



FUZZY MARKOV CHAINS MODELING OF AGGREGATION PROCESSES

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ABSTRACT. In this paper, the fuzzy Markov chain method is proposed as a new discrete solution of a population balance equation for an aggregation process. In order to validate the proposed method, analytical solution of an aggregation equation is compared with the fuzzy Markov chain method for the constant aggregation kernel. According to the results, if the size range of the system is divided into a sufficient number of states and an appropriate transition time step is chosen, then the fuzzy Markov chain method displays a good approximation for the particle size distribution (PSD) while the main equation is driven by a constant aggregation kernel.

1. INTRODUCTION

Population balance modelling which is a well-known model to describe the size distribution of particulate systems, is widely used in the food, pharmaceutical and chemical industries. Population balance equations (PBEs) are underpinned by the law of mass conservation. A continuous number density function in PBEs makes the model quite powerful in analyzing the dynamics of a process. However, the structure of these equations is complex due to the intrinsic partial integro-differential equations and hence analytical solutions may not be possible except for simple cases. Nonetheless there are a number of numerical methods described in the literature which allow for PBEs to be solved. [1] contributed numerical solutions of PBEs, [2] developed a discrete method to predict a single property of the particle such as total number or volume and [3] published a discrete solution of PBEs for a crystallization process. Moreover, [4] presented a new discretization procedure for an agglomeration equation where they mitigated the intrinsic problems encountered through discretization by using proper probability density functions. On the other hand, [5] discussed the characteristics of approximate methods for modelling aggregation by

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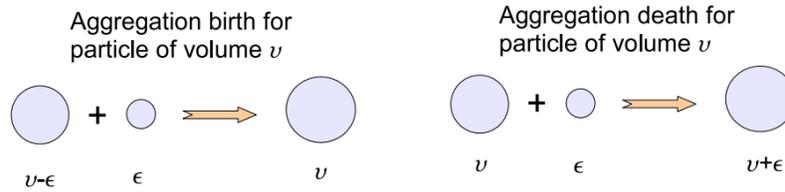


FIGURE 1. An illustration of the aggregation birth and death

considering their accuracy, ability to produce error estimates, ease of implementation and speed. A comprehensive discussion about various applications is available in [6]. The term aggregation is used for size enlargement processes whereby small particles are gathered into larger, relatively permanent masses in which the original particles can still be distinguished [7]. It is encountered among many diverse sectors of industry including pharmaceuticals, fertilizers and chemicals.

1.1. Continuous population balances. A general form of the continuous aggregation equation for a batch system can be given as [6];

$$\frac{\partial f(v, t)}{\partial t} = \frac{1}{2} \int_0^v \beta(v - \epsilon, \epsilon, t) f(v - \epsilon, t) f(\epsilon, t) d\epsilon - f(v, t) \int_0^\infty \beta(v, \epsilon, t) f(\epsilon, t) d\epsilon \quad (1)$$

where $f(v, t)$ is the number distribution of particles as a function of particle volume and time, $\beta(v, \epsilon)$ is the aggregation kernel of aggregating particles of volume v and ϵ .

Equation 1 states that the change in the number of particles of volume v over an incremental time step at time t depends on the number of new particles of volume v produced by aggregation of particles smaller than v , and depends on the average number of particles lost by aggregation of particles of volume v .

The first integral part of the right hand side of equation 1 represents *aggregation birth* for particle of volume v . That is, particles of volume $v - \epsilon$ and ϵ aggregate to form a new particle of volume v . The second integral part of the right hand side of equation 1 represents *aggregation death* for particle of volume v . In other words, particles of volume v and ϵ aggregate to produce a new particle of volume $v + \epsilon$. A schematic representation of aggregation birth and death is given in Figure 1.

