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Short communication

Correcting the multi-Monte Carlo method for particle coagulation

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ABSTRACT

Multi-Monte Carlo (MMC) method does not correctly treat the results of coagulation event between two differentially-weighted simulation particles, resulting in comparatively large error in predicting the evolution of particle size distribution. This study corrected the consequential treatment of coagulation event, satisfying the basic laws of mass conservation and number depletion and then resulting in good prediction for particle size distribution. The corrected MMC method can complete with any available Monte Carlo methods with respect to computational accuracy.

1. Introduction

Among many kinds of solution for population balance modeling, Monte Carlo (MC) method has attracted more and more attentions because of its simplicity, and its ability to deal with high-dimensionality problems in a straightforward manner, and, more importantly, their stochastic and discrete nature that adapts naturally to dynamic events which have same nature.

In order to overcome the conflict between large numbers of real particles in a reasonably sized systems (e.g., 10^{20} in an aerosol reactor) and limited CPU speed and memory capacity of common computer, a subsystem of the total system, which contains 10^4 –10^7 real particles, is simulated in most of MCs [1]. The subsystem concept indicates simulation particles are equally weighted with value of V/V_s, where V and V_s are volumes of the total system and the subsystem, respectively. These equally-weighting MCs focus on how to keep the number of simulation particles within bounds by regulating the subsystem domain periodically (e.g., in stepwise constant volume method [2]) or continuously (e.g., in constant number method [3]). As we know, the statistical accuracy of MC for the time evolution of particle size distribution (PSD) depends on not only the total number of simulation particles but also the number of simulation particles in each size interval. These equally-weighting MCs cause large statistical noise for particles in those size sections where number concentration of real particles is so low that only several simulation particles or even no simulation particle are assigned to represent these real particles, for example, the edges of PSD.

Multi-Monte Carlo (MMC) method [4], which is based on time-driven technique and has the characteristics of constant number and constant volume, is the first differentially-weighting MC and is capable of simulating the size distribution of particles over the full size range [5]. The MMC method, however, cannot correctly deal with the result of a coagulation event between two differentially-weighted simulation particles (Section 2). Thus the method demonstrates comparatively low accuracy on, especially, the moments of particle size distribution. The paper aims to correct the consequential treatment of a coagulation event between two differentially-weighted simulation particles to improve computational accuracy of the MMC method.

2. The corrected multi-Monte Carlo method

The differentially-weighting scheme is usually adopted to simulate trace species of gas or plasma in traditional direct simulation Monte Carlo (DSMC). As for a polydispersed particle population, those particles within a size interval may have low number density and then can be considered as trace specie of particle population. Thus the differentially-weighting scheme is useful for population balance in order to simulate the size distribution of particles over the full size range and fatigue against statistical noise of MC. In fact, once the differentially-weighting scheme is adopted, MC is capable of simulating the dynamic evolution of the total system rather than the subsystem. In the differentially-weighting scheme, particles having same or similar size (i.e., in the same interval of particle size distribution) are considered to have similar dynamic behavior and then are represented by some simulation particles. The weight of a simulation particle i in a size interval k, w_i, is equal to N_k/N_{sk}, where N_k and N_{sk} are the numbers of real particles and simulation particles in size interval i, respectively. Size intervals where number density of real particles is high have thus larger values of mean weight and

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simulation particle number than those size intervals where number density is low.

During the time-driven simulation, the total coagulate rate of simulation particle i is calculated as:

$$C_i = \frac{1}{V^2}\sum_j \left(\beta_{ij} w_j\right) = \frac{1}{V^2}\sum_j \beta'_{ij}, \tag{1}$$

where β_{ij} is the coagulation kernel between particle i with volume of v_i and particle j with volume of v_j. And the time step is calculated as $\Delta t = \alpha / \max_i(VC_i)$, where the multiplicative constant α has a value of 0.01 or less.

The process of dynamic evolution due to particle coagulation is considered as a Markov process, and the coagulation probability of simulation particle i within Δt and V is represented by a exponential function: $P_i(\Delta t) = 1 - \exp(-VC_i\Delta t)$. Since a coagulation event is related with two particles, the occurrence probability of a coagulation event of simulation particle i within Δt and V is $P_{E,i}(\Delta t) = 1 - \exp(-VC_i\Delta t/2)$. Note that $P_i(\Delta t)$ is approximately twice of $P_{E,i}(\Delta t)$. MC examines each simulation particle in turn to determine whether the particle coagulates and who is its coagulation partner according to $P_i(\Delta t)$ or $P_{E,i}(\Delta t)$.

