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The event-driven constant volume method for particle coagulation dynamics

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Monte Carlo (MC) method, which tracks small numbers of the dispersed simulation particles and then describes the dynamic evolution of large numbers of real particles, constitutes an important class of methods for the numerical solution of population balance modeling. Particle coagulation dynamics is a complex task for MC. Event-driven MC exhibits higher accuracy and efficiency than time-driven MC on the whole. However, these available event-driven MCs track the "equally weighted simulation particle population" and maintain the number of simulated particles within bounds at the cost of "regulating" computational domain, which results in some constraints and drawbacks. This study designed the procedure of "differently weighted fictitious particle population" and the corresponding coagulation rule for differently weighted fictitious particles. And then, a new event-driven MC method was promoted to describe the coagulation dynamics between differently weighted fictitious particles, where "constant number scheme" and "stepwise constant number scheme" were developed to maintain the number of fictitious particles within bounds as well as the constant computational domain. The MC is named event-driven constant volume (EDCV) method. The quantitative comparison among several popular MCs shows that the EDCV method has the advantages of computational precision and computational efficiency over other available MCs.

population balance modeling, stochastic solution, coagulation, particle size distribution, numerical simulation

The main function of population balance modeling (PBM) is to describe the dynamic evolution in the dispersed systems. The involved dynamic events in PBM include collision, coagulation (or agglomeration), (heterogeneous) condensation/evaporation (or surface growth/dissolution), (homogeneous) nucleation, deposition (or scavenging), and chemical reaction of discrete elements (for example, solid particles, droplets, or bubbles in multiphase flows). Particle size distribution function (PSDF) is one of the key issues for the dynamic evolution of the dispersed systems. PBM focuses on the dynamic evolution of PSDF, which is dominated by some kinds of dynamic events.

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When a dispersed system is considered as a spatially homogeneous zero-dimensional system, it is called zero-dimensional PBM, where the details of the multiphase flows and the space dispersion of PSDF are ignored, and only the time evolution of PSDF dominated by those dynamic events are obtained; on the other hand, as far as the temporal-spatial evolution of PSDF is considered, it is known as multi-dimensional PBM, where the zero-dimensional PBM are coupled into the models of multiphase turbulent flow.

PBM has attracted significant coverage from many different fields of science and engineering including combustion, chemical engineering, aerosol, nanoparticle, and atmospheric physics. A large number of scientists and engineers have made great efforts to assist in further development of the methodology. The academic and industrial researches on PBM have focused on three broad groups during the past twenty years[1]: the numerical solutions of the population balance equation (PBE), the theoretical or experimental researches on the kernels of dynamic events, and the application of PBM to complex dispersed systems. Owing to the importance and generalization of the particle coagulation event (that is, two particles collide each other and bond together to result in a new bigger particle) in the dispersed systems, the event is one of the most important areas in PBM; furthermore, the coagulation event is also one of the most intractable tasks in PBM because a coagulation event is related to two or more particles (in most references and also in the paper, only two-particle coagulation event is considered). The difficulty of PBM for particle coagulation is mostly associated with the partial integro-differential nature of the PBE. Based on the postulates of rare particles and molecular chaos, the PBE for coagulation, which describes the time evolution of PSDF, is represented as the following mathematical equation[2]:

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

where $n(v, t)$ with dimension $\mathrm{m}^{-3}\cdot\mathrm{m}^{-3}$ is the PSDF at time t, so that $n(v,t)\mathrm{d}v$ is the number concentration of particles with size range between v and $v+\mathrm{d}v$ per unit volume at time t; $\beta(v, u, t)$ is coagulation kernel for two particles of volumes v and u at time t, $\mathrm{m}^3\cdot\mathrm{s}^{-1}$. The model of coagulation kernel can be obtained by theoretical analysis, the mesoscopic or microscopic numerical simulation, or experimental research, and some successful models are available at present, including some kinds of Brownian coagulation kernels and turbulent coagulation kernels. The first term on the right-hand of eq. (1) is the birth term, accounting for the formation of a particle of volume v due to the coagulation event between a particle of volume u (smaller than v) and a particle of volume $(v–u)$; and the coefficient 1/2 is raised from the fact that one coagulation event is related to two particles. The second term on the right-hand of eq. (1) is the death term, representing the disappearance of a particle of volume v due to its coagulation with any other particle. It is worth noting that the inter-collision of particles does not influence the time evolution of PSDF, but affects the space evolution of PSDF. So the zero-dimensional PBM does not take any collision event into account. This study focuses on the zero-dimensional PBM for particle coagulation.

The deterministic scheme[3] such as method of moments and sectional method is capable of solving eq. (1) by means of the direct integral-differential operation on high-dimensional phase space based on the Eulerian coordinates, however there exist some difficulties such as complicated mathematical models and discrete errors. The stochastic scheme, which describes directly the dynamic evolution of a particle population in dispersed systems and then solves indirectly the PBE, is a good alternative. The discrete nature of Monte Carlo (MC) method adapts itself naturally to

discrete process (i.e., the discrete particle population and the discrete dynamic events). As a Lagrangian method, MC can gain the information about particle history and trajectory crossing, and then obtain the details of the dynamic evolution of particles and describe multi-dimensional, multi-component, and polydispersed (spectrum-widened) particle population. Furthermore, the algorithm of MC is comparatively easy and is programmed in a simple manner. Owing to the above-described advantages, MC constitutes an important class of methods for the numerical solution of the PBE.

Generally speaking, MC methods can be classified[3] either according to the treatment of the time-step, namely, "time-driven" MC and "event-driven" MC, or according to the scheme that maintains the total number of simulation particles and the volume of computational domain, i.e., constant-volume MC and constant-number method. In the time-driven MC[4], possible events such as coagulation are treated within one adjustable time-step, based on the assumption that any dynamic events of the same or different simulation particles are uncoupled with each other within a sufficiently small time-step. Within the time-step, a simulation particle is constrained to participate in one specified event at most, although it is permitted to participate in several different kinds of events. In the event-driven MC[5], special events are stochastically implemented with probabilities derived from the mean-filed rates of corresponding dynamic processes. Then time is advanced by the waiting time between two events, which adjusts itself to the rates of various event processes. The constant-volume method[5] tracks a constant domain and thus the total number of simulation particles grows or shrinks in direct proportion to the number concentration of a physical system. A practical difficulty with this implementation is that a prolonged simulation causes the number of particles to exceed the bounds of the simulation box. If the number of simulation particles is superabundant, the computational cost is badly burdened; on the other hand, insufficient samples can result in low stochastic precision of MC. The constant-number method[6], however, contracts (when the number concentration of particles increases) or expands (when the number concentration decreases) the domain volume so that a constant number of simulation particles is contained. Some other MCs[4,7,8] adopt the stepwise "regulating" to compromise the maintenance of both the number of simulation particles and the computational domain, where the doubling or halving action of the domain volume is ignited to restore the number of simulation particles once the simulation particle number exceeds to half or double of the initial number.

