The following publication Liu, S. and Chan, T.L. (2017), "A stochastically weighted operator splitting Monte Carlo (SWOSMC) method for the numerical simulation of complex aerosol dynamic processes", International Journal of Numerical Methods for Heat & Fluid Flow, Vol. 27 No. 1, pp. 263-278 is published by Emerald and is available at https://doi.org/10.1108/HFF-08-2015-0335.

A stochastically weighted operator splitting Monte Carlo (SWOSMC) method for the numerical simulation of complex aerosol dynamic processes

Shuyuan Liu[1] and Tat Leung Chan[1,2,*]

1. Department of Mechanical Engineering, The Hong Kong Polytechnic University, Kowloon, Hong Kong

2. The Hong Kong Polytechnic University, Shenzhen Research Institute, Shenzhen, PR China

*Corresponding author at mmtlchan@polyu.edu.hk (T. L. Chan)

ABSTRACT

Purpose - The purpose of this paper is to study the complex aerosol dynamic processes by using this newly developed stochastically weighted operator splitting Monte Carlo (SWOSMC) method.

Design/methodology/approach – Stochastic weighted particle method and operator splitting method are coupled to formulate the SWOSMC method for the numerical simulation of particle-fluid systems undergoing the complex simultaneous processes.

Findings - This SWOSMC method is first validated by comparing its numerical simulation results of constant rate coagulation and linear rate condensation with the corresponding analytical solutions. Coagulation and nucleation cases are further studied whose results are compared with the sectional method in excellent agreement. This SWOSMC method has also demonstrated its high numerical simulation capability when used to deal with simultaneous aerosol dynamic processes including coagulation, nucleation and condensation.

Originality/value –There always exists conflict and tradeoffs between computational cost and accuracy for Monte Carlo based methods for the numerical simulation of aerosol dynamics. Operator splitting method has been widely used in solving complex partial differential equations while stochastic weighted particle method has been commonly used in numerical simulation of aerosol dynamics. However, the integration of these two methods has not been well investigated.

Keywords: Stochastic weighted particle method, operator splitting, simultaneous aerosol dynamics

Paper type: Research paper

1. Introduction

Numerical simulation has gained increasing attention in dealing with comprehensive engineering and scientific problems which are governed by the aerosol dynamics e.g. the production of industrial nanoparticles (Chamkha and Rashad, 2012; Hao *et al.*, 2013; Tu and Zhang, 2014) and the control of aircraft particulate emission (Vacassel *et al*., 2014). Aerosol dynamics involves complex physical or chemical processes such as coagulation, nucleation and condensation (Zhang *et al*., 1999) and surface reactions (Frenklach and Wang, 1991). Various numerical methods are thus developed to deal with these complex aerosol dynamic processes (Efendiev, 2004; Yu *et al*., 2009; Chan *et al*., 2010; Zhou and Chan, 2011; Geng *et al*., 2013; Yin and Liu, 2013; Zhou and Chan, 2014; Yu and Chan, 2015; Fede *et al*., 2015; Zhang and You, 2015). Reducing the modelling complexity and improving numerical simulation accuracy have always been the research focus of aerosol dynamics (Kumar *et al*., 2011). Monte Carlo based methods offer an option to solve the particle General Dynamics Equation (GDE) through the stochastic probability events (Gillespie, 1972; Debry *et al.*, 2003; Zhou and He, 2014; Xu et al., 2015) and obtain the evolution history of particles. Generally, no input information of initial particle size distribution shape is required for Monte Carlo simulation except for some special problems. Another advantage of Monte Carlo based methods lies in that any number of particle properties can be described as a combination between Lagrangian transport, particle interaction due to coagulation and individual particle events (Kruis *et al.*, 2012). However, high computational cost and limited accuracy have been the major shortcomings of Monte Carlo based methods (Liffman, 1992; Wei and Kruis, 2013) since the computational accuracy of Monte Carlo based methods is highly dependent on the number of numerical particles that are used. It also implies that a large number of numerical particles is required for obtaining high computational accuracy. Instead of selecting all aerosol dynamic processes (i.e. nucleation, condensation and coagulation) randomly at a time as in traditional Monte Carlo simulation, the operator splitting concept is used in recently developed methods (Patterson *et al*. 2006 and Zhou *et al*. 2014). The main idea of operator splitting method is to separately deal with coagulation and other deterministic processes such as nucleation with different methods, respectively. Additionally, the weighting particles are also used for the numerical simulation of spatially resolved particle-fluid systems. In contrast to the fixed and unique weights of numerical particles in conventional Monte Carlo based methods, different and variable weights are introduced to numerical particles in weighted Monte Carlo based methods to reduce the numerical diffusion caused

by the spatial inhomogeneity or wide spectrum of particle size distribution (Rjasanow and Wagner, 1996; Zhao *et al*., 2009; Patterson *et al*., 2011). A new Stochastically Weighted Operator Splitting Monte Carlo (SWOSMC) method is proposed in the present study to solve the complex aerosol dynamics with high computational efficiency and accuracy. This new SWOSMC method is first validated by both analytical solutions and sectional method (Prakash *et al*., 2003) and then is applied to typical cases study including simultaneous coagulation, nucleation and condensation processes.

2. Methodology

2.1 General dynamics equation

The governing equation of the time dependent evolution of number density $n(v,t)$ for a single component aerosol can be written as (Debry *et al*., 2003):

$$\begin{aligned}\frac{\partial n}{\partial t}(v,t) &= 1/2\int_{v_0}^{v-v_0} K(u,v-u)n(u,t)n(v-u,t)du \\ &-n(v,t)\int_{v_0}^{\infty} K(u,v)n(u,t)du + \frac{\partial(I_0 n)}{\partial v}(v,t) + \delta(v_0,v)J_0(t)\end{aligned} \tag{1}$$

where $K(u,v)$, $I_0(u,v)$ and $J_0(t)$ are the coagulation, condensation and nucleation kernels, respectively. Compared to the original Smoluchowski's equation which deals only with coagulation, the extended Smoluchowski's equation describes more physical processes which exchange mass between individual particles and the environment such as the fluid in which particles are contained and new particles are also introduced into the particle population (Patterson *et al*., 2011).