1.2. Discrete population balances. The underlying basis of discretization is to turn the main population balance of integro-differential equations into ordinary differential equations. To achieve this, the integral signs of the continuous equation 1 are turned to summation signs, and continuous functions are replaced by

corresponding discrete functions;

$$\frac{dN_i}{dt} = \frac{1}{2} \sum_{j=1}^{i-1} \beta_{j,i-j} N_j N_{i-j} - N_i \sum_{j=1}^{\infty} \beta_{i,j} N_j \quad (2)$$

where N_i is the number of particles in the interval i and $\beta_{i,j}$ is the discrete aggregation kernel of the particles in the intervals i and j .

There should be a finite state space to model the system, that is j cannot be increased to infinity in the second part of right hand side of equation 2. On this basis, two realistic approaches can be stated as follows;

- i) A maximum volume size v_{max} that can appear in the process is assigned. That is, any two particles cannot aggregate if their total volume is greater than v_{max} . This kind of limitation is possible if aggregation occurs in a pipe or similar geometrical places where physical parameters mostly determine the maximum volume size. Let the maximum state be represented by n^{th} state, then equation 2 can be rewritten as;

$$\frac{dN_i}{dt} = \frac{1}{2} \sum_{j=1}^{i-1} \beta_{j,i-j} N_j N_{i-j} - N_i \sum_{j=1}^{n-i} \beta_{i,j} N_j \quad (3)$$

Equation 3 produces some restrictions on particle aggregation. If the maximum state number $n=40$ and $i=5$, a particle in the interval 5 cannot aggregate with another particle in the interval j when $i + j > 40$, such as 36, 37, 38, 39, 40.

- ii) A maximum volume size v_{max} which is represented by interval n that can join to aggregation process is defined. Then, the maximum particle size can be $2v_{max}$ and if the intervals created by using a uniform method, then the total number of intervals will be $2n$. Thus, equation 2 can be defined as;

$$\frac{dN_i}{dt} = \frac{1}{2} \sum_{j=1}^{i-1} \beta_{j,i-j} N_j N_{i-j} - N_i \sum_{j=1}^n \beta_{i,j} N_j \quad (4)$$

2. MARKOV PROCESSES

The basic property of a Markov process is that it has no memory. That is, the past and the future of the process are mutually independent and only the present can influence the future [8]. If the process has a finite or countably infinite set of states, then it is referred as Markov chain. Classification of the Markov processes referring to their time and property behaviour is given in Table 1.

2.1. Discrete time Markov chains. If the total processing time is divided into the finite intervals of duration τ , then a representation of the process which is discrete in time is obtained, which is called Discrete Time Markov Chains (DTMC) [9]. In the rest of this study, the term Markov chains refers to DTMC.

TABLE 1. Classification of Markov processes (adopted from [8])

Markov Processes		
Time	Property	Governing Equation
Continuous	Continuous	Partial differential equations
Continuous	Discrete	Ordinary differential equations.
Discrete	Discrete	Matrix algebra

To constitute a Markov chain, three important notations are used such as a state vector $\mathbf{a}(t)$, a transition time step τ and a transition matrix \mathbf{P} . The vector $\mathbf{a}(t)$ with components $(a_1(t), a_2(t), a_3(t), \dots)$ is called the state probability distribution vector of the system at time t . If the probability for an entity currently in the state j at time t is denoted by $a_j(t)$ then the state probability distribution of state i for the next time step, $a_i(t+1)$ is given by the sum of product of all probabilities. This is formulated as follows;

$$a_i(t+1) = \sum_{j=1}^n p_{ji} a_j(t) \quad (5)$$

If equation 5 is written down for all states, then it is possible to show transitions between the states using a square matrix which is called Markov or transition matrix. The transition matrix \mathbf{P} has entries p_{ij} which represents transition probabilities from state i to state j at each time step. On this basis, the transition matrix is given by matrix as follows [10];

$$\mathbf{P} = \begin{pmatrix} p_{11} & p_{12} & \dots & p_{1n} \\ p_{21} & p_{22} & \dots & p_{2n} \\ \dots & \dots & \dots & \dots \\ p_{n1} & p_{n2} & \dots & p_{nn} \end{pmatrix}_{n \times n}$$

Properties of the transition matrix are;

- The sum of all probabilities of state i equals 1, i.e., $p_{i1} + p_{i2} + \dots + p_{in} = 1 \quad \forall i = 1, \dots, n$
- Each row of the transition matrix has at least one nonzero element.
- Entities of the transition matrix are nonnegative, since p_{ij} is a probabilistic ratio, i.e. p_{ij} values lie between 0 and 1.

