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Fast Monte Carlo methodology for multivariate particulate systems—I: Point ensemble Monte Carlo

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Abstract

A fast Monte Carlo methodology for particulate processes is introduced. The proposed methodology combines concepts from discrete population balance equations and dynamic Monte Carlo simulations of chemical kinetics to construct a new jump Markov model that approximates the population balance dynamics. The Markov model consists of a definition of a new type of reaction channel, in which the reaction product is a stochastic process by itself. One feature of this model is that, although a coarse view of the process is taken, it still conserves the history of individual particles. This is a very important aspect for effective modeling of multivariate models, especially when part of the goal is to study the evolution of the internal states of the particles (i.e., composition, phase behavior, etc.).

Numerical experiments show that this algorithm can improve the computational load of the exact method by orders of magnitude without sacrificing computational accuracy. The methodology is useful especially in stochastic optimization applications where many function calls (simulations) are required. Possible applications are optimization and dynamic optimization using an artificial chemical process algorithm, genetic algorithm, or simulated annealing among others.

Keywords: Population balance; Multivariate population balance; Monte Carlo simulation; Aggregate coagulation; Aggregate sintering; Nonlinear dynamics

1. Introduction

The continuous population balance equation, PBE, is a transport equation for the temporal evolution of a population density as a result of different particle mechanisms like nucleation, growth, aggregation and breakage. The PBE has diverse applications, like crystallization, precipitations, aerosol dynamics, microbial fermentors and polymer reactors among others (Ramkrishna, 1985). Due to the importance of PBE, many numerical solution methodologies have been proposed to solve the one dimensional PBE. A partial list consists of discretization methods (Hounslow et al., 1990; Kumar and Ramkrishna, 1996; Ramkrishna, 2000), finite elements (Nicmanis and Hounslow, 1996; Mahoney and Ramkrishna, 2002; Roussos et al., 2005; Alexopoulos and Kiparissides, 2005), moments methods (Diemer and Olson, 2002a, b) generalized method of moments(McGraw, 1997; Marchisio et al., 2003). A comparative study between the different techniques is given in Alexopoulos et al. (2004), Alexopoulos and Kiparissides (2005), and Roussos et al. (2005). Using some of these methods to solve multidimensional problems becomes a challenging task.

In many cases of industrial interest, other particle properties like surface area, chemical composition, crystallinity, etc. are needed in addition to particle size (volume), to fully characterize the powder. These multidimensional problems are very difficult to solve using a direct numerical solution. A practical approach to model these systems is to use Monte Carlo simulations, which can deal with multiple state variables in a natural way (Tandon and Rosner, 1999; Efendiev and Zachariah, 2002). There are many efficient Monte Carlo algorithms for the solution of population balance equations in the literature (Gillespie, 1975; Garcia et al., 1987; Liffman, 1992; Kruis et al., 2000; Maisels et al., 2004; Smith and Matsoukas, 1998; Ramkrishna, 2000). These methods consist in an artificial realization of the population dynamics of a finite system (finite number of particles in a very small volume) to estimate the properties of the

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whole system. The estimate becomes exact as the number of particles approaches infinity.

One drawback of the MC methods is the large computational load associated with them. The computational cost may become an issue in some applications. One example is the solution of optimization and dynamic optimization of systems modeled by MC methods (Irizarry, 2005, 2006). Another example where computational cost may be a limiting factor is when the transport due to the flow field is incorporated in the PBE model. The goal of this work is to develop a general-purpose approximated MC methodology with negligible loss of accuracy and lower CPU time than existing exact MC methods are able to offer. The methodology is effective in solving complex multivariate problems with competing mechanisms including complex coagulation and breakage kernels. Furthermore, this accelerated MC strategy makes solutions of optimization and dynamic optimization using a stochastic optimization algorithm feasible in terms of CPU time.

The paper is organized as follows. In Section 2, the stochastic simulation algorithm for chemical kinetics and the Monte Carlo methods for the solution of PBE are briefly reviewed. The construction of the new Jump Markov model is described in Section 3. The proposed MC simulation is introduced in Section 4. In Section 5, the performance of the approximated method is studied with a set of study cases. In Section 6, competitive reactions in droplets are studied. In Section 7, the problem of aggregation with a finite coalescing rate is discussed. Finally, in Section 8, the performance of the algorithm for physically relevant kernels is compared with the best results in the literature.