In fact, most of the available stochastic methods for PBM exhibit several characteristics. For example, the MC by Garcia et al.[5] is simultaneously event-driven and constant-volume MC; the MC by Liffman[4] performs the time-driven and stepwise constant-volume characteristics; stepwise constant volume method proposed by Maisels et al.[7,8] is event-driven with periodic regulation of number of particles; constant number method by Matsoukas and his co-workers[6] is also event-driven technique with continuous regulation of particle number; multi-Monte Carlo (MMC) method by us[9] demonstrates the characteristics of time-driven technique, constant volume and constant number.

We established the quantitative evaluation method of MC accuracy for the time evolution of PSDF[10], and then compared in terms of numerical precision and computational cost the time-driven direct simulation Monte Carlo (time-driven DSMC)[4], stepwise constant volume method[8], constant number method[6], and MMC method[9] in the case of zero-dimensional PBM. It is found that: (1) Event-driven MC shows higher precision and lower cost than time-driven MC; (2) the forcible maintenance of particle number results in comparatively low precision and efficiency;

(3) the constant volume is necessary for the purpose of considering the space evolution of the PSD. Nevertheless, these available MCs in references exhibit some drawbacks, more or less. In these MCs such as stepwise constant volume method and constant number method, a subsystem of the whole system is sampled, either implicitly or explicitly, based on the assumptions of the spatially-homogeneous system and the periodic boundary conditions. The simulation particles that have the same number weight are tracked by these MCs, which means the procedure of "equally weighted simulation particle population" is adopted to initialize real particle population and the equally weighted simulation particles are tracked; and then, these MCs keep the number of simulation particles within prescribed bounds at the cost of "regulating" the domain volume. The above-mentioned drawbacks in these MCs makes themselves into some difficulties such as the treatment of boundary conditions, the coupling with multiphase flow models and grid-plotting techniques, and then the space evolution of PSDF (the multi-dimensional PBM). Based on those understandings, we developed a new event-driven MC method for the zero-dimensional PBM, which is based on the new procedure of "differently weighted simulation particle population".

1 Methodology

1.1 Procedure of differently weighted fictitious particle population

The MMC method[11,12] introduced the procedure of "differently weighted fictitious particle population" (abbreviated to the differently weighted procedure in the following text) for the first time. From the point of view of the whole system, the procedure initializes directly real particle population and then generates the differently weighted fictitious particle population, where no any assumptions are adopted. In the procedure, real particles that have same or similar volumes are considered to have same properties and, hence, same dynamic behaviors. These real particles are represented by one or several weighted fictitious particles, where the fictitious particle is an indicator of these represented real particles. The number weight of a fictitious particle is just the number of real particles represented by the fictitious particle. The dynamic evolution of real particle population is described through tracking the fictitious particle population. The fictitious particle population has the same statistical property of the key parameter, i.e., particle size distribution function, as the real particle population. Factually, the fictitious particles of the same class (or, the same or similar volumes) have the same value of number weight, however the different values for the different classes. The number weight and volume of these fictitious particles participating in dynamic events are adjusted to describe the events, which will benefit the maintenances of computatonal domain and simulation particle number. A pictorial representation of weighted fictitious particle is demonstrated in refs. [9, 11, 12]. In the paper, the new stochastic method is also based on the differently weighted procedure.

In fact, the procedure of "equally weighted simulation particle population" (abbreviated to the equally weighted procedure in the following text) adopted by general MCs is a special case of the differently weighted procedure. The latter has at least the following three advantages over the former. (1) The equally weighted procedure may introduce some discrete errors. The real particle population needs to be initialized to generate the simulation particle population before any MC simulation is started. No matter which procedure is adopted, any MC expects that the tracked simulation particle population duplicates the details of PSDF from the real particle population as much as possible. As for an initial monodispersed real particle population, the equally weighted procedure is capable of duplicating exactly any details of PSDF from the real particle population.

However, as far as an initial polydispersed real particle population is considered, the equally weighted procedure may lose some detailed PSDF information of the real particle population. It is because in the equally weighted procedure these particle classes with small number concentrations can not be represented by even one simulation particle and then some PSDF details are lost during the initialization process. On the other side, the differently weighted procedure duplicates the real particle size distribution almost exactly by means of assigning some fictitious particles with small number weights in a simple manner to represent these particle classes with small number concentrations. (2) the equally weighted procedure may deteriorate the statistical precision of stochastic approach. In order to obtain PSDF at any specified time-point, the number concentration of every particle section must be counted statistically. The statistical precision is inversely proportional to the square root of the number of simulation particles in the particle section. If only several simulation particles represent the particle section, a big statistical error appears. The case usually happens in the equally weighted procedure. The differently weighted procedure, however, is capable of protecting against statistical fatigue and updating the numerical precision of MC method, in which more simulation particles are utilized to represent every bin of real particle population by means of adjusting the private number weight of these simulation particles. (3) The equally weighted procedure constraints greatly the scope of MC method. For example, if a breakage event leads to some differently sized particles, as it surely happens in nature, several differently sized simulation particles with different number weights and different volumes should be used to represent these fragments. If a nucleation event produces some unequally sized particles, the same problem is solved by the differently weighted procedure.

1.2 Flowchart of the event-driven constant volume method

A new MC method is proposed based on the above differently weighted procedure. It is an event-driven technique and keeps the computational domain constant during the MC simulation. The MC is therefore named event-driven constant volume (EDCV) method. The flowchart is shown in Figure 1. Since the equally weighted procedure happens naturally in some special cases (for example, the monodispersed or discrete-distributed particle population) and possesses some advantages of precision and efficiency in these cases (which will be discussed in the following text), the EDCV method in the paper has two versions, i.e., EDCV-1 which is based on the differently weighted procedure and EDCV-2 which is based on the equally weighted procedure. The details of the two versions are shown in Figure 1. In the following, the key issues of the EDCV method are introduced in details.

1.3 Rate of dynamic events

As far as an event-driven MC method is considered, the rate of dynamic event, which represents the probability of dynamic event per unit time and per unit volume, must be first calculated to determine the waiting time (or the interval of quiescence) between two dynamic events and to select the main event (the dynamic event which happens after the waiting time) and the main particle(s) (the fictitious particle(s) which participate(s) in the main event). In the paper the coagulation rate of fictitious particle population depends on the different procedures.

