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A Monte Carlo method for the simulation of coagulation and nucleation based on weighted particles and the concepts of stochastic resolution and merging

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ABSTRACT

Monte Carlo simulations based on weighted simulation particles can solve a variety of population balance problems and allow thus to formulate a solution-framework for many chemical engineering processes. This study presents a novel concept for the calculation of coagulation rates of weighted Monte Carlo particles by introducing a family of transformations to non-weighted Monte Carlo particles. The tuning of the accuracy (named 'stochastic resolution' in this paper) of those transformations allows the construction of a constant-number coagulation scheme. Furthermore, a parallel algorithm for the inclusion of newly formed Monte Carlo particles due to nucleation is presented in the scope of a constant-number scheme: the low-weight merging. This technique is found to create significantly less statistical simulation noise than the conventional technique (named 'random removal' in this paper). Both concepts are combined into a single GPU-based simulation method which is validated by comparison with the discrete-sectional simulation technique. Two test models describing a constant-rate nucleation coupled to a simultaneous coagulation in 1) the free-molecular regime or 2) the continuum regime are simulated for this purpose.

1. Introduction

The processes occurring during particle production are usually modeled with the help of the population balance equation (PBE) which is described in detail by [1]. This modeling approach plays a crucial role for the analysis of aerosol reactors [2], fluidized beds [3,4], crystallizers [5,6] and many other processes used in the field of chemical engineering [7].

A variety of methods exists allowing to solve the PBE numerically. However, the correct description of simultaneous processes like nucleation, coagulation, growth, breakage, mixing, sintering and other processing mechanisms poses a major problem, which has not been solved to a satisfactory extent, yet.

The classical moment methods [8,9] and the newer direct quadrature method of moments (DQMOM) [10] are only able to render the time-dependency of certain values for the moments of the particle size distribution (PSD), and not the full PSD. Sectional methods [11] face problems when combined with growth processes in the form of numerical diffusion [12] and make the extension to multiple dimensions complicated [13]. Finite element methods avoid the problem of numerical

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diffusion [14], but they are computationally demanding for time-depending coagulation kernels [15] and necessitate special treatment of discontinuities, which are typically encountered in nucleation processes [16].

The Monte Carlo (MC) simulation [17] avoids per se the problem of numerical diffusion for the growth process [18] and is easily extensible to multiple particle properties – making it thus a most promising approach in order to describe all of the processes mentioned above. The disadvantages of the MC simulation are long simulation times and the inherent stochastic noise of the method.

The description of the nucleation process poses a fundamental problem for the MC simulation, hence the continuous incorporation of newly nucleated particles has to be reconciled with limited computational capacities. The here presented work focuses on the simulation of coagulation and nucleation, only, in order to investigate the extent of the amount of the statistical noise stemming from different particle insertion techniques.

The traditional two solutions for the problem of including new simulation particles into the simulation are 1) the resizing approaches also called 'topping-up' [19] or 2) the so called constant-number approaches, introduced to coagulation first [20] and extended to the joined simulation of coagulation and nucleation later [21], this approach will be called 'random removal' in the following. An alternative approach is the rarely applied merging of simulation particles, which is used by [22].

A novel merging strategy, the low-weight merging, for the inclusion of newly formed particles is proposed in this paper which is able to preserve all particle properties in contrast to the merging technique by [22], which preserves the properties only, if two simulation particles with equal properties can be found. It is shown that simulations using the low-weight merging technique produce far less statistical noise than the ones using the traditional random removal method [21].

The presented low-weight merging technique resorts to the use of weighted MC particles. The use of differentially weighted simulation particles (a particle i with statistical weight W_i represents a concentration W_i of real particles) has several advantages: it allows to describe the interaction between simulation particles having different concentrations coming from different cells or compartments and it can also be used as a tool to control the number of simulation particles (e.g. to gain numeric accuracy). The combination of stochastically formulated coagulation events with an ordinary differential equation(ODE)-based growth and nucleation processes [18,23], based on the operator splitting technique, or [2] with less formal framework, is facilitated, because no restrictions are posed upon the statistical weight of the particles included into the simulation due to nucleation.

The difficulty associated with weighted MC particles is the correct description of the coagulation rates and schemes. The mathematically complex stochastic weighted algorithm developed by [24] and further refined by [25] has been suggested in this context. The recently proposed Markov jump models [26] describe the coagulation by resorting to constant-number methods introduced by the Matsoukas group [20]. An alternative approach has been elaborated by [27] leading to the concept of 'fictitious particles', the attribution of two distinct rates or probabilities for the same coagulation event are conceptually difficult to understand.

We introduce an alternative approach for the derivation of the coagulation rates for various coagulation schemes by elaborating the theory of 'stochastic resolution'.

2. Theoretic concepts for weighted simulation entries

In order to provide more clarity, the differentially weighted MC particles (also named 'stochastic particles' [25], or 'fictitious particles' [27] in other publications) will be called simulation entries (SE) for which the statistical weight (a concentration in units m^{-3}) and the properties of the SEs are stored on the computer.

2.1. Operator splitting technique

The simultaneous nucleation and coagulation processes are described by the following PBE:

$$\frac{\mathrm{d}n(v)}{\mathrm{d}t} = \frac{1}{2}\int_0^v \beta(v-v', v')n(v-v')n(v')dv' - n(v)\int_0^\infty \beta(v, v')n(v')dv' + \delta(v_{\mathrm{nuc}} - v)R_{\mathrm{N}}(t)\,. \tag{1}$$

The first two terms on the r.h.s. of the equation describe the coagulation process. $n(v)$ is the number density of particles with the property v and $\beta(v, v')$ is the coagulation kernel, describing the rate of coagulation between particles of types v and v' (the newly formed particle has the type $v + v'$). The particle properties are defined by the type space considered in the problem and this can be univariate or multivariate. The last term of Eq. (1) describes the insertion of particles with the property v_{nuc} with the nucleation rate $R_{\mathrm{N}}(t)$. For a given time-step τ_{mc}, the concentration of the nucleated particles can be written as:

$$W_{\mathrm{nuc}} = \int_{t_{\mathrm{start}}}^{t_{\mathrm{start}}+\tau_{\mathrm{mc}}} R_{\mathrm{N}}(t)\mathrm{d}t\,. \tag{2}$$

The nucleation rate R_{N} as well as the volume (or multivariate properties) of the nucleated particles, v_{nuc}, can be time-dependent, especially in the context of a physically induced homogeneous nucleation (e.g. [28,29]) due to the dependence