A Markov graph shown in Figure 2 frequently is used to figure the state space of the process. Such a graph has n nodes and a number (between n and n^2) of lines directed from i to j if p_{ij} is not equal to zero. the changes that happen in direction shown by the arrows are called transition. There would be n^2 lines if any p_{ij} is not zero.

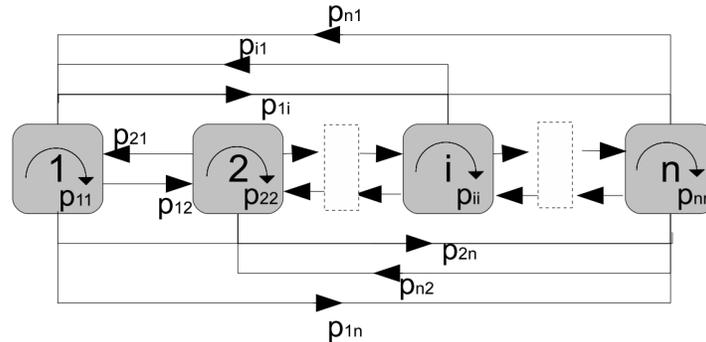


FIGURE 2. Transition probabilities' illustrations using a Markov graph

Both equation 3 and equation 4 are convenient for the fuzzy Markov chain method in relation to operation parameters. In this paper equation 4 will be used for the Markov chains method as an illustrative example. Note that in equation 4 $\beta_{i,j}$ is the discrete aggregation kernel and represents the case where a particle in the interval i and another particle in the interval j aggregate, whereby a new particle in the interval $i+j$ appears. It is obvious that $\beta_{i,j} = 0$ if $i > n$ or $j > n$ since particles bigger than the size of the interval n cannot aggregate.

2.2. Fuzzy Markov chains. Let S be a finite state space and take $S = 1, 2, \dots, n$. A finite fuzzy set on S is defined by a mapping z from S to $[0, 1]$ denoted by a vector $z = [z_1, z_2, \dots, z_n]$. A fuzzy relation \tilde{P} is defined as a fuzzy set on the $S \times S$. \tilde{P} , namely the fuzzy transition matrix, consists of $\tilde{p}_{i,j}$ where $0 \leq \tilde{p}_{i,j} \leq 1$. The entities of \tilde{P} are assigned as possibility distributions instead of probability distributions in classical Markov chains. In this paper, α -cut fuzzy numbers are employed to define $\tilde{p}_{i,j}$. Let $\tilde{M}[\alpha]$ be an α -cut fuzzy set, then $\tilde{M}[\alpha] = x | \tilde{M}[\alpha](x) \geq \alpha$, where $0 < \alpha \leq 1$. Triangular fuzzy numbers can be explained using three parameters c_1, c_2, c_3 such that the membership function, $\mu(z)$, has a triangular shape where the vertex of the triangle are located at $(c_1, 0), (c_2, 1), (c_3, 0)$. Let define the lowest end of the $\tilde{p}_{i,j}$, $\tilde{p}_{i,j,min}$, and the highest end of the $\tilde{p}_{i,j}$, $\tilde{p}_{i,j,max}$ as;

$$\begin{aligned} \tilde{p}_{i,j,min} &= \tilde{p}_{i,j} - \eta\alpha \\ \tilde{p}_{i,j,max} &= \tilde{p}_{i,j} + \eta\alpha \end{aligned} \tag{6}$$

