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Chemical Engineering Science 60 (2005) 963–967

Chemical Engineering Science

www.elsevier.com/locate/ces

Single-particle method for stochastic simulation of coagulation processes

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Received 3 December 2003; received in revised form 22 September 2004; accepted 28 September 2004
Available online 23 November 2004

Abstract

A Monte Carlo stochastic simulation algorithm based on a single-particle method is suggested to describe steady-state particle coagulation processes. The method does not require any information on nearby particles; instead a fictitious coalescence partner with a given size is generated. The main drawback that limited applicability of this method in the past was that for each control volume the particle size distribution function had to be sampled and stored. In the present study we applied a discrete representation of the distribution function that requires only small memory resources and allows fast updating.

Keywords: Coagulation; Population balances; Monte Carlo

1. Introduction

Coagulation and breakage of particles, droplets and bubbles that are suspended in complex recirculating flows are typical processes in numerous industrial and environmental systems. Due to the complexity and multidimensional nature of the processes involved, Monte Carlo statistical simulations have become one of the most efficient and maybe the only accessible numerical technique. The analogy between particle collision in suspensions and molecular collisions enables the application of methods previously developed for rarefied gas dynamics (Pai, 1974; Bird, 1976; Kitron et al., 1991).

The direct simulation Monte Carlo (DSMC) method can be formulated as follows. The flow volume is divided into cells. The particle ensemble is represented by computational particles such that a group of identical particles in the physical system is substituted by one computational particle. Provided that the particle sizes, positions and other necessary parameters are known at time t, the particle distribution at time $t + \Delta t$ is calculated by an operator-splitting technique which comprises free flow and a collision step.

In the free flow phase the particles move, without any collisions occurring, during the time interval Δt. Their positions, velocities, sizes, temperatures, etc., are determined from the equations of motion, heat and mass transfer. In the second splitting step a new particle ensemble is calculated by simulating spatially homogeneous coagulation in each cell, when binary collisions between particles are sampled randomly. At this step, a particle can collide only with those particles that are in the same cell irrespective of the relative positions of the particles within the cell. The overall solution is thus accurate to first order in Δt. This can be improved by higher order splitting schemes, such as the Strang splitting scheme (Strang, 1968).

An accurate spatial discretisation ranges from 10^3 to 10^4 cells in the computational domain in the two-dimensional case, while a typical three-dimensional flow is usually resolved with 10^5–10^6 cells. For a reasonable representation of a polydispersed particle ensemble, one needs 10–100 computational particles in each spatial cell. Thus, modelling a spatially inhomogeneous polydispersed system requires simultaneous tracking of 10^4–10^8 particles. Given that the particle number density is nonuniform over the flow region, it is either necessary to increase the total number of

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0009-2509/$ - see front matter © 2004 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ces.2004.09.062

computational particles, or use a weighted particle method with splitting and termination of particle trajectories (Rjasanow and Wagner, 1998) to resolve low-density regions. Both approaches are time-consuming.

However, a significant simplification can be achieved for steady flows. A single-particle (or alternatively, test particle) method (SPM) can be applied. The spatially homogeneous coagulation step does not require any information about neighbouring particles; instead a particle (which is referred to as test particle) coagulates with a fictitious collision partner that is generated according to the local particle distribution (Sommerfeld, 2001). Thus, the particle always has a collision partner even in a low-density region. As soon as the particle leaves the system, the new particle distribution function is recalculated. Since a particle visits many cells (especially if the flow has recirculation zones) as it crosses the flow region, a relatively small number of the particles is sufficient in order to update the new distribution function. In our work (Vikhansky and Kraft, 2004) the SPM has been used to calculate droplets coagulation and fragmentation in an axisymmetric rotating disc contactor. Less than 5000 particles were sufficient to reach a steady-state solution.

This procedure is iterative until convergence is achieved. Note that if the coagulation submodel is integrated in a computational fluid dynamics (CFD) code, the iterative nature of the above-described method should not be considered as a drawback. Most CFD methods use iterations to calculate a steady solution, i.e., an intermediate velocity field is used to calculate an approximated temperature distribution, etc. Thus the coagulation SPM step naturally fits the general iterative strategy.

