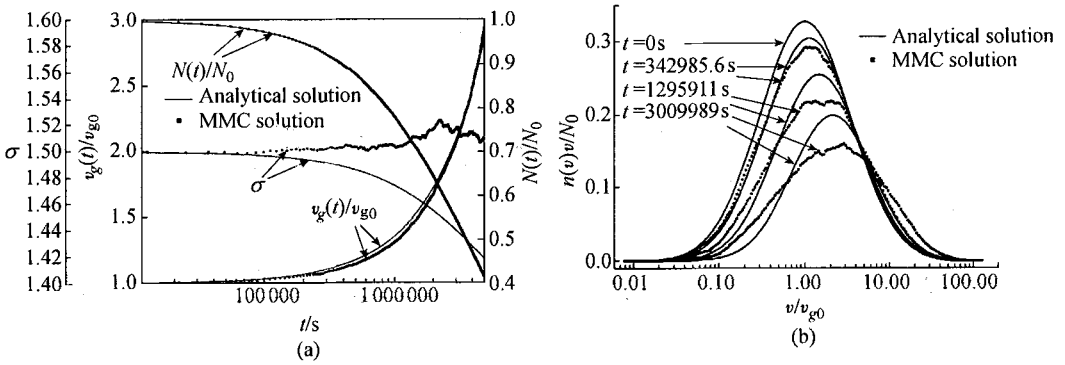

Fig. 4 The time evolution of parameters for Case 5

Ordinary Monte-Carlo methods have to increase the number of simulation particles because of the continuous decreasing of simulation particles along with time evolution, however MMC method will save computation cost because of less number of fictitious particles. The setting evolution time for Case 1 through Case 5 is 1 000 s, 8 s, 10 s, 10^8 s and 5×10^6 s, respectively. The corresponding computation cost of MMC method is 1 091 s, 763 s, 94 s, 1 211 s and 1 105 s, respectively (CPU is Athlon Xp 2 500 +).

4 Discussions

The different kinds of coagulation kernel have the different influence on the process of coagulation. As for constant coagulation kernel case, those particles with different volume have the same coagulation probability, says, constant coagulation kernel has the same

influence on small particles and big particles at the same moment; However, along with the evolution of time, the total number of real particles decreases, which leads to C_i, the total coagulation probability per unit time of fictitious particle i with any fictitious particle, smaller and smaller and then time step Δt bigger and bigger. As for both linear and quadratic coagulation kernel cases, coagulation probability of those small particles will be less than that of those big particles, and coagulation probability for both cases has cumulative effect (linear cumulation or quadratic cumulation), says, the bigger tracked particle or partner has bigger coagulation probability; Along with the advancement of time, both the depletion of real particles and the enlargement of mean particle volume have complicated influence on C_i. It is found during numerical simulation that coagulation probability C_i will increase continuously as for both linear and quadratic coagulation kernel cases, which also makes time step Δt decrease continuously. Comparatively, C_i in linear coagulation kernel case increases faster than C_i in quadratic coagulation kernel case in the paper. As far as Brownian coagulation kernel in continuum regime is considered, the kernel is weakly size-dependent because the kernel is the function of the radio between the diameters of two particles. Furthermore, although the mean particle mass in Case 5 increases continuously, the range of particle size distribution does not expand obviously and both big and small particles exist, which also proves Brownian coagulation kernel in continuum regime has weak correlation with particle size distribution. So constant coagulation kernel is similar with Brownian coagulation kernel in continuum regime and can be regarded as approximation or limit case of Brownian coagulation kernel in continuum regime. In a word, constant coagulation kernel and Brownian coagulation kernel in continuum regime affect small particles much more than linear and quadratic coagulation kernel, whereas affect big particles much less than linear and quadratic coagulation kernel.