2.2 Operator splitting

For complex aerosol dynamic processes, the terms on the right hand side of Equation (1) may include terms of different physical processes besides coagulation term. Operator splitting is very efficient in solving such complex equation. Instead of integrating all these aerosol dynamic processes together in one step, the operator splitting method separates the integration into multiple steps, such as:

$$\exp(\Delta t X) = \exp(\Delta t X_d)\exp(\Delta t X_s) + O(\Delta t^2) \tag{2}$$

$$= \exp(1/2\Delta t X_d)\exp(\Delta t X_s)\exp(1/2\Delta t X_d) + O(\Delta t^3) \tag{3}$$

$$= \exp(1/2\Delta t X_s)\exp(\Delta t X_d)\exp(1/2\Delta t X_s) + O(\Delta t^3) \tag{4}$$

where X_d denotes nucleation and condensation processes which are solved by deterministic integration

method, while X_s denotes coagulation process which is solved by stochastic method (Monte Carlo method). Equation (2) is of first-order accuracy while Equations (3) and (4) are of second-order accuracy.

2.3 Aerosol dynamics kernels

For free molecule regime, the coagulation kernel, $K(u,v)$ can be written as (Zhou *et al*., 2014):

$$K(u,v) = (\frac{6}{\pi})^{\frac{2}{3}}(\frac{\pi k_B T_K}{2\rho})^{\frac{1}{2}}(\frac{1}{u}+\frac{1}{v})^{\frac{1}{2}}\left(u^{\frac{1}{3}}+v^{\frac{1}{3}}\right)^2 \tag{5}$$

where u and v are the volume of the spherical colliders; T_K is the temperature, k_B is the Boltzmann's constant and ρ is the density of particles.

In continuous regime, for spherical particles of size u and v, the coagulation kernel can be written as (Debry *et al*., 2003):

$$K(u,v) = 2k_B T_K / 3\mu_{air}[2+(v/u)^{1/3}+(u/v)^{1/3}] \tag{6}$$

where μ_{air} is the viscosity of air and the other parameters have the same physical meanings as that in Equation (5).

The homogeneous nucleation rate can be generally written as (Seinfeld and Pandis, 1998):

$$J_0(t) = C\exp(-\frac{\Delta G^*}{k_b T}) \tag{7}$$

where ΔG^* is the free energy that is required to form a stable nucleus and C is a constant which is related to vapor pressure.

As condensation/evaporation process involves the relaxation to an equilibrium state between aerosol and gas phases for one chemical species, the kernels are thus proportional to the pressure difference between the bulk gas and the equilibrium pressure:

$$I_0(v,t) = C_I \exp(p_i^{\infty} - p_i^{eq}) \tag{8}$$

where C_I is a constant which is related to the diffusion species and temperature, p_i^{∞} and p_i^{eq} are the vapor pressure and the equilibrium vapor pressure of species i from the particle, respectively.

2.4 Non-dimensionalization

The GDE Equation (1) is non-dimensionalized based on the relative mass density, $\tilde{q}(v,t)$ of aerosol particles (Debry *et al*., 2003) which can be expressed as Equation (9):

$$\tilde{q}(v,t)=\frac{q(v,t)}{Q_0},\quad Q_0=\int_{v_0}^{\infty}q_0(v)dv \tag{9}$$

where $q(v,t)$ is the mass density of aerosol particles and Q_0 is the total initial mass of aerosol particles.

Substituting $n(v,t)$ into Equation (1) with $\tilde{q}(v,t)$, it can be expressed as (Debry *et al*., 2003):

$$\begin{aligned}&\frac{\partial\tilde{q}}{\partial t}(v,t)=1/2\int_{v_0}^{v-v_0}Q_0\frac{K(u,v-u)}{v-u}\tilde{q}(u,t)\tilde{q}(v-u,t)du\\&-\tilde{q}(v,t)\int_{v_0}^{\infty}Q_0\frac{K(u,v)}{u}\tilde{q}(u,t)du+\frac{I_0(v,t)\tilde{q}(v,t)}{v}\\&-\frac{\partial(I_0\tilde{q})}{\partial v}(v,t)+\frac{v_0J_0(t)}{Q_0}\delta(v_0,v)\end{aligned} \tag{10}$$

2.5 Algorithm formulation

The main idea of the present method is to introduce stochastic weights to various numerical particles according to the mass change caused by different aerosol dynamic processes in order to increase the numerical stability of Monte Carlo method. Operator splitting technique is used to treat stochastic process (i.e., coagulation) and deterministic processes (i.e., condensation, nucleation etc.) separately with corresponding methods to reduce computational time for the simulation of complex aerosol dynamics. The idea of using numerical particles with varying mass weights (Debry *et al*., 2003) is adopted herein, in which the *i*-th numerical particle is associated with a varying mass weight, $w_i(t)$ of real aerosol particle of size, $y_i(t)$, thus the *i*-th numerical particle now stands for a number of $w_i(t)/y_i(t)$ of real aerosol particles. The introduction of such a varying mass weight to numerical particles in stochastic simulation of simultaneous aerosol dynamic processes is necessary and well justified. When mass weights are adhered to numerical particles i.e. numerical particles are connected with a certain mass of real aerosols, the total number of numerical particles remains constant and no re-sampling is needed for coagulation process (Eibeck and Wagner, 2001). This is because the total mass of real aerosol particles remains constant in coagulation process although the total number of real aerosol

particles decreases. If the numerical particles are directly connected with the number of real aerosol particles and no further up sampling (adding new particles to the particle-fluid system) is conducted, the decreasing number of numerical particles may cause severe numerical diffusion as coagulation converges on infinite particle number. However, for some other aerosol dynamic processes including nucleation, condensation, evaporation, deposition and removal, the total mass of aerosol particles in the particle-fluid system actually varies with time. By introducing varying mass weights to numerical particles, the weights, $w_i(t)$ will evolve with time for mass-varying process such as condensation and evaporation. In here, nucleation is conducted by creating a certain mass of new particles according to nucleation rate which is independent of pre-existing particles. The integration details for individual process will be presented below. The main algorithm of stochastically weighted operator splitting Monte Carlo (SWOSMC) method over a time period [0, T] is presented as follows:

(1) Initialization: setting of the following quantities:

$$[\left(y_i^0, w_i^0\right),\ i = 1, 2, ..., N_0];$$

(2) Operator splitting over time loop [0, T]:

Integration of Equation (10) from t_k to $t_{k+1} = t_k + \tau_k$, where τ_k is the time step determined previously:

(i) Integration of coagulation based on Monte Carlo based methods (Gillespie, 1972);

(ii) Integration of condensation using an ODE solver from (Zhou *et al*., 2014);

(iii) Integration of nucleation: creation of new particles, J;

(3) Updating the particle-fluid system; and

(4) When $t > T$, STOP and averaging on the results.