In the original MMC method, once a random number r from a uniform distribution in the interval [0, 1] is less than $P_i(\Delta t)$, i will be decided to coagulate. Its partner j is further determined with probability $\beta'_{ij} / \sum_j \beta'_{ij}$ using the cumulative probabilities method or the acceptance-rejection method. In fact, the original MMC method allows double counting of the i–j coagulation to occur, that is, not only can i coagulates with j via the probability $P_i(\Delta t)$, but it is possible that j will coagulate with i via the probability $P_j(\Delta t)$. The method halves the weight of i (when i coagulates with j) or the weight of j (when j coagulates with i) to treat the result of the i–j coagulation as well as keep the total number of simulation particles constant, as follows:

$$\begin{aligned} &\text{the first count: } w_i^* = w_i/2;\ v_i^* = v_i + v_j; \\ &\text{the second count: } w_j^* = w_j/2;\ v_j^* = v_i + v_j; \end{aligned} \tag{2}$$

where the asterisk indicates a new value of weight or size after the coagulation event. In fact, the MMC considers the coagulation of i and j results in a new simulation particle with volume of $(v_i + v_j)$ and weight of $(w_i + w_j)/2$. The approximate scheme for the consequential treatment does not have a closed mass balance for coagulation [6], resulting in a poor PSD compared with other MCs [5,7].

As known, the total number of real coagulation events is $w_i \times w_j$ once i coagulates with j. Since the total number of real particles from i and j is $(w_i + w_j)$ and one coagulation event is involved with two particles, the mean number of real coagulation events per real particle from i or j is

$$\Omega = 2w_i w_j / \left(w_i + w_j\right). \tag{3}$$

Note that $\min(w_i,w_j) \le \Omega \le \max(w_i,w_j)$, and $\Omega = w$ if $w_i = w_j = w$. Thus, the coagulation of i and j results in a new simulation particle with volume of $(v_i + v_j)$ and weight of Ω.

In the corrected MMC method, the coagulation event of simulation particle i is once counted via the probability $P_{E,i}(\Delta t)$, that is, if the random number r is less than $P_{E,i}(\Delta t)$ the coagulation event of i is calculated. The partner of i is similarly determined through probability