If the number weights of all fictitious particles are the same and equal to 1, then every real particle is tracked. In the case, the coagulation rate R_{coag} (with dimension of $\text{s}^{-1} \cdot \text{m}^{-3}$) is calculated by[6]

$$R_{\text{coag}} = \frac{1}{2V_{\text{s}}^2} \sum_{i=1}^{N_{\text{f}}} \sum_{j=1, i \neq j}^{N_{\text{f}}} \beta_{ij} = \frac{1}{V_{\text{s}}^2} \sum_{i=1}^{N_{\text{f}}} \sum_{j=i+1}^{N_{\text{f}}} \beta_{ij} , \tag{2}$$

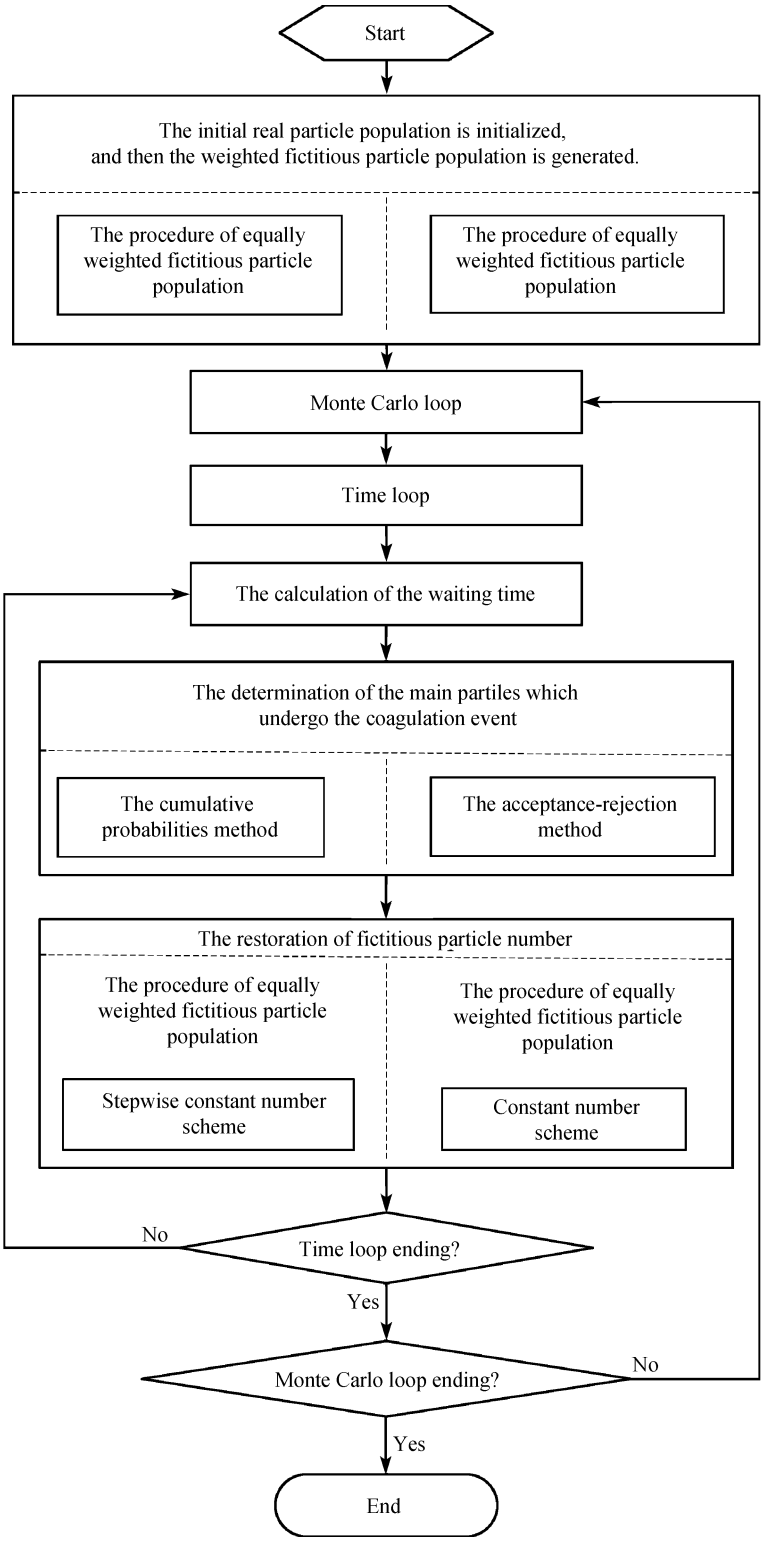

Figure 1 Flowchart of event-driven constant number method.

where V_s is the volume of computational domain; N_f is the total number of fictitious particles, and β_{ij} is coagulation kernel of two particles i and j.

We further discuss the general case in the equally weighted procedure, where the number weight of fictitious particles is same however the common number weight w is unequal to 1. The coagulation of two fictitious particles i and j (their number weights are w_i and w_j, respectively; w_i=w_j=w; their volumes are v_i and v_j, respectively) is considered as the interaction between two groups of real particles. Thus the number of real coagulation events occurring among ith-group real particles (those real particles are represented by fictitious particle i, and number concentrations is w_i/V_s) and jth-group particles (those real particles are represented by fictitious particle j, and number concentrations is w_j/V_s) per unit time and per unit volume is given by

$$N'_{ij} = \beta_{ij} \times \frac{w_i}{V_s} \times \frac{w_j}{V_s}, \tag{3}$$

and N'_{ij} has the dimension of $\mathrm{m}^{-3} \cdot \mathrm{s}^{-1}$.

Thus, the total number of real coagulation events occurring between a real particle represented by fictitious particle i and any other real particle per unit time and per unit volume, N'_i, is then calculated as follows:

$$N'_i = \sum_{j=1, j\neq i}^{N_f} N'_{ij} = \frac{1}{V_s^2} \sum_{j=1, j\neq i}^{N_f} \left(\beta_{ij} \times w_i \times w_j\right). \tag{4}$$

Since fictitious particle i represents w_i real particles, each real particle of fictitious particle i participates in C'_i real coagulation events per unit time on the average, i.e.,

$$C'_i = \frac{N'_i}{w_i} = \frac{1}{V_s^2} \sum_{j=1, i\neq j}^{N_f} \left(\beta_{ij} \times w_j\right). \tag{5}$$

Since fictitious particle i is just an indicator of these represented real particles, the coagulation probability of fictitious particle i is then equal to C'_i. Then, the coagulation probability occurring between two fictitious particles per unit volume and per unit time, R'_{coag}, is calculated as

$$R'_{\mathrm{coag}} = \frac{1}{2}\sum_{i=1}^{N_f} C'_i = \frac{1}{2V_s^2}\sum_{i=1}^{N_f}\sum_{j=1, i\neq j}^{N_f}\left(\beta_{ij} \times w_j\right) = \frac{1}{2V_s^2}\sum_{i=1}^{N_f}\sum_{j=1, i\neq j}^{N_f}\left(w\beta_{ij}\right) = \frac{w}{V_s^2}\sum_{i=1}^{N_f}\sum_{j=i+1}^{N_f}\beta_{ij}. \tag{6}$$

The coagulation rate R'_{coag} in the equally weighted procedure is naturally based on the so-called coagulation rule for equally weighted fictitious particles (abbreviated to the equally weighted rule in the following). It means that each real particle represented by fictitious particle i undergoes a real coagulation event with probability of 100% once the fictitious particle is involved in one coagulation event. The rule can not describe the coagulation event between two differently weighted fictitious particles in a right manner. The paper, therefore, establishes a new coagulation rule for two differently weighted fictitious particles, which is abbreviated to the differently weighted rule in the following. In the new coagulation rule, it is thought that the number of real coagulation events between fictitious particles i and j is $\min(w_i, w_j)$. That is, each real particle of fictitious particle i may coagulate with any one among $\min(w_i, w_j)$ real particles of fictitious particle j; vice versa. Thus, each real particle of fictitious particle i undergoes a real coagulation event

with probability of $\min(w_i, w_j)/w_i$, and each real particle of fictitious particle j undergoes a real coagulation event with probability of $\min(w_i, w_j)/w_j$. The two probabilities are less than or equal to 100%.