Then;

$$\tilde{p}_{i,j[\alpha]} = [\tilde{p}_{i,j,min}, \tilde{p}_{i,j,max}] \tag{7}$$

The transition possibility of a fuzzy Markov chain is defined by the fuzzy relation \tilde{P} as;

$$z_j(t+1) = \max\{z_i(t) \wedge \tilde{p}_{i,j}\} \quad (8)$$

$\mathbf{z}(0)$ is given as an initial fuzzy set representing the initial distribution. Equation 8 can be given in matrix notation as;

$$\mathbf{z}(\tau) = \mathbf{z}(0) \circ \tilde{P}(\tau) \quad (9)$$

where \circ denotes max-min composition.

The similarity between the classical Markov chains and the fuzzy Markov chains is obvious referring to Equations 5 and 9. The algebraic summation and multiplication are used to define the classical Markov chains, whereas max-min composition is used in the fuzzy Markov chains.

2.2.1. *Defuzzification.* There are several approaches to defuzzify the fuzzy numbers in literature [11]. The centroid method, it is also known as center of gravity, was employed in this study.

$$z^* = \frac{\int \mu_{\tilde{M}}(z)zdz}{\int \mu_{\tilde{M}}(z)dz} \quad (10)$$

2.2.2. *Simulation algorithm.* The simulation algorithm which is used in this paper can be summarized as follows;

- (1) Insert the aggregation kernel, $\beta(v, \epsilon)$
- (2) Insert the transition time step, τ
- (3) Insert the total simulation time t , $t = k\tau$, where k is a positive integer
- (4) Insert initial distribution of numbers in a row vector $\mathbf{z}(0)$
- (5) Calculate the transition probability matrix \mathbf{P} using τ , $\beta(v, \epsilon)$ and \mathbf{N}
- (6) Calculate the transition possibility matrix $\tilde{\mathbf{P}}$ using \mathbf{P} , α and η
- (7) Find the number possibility distribution $\mathbf{z}(t)$ after a transition step time τ ;
- (8) Defuzzification, then go to step 7
- (9) Continue the simulation until one finds the distribution of numbers for the desired time, i.e. repeat to find \mathbf{N}_{next} up to k times

This algorithm was applied to simulate the fuzzy Markov chain method results using Matlab.

3. RESULTS AND DISCUSSION

In this section, the simulation result of the fuzzy Markov chain method is compared with the analytical solution of the continuous equation 1. A simple constant aggregation kernel is used and the initial distribution functions are chosen in the exponential form in order to find analytical solutions.

TABLE 2. States vs. size ranges for 1000 state space

	Size range	Average size
state1	0.00 - 0.1	0.05
state2	0.1 - 0.2	0.15
state3	0.2 - 0.3	0.25
.	.	.
.	.	.
state998	199.7 - 199.8	199.75
state999	199.8 - 199.9	199.85
state1000	199.9 - 200.00	199.95

3.1. Case study. The continuous population balance equation, with a constant aggregation kernel $\beta(v, \epsilon) = \beta$, can be written as;

$$\frac{\partial f(v, t)}{\partial t} = \frac{1}{2} \int_0^v \beta f(v - \epsilon, t) f(\epsilon, t) d\epsilon - f(v, t) \int_0^\infty \beta f(\epsilon, t) d\epsilon \quad (11)$$

given an initial distribution of

$$f(v, 0) = e^{-v} \quad (12)$$

where v is the particle volume, the analytical solution of equation 11 is found using *Laplace transform*¹ as;

$$f(v, t) = \frac{4e^{\frac{-2v}{2+\beta t}}}{(2+\beta t)^2} \quad (13)$$

The solution of the aggregation process with the constant aggregation kernel, which is $\beta = 0.1 \text{ s}^{-1}$ and the transition time step $\tau = 1 \text{ s}$ will be carried out in the next section by applying the fuzzy Markov chain method to the same initial condition of equation 13.

