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Solution of the Bi-Variate Dynamic Population Balance Equation in Batch Particulate Systems

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Abstract

This paper presents a comparative study on the application of different numerical methods for the solution of the dynamic bi-variate population balance equation (PBE) in batch particulate systems. Specifically, particulate processes operating under the combined action of particle aggregation and/or particle growth are examined. In order to solve the bi-variate PBE the Galerkin finite element method, the sectional grid technique, and the stochastic particle model are employed. The performance of these numerical methods (i.e., their accuracy and stability) is assessed by a direct comparison of the calculated bi-variate particle size distributions to available analytical solutions.

Keywords: bi-variate, PSD, growth, aggregation, finite elements, stochastic.

1. Introduction

An important property of many industrial particulate processes is the particle size distribution (PSD) that controls key aspects of the process as well as the end-use properties of the product. In many processes, the dynamic PSD involves the distribution of one or more internal particle variables, i.e., number of radicals, amount of adsorbed species, porosity, etc. The quantitative determination of the evolution of such multi-variate PSDs in particulate processes is a rather complex numerical problem for it presupposes good knowledge of the particle nucleation, growth, and aggregation mechanisms in addition to the reaction kinetics (Ramkrishna and Mahoney, 2002).

The evolution of the PSD in particulate processes is commonly obtained by the solution of a population balance equation (Ramkrishna, 1985). In the bi-variate population balance equation (PBE), the distribution of particles is considered to be continuous over the volume v and an internal property x and is described by a number density function, n(v,x,t), that represents the number of particles, per unit volume, in a differential particle volume, v to v+dv, and internal property, x to x+dx, size range. The general bi-variate PBE for a batch particulate system is given by:

$$\frac{\partial n(v,x)}{\partial t} + \frac{\partial}{\partial v} G_v(v,x) n(v,x) + \frac{\partial}{\partial x} G_x(v,x) n(v,x) = S(v,x,t) + \int_0^{v/2} \int_0^{x/2} \beta(v-u, x-z, u, z) n(v-u, x-z) n(u,z) du dz - n(v,x) \int_0^{\infty}\int_0^{\infty} \beta(v,u,x,z) n(u,z) du dz \quad (1)$$

where β(v,u,x,z) is the aggregation rate kernel between particles of volumes v and u and internal properties x and z, S(v,x,t) is the nucleation rate function, and $G_v(v,x)$ and $G_x(v,x)$ are the growth rate functions for the particle volume and the internal property, respectively. In general, the initial condition of the PBE is $n(v,x,0) = n_0(v,x)$ and the boundary conditions are $n(0,x,t) = n_x(x,t)$ and $n(v,0,t) = n_v(v,t)$.
Despite the increased interest in solving the dynamic bi-variate PBE, the majority of the numerical methods described in the open literature have not been adequately tested, especially for long simulation times. The present study describes the numerical solution of the dynamic bi-variate PBE under the combined action of particle growth and aggregation. The bi-variate PSDs are determined for a number of simple test problems including aggregation as well as combined aggregation and growth. The calculated solutions are compared to the corresponding analytical solutions for the PSD and/or the PSD moments.

2. Numerical Solution of the Bi-variate PBE

This section describes the implementation of the finite element, the sectional grid, and the stochastic particle methods to the solution of the dynamic bi-variate PBE.

2.1 The Galerkin finite element method

A two-dimensional Galerkin finite element method (GFEM) was employed to solve the bi-variate PBE. Following the developments of Roussos et al. (2004), the GFEM formulation results in the following matrix equation:

$$[A]_{ij} \frac{\partial}{\partial t} n_j + [B]_{ij} n_j = h_B(n_i) - h_D(n_i) \tag{2}$$

where n_j is the vector of unknowns in global coordinates, and h_B and h_D denote the birth and death integrals, respectively. Bi-quadratic isoparametric elements were employed and the double integrals were determined based on Simpson's rule.
In order to reduce the interpolation error and increase numerical stability, a modified Lagrangian interpolation formula was adopted. Moreover, moving discontinuities in the PSD (typically arising in problems involving particle growth) were effectively alleviated by the use of an artificial diffusion term (Alexopoulos et al. 2004).