In order to calculate the coagulation rate in the differently weighted rule, it is necessary to relate the differently weighted rule to the equally weighted rule. Firstly we investigate the coagulation process of two equally weighted fictitious particles. In the equally weighted rule, one real particle of fictitious particle i may interact with any one among w_j real particles of fictitious particle j; in the same way, there exist w_i candidate particles which may coagulate with one real particle of fictitious particle j. So, as for any one real particle of fictitious particle i or j, the average number of its candidate particles is calculated by

$$c' = \frac{w_i \times w_j + w_i \times w_j}{w_i + w_j} = \frac{2w_i w_j}{w_i + w_j}. \tag{7}$$

If w_i=w_j=w, $c' = w$. Let τ' be the time increment of the coagulation event in the equally weighted rule. Then, dominated by the coagulation rule, the average coagulation rate of any real particle is given by c'/τ'. In the differently weighted rule, one real particle of fictitious particle i or j may interact with any one among $\min(w_i, w_j)$ real particle of fictitious particle j or i. And then, with respect to a real particle of two interacting fictitious particles i and j, the average number of its candidate particles in the new coagulation rule, c'', is equal to $\min(w_i, w_j)$. If w_i=w_j=w, $c'' = w$. Let τ'' be the time increment of the coagulation event; then the average coagulation rate of any real particle in the differently weighted rule is calculated by c''/τ''. Because the average coagulation rate of any real particle in the two coagulation rules is equivalent, we relate the two coagulation rules to the following equations:

$$\frac{c'}{\tau'} = \frac{c''}{\tau''}, \quad \text{or,} \quad \frac{\dfrac{2w_i w_j}{w_i + w_j}}{\tau'} = \frac{\min\left(w_i, w_j\right)}{\tau''}. \tag{8}$$

So

$$\tau'' = \frac{\left(w_i + w_j\right) \times \min\left(w_i, w_j\right)}{2w_i w_j} \tau' \leqslant \tau'. \tag{9}$$

And then, the number of coagulation events occurring among ith-group and jth-group real particles per unit time and per unit volume in the differently weighted rule, N''_{ij}, associates with N'_{ij} through the relation

$$N'_{ij} \times V_s \times \tau' = N''_{ij} \times V_s \times \tau'', \tag{10}$$

that is,

$$N''_{ij} = \left[\beta_{ij} w_i w_j \frac{2w_i w_j}{\left(w_i + w_j\right) \times \min\left(w_i, w_j\right)} \right] \Big/ V_s^2. \tag{11}$$

So, similar to the calculation of coagulation rate R'_{coag} in the equally weighted rule, the coagulation rate per unit volume in the differently weighted rule is calculated by

$$R''_{\text{coag}} = \frac{1}{2V_{\text{s}}^2}\sum_{i=1}^{N_{\text{f}}}\sum_{j=1,j\neq i}^{N_{\text{f}}}\left[\beta_{ij}w_j\frac{2w_iw_j}{\left(w_i+w_j\right)\times\min\left(w_i,w_j\right)}\right] = \frac{1}{V_{\text{s}}^2}\sum_{i=1}^{N_{\text{f}}}T_i\,, \tag{12}$$

$$T_i = \sum_{j=1,j\neq i}^{N_{\text{f}}}\left[\frac{\beta_{ij}w_iw_j^2}{\left(w_i+w_j\right)\times\min\left(w_i,w_j\right)}\right]. \tag{13}$$

If w_i=w_j=w, $R''_{\text{coag}} = R'_{\text{coag}}$; especially, if w_i=w_j=w=1, $R''_{\text{coag}} = R'_{\text{coag}} = R_{\text{coag}}$. Thus, the equally weighted rule is a special case of the differently weighted rule.

1.4 Waiting time between two events

Both time-driven MC and event-driven MC consider the dynamic evolution in the dispersed system as a standard Markov process, in which the interval of quiescence of two events is distributed by an exponential function. According to Shah et al.[13], the waiting time after (k–1) events, Δt_k, is inversely proportional to the coagulation rate $R''_{\text{coag},k}$ in the event-driven MC, that is,

$$\Delta t_k = 1\Big/\left(V_{\text{s}}R''_{\text{coag},k}\right). \tag{14}$$

1.5 Selection of main particle(s)

If only one kind of dynamic event is considered in the dispersed system, the main event is just the considered event. In the paper, the main event is the coagulation event. The next issue is the determination of two main particles that participate in the main event. The probability of the coagulation event between fictitious particles i and j satisfies the following distribution:

$$\beta\left(v_i,v_j\right)\Big/\sum_i\sum_{j,j\neq i}\beta\left(v_i,v_j\right). \tag{15}$$

This selection can be done using either the cumulative probabilities method or the acceptance-rejection method, depending on the particulars of the problem. The two methods satisfy the distribution of coagulation probability. In the cumulative probabilities method, the following processes are adopted. Firstly, the coagulation event of fictitious particle i will be calculated when the random number r satisfies the following condition (just shown in Step 1 of Figure 2):

$$\frac{\Delta t_k}{V_{\text{s}}}\sum_{m=1}^{i-1}T_m < r \leqslant \frac{\Delta t_k}{V_{\text{s}}}\sum_{m=1}^{i}T_m\,. \tag{16}$$

And then, the coagulation partner of fictitious particle i, fictitious particle j, is determined by the following circular procedure (see Step 2 of Figure 2):

$$\frac{\Delta t_k}{V_{\text{s}}}\left(\sum_{m=1}^{i-1}T_m + \sum_{m=1,m\neq i}^{j-1}\frac{\beta_{im}w_iw_m^2}{\left(w_i+w_m\right)\times\min\left(w_i,w_m\right)}\right) < r \leqslant \frac{\Delta t_k}{V_{\text{s}}}\left(\sum_{m=1}^{i-1}T_m + \sum_{m=1,m\neq i}^{j}\frac{\beta_{im}w_iw_m^2}{\left(w_i+w_m\right)\times\min\left(w_i,w_m\right)}\right) \tag{17}$$

The cumulative probabilities method may cause big computational cost to realize the circular procedure, especially in the case of the large particle number. The acceptance-rejection method, however, exhibits high efficiency in some cases. In the acceptance-rejection method, fictitious particles i and j are selected entirely at random. Then if a random number r satisfies the following condition, fictitious particles i and j are the main particles which undergo the same coagulation event.