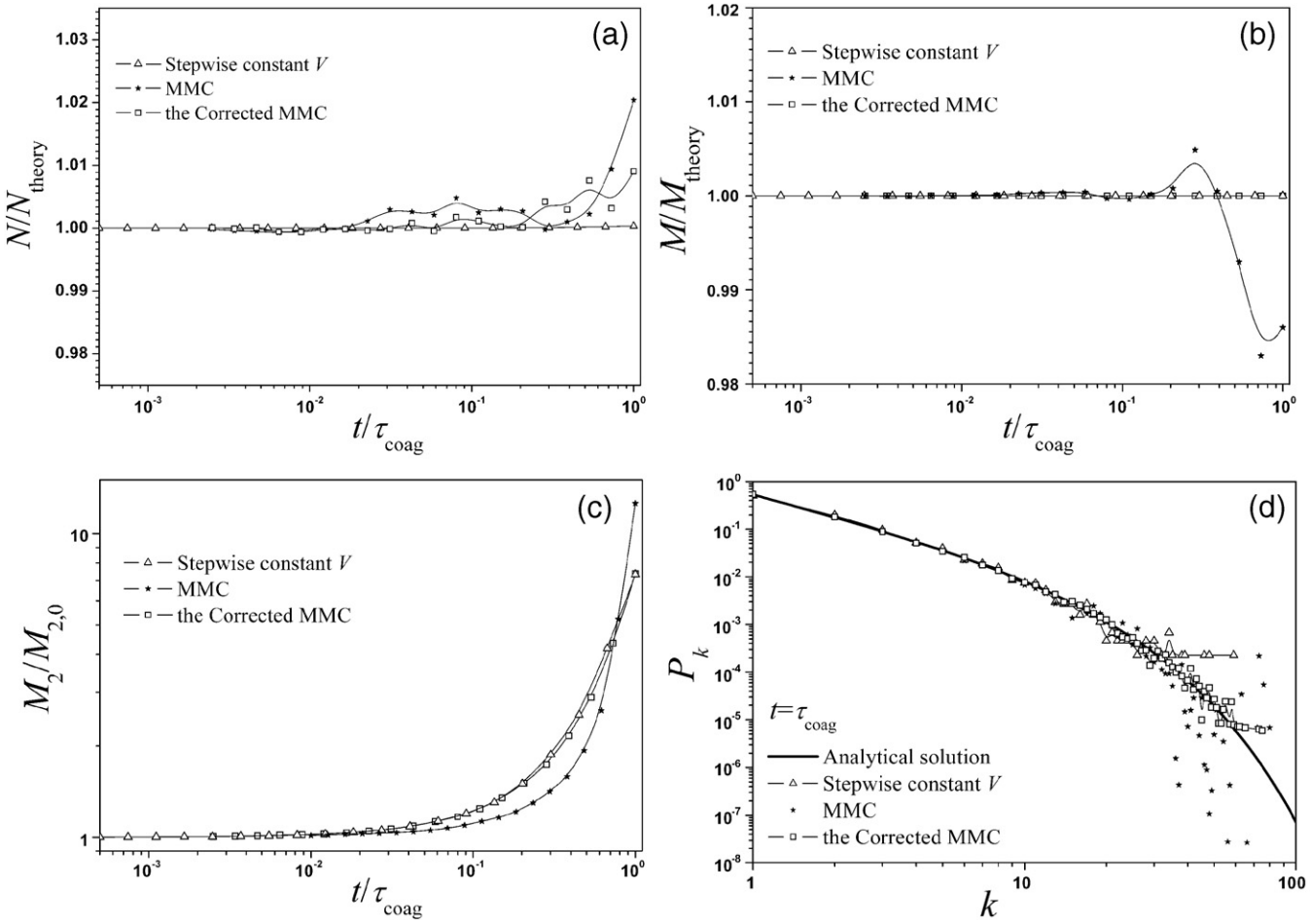

Fig. 1. Results for linear kernel (Case2): (a) number concentration; (b) mass concentration; (c) the second-order moment; (d) probability distribution.

distribution $\beta'_{ij} / \sum_j \beta'_{ij}$. The following measure is taken to treat the result of the i–j coagulation event: a new simulation particle with volume of $(v_i + v_j)$ and weight of Ω is stored in the position of i, and the position of j is replaced by the last simulation particle in the simulation particle array.

According to the consequential treatment, the occurrence of coagulation event results in net depletion of the total number of simulation particles. It is necessary for the MC to restore statistical sample in order to overcome statistical noise. When the number of simulation particles reaches to $N_{s,0}/2$ ($N_{s,0}$ is the initial number of simulation particle), the surviving particles are duplicated and added into simulation particle population to restore the number of simulation particles. As a result, the weight of each simulation particle is halved.

3. Numerical results

We first calculated three special cases in which analytical solutions exist[8]: (1) Case1, constant kernel, $\beta_{ij} = A$; (2) Case2, sum kernel, $\beta_{ij} = B(v_i + v_j)$; and (3) Case3, product kernel, $\beta_{ij} = Cv_iv_j$. Three cases start from a monodisperse initial condition, and all of MC simulations track 1000 (for Case1) or 2000 (for Case2 and Case3) simulation particles with same weight and same volume in the initial stage. Fig. 1(a)–(d) shows the zero-order moment (N), the first-order moment (M), the second-order moment (M_2) of size distribution, and the probability distribution (P_k) of aggregates having k primary particles ($t = \tau_{coag}$, where τ_{coag} is the characteristic coagulation time) for the linear kernel (Case2), and Table 1 lists the time-mean standard deviations of N, M and P_k for all of cases, which are considered as reference for evaluating MC methods [5] and are calculated as following

$$\sigma_\xi(t) = \frac{1}{Q}\sum_{i=1}^{Q} \sqrt{\frac{1}{t}\int_0^t \left[\delta_\xi^{(i)}(t)\right]^2 \mathrm{d}t}. \quad (4)$$

In Eq. (4), t is the length of time-interval; Q is the total amount of MC repetitions ($Q = 3$ in the paper); $\delta_\xi^{(i)}(t)$ is relative error of ξ (which may be N, or M, or P_k) at time t and the i-th MC repetition.

From the results in Fig. 1 and Table 1, the corrected MMC method has the least σ_{P_k}; the corrected MMC method and the constant-N method have the least σ_M; and the stepwise constant-V method has the least σ_N. The corrected MMC method has a remarkable advantage of describing N, M or P_k with respect to the original MMC method. Note that the original MMC method performs large numerical error for high-order moments such as the second-order moment (M_2). On the contrary, the obtained M_2 from the corrected MMC method is in very good agreement with the benchmark solution (the stepwise constant-V method), as shown in Fig. 1(c). And it is worth noting that the original and corrected MMC methods, which adopt the differentially-weighting scheme, are capable of tracking these large-scale aggregates ($k > 60$ in Fig. 1(d)); here the result of the corrected MMC method is closer to analytical solution.