2.2 The bi-variate sectional technique

The bi-variate sectional grid technique (SGT) is an extension of the uni-variate moving pivot technique of Kumar and Ramkrishna (1997) to a particulate process with an internal particle property, x. According to the sectional approach (Kumar and Ramkrishna, 1996) the PBE is expressed in terms of the number distribution $N_{i,j}$ given by:

$$N_{i,j} = \int_{v=v_i}^{v=v_{i+1}} \int_{x=x_j}^{x=x_{j+1}} n(v,x)\, dx\, dv \tag{3}$$

The following bi-variate sectional PBE can then be easily derived:

$$\frac{dN_{il}}{dt} = \sum_{j,k} \sum_{m,n} \eta_{i,j,k}\, \eta_{l,m,n}\, \beta_{j,k,m,n}\, N_{j,m}\, N_{k,n} - N_{i,l} \sum_{j,m} \beta_{i,j,l,m}\, N_{j,m} \tag{4}$$

where $\beta_{i,j,m,n}$ is the aggregation rate kernel between particles of volume v_i and v_j and internal property x_m and x_n. The aggregation matrices η are given by (Kumar and Ramkrishna, 1996):

$$\eta_{i,j,k} = \begin{cases} (v_{i+1} - v)/(v_{i+1} - v_i) & \text{for} \quad v_i < v < v_{i+1} \\ (v - v_{i-1})/(v_i - v_{i-1}) & \text{for} \quad v_{i-1} < v < v_i \end{cases} \tag{5}$$

where $v = v_j + v_k$. In the presence of particle growth, the particle volume and internal property grids, v and x, move according to the growth rate functions:

$$\frac{dv}{dt} = G_v(v, x) \qquad \text{and} \qquad \frac{dx}{dt} = G_x(x, v) \tag{6}$$

The values of η are updated periodically and the aggregation part of the problem is solved just as in the zero-growth problem. The incorporation of nucleation in the moving grid formulation follows the procedure outlined in Roussos et al. (2004).

2.3 The stochastic particle method

Recently, several stochastic Monte-Carlo methods have been employed to describe the dynamic evolution of multi-variate PSDs. However, very little work has been done to validate these methods, especially with respect to their applicability to bi-variate problems over a wide range of conditions. In the stochastic particle formulation (SPM), a number of particles is initially sampled which then evolves under the influence of aggregation and/or growth. The sample particle number is kept above a minimum value, N_{min}, by increasing the size of the sample volume, V_S. The probability of an aggregation event occurring during a time interval, Δt, is given by:

$$p_{i,j,m,n} = \beta_{i,j,m,n} V_S / \Delta t$$

If the sample size of the bi-variate PSD is sufficiently large, the discrete bi-variate number distribution, $N_{i,j}$, can be obtained.

3. Simulation Results

The Galerkin FEM as well as the sectional grid technique and the stochastic particle methods were employed to determine the solution of the general bi-variate PBE for a size independent aggregation processes as well as for two different combined aggregation and growth processes. In all the simulations, the x-v domain was discretized into a number of elements (e.g., 20-50 in each direction) using a logarithmic discretization rule. The numerical methods were validated by comparison of the calculated bi-variate PSDs and/or PSD moments with the available analytical solutions.

3.1 Solution of the bi-variate PBE for pure particle aggregation

In the pure size-independent particle aggregation case, the bi-variate PBE was solved for a constant aggregation kernel $\beta(v,u,x,z) = \beta_0$. The initial distribution was equal to:

$$n(v, x, 0) = n_0 \exp(-v/v_0) \exp(-x/x_0) \tag{7}$$

where n_0, v_0 and x_0 are the characteristic number density function, particle volume and particle internal property values, respectively. The analytical solution is given by: (Gelbard and Seinfeld, 1978)