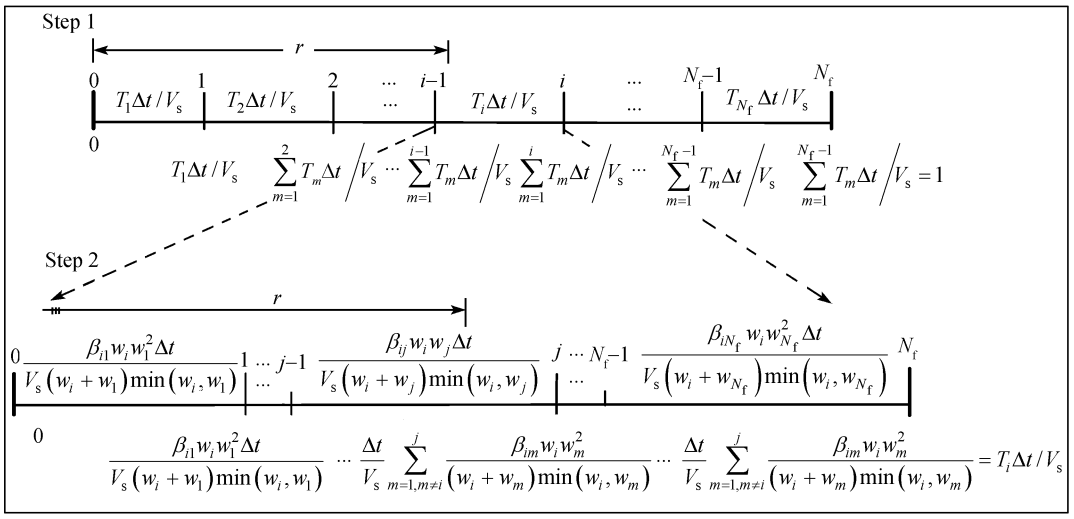

Figure 2 Selection of main particles by the cumulative probabilities method.

$$r \leqslant \frac{\beta_{ij}^{\mathrm{o}}}{\beta_{\max}^{\mathrm{o}}} = \frac{\beta_{ij} w_j \dfrac{2 w_i w_j}{\left(w_i + w_j\right) \times \min\left(w_i, w_j\right)}}{\max\limits_{\forall k, \forall m} \left\{ \beta_{km} w_k \dfrac{2 w_k w_m}{\left(w_k + w_m\right) \times \min\left(w_k, w_m\right)} \right\}}, \tag{18}$$

where β_{ij}^{o} is the specified coagulation kernel of fictitious particles i and j, and $\beta_{\max}^{\mathrm{o}}$ is the maximum over all particle pairs in the differently weighted rule . If $\beta_{\max}^{\mathrm{o}}$ is overestimated, the acceptance-rejection method still describes the Markov process exactly but less efficiently.

1.6 Restoration of fictitious particle number and maintenance of computational domain

In any MC, the total number of simulation particles and the volume of computational domain are two key issues that need particular attention. The excessive simulation particles will burden computation cost of MC simulation; however, the insufficient number of simulation particles will deteriorate computational precision of MC simulation. On the other hand, expansion or contraction of the domain results in bad expansibility and applicability of MC. In the EDCV method computational domain is always maintained and the fictitious particle number is kept within a prescribed bound.

1.6.1 Procedure of differently weighted fictitious particle population. When two differently weighted fictitious particles, i and j, coagulate, it is thought that $\min(w_i, w_i)$ real particles of fictitious particle i coagulate with $\min(w_i, w_i)$ real particles of fictitious particle j, and then $\min(w_i, w_i)$ real particles with volume of (v_i+v_j) are born. After the coagulation event between fictitious particles i and j, all of consequent real particles are divided into two parts: those "coagulated" real particles and those "non-coagulated" real particles. The two parts of real particles are re-represented by the two fictitious particles i and j respectively. If the number weight of i before the coagulation event, $(w_i)_{\mathrm{old}}$, is greater than that of j, $(w_j)_{\mathrm{old}}$, then the new number weights and volumes of i and j after the event are given by

$$\begin{cases} (w_i)_{\text{new}} = (w_i)_{\text{old}} - (w_j)_{\text{old}}; \ (v_i)_{\text{new}} = (v_i)_{\text{old}}, \\ (w_j)_{\text{new}} = (w_j)_{\text{old}}; \ (v_j)_{\text{new}} = (v_i)_{\text{old}} + (v_j)_{\text{old}}. \end{cases} \tag{19}$$

By the scheme the total number of fictitious particles is automatically maintained, although the real particle number decreases continuously along the coagulation process. The scheme is named "constant number scheme".

1.6.2 Procedure of equally weighted fictitious particle population. In the constant number scheme, the number weight of fictitious particles is changed continuously to describe the consequence of coagulation events, where the EDCV exhibits automatically the characteristics of constant number and constant volume. However, we found that[10] the disturbance of number weight of particle ensemble brings inevitably the so-called "constant number error" and the low efficiency of MC. In fact, in some case, for example, initial monodispersed particle population and some initial discrete-distributed polydispersed particle population, the fictitious particle population may possess the common number weight at the initial stage. It is possible for these cases to keep the number weights of all fictitious particles the same at any time, at the cost that the total number of fictitious particles always lies between $N_{\text{f0}}/2$ and $2N_{\text{f0}}$ (N_{f0} is the initial total number of fictitious particles). By the scheme the disturbance of number weight of particle ensemble is avoided as much as possible, and then the computational precision and efficiency of MC are improved in these special cases. In the scheme, the binary coagulation event between fictitious particles i and j results in a new fictitious particle k with volume of v_k ($=v_i+v_j$). The number weight of the fictitious particle k is still equal to the common value w. The old fictitious particles j is abandoned, and the new fictitious particle k is stored in the position of fictitious particle i. The direct consequence of coagulation event is net depletion of the number of fictitious particles.

Along with the occurrence of coagulation events, the total number of fictitious particles decreases, which may deteriorate badly the statistical precision of MC. The MC method develops the so-called "stepwise constant number scheme" to restore samples and then maintain the statistical precision and computational cost. When the number of fictitious particles reaches to $N_{\text{f},0}/2$, the surviving particles are duplicated and added into fictitious particle population to restore the number of fictitious particles. As a result, the number weight of each fictitious particle is halved, and other properties such as volume are kept.

Until now, the coagulation dynamics is described by the EDCV method according to Figure 1. In the next section, the performance of the MC is evaluated by comparing it with other popular MCs.

2 Results

The precision and efficiency of MC for coagulation dynamics do not depend on the forms of kernel, but the algorithmic nature of the MC. Thus, some special cases in which analytical solutions exist are usually used to evaluate the performance of MC, and then some excellent MCs are applied to some general cases. In the paper, the constant coagulation cases, where the coagulation probability does not depend on the particle size, are utilized to evaluate the performance of the EDCV method.