3. Numerical setup

3.1 *Time step*

According to Debry *et al*., (2003), the time scales for different physical processes can be calculated as Equations (11-13) in order to allow an accurate integration result as well as avoid too much computational time:

(1) For coagulation between aerosols of size y_i^k, y_j^k:

$$\Delta t_{coag} = \frac{y_j^k}{Q_0 w_j^k K(y_i^k, y_j^k)} \tag{11}$$

(2) For condensation/evaporation of aerosols of size y_j^k

$$\Delta t_{cond/evap} = \frac{y_j^k}{I_0(y_i^k, t_k)} \tag{12}$$

(3) For nucleation of aerosols :

$$\Delta t_{nucl} = \frac{Q_0}{v_0 J_0(t)} \tag{13}$$

To ensure an accurate integration, the time step should be less than the minimum of all the time scales from Equations (11) to (13) is set as the minimum time step.

3.2 Integration details

The system state after a time step is calculated by integrating over a time step. Noting that a varying mass weight is used, only condensation/evaporation processes that cause the mass change to the particle-fluid system will have numerical particles with varied weights.

(1) For integration of coagulation, the collision criteria should be met. The volume of particle *i* after a splitting time step becomes the total volume of particle *i* and its collision partner while the weight of *i*-th numerical particle remains unchanged. For the *i*-th numerical particle at *k*-th time step, the integration procedure is written as:

$$y_i^{k+1/2} = y_i^k + y_{Ji}^k, \text{ if } r \le \frac{Q_0 w_{Ji}^k K(y_i^k, y_{Ji}^k)}{y_{Ji}^k} \tau_k, \quad \tau_k = \min\left(\frac{y_{Ji}^k}{Q w_{Ji}^k K(y_i^k, y_{Ji}^k)}\right) c \tag{14}$$

where $y_i^{k+1/2}$, y_i^k and y_{Ji}^k are the volume size after the collision step, the volume of *i*-th particle and the volume collided with *i*-th particle at k-th time step, respectively. For the criterion part, r is a random number uniformly distributed over [0, 1], τ_k is the defined time step based on the minimum coagulation time scale, $\frac{Q_0 w_{Ji}^k K(y_i^k, y_{Ji}^k)}{y_{Ji}^k} \tau_k$ is the collision probability and c is a constant which is usually equal to 0.1 (Debry et al., 2003). τ_k is so defined that the collison probability is within [0, 1].

(2) For integration of condensation, it is performed via integration using a self-adaptive fifth order Runge-Kutta method (Zhou *et al*., 2014) over the splitting time step of varying weight function, which is determined by the mechanistic rate of condensation. For the *i*-th numerical particle at *k*-th time step, the results obtained from the above coagulation step are used as input for this step, the integration procedure is written as:

$$\begin{aligned} &\frac{\mathrm{d}y_i}{\mathrm{d}t} = I_0(y_i,t),\ \frac{\mathrm{d}w_i}{\mathrm{d}t} = w_i\frac{I_0(y_i,t)}{y_i} \\ &y_i^{k+1} = y_i^{k+1/2} + \Delta t I_0(y_i^{k+1/2},t_k),\ w_i^{k+1} = w_i^{k+1/2}\frac{y_i^{k+1}}{y_i^{k+1/2}} \end{aligned} \tag{15}$$

where y_i^{k+1} and $y_i^{k+1/2}$ are the volume size, w_i^{k+1} and $w_i^{k+1/2}$ are the weights, $I_0(y_i^{k+1/2},t_k)$ and $I_0(y_i,t)$ are the condensation rates after the condensation step and that obtained from the previous coagulation step for the *i*-th particle and the *k*-th time step, respectively. Δt is the integration time step.

(3) For integration of nucleation, only a certain mass of new particles with the minimum nucleus volume v_0 are created and added to the particle-fluid system. For the *i*-th newly created particle, it is defined as:

$$y_i^{k+1} = v_0,\ w_i = 1,\ m_c = v_0 J_0(t_k)\Delta t \tag{16}$$

where y_i^{k+1} is the volume size of the *i*-th newly created particle, w_i is the weight of the *i*-th newly created particle, $J_0(t_k)$ is the nucleation rate mass of of the *i*-th newly created particle, m_c is the total mass of all the newly created particles within one time step and v_0 is the initial volume size of the newly created particles.

3.3 Initial conditions and cases with analytical solutions

3.3.1 Initial conditions

The initial particle number density is 1.0/m^3 and initial particle dimeter is set as 1.24 nm for Case 1 (constant rate coagulation and linear rate condensation case) and Case 2 (constant rate coagulation and

nucleation) so that the following simple dimensionless expressions for moments can be obtained. The initial conditions of the other cases in the present study can be found in Frenklach and Harris (1987).

3.3.2 Constant rate coagulation and linear rate condensation

Analytical solutions to the GDE Equation (1) is only available for very limited cases, among which the case of constant rate coagulation and linear rate condensation is selected as the first validation case. For constant rate coagulation and linear rate condensation, when the coagulation and condensation kernel are both set as unity for simplicity, the number density $n(v,t)$ and the dimensionless zeroth moment, M_0 and first moments, M_1 with respect to the particle volume can be derived as (Ramabhadran *et al*., 1976):

$$\begin{aligned} n(v,t) &= \frac{M_0(t)^2}{M_1(t)} \exp(-\frac{M_0(t)}{M_1(t)} v) \\ M_0(t) &= \frac{2}{2+t}, \\ M_1(t) &= \exp(t), \end{aligned} \tag{17}$$

3.3.3 Constant rate coagulation and nucleation

If the coagulation rate and nucleation rate are constant (both are set as unity for simplicity in the present study), an analytical solution is available to Equation (1). The analytical solutions of relative particle number density and relative particle volume concentration can be expressed as (Maisels *et al*. 2004):

$$N / N_0 = B \frac{1 + B \tanh(\tau_0 / 2)}{B + \tanh(\tau_0 / 2)} \tag{18}$$

$$V / V_0 = 1 + \frac{B}{2} \tau_0 \tag{19}$$

where V and V_0 are the total volume of particles at time t and at initial time, respectively. N and N_0 are the number density of particles at time t and at initial time, respectively. B, E and τ_0 are dimensionless parameters determined by the initial conditions and the detailed expressions can be found in Maisels *et al*. (2004).

3.3.4 Simulataneous coagulation, nucleation and condensation

For simultaneous aerosol dynamic processes, if the nucleation rate, coagulation rate, condensation rate and monomer concentration are constant, the analytical solution to these simultaneous processes including coagulation, nucleation and condensation ***exists*** (Maisels *et al*., 2004). The analytical expression of relative particle number denstity is given in Equation (18) since condensation involves no change in the particle number. The analytical expression of the relative particle volume concentration for simulataneous coagulation, nucleation and condensation is written as:

$$V/V_0=1+[(\frac{B}{2}+E)\tau_0+2E\ln(\frac{1+\exp(-\tau_0)}{2}+\frac{1-\exp(-\tau_0)}{2B})] \tag{20}$$

where all the parameters in Equation (20) have the same physical meanings with those in Equation (18) and (19). More information of this case study can be found in Liu *et al.* (2015).