3.1.1. Fuzzy Markov chain method. The maximum size v_{max} that can aggregate is chosen as 50 mm^3 which is an arbitrary size to simulate the model. Thus, the maximum aggregate size is 100 mm^3 . The size range $[0, 100] \text{ mm}^3$ is divided into 1000 intervals using uniform method with $r = 0.1 \text{ mm}^3$ where the lowest size of first interval v_0 is 0 mm^3 . A representative volume size of each interval is calculated by using arithmetic mean, i.e. $\hat{v}_i = \frac{v_i + v_{i-1}}{2}$ (see Table 2). Following the simulation algorithm which is described in section 2.1.2, the aggregation process is simulated applying the fuzzy Markov chain method based on the transition matrix $P_{1000 \times 1000}$ and initial probability distribution $\mathbf{a}(0)_{1 \times 1000}$.

¹Details are available in Appendix section

3.1.2. *Discussion.* The analytical solution obtained in equation 13 for $\beta = 0.1 \text{ s}^{-1}$ is displayed in Figure 3. In this figure, the mean volume size of particles is monotonically increasing with increasing time. The mean size is 1 mm^3 at $t = 0$, but it increases to 11 mm^3 after 200 time steps as might be expected from an aggregation process. In addition, the variance of the distribution of numbers is increasing with time. Mathematical expressions of mean and variance functions with respect to time can be formulated as

$$v_{mean}(t) = \int_0^{\infty} v f(v, t) dv = 1 + \frac{t}{20} \quad (14)$$

$$v_{var}(t) = \int_0^{\infty} (v - v_{mean})^2 f(v, t) dv = 1 + \frac{t}{10} + \frac{t^2}{400} \quad (15)$$

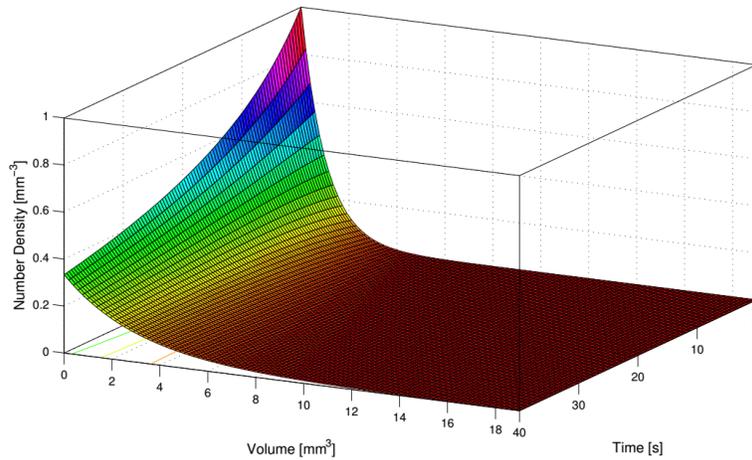


FIGURE 3. 3D visualization of analytical solution of equation 13

Since it is an aggregation process, the total number of particles in the system is expected to decrease in time. Let \mathcal{N}_0 be the initial total number of particles at $t = 0$, then total number function can be written as

$$\mathcal{N}(t) = \mathcal{N}_0 \int_0^{\infty} f(v, t) dv = \frac{2\mathcal{N}_0}{2 + \beta t} \quad (16)$$

The mean sizes obtained from the fuzzy Markov chain method for various time steps are calculated using following equation

$$\mu_v(t) = \sum_{i=1}^n \hat{v}_i \mathbf{N}_i(t) \quad (17)$$

Similarly, the variances over time in particle size are calculated as

$$\sigma_v^2(t) = \sum_{i=1}^n \mathbf{N}_i(t) (\hat{v}_i - \mu_v(t))^2 \quad (18)$$

The standard deviation denoted by σ , is the square root of the variance.

