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Stochastic solution of population balance equations for reactor networks

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ABSTRACT

This work presents a sequential modular approach to solve a generic network of reactors with a population balance model using a stochastic numerical method. Full-coupling to the gas-phase is achieved through operator-splitting. The convergence of the stochastic particle algorithm in test networks is evaluated as a function of network size, recycle fraction and numerical parameters. These test cases are used to identify methods through which systematic and statistical error may be reduced, including by use of stochastic weighted algorithms. The optimal algorithm was subsequently used to solve a one-dimensional example of silicon nanoparticle synthesis using a multivariate particle model. This example demonstrated the power of stochastic methods in resolving particle structure by investigating the transient and spatial evolution of primary polydispersity, degree of sintering and TEM-style images.

1. Introduction

Population balance modelling has traditionally been applied to modelling particle formation in a batch or plug-flow reactor [4,32,30]. In the former case, the equations governing particle growth can be simplified to exclude terms for particle transport to- and from the reactor. For the latter case, the reactor model typically represents an axial streamline through a plug flow reactor (PFR). However, there are many systems found in engineering where advective or diffusive particle transport is important [25,35]. This typically requires use of computational fluid dynamics (CFD) or reactor network approach, coupled with a population balance model to accurately capture particle dynamics.

Solutions for the population balance equation in these coupled systems may be grouped into several classes, the most common of which are monodisperse [22,11], moment [18,2,1,27] and sectional [40] methods. All of these methods yield a set of differential equations which may solved within the framework of the differential equations describing fluid transport and/or chemical reactions [25].

Many commercial packages now include particle population balances using the aforementioned methods, for example STAR-CCM+ [12], CHEMKIN-PRO [41] and gPROMS [5]. The major drawback of using these methods to solve the population balance equations is that they either do not resolve the particle size distribution (e.g. moments) or utilise a simplified particle model such as a spherical or surface-volume type description. This can cause errors when particle aggregates are formed through coagulation and sintering [29].

There is increasing recent interest in application of stochastic (or Monte Carlo) methods to such systems [25,35], after the initial work of Garcia et al. [19]. Stochastic methods approximate a real volume of particles by a sample volume of

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computational particles, where each computational particle represents a certain number of real particles [48,38]. The idea of a computational particle in the context of a stochastic method is general; and as such, any number of independent particle properties can simulated, for example internal aggregate structure [47,29,43] or soot particle composition [33,44,15].

Stochastic methods have been applied for simulation of the Boltzmann equations for colliding gas molecules on spatially inhomogeneous domains [9] and adapted to transport of electrons [34]. The key challenge in extending stochastic methods to solve population balances in spatially inhomogeneous systems lies in the simulation of the Smoluchowski coagulation process [35]. In contrast to Boltzmann collisions, Smoluchowski coagulation events reduce the number of computational particles, causing numerical issues.

Implementation of stochastic population balance solvers for flow systems requires discretising the reactor geometry into a mesh composed of reactor 'cells' [35,25]. These connected cells can be considered a type of reactor network. Kruis et al. [25] developed a 'cell-based weighted random walk' method to account for the movement of particles between cells in a one- and two-dimensional reactor geometry. While reasonably good agreement with a moments code was reported, it was observed that the method was sensitive to stochastic noise, a result of using too few computational particles.

Patterson and Wagner [35] presented a stochastic method for coagulation–advection problems. The performance in accounting for coagulation of the conventional direct simulation algorithm (DSA, [16]) against stochastic weighted algorithms (SWAs, [38]) was evaluated. It was reported that DSA is particularly susceptible to stochastic noise, which eventually manifests itself as a systematic variance from the true solution.

A study by Zhao and Zheng [52] gave a three-dimensional adaptation of a stochastic population balance solver (using an SWA [53]) coupled with a CFD code. An algorithm was presented to de-couple the fluid flow, particle flow and particle dynamics by choosing an appropriate timestep size. In a similar vein to [25], limits associated with computational cost restricted the maximum number of computational particles, preventing precise agreement with direct numerical simulation (DNS).

'Multizonal' [7,6] or 'compartmental' models [26,21,3,51] are models in which the fine mesh of CFD is simplified to a coarse network of reactors with a specified flow between each node. Such techniques typically require orders of magnitude fewer reactor cells, thus reducing computational cost, however this comes at the price of decreased spatial resolution of particle dynamics.

Only recently have compartmental models begun to be investigated using stochastic methods: Braumann et al. [10] used a two-reactor dead-zone model and a stochastic algorithm to simulate granulation. Irizarry [21] recently presented an approach to solve a compartmental model with a population balance using a stochastic method. A new algorithm to move particles between cells ('particle bundle flow' method) was reported, in which particles were restricted from jumping more than one compartment by choosing a suitably small timestep. The numerical properties of some test systems were evaluated as a function of the timestep, however the effect of other numerical parameters was not addressed.

There are, however, further open questions in solving reactor networks with stochastic population balance solvers. In order to maximise efficiency of these algorithms, it is necessary to understand the convergence of these algorithms with respect to the number of computational particles and independent runs [46]. Further, it is important to quantify and reduce stochastic noise in the system, as this was identified as a potential issue in [25]. Finally, the coupling of a such a system to a reacting gas-phase has not yet been investigated: this is critical if particle synthesis in real reactors are to be simulated [13,14].

The purpose of this paper is to extend the work of Irizarry [21], Kruis et al. [25] and Zhao and Zheng [52] by developing an algorithm for solving a fully-coupled gas-phase ODE/particle population balance network. The convergence properties of the stochastic population balance will first be investigated, and methods to reduce statistical error will be considered. The flexibility of stochastic methods in solving the population balance equation will be demonstrated by use of a detailed multivariate particle model to simulate silicon nanoparticle growth in a plug-flow reactor.

The structure of this paper is as follows. In Section 2, the reactor models are presented. The stochastic solution methodology and model used for generating reference solutions are discussed in Sections 2.1 and 2.2 respectively. The numerical test cases and their results are discussed in Section 3. Finally, Section 4 illustrates one of the many ways in which reactor networks can be applied to model a real reactor system.

2. Model description

This work will study the numerical and physical behaviour of a constant-pressure network of continuous-stirred tank reactors (CSTRs; or perfectly-stirred reactors, PSRs). It is assumed that each of these reactors has a characteristic residence time $\tau_{\text{CSTR}} = Q/V$ associated with it, where Q is the volumetric flowrate and V is the reactor volume. The creation and interaction of particles with each other in these reactors is described by a population balance equation. The evolution of the number density of particles $n(P)$ of type P is given by

$$\frac{\mathrm{d}}{\mathrm{d}t}n(P) = R_{\text{incep}}(\mathbf{c}, P) + R_{\text{coag}}(\mathbf{n}, P) + \sum_{j=1}^{N_{\text{SG}}} R_{\text{SG},j}(\mathbf{c}, P) + \frac{1}{\tau_{\text{CSTR}}}\left(\sum_{j=1}^{N_{\text{in}}} f^{[j]} n^{[j]}(P) - n(P)\right) - \psi(\mathbf{c}, \mathbf{n}, T)n(P) \tag{1}$$