3.4 Calculation of maximum relative quadratic error

The maximum relative quadratic error used to evaluate the numerical simulation results is defined as:

$$\varepsilon=\{\sqrt{[(X(t)-X_0(t))/X_0(t)]^2}\}_{\max} \tag{21}$$

where ε is the maximum relative error, $X(t)$ is the numerical simulation results obtained with the proposed SWOSMC method, and $X_0(t)$ is the reference value for comparison.

4. Results and discussion

4.1 Initial validation

The SWOSMC method is first validated for constant rate coagulation and linear rate condensation processes. The dimensionless zeroth moment, M_0 which is proportional to the number density of particles, is shown in Figure 1. An increasing number of numerical particles are used for the numerical simulation. It can be seen that the numerical simulation results agree well with the analytical solution when only 1000 numerical particles are used, which shows the good capability of the SWOSMC method in dealing with simultaneous coagulation and condensation processes.

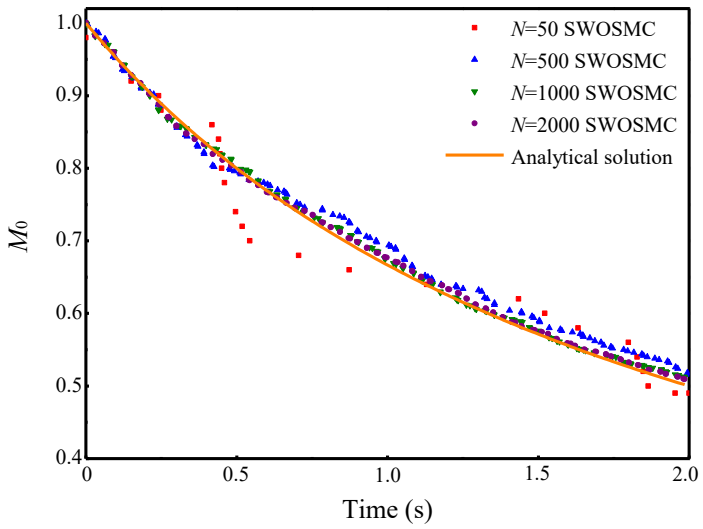

Figure 1. Zeroth moment, M_0 under coagulation and condensation processes for SWOSMC versus analytical solution (Ramabhadran *et al*., 1976) (N is the number of numerical particles (used in each simulation run).

An excellent agreement between the dimensionless first moment, M_1 obained by the SWOSMC method and analytical solution is also observed is Figure 2. The exponentially increasing M_1 with respect to simulation time represents the rapid increase of the total volume of particles in the numerical simulation particle-fluid system due to condensation.

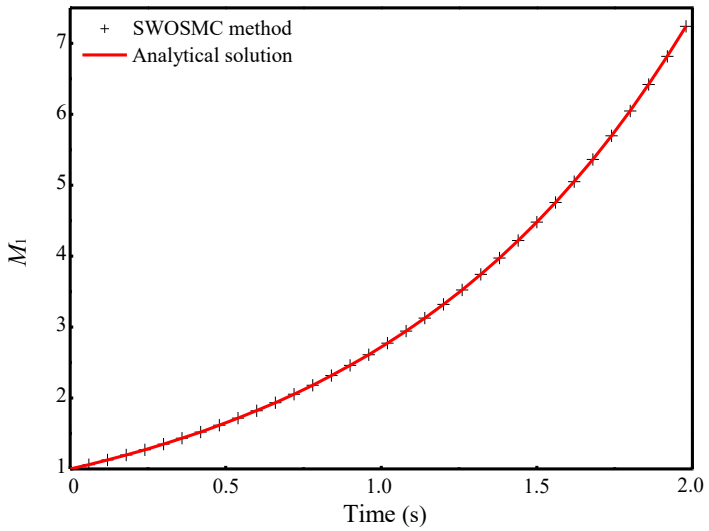

Figure 2. First moment, M_1 under coagulation and condensation processes for SWOSMC versus analytical solution (Ramabhadran *et al*., 1976) (The number of numerical particles used in each simulation run is 1000).

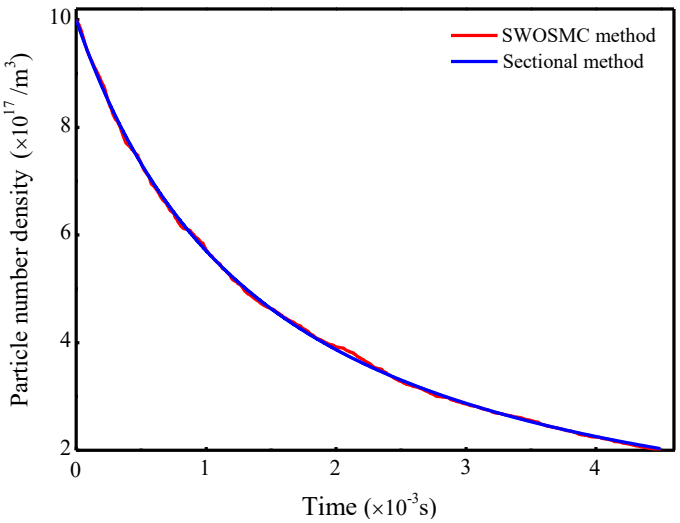

Figure 3. Particle number density under free molecule regime coagulation for SWOSMC versus sectional method (Prakash *et al*., 2003) (The number of numerical particles used in each simulation run is 1000).

Figure 3 shows another application of the SWOSMC method to free molecule regime coagulation and the numerical simulation results is validated by comparing to the sectional method (Prakash *et al*., 2003). Excellent agreement can be also observed between the particle number density obtained by these two methods with the maximum relative quadratic error (taking the sectional method as reference) less than 1% during the whole simulation time.

4.2 Constant rate coagulation and nucleation

For constant rate coagulation and nucleation, an increasing number of numerical particles are used in the SWOSMC method. Figure 4 shows the relative particle number density N/N_0 obtained via the SWOSMC method and the analytical solution (Maisels *et al*., 2004), respectively. It can be seen that the SWOSMC method agrees well with the analytical solution with the increase of the number of numerical particles. The increasing relative particle number density implies the nucleation is dominant within the simulation time. This is because the initial particle number density is set as low as 1.0 /m^3 for simplicity of the validation. As the simulation proceeds, some statistical fluctuations can be observed, but the maximum relative quadratic error compared with the analytical solution remains less than 2% during the whole simulation time, which also proves the reliability of this newly proposed SWOSMC method.