The total number of the particles is found as

$$\mathcal{N}(t) = \mathcal{N}_0 \sum_{i=1}^n \mathbf{N}_i \quad (19)$$

The expected mean of the particle volume and the variance expectations are shown in Figure 4. Also, mean size prediction of the fuzzy Markov chain method is very close to the analytically expected mean size during 100 time steps. Both curves of the variance expectations based on the analytical solution and the fuzzy Markov chain method increase quadratically with time and are in agreement. In equation 16, it is shown that the total number of particles in the system is a function of time for the constant kernel. As illustrated in Figure 5, the total number of particles predicted by the fuzzy Markov chain method is almost identical to the analytical solution. This signifies that the fuzzy Markov chain method is capable of adequately predicting the analytical solution.

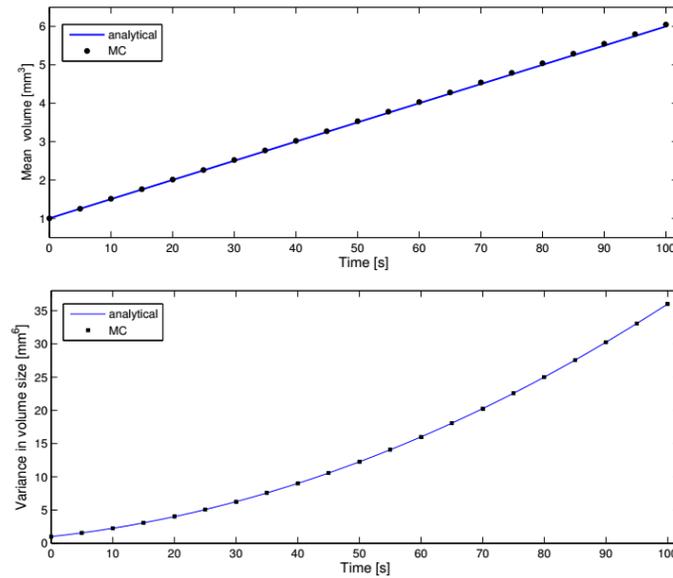


FIGURE 4. Changes in the mean size and in variances with respect to time, case study 1

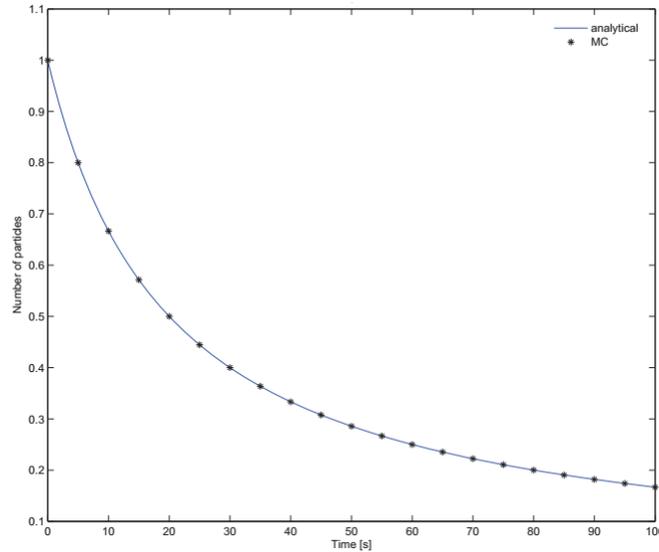


FIGURE 5. Total number of particles in the system with respect to time, case study 1

In Figure 6, simulation results of the fuzzy Markov chain method and the analytical solution are illustrated at different time steps. Figure 6 shows that the results of the fuzzy Markov chain method exactly overlap the analytical solution for size intervals which are bigger than 5 mm^3 . The reason for the slight mismatch of the smaller size volumes is that the uniform discretization does not perfectly cover the sensitivity of the exponential function where the particle volume is smaller than 5 mm^3 . Yet the agreement between the analytical solution and the fuzzy Markov chain method is more than 95% according to Kolmogorov-Smirnov test at $t = 200$ seconds, i.e. 200 time steps after starting.