Note that SPM for the Boltzmann equation has a history longer than the DSMC method (Haviland and Lavin, 1962). Recently this method has been applied for the spatially homogeneous coagulation–fragmentation problem (Ramkrishna et al., 1995; Ramkrishna, 2000). The main obstacle that limited wide application of SPM in the past was the necessity to store and update the particle distribution function in each cell of the computation domain. Haviland and Lavin (1962) stored the particle distribution function as a histogram, while Sommerfeld (2001) used an appropriate parametrisation. It was noted by Ramkrishna et al. (1995) that recalculation of the particle distribution function is the most time-consuming element of the method. This difficulty has been resolved by Vlasov (1966). According to this approach only the number density and parameters of few (maybe even one) particles are stored in each cell. These particles are referred to as field (or target) particles. When a test particle crosses a cell, one of the field particles is replaced by the test particle with a probability p that is proportional to the residence time t_{res} of the test particle in the cell. The number density of the target particles is also updated according to t_{res}. In the present study we investigate the applicability of the SPM to coagulation processes and discuss associated numerical issues.

2. Description of the single-particle method

To proceed further, consider a control volume V. The number of particles with size x that enter V is $n_{\mathrm{in}}(x)$, and the size-dependent residence time of a particle is $\theta(x)$. The Smoluchowski coagulation equation reads

$$\frac{\partial n(t,x)}{\partial t} = \tfrac{1}{2}\int_0^x K(x-x',x')n(t,x')n(t,x-x')\,\mathrm{d}x' + \frac{n_{\mathrm{in}}(x)}{V} - \int_0^\infty K(x,x')n(t,x')n(t,x)\,\mathrm{d}x' - \frac{n(t,x)}{\theta(x)}, \tag{1}$$

where $n(t,x)$ is the number density of the particles that have mass x at time t. The probability that two particles with masses x and x', respectively, coalesce during a small time interval $\mathrm{d}t$ is $K(x,x')\,\mathrm{d}t$.

Let us reformulate Eq. (1) in terms of mass density. Advantages of this formulation are discussed in Ramkrishna et al. (1995), Babovsky (1999) and Eibeck and Wagner (2001); note also that the description of particle distributions according to their mass is encountered in technological applications more frequently than number distributions. The mass density of the particles that have mass x at a time t is $m(t,x) = xn(t,x)$, the total mass density that V contains is $M = \int m(t,x)\,\mathrm{d}x$, and the mass of particles with size x that enter V is $m_{\mathrm{in}}(x) = xn_{\mathrm{in}}(x)$. The total flow rate of particles through V is $Q_{\mathrm{in}} = \int m_{\mathrm{in}}(x)\,\mathrm{d}x$. In order to reformulate the collision equation (1) in terms of mass density, we express $n(t,x)$ as $m(t,x)/x$, substitute it into (1) and multiply the equation by x. Note that if $K(x,x') = 0$ for $x \leqslant 0$ or $x' \leqslant 0$, the limits of integration in (1) can be extended from $-\infty$ to ∞. After some algebra we obtain (Ramkrishna, 2000; Eibeck and Wagner, 2001)

$$\frac{\partial m(t,x)}{\partial t} = \int \frac{K(x-x',x')}{x'} m(t,x')m(t,x-x')\,\mathrm{d}x' + \frac{m_{\mathrm{in}}(x)}{V} - \int \frac{K(x,x')}{x'} m(t,x')m(t,x)\,\mathrm{d}x' - \frac{m(t,x)}{\theta(x)}. \tag{2}$$

The factor $\frac{1}{2}$ before the first integral in Eq. (2) disappears because coagulation reduces the number of particles but does not affect their mass. Eq. (2) can be solved by the mass flow algorithm (MFA) (Babovsky, 1999; Eibeck and Wagner, 2001), which simulates evolution of N-particles until convergence to a steady state. Below we will use the MFA to validate the results obtained by SPM.