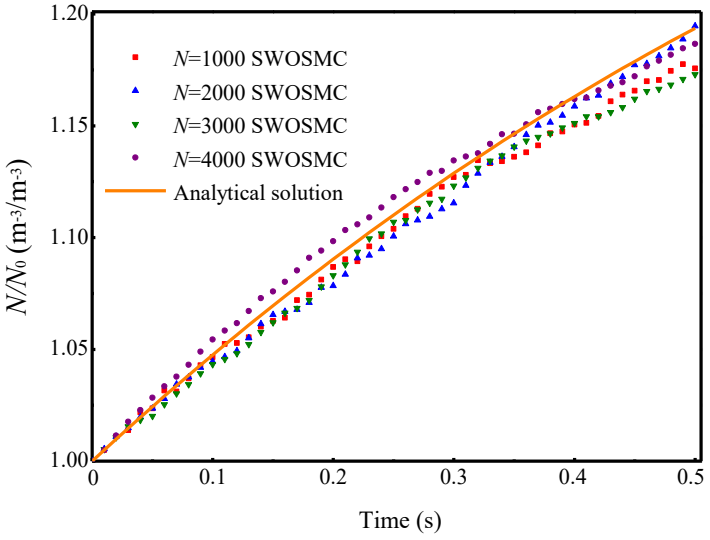

Figure 4. Relative particle number density, N/N_0 under constant rate coagulation and nucleation processes for SWOSMC versus the analytical solution (Maisels *et al*., 2004) (*N* is the number of numerical particles used in each simulation run).

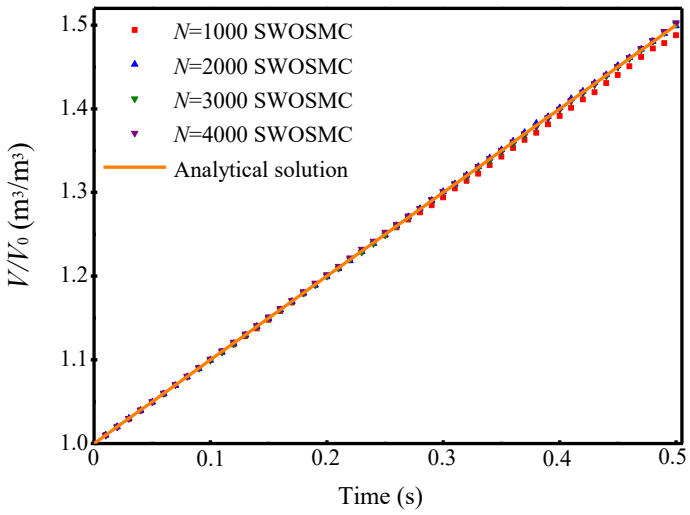

Figure 5. Relative particle volume concentration, V/V_0 under constant rate coagulation and nucleation processes for SWOSMC versus the analytical solution (Maisels *et al*., 2004)) (*N* is the number of numerical particles used in each simulation run).

The relative particle volume concentration, V/V_0 under the constant rate coagulation and nucleation is shown in Figure 5. The agreement between the SWOSMC method and the anlytical solution is so excellent that even for 1000 numerical particles, the maximum relative quadratic error is less than 1%. The linear increase in relative particle volume concentration can be well explained by the constant rate nucleation that continously creates new particles in the particle-fluid system. The SWOSMC method

has demonstrated the ability to reach high numerical simulation accuracy with acceptable number of numerical particles.

4.3 Free molecule regime coagulation and constant rate nucleation

Figure 6 shows the evolution history of particle number density of simultaneous free molecule regime coagulation and constant rate nucleation obtained via the SWOSMC method and the sectional method (Prakash *et al*., 2003). A very satifactory agreement can be found between the two methods for even 100 numerical particles used for the SWOSMC method, which demonstrates the good applicability and computational efficency of the SWOSMC method in solving simultaneous free molecule regime coagulation and constant nucleation problem. As the number of numerical particles increases from 100 to 2000, the maximum relative quadratic error between the two methods remains basically the same, which implies that 100 numerical particles is already enough to reach very high computational accuracy when compared with the sectional method. Some fluctuations can be found for numerical simulation with different number of numerical particles e.g. the simulation results with 500 numerical particles are closer to the results via the sectional method than that using 1000 particles at the initial stage of simulation, specifically before 2×10^{-3}s. This may be explained by the statistical unstability of the particle-fluid system at the initial stage. Similar fluctuations can also be found in Figure 7.

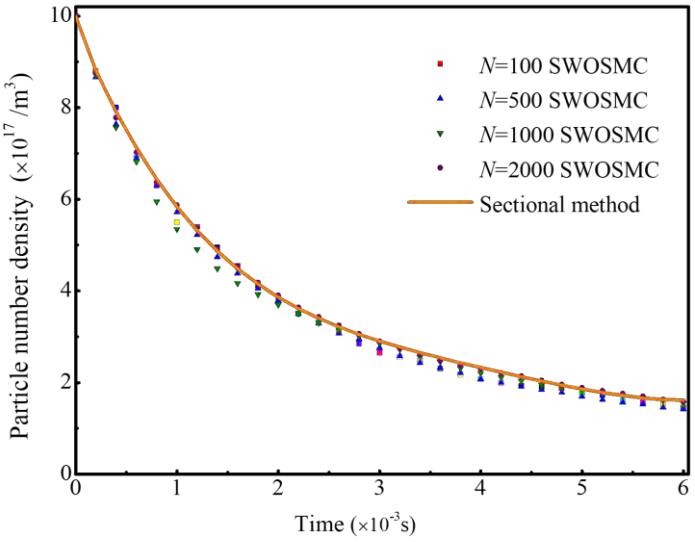

Figure 6. Particle number density under free molecule regime coagulation and constant rate nucleation for SWOSMC versus sectional method (Prakash *et al*., 2003) (*N* is the number of numerical particles used in each simulation run).

The evolution history of number average diameter of particles, d_{ave} are tracked and shown in Figure 7. An increasing number of numerical particles from 100 to 2000 are used in the SWOSMC simulation. The number average diameter of particles shows good agreement between the SWOSMC method and the sectional method. A rapid increase of the average diameter is observed duet to nucleation and coagulation. With the increase of the number of numerical particles, the maximum relative error between the two methods signifcantly decreases, reaching far less than 1% when only 500 numerical particles are used.