4. CONCLUSION

In this study, the fuzzy Markov chain method is used to solve the continuous population balance equation for an aggregation process. The results show that if the particle size range of the system has been divided into a sufficient number of states and an appropriate transition time step is chosen, then the fuzzy Markov chain method exhibits very good agreement with the analytical solution.

Some process systems have a natural periodicity such as rotating mixers (period of rotation), vibratory conveyors (period of excitation) for which the transition time step can be chosen from basic physics. For systems which have no period, the time step should be chosen taking into consideration process kinetics and total

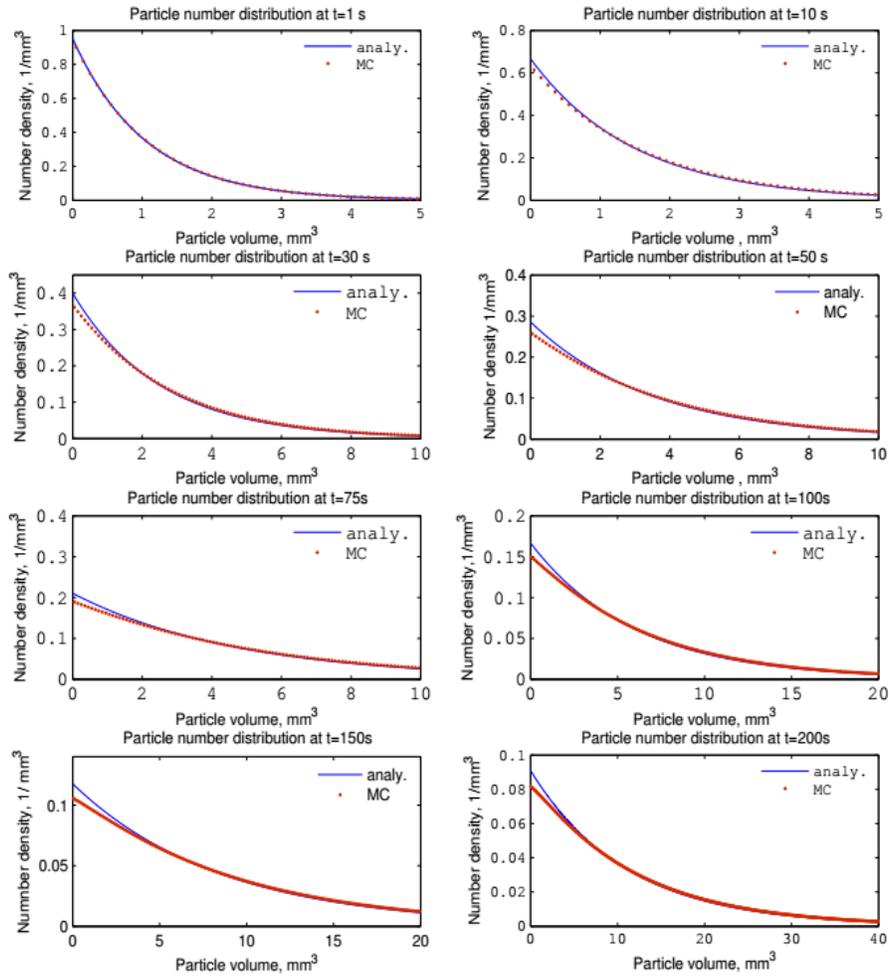


FIGURE 6. Comparison of Markov Chains simulation results and analytical solution, case study 1

processing time. The sufficiency of the number of states is dependent of the process kinetics. The authors suggest to divide the state property under analysis into 30 intervals as an initial trial. Ultimately, a good pair of (τ, n) should be selected to represent the time and particle property in discrete form efficiently.

Provided that the aggregation kernel $\beta(v, \epsilon)$ is time independent, Markov chains can be used as a new discrete method for the solution of the aggregation equation.