2. Stochastic simulation algorithm

2.1. Chemical kinetics

The exact stochastic simulation algorithm (SSA) is a general methodology for solving Jump Markov processes. A Jump Markov process is a class of stochastic process with a probability of discontinuous transition from one state to another. Other names in the literature for SSA are dynamic Monte Carlo (DMC), kinetic Monte Carlo (KMC), dynamic simulation Monte Carlo (DSMC). The SSA of a well mixed chemical reaction system has been developed by Gillespie (1976, 1977). Let $S = (S_1, \ldots, S_M)$ be the vector of chemical species and $N = (N_1, \ldots, N_M)$ the vector of number of molecules for each specie in the simulation volume. M is the total number of species in the system and T the total number of reactions. For example consider the parallel reaction system: $NaOH + HCl \rightarrow NaCl + H_2O$ and $NaOH + CH_2ClCOOC_2H_5 \rightarrow CH_2ClCOONa + C_2H_5OH$. This system has six species ($M = 6$) NaOH (S_1), HCl (S_2), $CH_2ClCOOC_2H_5$ (S_3), NaCl (S_4), $CH_2ClCOONa$ (S_5), C_2H_5OH (S_6) with the following two reactions ($T = 2$).

$$E_1 : S_1 + S_2 \rightarrow S_4, \tag{1}$$

$$E_2 : S_1 + S_3 \rightarrow S_5 + S_6. \tag{2}$$

Each reaction is considered an event E_i that can change the system from the current state, N, to a new state. The SSA consists in answering the following question: for a system in a given state what reaction occurs next, E_f, and when does it occur, τ_{micro}? The simulation generates random number pairs (τ_{micro}, E_f) from conditional probabilities which are functions of the propensity function for each reaction, $R(E_s)$, defined as

$$R(E_s)\,\mathrm{d}t \equiv \text{the probability that reaction } E_s \text{ occurs in the time interval } [t, t + \mathrm{d}t).$$

In each iteration of the SSA algorithm, two random numbers r_1, r_2 are selected from the uniform distribution (0, 1). The time for the next reaction is calculated as

$$\tau_{\text{micro}} = \frac{-\log(r_1)}{\sum_{j=1}^{T} R(E_j)}. \tag{3}$$

The time is moved to $t + \tau_{\text{micro}}$. The reaction fired at $(t + \tau_{\text{micro}})$ is selected by solving the following equation

$$\sum_{s=1}^{f} R(E_s) < r_2 \sum_{s=1}^{T} R(E_s) \leqslant \sum_{s=1}^{f+1} R(E_s). \tag{4}$$

The selected event is E_f. These steps are repeated until a final time is reached. Because one reaction is fired at a time in a microscopic time interval, τ_{micro}, the method can be slow in some cases, for example, if the concentrations are high or if the number of reactions is very large.

2.2. MC simulation of PBE

Another application where SSA type simulation is very useful is in the simulation of a Jump Markov process of a system of coalescing particles (Gillespie, 1975; Garcia et al., 1987; Liffman, 1992; Kruis et al., 2000, Maisels et al., 2004) or particles undergoing breakage (Ramkrishna, 2000). The SSA of these Markov processes reproduces the correct dynamic of the macroscopic PBE. Garcia et al. (1987) describes two procedures for the coagulation process. The first one, called the "inverse method," is the exact DSMC method applied to a coalescing system of particles. Since the inverse method calculates all possible collision rates of all possible particles pairs (i.e., solving an equation analogous to Eq. (4)), they are time-consuming. Garcia et al. proposed a second method based on acceptance-rejection (AR) method (Bird, 1976). Different from exact MC, the AR is an approximated MC method in which performance and accuracy are problem dependent (Kruis et al., 2000; Smith and Matsoukas, 1998). Kruis et al. (2000) proposed a streamlined version of the exact inverse method of Garcia et al. (1987). Smart book-keeping eliminates re-calculation of most of the collision rates not participating in the event, making the method much faster, and without any approximations like in the AR method. They called this method fast DSMC. Notice that this bookkeeping is limited to coagulation problems. We will call the exact methods (inverse method, DSMC, fast DSMC, etc.) "exact MC" and the acceptance-rejection method "AR".

In the next section, a construction of a new Jump Markov model that approximates the dynamics of the PBE is introduced.