2.1 Case1: Constant coagulation kernel, initial monodispersed population

In the case, the initial volume v_0=1 (dimensionless), the initial number concentration $N_0=10^{10}$ m^{-3}, the coagulation kernel $\beta_{ij}=A=10^{-10}$ m$^3\cdot$s^{-1}, and the characteristic coagulation time is defined as

$\tau_{coag}=1/(N_0A)$. The analytical solutions of number concentration, mass concentration and PSDF at some specified time-points are available in ref. [14].

In the case of initial monodispersed particle population, we established the quantitative evaluation method of MC accuracy for the time evolution of PSDF, where standard deviations of key properties of particle size distribution (particle number concentration, particle mass concentration and PSDF)[10] are calculated. The associated errors of number concentration N and mass concentration M over several MC loops, σ_N and σ_M, are calculated as follows, respectively:

$$\sigma_N(t)=\frac{1}{Q}\sum_{i=1}^{Q}\sigma_{N,i}(t),\ \ \sigma_{N,i}(t)=\sqrt{\frac{1}{L}\sum_{j=1}^{L}\left(\frac{N_j-N_{j,\text{theory}}}{N_{j,\text{theory}}}\right)^2}, \tag{20}$$

$$\sigma_M(t)=\frac{1}{Q}\sum_{i=1}^{Q}\sigma_{M,i}(t),\ \ \sigma_{M,i}(t)=\sqrt{\frac{1}{L}\sum_{j=1}^{L}\left(\frac{M_j-M_{j,\text{theory}}}{M_{j,\text{theory}}}\right)^2}, \tag{21}$$

where Q is the total amount of MC loops; $\sigma_{N,i}$ and $\sigma_{M,i}$ are the standard deviations of N and M at time t and in the i-th MC loop, respectively; L is the number of discretized time-steps within the time bound $0-t$; N_j and M_j are the MC solutions of N and M in the j-th time-step, i.e., at the time-point $t=\sum_{m=1}^{j}\Delta t_m$, respectively; and $N_{j,\text{theory}}$ and $M_{j,\text{theory}}$ are the corresponding analytical solutions of N and M.

Similarly, the overall error in the size distribution, σ_d, is calculated in the case of initial monodispersed particle population as follows:

$$\sigma_d(t)=\frac{1}{Q}\sum_{i=1}^{Q}\sigma_{d,i}(t),\ \ \sigma_{d,i}(t)=\sqrt{\frac{1}{k_{\max}-k_{\min}+1}\sum_{j=k_{\min}}^{k_{\max}}\left(\frac{P_j(t)-P_j(t)_{\text{theory}}}{n_{\text{total}}}\right)^2}, \tag{22}$$

where $P_j(t)$ is the probability of obtaining a cluster containing j primary particles; n_{total} is the dimensionless number concentration of particles, $n_{\text{total}}=1/\overline{v}=N(t)_{\text{theory}}\,v_0\big/M(t)_{\text{theory}}$, $\overline{v}$ is the dimensionless particle average volume; $k_{\max}$ and $k_{\min}$ are the total number of the primary particles in the largest- and smallest-sized particles, respectively.

The EDCV method is quantitatively compared with stepwise constant volume method[7], constant number method[8], and MMC method[9,11,12]. EDCV-1 adopts the differently weighted procedure and the constant number scheme, and EDCV-2 is based on the equally weighted procedure and the stepwise constant number scheme. The case is for the purpose of validating the differently weighted procedure and the differently weighted rule. In the two EDCV methods, the acceptance-rejection method is adopted to select the main particles. In all of MCs, the initial total number of simulation particles is 1000. The multiplicative constant in the MMC method, α, is set as 0.01. The dynamic evolution in the dispersed system is advanced by 1000 s. The numerical results are shown in Figure 3, and CPU times of MCs, T_{CPU}, at the same computer (Althon 2500+, 512 M) are listed in Table 1.

Table 1 Comparison of computational costs

Parameter	Stepwise constant volume method	Constant number method	MMC	EDCV-1	EDCV-2
CPU time/s, Case 1	18.07	45.15	13.01	331.04	20.27
CPU time/s, Case 2	-	-	43.83	8392.72	144.02

As shown in Figure 3, the results of EDCV-2 method are very similar to those of the stepwise constant volume method, indicating that the stepwise constant volume method is a special case of the EDCV method. In fact, the EDCV-2 method tracks the whole system, and every fictitious particle is assigned with the same number weight, in which the halving of the common weight is adopted to restore the number of fictitious particles in the case of coagulation event. As far as the stepwise constant volume method is concerned, the subsystem is tracked and the simulation particles in the subsystem are equally weighted with value "1". However, from the point of view of the whole system (with volume V), every simulation particle has the common weight of $w=V/V_s$. The stepwise constant volume method restores the number of simulation particles by means of the doubling action of the subsystem domain V_s in the coagulation case, that is, the halving of the common weight w from the point of view of the whole system. So the two MC methods are equivalent in some sense, and the differences lie in the tracked computational domain and the scheme of sample restoration.