Although, a large number of states makes the computation difficult, this can be readily handled using modern computer software packages. Thereafter, for calculating transition matrix $\tilde{\mathbf{P}}$, the particle size distribution of the system can be easily evaluated using the fuzzy Markov chain method.

APPENDIX

The Laplace transform of a function $f(v, t)$ which is defined $\forall v \in \mathfrak{R}^+$, is the function $\hat{f}(s, t)$ defined by

$$\hat{f}(s, t) = \mathcal{L}\{f(v, t)\} = \int_0^{\infty} e^{-sv} f(v, t) dv \quad (20)$$

Thus, the function $f(v, t)$ is transformed from v domain to s domain.

The continuous population balance equation with a constant aggregation kernel $\beta(v, \epsilon) = \beta$ can be written as

$$\begin{aligned} \frac{\partial f(v, t)}{\partial t} = & \frac{1}{2} \int_0^v \beta f(v - \epsilon, t) f(\epsilon, t) d\epsilon \\ & - f(v, t) \int_0^{\infty} \beta f(\epsilon, t) d\epsilon \end{aligned} \quad (21)$$

The solution procedure of the continuous population balance equation with the Laplace transform method is summarized step by step as

i) *Laplace transform*

When the Laplace transform is applied aiming to transform the function $f(v, t)$ from v domain to s domain, i.e. $\mathcal{L}\{f(v, t)\} = \hat{f}(s, t)$, then the continuous population balance equation 21 becomes

$$\begin{aligned} \frac{\partial \hat{f}(s, t)}{\partial t} = & \frac{1}{2} \int_0^{\infty} e^{-sv} dv \int_0^v \beta f(v - \epsilon, t) f(\epsilon, t) d\epsilon \\ & - \int_0^{\infty} e^{-sv} f(v, t) dv \int_0^{\infty} \beta f(\epsilon, t) d\epsilon \end{aligned} \quad (22)$$

Note that, the first integral part of the right hand side of equation 22 is a convolution integral. Moreover, $\int_0^{\infty} f(\epsilon, t) d\epsilon$ gives the total number of particles in the system which equals $\frac{2}{2 + \beta t}$ in the second integral part of the right hand side of equation 22. Then equation 22 can be simply written as

$$\frac{\partial \hat{f}(s, t)}{\partial t} = \frac{\beta}{2} \hat{f}^2(s, t) - \frac{2\beta}{2 + \beta t} \hat{f}(s, t) \quad (23)$$

ii) *Solution of the partial differential equation*

The solution of the partial differential equation 23 is

$$\hat{f}(s, t) = \frac{2}{2 + \beta t + 8G(s) + 8G(s)\beta t + 2G(s)\beta^2 t^2} \quad (24)$$

where $G(s)$ is a function of s .

iii) *Invoking the initial condition to find the $G(s)$ function*

The Laplace transform of the initial particle size distribution function which is given in equation 15 is

$$\hat{f}(s, 0) = \frac{1}{s+1} \quad (25)$$

which should equal equation 24 at $t = 0$, then

$$\frac{1}{s+1} = \frac{2}{2+8G(s)} \quad (26)$$

which gives

$$G(s) = \frac{s}{4} \quad (27)$$

iv) *Invoking the $G(s)$ function*

If the $G(s)$ function is inserted in equation 24, it becomes

$$\hat{f}(s, t) = \frac{2}{2 + \beta t + 2s + 2s\beta t + \frac{s}{2}\beta^2 t^2} \quad (28)$$

v) *Inverse Laplace transform*

As a final step, if the inverse Laplace transform is applied to equation 28 to be transformed in v domain back, then the analytical solution of the population balance equation is found as

$$f(v, t) = \frac{4e^{\frac{-2v}{2+\beta t}}}{(2 + \beta t)^2} \quad (29)$$

which is given in equation 16.

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