A real coagulation case, Brownian coagulation in free-molecular regime [9], is further simulated, where $\beta_{ij} = K(1/v_i + 1/v_j)^{1/2}(v_i^{1/3} + v_j^{1/3})^2$. Although the initial particle population satisfies a monodispersed

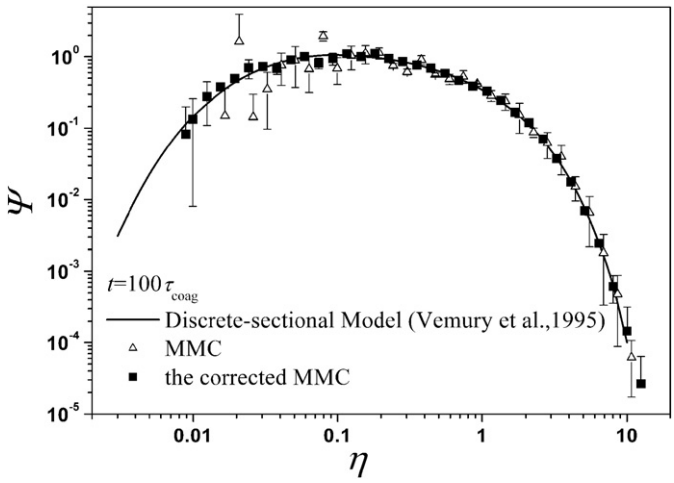

Fig. 2. The self-preserving particle size distribution for Brownian coagulation in free-molecular regime.

distribution, the self-preserving PSD with less populated edges and densely population centre will reach after a time-lag. In the self-preserving formulation, the dimensionless particle volume is defined as $\eta = Nv/M$, and the dimensionless distribution as $\psi = Mn(v,t)/N^2$[10], where $n(v,t)$ is the particle size distribution function (PSDF) at time t. The MC results shown in Fig. 2 represent the average of 3 MC repetitions. The original MMC exhibits large numerical errors within the dimensionless volume range of 0.01 –0.1, although the number density in these size intervals is comparatively high and thus should have enough statistical samples in theory. In fact, as for the i–j coagulation event, the original MMC makes large particles with number of $(w_i + w_j)/2$, which is greater than the right value, i.e., $2w_iw_j/(w_i + w_j)$. The consequential treatment in the original MMC leads to that the MMC is unable to correctly simulate these small-scale particles and overestimates number concentration of these large-scale particles. On the contrary, the present method correctly treats the coagulation event between two differentially-weighted simulation particles and then performs good precision with respect to the dynamic evolution of not only large-scale particles but also small-scale particles.

4. Conclusions

The original MMC method exhibits comparatively high numerical errors, which is mainly originated from the incorrect consequential treatment of a coagulation event. From this point, the paper overcame the conceptual difficulty of imagining a coagulation event between two differentially-weighted simulation particles and then correctly dealt with the result of a coagulation event, satisfying the basic laws of mass conservation and number depletion and then resulting in good prediction for particle size distribution. The corrected MMC method can further applied in multivariate and multidimensional population balance modelling.

Table 1
The mean standard deviations of mass concentration, mass concentration and size distribution in several MC methods.

Cases	Time-averaged standard deviations	Stepwise constant-V	Constant-N	Time-driven DSMC	MMC	The corrected MMC
Constant kernel	$\overline{\sigma}_N(t = 1000\tau_{coag})$	3.102×10^{-15}	2.097×10^{-2}	2.106×10^{-2}	4.803×10^{-2}	4.648×10^{-2}
	$\overline{\sigma}_M(t = 1000\tau_{coag})$	0	0	7.648×10^{-3}	1.056×10^{-1}	0
	$\overline{\sigma}_{P_k}(t = 1000\tau_{coag})$	1.677×10^{-3}	1.633×10^{-3}	1.695×10^{-3}	2.097×10^{-3}	1.404×10^{-3}
Linear kernel	$\overline{\sigma}_N(t = \tau_{coag})$	1.965×10^{-4}	1.006×10^{-2}	2.019×10^{-2}	1.238×10^{-2}	8.776×10^{-3}
	$\overline{\sigma}_M(t = \tau_{coag})$	0	0	1.530×10^{-3}	1.366×10^{-2}	0
	$\overline{\sigma}_{P_k}(t = \tau_{coag})$	4.159×10^{-3}	3.306×10^{-3}	5.633×10^{-3}	2.704×10^{-3}	2.134×10^{-3}
Product kernel	$\overline{\sigma}_N(t = \tau_{coag})$	5.398×10^{-4}	9.609×10^{-3}	1.886×10^{-2}	6.733×10^{-3}	6.307×10^{-3}
	$\overline{\sigma}_M(t = \tau_{coag})$	1.424×10^{-4}	0	3.321×10^{-4}	1.676×10^{-2}	0
	$\overline{\sigma}_{P_k}(t = \tau_{coag})$	5.126×10^{-3}	2.652×10^{-3}	3.532×10^{-3}	1.709×10^{-3}	1.490×10^{-3}

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