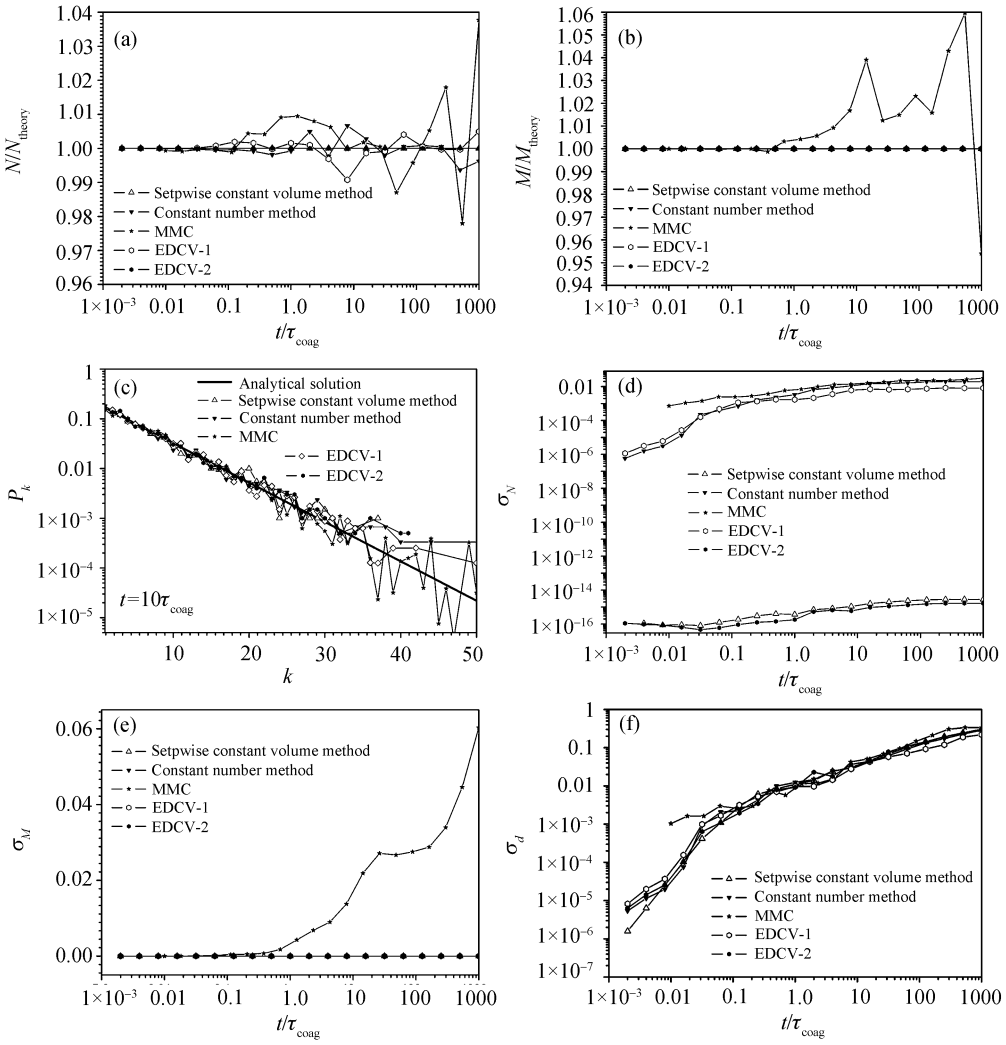

Figure 3 Case 1: Constant coagulation kernel with initial monodispersed population. (a) Number concentration; (b) mass concentration; (c) size distribution; (d) error in number concentration; (e) error in mass concentration; (f) error in size distribution.

However, with respect to the number concentration N, the EDCV-1 method, which keeps the number of fictitious particles constant, exhibits a higher precision than the MMC method and the constant number method but the lower precision than the EDCV-2 method. It is because the disturbance of the number weight of fictitious particles results in some numerical errors when automatically conserving the number of fictitious particles and the volume of computational domain in the differently weighted procedure. Less disturbance and smaller disturbance result in less error. As known, the stepwise constant number scheme restores periodically the simulation particle number (in the stepwise constant volume method, the doubling of the domain is realized; in the EDCV-2, the halving of the number weight is adopted), which results in so-called "stepwise error"; however, the constant scheme restores continuously the simulation particle number (in the EDCV-1 and the MMC method, the number weight of these involved fictitious particles is adjusted; in the constant number method, the volume of computational domain is adjusted in real-time), which results in so-called "constant number error". As for the moments of PSDF (for example, the number concentration and the mass concentration), the "stepwise error" is comparatively less than the "constant number error". As shown in Figures 3(b) and 3(e), the two EDCV methods and two other event-driven MC methods (the stepwise constant volume method and the constant number method) describe perfectly the mass concentration M, which contrasts with the lower precision of the MMC method. In fact, time-driven MCs (including the MMC method) adopt the uncoupling assumption in nature and then perform so-called "uncoupling error"; however event-driven MCs avoid naturally the concept of uncoupling.

As shown in Figures 3(c) and 3(f), the five MCs possess nearly the same precision on the time evolution of PSDF. In fact, the precision of PSDF in MC depends mainly on the number of simulation numbers in the MC. Using reasonably small numbers of simulation particles, $O(10^3)$, very good precision of PSDF can be achieved in these MCs as for the same initial simulation particle number, although the simulation particle number is not the same in the simulations of these MCs. It is worth noting that the EDCV-1 and the MMC methods, which are based on the differently weighted procedure, are capable of tracking the size distribution of larger sizes compared to all other MCs, as shown in Figure 3(c). This is because the differently weighted procedure allows more fictitious particles with lower number weight to represent these rare real particles at the high-end of the size distribution.

The computation costs of these MCs are listed in Table 1. The EDCV-1 method shows the lowest efficiency, which is attributed to two factors at least. On the one hand, the EDCV-1 method, which adopts the differently weighted procedure, takes the CPU time of $O(N_f^2)$ to calculate the coagulation rate $R_{i,\mathrm{coag}}$; however, those MC methods that adopt the equally weighted procedure cost only one half of CPU time. On the other hand, the number of the tracked fictitious particles is always N_{f0} in the EDCV-1 method but between $N_{f0}/2 \sim N_{f0}$ in the stepwise constant volume method and the EDCV-2 method.

As for the coagulation case of initial monodispersed particle population, the EDCV-2 has advantages of computational precision and efficiency over the EDCV-1 on the whole. Except the above-mentioned factors, the high precision and efficiency in the EDCV-2 are also attributed to the absence of the "discrete error" when transforming the initial monodispersed real particle population into the fictitious particle population. However, the standard deviation of particle population will increase and the difference among particle sizes will be bigger and bigger along the time evolution. It is concluded that the EDCV-1 will exhibit higher precision on the whole than the

EDCV-2 because the former can describe more accurately the coagulation dynamics of these particles with small number concentrations. Therefore, the EDCV-2, which adopts the equally weighted procedure, is commended to describe the coagulation dynamics of initial monodispersed particle population within comparatively short time bounds.

2.2 Case2: Constant coagulation kernel, initial exponential distributed particle population

In the case, the initial particle size distribution is represented by an exponential function:

$$n(v,0) = N_0 / v_{g0} \times \exp\left(-v / v_{g0}\right), \tag{23}$$

where the initial mean volume v_{g0}=0.029 μm^3, N_0=10^6 cm^{-3}, β_{ij}=A=6.405×10^{-10} cm^3/s, and τ_{coag}=2/(AN_0)≈1561.3 s. The case is utilized to validate the performance of the EDCV method in the case of initial polydispersed particle population. Here, the two EDCV methods utilize the cumulative probabilities method to select the main particles; The numerical results of the two EDCV methods and the MMC method[12] are compared with analytical solutions[14]. When initializing the continuous-distributed particle population, the initial number of fictitious particles is set as 3000. The continuous size distribution is divided into 200 bins by the logarithmically spaced law between 10^{-5}—1 μm^3, and the fictitious particle number of every bin is greater than 10. Therefore the EDCV-1 and MMC methods track fictitious particle population with number of 4040 from beginning to end, and the EDCV-2 tracks fictitious particle population with number of 3160 in the first stage and the particle number fluctuates between 1560—3160 during the coagulation dynamics.