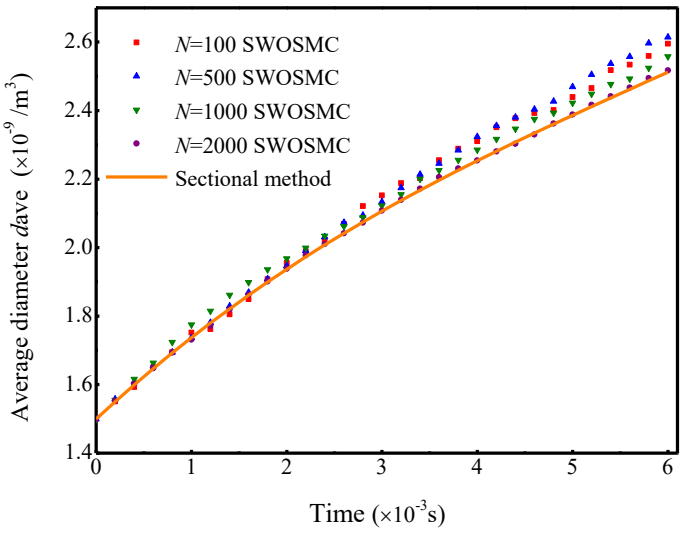

Figure 7. Average diameter of the numerical particles, d_{ave} under free molecule regime coagulation and constant rate nucleation via SWOSMC and the sectional method (Prakash *et al*., 2003) (N is the number of numerical particles particles used in each simulation run).

The second moment, M_2 with respect to the particle size distribution (PSD) is shown in Figure 8. As particles are continually created by nucleation process, the second moment shows rapid increase once the nucleation process begins. With the increase of the number of numerical particles, the maximum relative quadratic error relative to the sectional method (Prakash *et al*., 2003) decreases rapidly to less than 1% when only 500 numerical particles are used. However, no significant improvement is achieved with the number numerical particles increasing from 500 to 2000. It indciates that 500 numerical particles are enough to obtain as high computational accuracy as that via the sectional method (Prakash *et al*., 2003). It can be seen from Figure 8 that the SWOSMC method is promising to capture the evolution of high-order moments with relatively low computational cost.

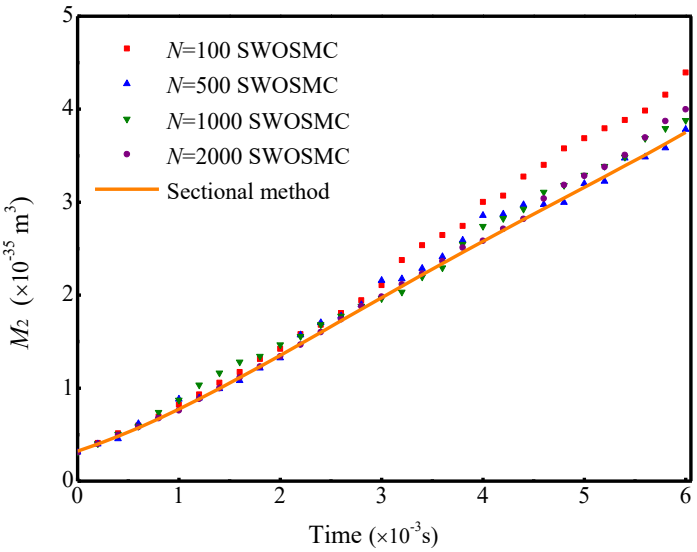

Figure 8. Second moment, M_2 of the particles under free molecule regime coagulation and constant rate nucleation processes for SWOSMC versus sectional method (Prakash *et al*., 2003) *(N* is the number of numerical particles used in each simulation run).

4.4 Simultaneous coagulation, nucleation and condensation processes

The details of this case study can be found in Liu *et al.* (2015). The particle volume concentration, V/V_0 of simultaneous aerosol dynamic processes is shown in Figure 9 and is validated with the analytical solution obtained from Maisels *et al*. (2004). Coagulation, nucleation and condensation processes are simulated simultaneously. It can be seen that the numerical simulation results fit very well with the analytical solution for the selected case**.** The particle volume concentration increases linearly with the dimensionless time, which is also consistent with the theoretical expression given in Equation (20). As the simulation time is very short, the nonlinear term in the theoretical expression in Equation (20) can be neglected, which yields a linear relationship between the particle volume concentration and dimensionless time, as shown in Figure 9.

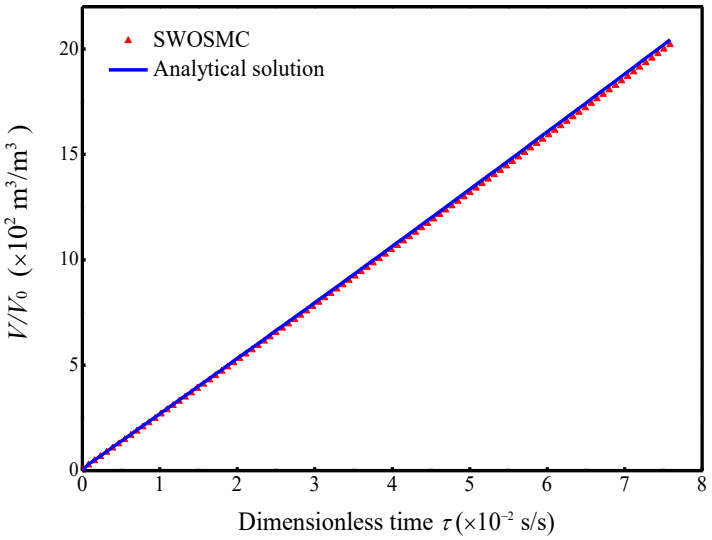

Figure 9. Particle volume concentration, V/V_0 under simultaneous aerosol dynamic processes for SWOSMC versus analytical solution from Maisels *et al*. (2004) (The number of numerical particles, N used in each simulation run is 4000).

The particle number density, N/N_0 of this simultaneous aerosol dynamic processes is shown in Figure 10. For the reason of short simulation time, the theoretical expression in Equation (18) also approximates a linear relationship between the particle number density and the dimensionless time. However, even with small simulation time and only 4000 numerical particles, the results obtained via the SWOSMC method agree well with the analytical solution, which shows the potential of this method in solving simultaneous full processes in complex aerosol dynamics.