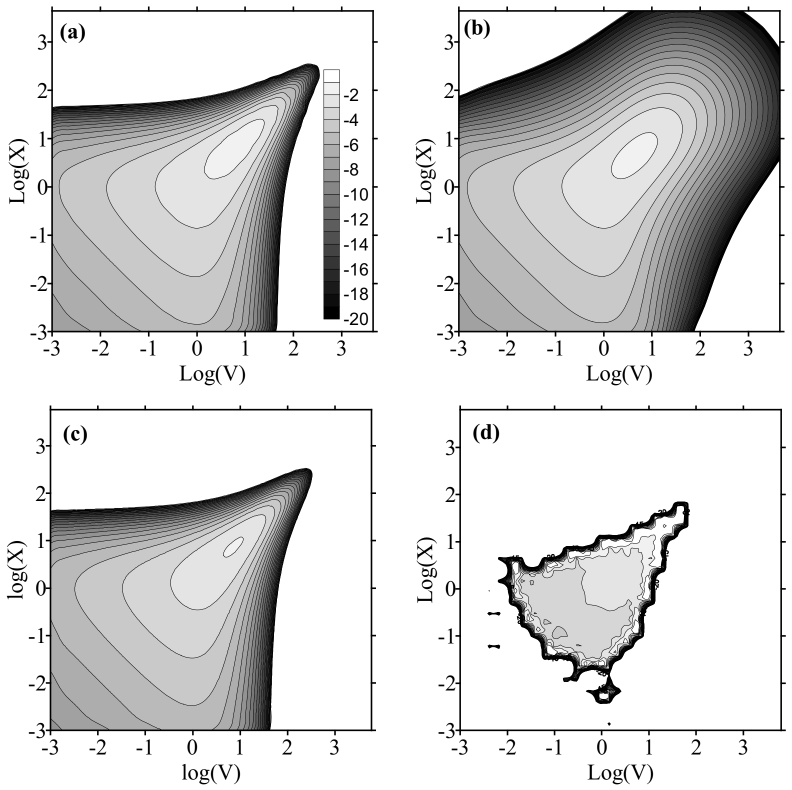

Figure 1. Contour plot of bi-variate PSD at τ = 10. (a) Analytical solution (b) SGT (20x20 elements) (c) GFEM (30x30 elements) (d) SPM (5000 particles)

$$n(v,x,t) \;=\; m_0^2\, n_0 \exp\left(-\frac{v}{v_0}-\frac{x}{x_0}\right) I_0\left[2\sqrt{(1-m_0)\frac{v}{v_0}\frac{x}{x_0}}\right] \qquad (8)$$

where: $m_0 = 2/(2+\tau)$ is the dimensionless total particle number and $\tau = \beta_0 N_0 t$ is the dimensionless aggregation time.

The bi-variate PBE was solved up to a dimensionless time of $\tau = 10$ using the numerical methods described in Section 2 (GFEM 30x30elements, SGT 20x20elements, SPM N_{min} = 10000). In Figure 1, the dimensionless number distributions, N_{ij} / N_0, are compared to the analytical solution in a series of contour plots. It is clear that the SGT displays significant diffusion error in the small number distribution region of the bi-variate PSD. However, the peak of the bi-variate PSD is well predicted (i.e., in terms of position and height) and the PSD moments are very accurate (i.e., errors < 0.5%). The most accurate PSDs were obtained by the GFEM but required more computational time. Although, the SPM produced accurate moments (to within 1% of analytical values) it could only resolve the peak region of the PSD. In Figure 2, the evolution of the SPM particle-cloud up to $\tau = 1000$ is shown. It is clear that the bi-variate PSD converges to a uni-variate distribution (i.e., of constant concentration x/v) due to aggregation-driven mixing. Note that at long times the stationary non-adaptive grids typically used in bi-variate sectional and FEM are very inefficient and, thus, the SPM becomes competitive.

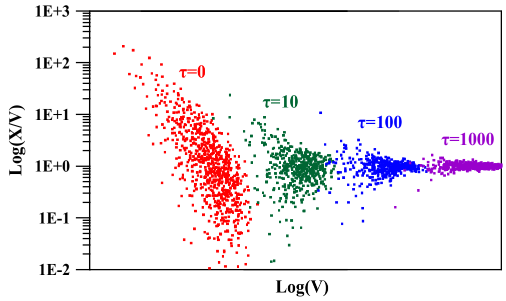

Figure 2. Evolution of the bi-variate PSD due to aggregation. SPM (2000 particles)

3.2 Solution of the bi-variate PBE for combined particle aggregation and growth.

The evolution of the PSD in a batch particulate process under the combined action of a size-independent particle aggregation kernel and a size-dependent particle growth rate was also examined. The following power law size-dependent growth rate functions, characteristic of many particulate processes, were employed:

$$G_v(v,x) = G_{0v}\,(v/v_0)^{\alpha} \qquad \text{and} \qquad G_x(v,x) = G_{0x}\,(x/x_0)^{\alpha} \tag{9}$$

where G_{0x} and G_{0v} are growth rate constants and α is a parameter.

For the initial condition given by eq. (7), the following analytical solution for n(v,x,t) can be obtained (Gelbard and Seinfeld, 1978):

$$n(v,x,t) = m_0^2\, n_0 \exp\left(-\frac{ve^{-\Lambda_1\tau}}{v_0} - \frac{xe^{-\Lambda_2\tau}}{x_0} - (\Lambda_1+\Lambda_2)\tau\right) I_0\left[2\sqrt{(1-m_0)\frac{v}{v_0}\frac{x}{x_0}e^{-(\Lambda_1+\Lambda_2)\tau}}\right] \tag{10}$$

where: $\Lambda_1 = G_{0v} / v_0\beta_0 N_0$, $\Lambda_2 = G_{0x} / x_0\beta_0 N_0$ and I_0 is the modified Bessel function of the first kind of zero order.