As shown in Figure 4, the two EDCV methods exhibit the high precision of the moments of particle size distribution and particle size distribution function at some specified time-points. The EDCV-1 method describes nearly perfectly the time evolution of the mass concentration; however it presents the "constant number error" for the number concentration. The equally weighted procedure adopted by the EDCV-2 has the obvious "discrete error" when transforming the initial polydispersed particle population into the equally weighted fictitious particle population. In fact, the equally weighted procedure can not duplicate fully the PSDF of the real particle population, and some information of real particle population on the lower and higher ends of the PSD are lost in the initial stage of numerical simulation. The "discrete error" deteriorates the precision of the EDCV-2 method for N, M and PSDF at the specified time-points. On the contrary, the differently weighted procedure adopted by the EDCV-1 and MMC methods describes the coagulation dynamics of particles of the whole size bounds, especially at the high- and lower-ends of PSD. As for the MMC method, which is based on the uncoupling assumptions and exhibits the uncoupling error in nature, its precision on N is lower than that of the EDCV-2 (as shown in Figure 4(a)), and its precision on M is lower than that of both the EDCV-1 and the EDCV-2 (as shown in Figure 4(b)), and its precision on PSDF is lower than that of the EDCV-1 on the whole (as shown in Figure 4(c), where the bigger and more fluctuations are observed in the MMC method).

The computational costs of several MCs in the case are also listed in Table 1. It is known that the EDCV-1 consumes more CPU time than the EDCV-2. This is also attributed to more cost in the calculation of coagulation rate and more fictitious particles tracked in the EDCV-1.

As for the coagulation case of initial polydispersed particle population, the EDCV-1 possesses higher precision than the EDCV-2 on the whole. This is mainly ascribed to the discrete error of the EDCV-2, which is raised in the first stage and is propagated and even enlarged along the coagulation dynamics. Thus, the EDCV-1, which is based on the differently weighted procedure, is more suitable for the coagulation case of initial polydispersed particle population.

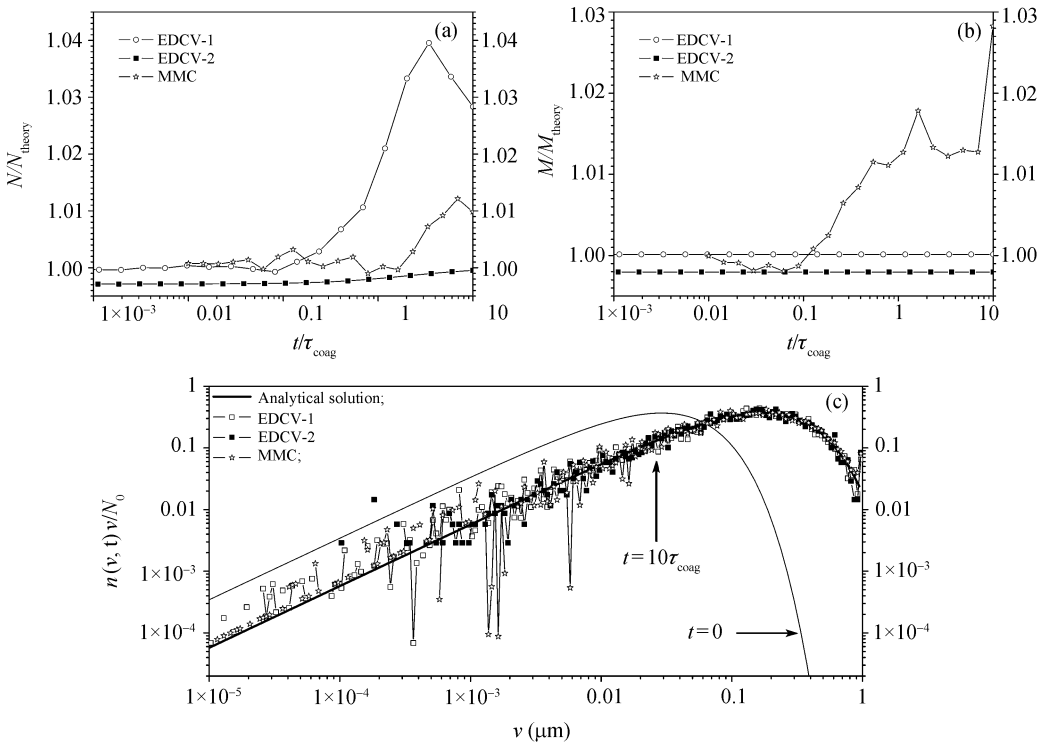

Figure 4 Case 2: Constant coagulation kernel with initial polydispersed population. (a) Number concentration; (b) mass concentration; (c) time evolution of PSDF.

3 Conclusions

With the understanding that event-driven MC demonstrates naturally the immunity to the uncoupling error and, therefore, high computational precision and efficiency, a new MC method, event-driven constant number (EDCV) method, is proposed to describe the time evolution of particle size distribution. The MC introduces the concept of "weighted fictitious particle" and exhibits the constant-volume characteristic. Furthermore, the equally weighted procedure and the differently weighted procedure, the equally weighted rule and the differently weighted rule, and the cumulative probabilities method and the acceptance-rejection method are unified within the same event-driven framework to describe the coagulation dynamics.

By the quantitative comparison of several popular MCs, it is concluded that the EDCV method is capable of describing the coagulation dynamics with high precision and efficiency in the case of initial monodispersed or polydispersed particle population. In the two versions of the EDCV, the EDCV-2 that adopts the equally weighted procedure avoids the disturbance of the number weight of particle ensemble and then performs comparatively less "stepwise error", and it also tracks fictitious particles with comparatively smaller number. The EDCV-2 performs comparatively higher precision and efficiency in some special coagulation cases such as initial monodispersed or discrete-distributed particles; however it can not describe accurately the coagulation case of the polydispersed particles of the whole size bounds, which usually happens in fields of natural and engineering. With respect to these general cases, the EDCV-1 has an advantage over the EDCV-2.

On the one hand, the EDCV-1 describes the coagulation dynamics of polydispersed particles of the whole size bounds with comparatively higher precision, and describes almost accurately the mass concentration. On the other hand, the EDCV-1 exhibits comparatively more "constant number error" due to the continuous disturbance of number weight of particle ensemble (for example, it possesses lower efficiency on the number concentration than the EDCV-2), and consumes more CPU time than the EDCV-2. On the whole, the EDCV-2 is a special case of the EDCV-1. The selection of the two versions of the EDCV method depends on the particular requirements. As for those coagulation cases of initial monodispersed or discrete-distribution particles, or polydispersed particles in which accurate computational precision is not needed, the EDCV-2 is recommended. As for the coagulation cases of polydispersed particles where all-sized particles must be considered, the EDCV-1 is more suitable. Generally speaking, numerical simulation shows the EDCV method has the same precision as or higher precision than other MC methods for zero-dimensional PBM.

Multi-dimensional PBM, which describes the temporal-spatial evolution of PSDF, has been the challenging and urgent requirement in science and engineering at present. It is interesting to investigate the multi-dimensional PBM by coupling the proposed EDCV method for zero-dimensional PBM and the models of two-phase turbulent flow. The difficulty may lie in how to integrate the event-driven framework for particle dynamics with the time-driven framework for two-phase flow, which will be researched in the next stage.

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