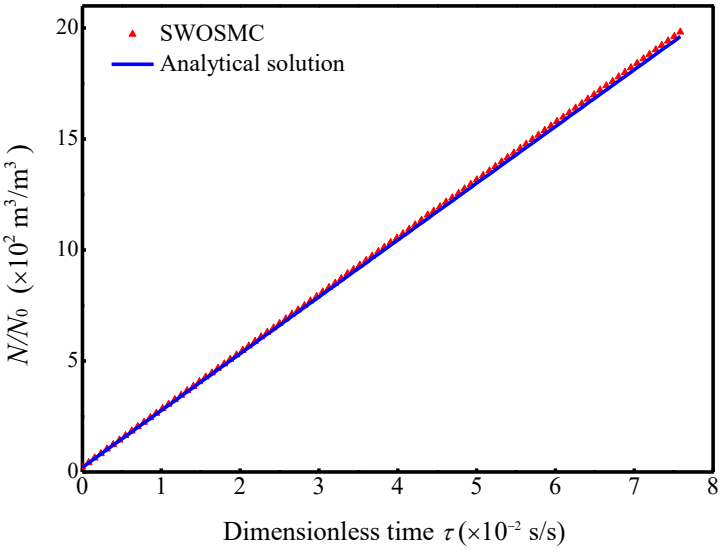

Figure 10. Particle number concentration, N/N_0 under simultaneous aerosol dynamic processes for SWOSMC versus analytical solution from Maisels *et al*. (2004) (The number of numerical particles, N used in each simulation run is 4000).

4.5 Parametric analysis of the studied cases

All the studied cases in the present study are listed in Table 1. These cases are selected to represent the common aerosol dynamic processes taking place in the actual particle-fluid systems such as particulate emission from vehicles, aerosol formation, collodial solution, particulate emission from industrial boilers etc. The cases are so arranged that the complexity of simulation cases increases from Case 1 to Case 4, which is used for the evaluation of simulation accuracy and efficiency of the SWOSMC method.

Table 1. Summary of the studied cases.

Cases	Description
Case 1	Constant rate coagulation and linear rate condensation for validation with analytical solution. (The free molecule regime coagulation is only shown a part of Case 1.)
Case 2	Constant rate coagulation and constant rate nucleation.
Case 3	Free molecule regime coagulation and constant rate nucleation.
Case 4	Simultaneous aerosol dynamic processes including coagulation, nucleation and condensation.

The main simulation parameters are presented in Table 2 for evaluation purpose of this proposed new SWOSMC method. It can be seen that with the increase of the complexity from Case 1 to Case 4, an increasing computational time is needed for a fixed number of numerical particles. However, for a certain case, the computational time is approximately proportional to the square root of the number of numerical particles, which is consistent with the results obtained by Liffman (1992) and Wei *et al*. (2013). It also suggests that further optimization of this new SWOSMC method is needed in order to improve the computauonal efficiency and accuracy for the complex particle-fluid systems. Compared with the sectional method (Prakash *et al*., 2003), the SWOSMC method takes shorter simulation time even with the largest number of numerical particles in Case 3. The SWOSMC method has

demonstrated its capability to obtain higher computaional accuracy with shorter simulation time than the sectional method for the same case.

Table 2. Analysis of main simulation parameters.

Cases	Number of numerical particles	Normalized computational time	Maximum relative quadratic error (%)
Case 1	50	1	11
	500	3	5
	1000	5	2
	2000	7.5	<1
Case 2	1000	5	3.7
	2000	7	3
	3000	9	2
	4000	11	<1
Case 3	100	1.5	6.5
	500	4	5.3
	1000	6	<1
	2000	9	<1
	Sectional method	>100	-
Case 4	4000	45	<1

Note: Cases 1, 2 and 3 are evaluated by the simulation results of zeroth moment, M_0 while the particle number concentration is considered for Case 4. The maximum relative error is calculated according to Equation (21). The normalized computaional time is defined as the ratio of any computaional time to the computational time of Case 1 with 50 numercial particles.

5. Conclusions

The simulation results of this new Stochastically Weighted Operator Splitting Monte Carlo (SWOSMC) method are fully validated with corresponding analytical solution and the sectional method (Prakash *et al*., 2003) for varied aerosol dynamic processes (i.e., coagulation, condensation and nucleation) in different flow regimes. This SWOSMC method also offers higher numerical simulation capability of solving simultaneous aerosol dynamic processes occurring in complex particle-fluid systems. Further testing and optimization of this method will be conducted to achieve higher computational efficiency and accuracy in solving complex particle-fluid system problems.

Acknowledgement

The first author, Shuyuan Liu is grateful to Professor Kun Zhou, Wuhan University of Science and Technology, China who kindly shared his developed source codes of the Operator Splitting Monte Carlo (OSMC) method in Zhou et al. (2014) to further develop this new SWOSMC method for the present study. This work was supported by the Central Research Grants and the research studentship of The Hong Kong Polytechnic University, Hong Kong. Last but not least, the authors also greatly appreciate The National Natural Science Foundation of China (Project No. 11572274) for allowing the further study of aerosol dynamics.

References

Chamkha, A. J. and Rashad, A. M. (2012), "Natural convection from a vertical permeable cone in a nanofluid saturated porous media for uniform heat and nanoparticles volume fraction fluxes", *International Journal of Numerical Methods for Heat & Fluid Flow*, Vol. 22 No. 8, pp. 1073-1085.

Chan, T.L., Liu, Y.H. and Chan, C.K. (2010), "Direct quadrature method of moments for the exhaust particle formation and evolution in the wake of the studied ground vehicle", *Journal of Aerosol Science*, Vol. 41 No. 6, pp. 553-568.

Debry, E., Sportisse, B. and Jourdain, B. (2003), "A stochastic approach for the numerical simulation of the general dynamics equation for aerosols", *Journal of Computational Physics*, Vol.184 No. 2, pp. 649-669.

Efendiev, Y. (2004), "Modelling and simulation of multi- component aerosol dynamics", *Computational & Applied Mathematics*, Vol. 23 No. 2-3, pp. 401-423.

Eibeck, A. and Wagner, W. (2001), "Stochastic particle approximations for Smoluchowski's coagulation equation", *Annals of Applied Probability*, Vol. 11 No. 4, pp. 1137-1165.

Fede, P., Simonin, O. and Villedieu, P. (2015), "Monte-Carlo simulation of colliding particles or coalescing droplets transported by a turbulent flow in the framework of a joint fluid-particle pdf approach", *International Journal of Multiphase Flow*, Vol. 74, pp. 165-183.

Frenklach, M. and Harris, S.J. (1987), "Aerosol dynamics modelling using the method of moments", *Journal of colloid and interface science*, Vol. 118 No. 1, pp. 252-261.

Frenklach, M. and Wang, H. (1991), "Detailed modeling of soot particle nucleation and growth", *Symposium (International) on Combustion*, Vol. 23 No. 1, pp. 1559-1566.

Geng, J., Park, H. and Sajo, E. (2013), "Simulation of aerosol coagulation and deposition under multiple flow regimes with arbitrary computational precision", *Aerosol Science and Technology*, Vol. 47 No. 5, pp. 530-542.