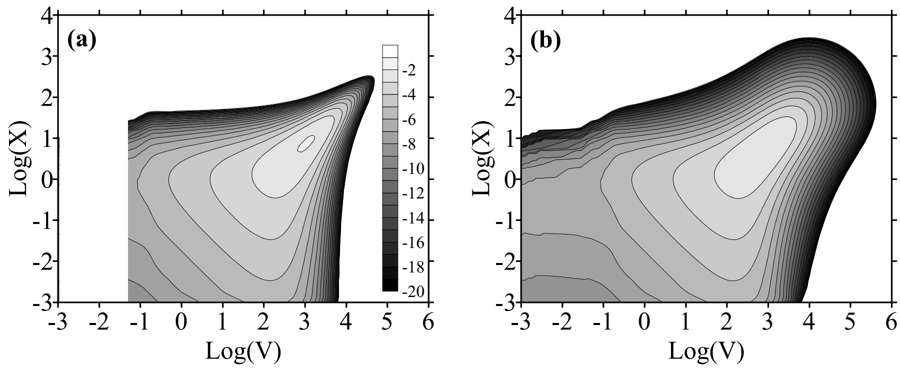

Figure 3. Bi-variate PSD at $\tau = 10$, $\Lambda_1=0.5$, $\Lambda_2=0$. (a) Analytical (b) SGT (30x30 elements)

The bi-variate PBE was solved up to a dimensionless time of $\tau = 10$ with $\beta_0 = 1$, $G_{0x}=0$ and $G_{0v} = 0.5$. In Figure 3, the number distribution calculated by the SGT is compared to the analytical solution. It is clear that the solution obtained by the SGT using a 30x30 grid displays less diffusion error than the solution obtained with a 20x20 grid in the zero-growth problem (Fig. 1b). From Table 1, it is also clear that the SGT is the most

Table 1. Comparison of numerical methods. Constant aggregation plus linear growth (τ=10, G_{0v}=0.5, G_{0x}=0, β_0=1, n_0=1)

Method	Δm_0, %	Δm_1, %	CPU (s)
SGT (30x30 elements)	0.00	0.09	1704
GFEM (30x30 elements)	0.52	3.83	3420
SPM (5000 particles)	0.40	2.33	449

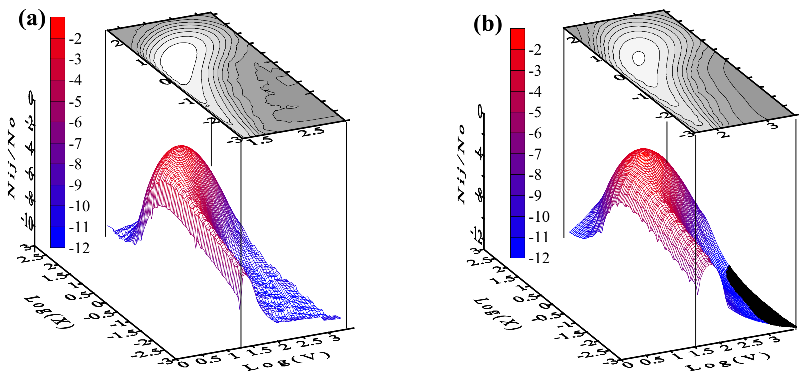

Figure 4: Bi-variate PSDs at τ=10 for combined aggregation and size-dependent growth (β_0=1, Λ_1=1, Λ_2=0) (a) α=1/3, (b) α=2/3. (SGT: 30x30 elements)

accurate method while the SPM is the most efficient computational method. In this case the stationary-grid GFEM is not as accurate as the moving SGT.

The bi-variate PBE was also solved using the SGT under the combined action of constant aggregation and different size-dependent growth functions. In Figure 4, the bi-variate PSDs at a dimensionless aggregation time of τ = 10 are displayed. It is observed that, due to the size-dependent nature of the growth function, the small-volume portion of the bi-variate PSD is "compressed" towards the main peak as the PSD evolves.

4. Conclusions

A bi-variate Galerkin FEM and sectional grid technique have been developed for solving the general dynamic bi-variate PBE. The bi-variate PSDs computed by the GFEM are compared to those calculated by the SGT for constant aggregation and combined constant aggregation and size-dependent growth problems. Overall, the GFEM and the SGT were found to be more accurate than the stochastic particle method but required more computational time.

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