Different from those known Monte-Carlo methods for coagulation, MMC method introduces the concept of "weighted fictitious particle", which not only makes it have receivable computation cost in engineering application, but also makes it possible to simulate the time and space evolution of PSD within the whole system discarding the concept of "subsystem"; Similar with constant number method[6-8], MMC method has high computation precision because of the constant number of simulation particles. In addition, MMC method conserves computational domain, which makes it convenient to be applied in engineering computation and scientific quantitative analysis. Last but not least, MMC method is based on time-driven technique and can expand to take into account other events such as condensation/evaporation, nucleation, deposition, breakage, *etc.* MMC method not only can consider space dispersion of PSD by means of plotting grid, but also can describe particle and medium velocity field by coupling Lagrange particle tracking algorithm and then two-phase flow model. MMC method can also be applied on consider GDE for multi-component, more-dimension and polydisperse particle population. Those works will be developed at the next stage.

Computational bias of MMC methods should be advanced by means of applicable measure of particle bin discretization, the opposite number of fictitious particles and the more times of MMC loop.

5 Conclusion

Multi-Monte-Carlo (MMC) method for general dynamic equation (GDE) considering particle coagulation is performed: MMC method introduces the concept of "weighted fictitious particle" and then tracks those fictitious particles, the number of which is far less than that of real particles. MMC method is based on "time-driven" Monte-Carlo technique and conservers the number of fictitious particles and computational domain constant during simulating. MMC method had been used to simulation five special cases with different coagulation kernels. The agreement between MMC solutions and analytical solutions is mostly good, which validates computation precision of MMC method. Furthermore computation cost of MMC method is receivable for engineering computation and general scientific quantitative analysis. Those may make MMC method a standard reference solution for solving GDE.

References:

[1] Tucker W G. An overview of PM 2.5 sources and control strategies[J]. *Fuel Processing Technology*,2000,**65**(1):379 - 392.

[2] Meng Z,Dabdub D,Seinfeld J H. Size-resolved and chemically resolved model of atmospheric aerosol dynamics[J]. *Journal of Geophysical Research*,1998,**103**(3):3419 - 3435.

[3] Debry E,Sportisse B,Jourdain B. A stochastic approach for the numerical simulation of the general dynamics equation for aerosols[J]. *Journal of Computational Physics*,2003,**184**(2): 649 - 669.

[4] Liffman K. A direct simulation Monte Carlo method for cluster coagulation[J]. *Journal of Computational Physics*,1992,**100**(1):116 - 127.

[5] Kruis F E,Maisels A,Fissan H. Direct simulation Monte Carlo method for particle coagulation and aggregation[J]. *AICHE Journal*,2000,**46**(9):1735 - 1742.

[6] Smith M,Matsoukas T. Constant-number Monte Carlo simulation of population balances[J]. *Chemical Engineering Science*,1998,**53**(9):1777 - 1786.

[7] Lee K,Matsoukas T. Simultaneous coagulation and break-up using constant-N Monte Carlo [J]. *Powder Technology*,2000,**110**(1/2):82 - 89.

[8] Lin Y, Lee K, Matsoukas T. Solution of the population balance equation using constant-number Monte Carlo[J]. *Chemical Engineering Science*,2002,**57**(12):2241 - 2252.

[9] Nanbu K. Direct simulation scheme derived from the Boltzmann equation I —Monocomponent gases[J]. *Journal of the Physical Society of Japan*,1980,**49**(11):2042 - 2049.

[10] Nanbu K,Yonemura S. Weighted particles in coulomb collision simulations based on the theory of a cumulative scattering angle[J]. *Journal of Computational Physics*,1998,**145**(2): 639 - 654.

[11] Zhao Haibo,Zheng Chuguang,Xu Minghou. Multi-Monte Carlo method for particle coagulation:description and validation[J]. *Applied Mathematics and Computation*,2005,**168**(2).

[12] Friedlander S K,Wang C S. The self-preserving particle size distribution for coagulation by Brownian motion[J]. *Journal of Colloid and Interface Science*,1966,**22**(1):126 - 132.

[13] Lee K W,Lee Y J,Han D S. The log-normal size distribution theory for Brownian coagulation in the low Knudsen number regime[J]. *Journal of Colloid and Interface Science*,1997, **188**(2):486 - 492.