Gillespie, D.T. (1972), "Stochastic coalescence model for cloud droplet growth", *Journal of the Atmospheric Sciences*, Vol. 29 No. 8, pp. 1496-1510.

Hao, X.M., Zhao, H.B., Xu, Z.W. and Zheng, C.G. (2013), "Population balance Monte Carlo simulation for gas-to-particle synthesis of nanoparticles", *Aerosol Science and Technology*, Vol.47 No.10, pp. 1125-1133.

Kruis, F.E., Wei, J.M., Zwaag, T. van der and Haep, S. (2012), "Computational fluid dynamics based stochastic aerosol modeling: combination of a cell-based weighted random walk method and a constant-number Monte-Carlo method for aerosol dynamics", *Chemical Engineering Science*, Vol. 70, pp. 109-120.

Kumar, P., Ketzel, Vardoulakis, M. S., Pirjola, L. and Britter, R. (2011), "Dynamics and dispersion modelling of nanoparticles from road traffic in the urban atmospheric environment-a review", *Journal of Aerosol Science*, Vol. 42 No. 9, pp. 580-603.

Liffman, K. (1992), "A direct simulation Monte-Carlo method for cluster coagulation", *Journal of Computational Physics*, Vol. 100 No. 1, pp. 116-127.

Liu S.Y., Chan T.L. and Zhou K. (2015), "A New Stochastically Weighted Operator Splitting Monte Carlo Method for Particle-fluid Systems", *ASME-ATI-UIT 2015 Conference on Thermal Energy Systems: Production, Storage, Utilization and the Environment (In Session: Computational Thermal-fluid Dynamics)*, May 17-20, 2015, Naples, Italy.

Maisels, A., Kruis, F.E. and Fissan, H. (2004), "Direct simulation Monte Carlo for simultaneous nucleation, coagulation, and condensation in dispersed systems", *Chemical Engineering Science*, Vol. 59 No. 11, pp. 2231-2239.

Patterson, R.I.A., Singh, J., Balthasar, M., Kraft, M. and Norris, J.R. (2006), "The linear process deferment algorithm: a new technique for solving population balance equations", *SIAM Journal of Scientific Computing*, Vol. 28 No. 1, pp. 303-320.

Patterson, R. I., Wagner, W., and Kraft, M. (2011), "Stochastic weighted particle methods for population balance equations", *Journal of Computational Physics*, Vol. 230 No. 19, pp. 7456-7472.

Prakash, A., Bapat, A.P. and Zachariah, M.R. (2003), "A simple numerical algorithm and software for solution of nucleation, condensation, and coagulation problems", *Aerosol Science and Technology*, Vol. 37 No. 11, pp. 892–898.

Ramabhadran, T.E., Peterson, T.W. and Seinfeld, J.H. (1976), "Dynamics of aerosol coagulation and condensation", *AIChE Journal*, Vol. 22 No. 5, pp. 840-851.

Rjasanow, S. and Wagner, W. (1996), "Stochastic weighted particle method for the Boltzmann equation", *Journal of Computational Physics*, Vol. 124 No. 2, pp. 243-253.

Seinfeld, J.H. and Pandis, S.N., (1998), Atmospheric Chemistry and Physics, Wiley-Interscience, New York, NY.

Tu, C. and Zhang, J. (2014), "Nanoparticle-laden gas flow around a circular cylinder at high Reynolds number", *International Journal of Numerical Methods for Heat & Fluid Flow*, Vol. 24 No. 8, pp. 1782-1794.

Vancassel, X., Mirabel, P. and Garnier, F. (2014), "Numerical simulation of aerosols in an aircraft wake using a 3D LES solver and a detailed microphysical model", *International Journal of Sustainable Aviation*, Vol. 1 No. 2, pp. 139-159.

Wei, J. and Kruis, F.E. (2013), "GPU-accelerated Monte Carlo simulation of particle coagulation based on the inverse method", *Journal of Computational Physics*, Vol. 249 No. 15, pp. 67-79.

Xu, Z., Zhao, H. and Zheng, C. (2015), "Accelerating population balance-Monte Carlo simulation for coagulation dynamics from the Markov jump model, stochastic algorithm and GPU parallel computing", *Journal of Computational Physics*, Vol. 281 No. 15, pp. 844-863.

Yin, Z. and Liu, H. (2014), "Numerical simulation of nanoparticles diffusion and coagulation in a twin-jet via a TEMOM method", *International Journal of Numerical Methods for Heat & Fluid Flow*, Vol. 24 No.6, pp. 1312-1320.

Yu, M.Z., Lin, J.Z. and Chan, T.L. (2009), "Numerical simulation for nucleated vehicle exhaust particulate matters via the TEMOM/LES method", *International Journal of Modern Physics C*, Vol. 20 No. 3, pp. 399-421.

Yu M.Z. and Chan T.L. (2015), "A bimodal moment method model for submicron fractal-like agglomerates undergoing Brownian coagulation", *Journal of Aerosol Science*, Vol. 88, pp. 19-34.

Zhang, Y., Seigneur, C., Seinfeld, J.H., Jacobson, M.Z., Binkowski, F.S. (1999), "Simulation of aerosol dynamics: a comparative review of algorithms used in air quality models", *Aerosol Science and Technology*, Vol. 31 No. 6, pp. 487-514.

Zhang, W. and You, C. (2015), "Numerical approach to predict particle breakage in dense flows by coupling multiphase particle-in-cell and Monte Carlo based methods", *Powder Technology*, Vol. 283, pp. 128-136.

Zhao, H., Kruis, F. E., and Zheng, C. (2009), "Reducing statistical noise and extending the size spectrum by applying weighted numerical particles in Monte Carlo simulation of coagulation", *Aerosol Science and Technology*, Vol. 43 No.8, pp. 781-793.

Zhou, K. and Chan, T.L. (2011), "Simulation of homogeneous particle nucleation in a free turbulent jet", *Aerosol Science and Technology*, Vol. 45 No. 8, pp. 973-987.

Zhou, K. and Chan, T.L. (2014), "Analytical approximation schemes for mean nucleation rate in turbulent flows", *Aerosol Science and Technology*, Vol. 48 No. 5, pp. 459-466.

Zhou, K. and He, Z. (2014)", "Monte Carlo simulation of aerosol evolution in a planar mixing layer", *International Journal of Numerical Methods for Heat & Fluid Flow*, Vol. 24 No.8, pp. 1769-1781.

Zhou, K., He, Z., Xiao, M. and Zhang, Z. (2014), "Parallel Monte Carlo simulation of aerosol dynamics", *Advances in Mechanical Engineering*, AN 435936 pp. 1-11.