498g On the Numerical Solution of Bi-Variate Population Balance Equations under the Combined Action of Nucleation, Growth and Aggregation Mechanisms

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An important property of many industrial particulate processes is the particle size distribution (PSD) which controls key aspects of these processes 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 population balance equations (PBE) which have been utilized in a diverse range of problems (Ramkrishna, 1985). In the bi-variate population balance equation, the distribution of particles is considered to be continuous over the volume, v, and an internal properties, 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 rate of change of the particle number density is described by the bi-variate PBE, which is a nonlinear integro-differential equation. The numerical solution of the PBE generally requires the discretization of the v and x particle domain into a small number of elements leading to a system of nonlinear ODE's or DAE's which is usually stiff. The PBE can be solved in its continuous form, e.g. by collocation on finite elements (Alexopoulos et al., 2004) or it can be solved in a discretized form (Kumar and Ramkrishna, 1996). Despite the increased interest in solving the dynamic multi-variate PBE, the majority of the numerical methods described in the open literature have not been adequately tested. In the present study the Galerkin finite elements method (GFEM), the sectional grid technique (SGT), and a Monte-Carlo (MC) stochastic method were employed to solve the dynamic bi-variate PBE under the combined action of particle growth, aggregation and nucleation. The bi-variate PSDs are determined for a number of simple test problems including aggregation, combined particle nucleation and aggregation, as well as combined particle growth and aggregation. The calculated solutions are compared to the corresponding analytical solutions for the PSD. For problems where analytical solutions are not available the numerical solutions are evaluated by comparison of the calculated to the analytical PSD moments. The bi-variate GFEM is an extension of the GFEM described in Roussos et al. 2005. Bi-quadratic iso-parametric elements were employed and the integral-terms 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). The bi-variate sectional grid technique (SGT) is an extension of the pivot technique of Kumar and Ramkrishna (1996) to a particulate process with an internal particle properties, x. The description of particle aggregation has been improved so that the bi-variate SGT is at least an order of magnitude faster than the original un-optimized version. Finally, a stochastic Monte-Carlo approach was employed in which a number of particles is initially sampled and then allowed to evolve under the influence of aggregation, nucleation and growth. The sample particle number is kept above a minimum value by increasing the size of the sample volume. The PSD as well as the PSD moments were calculated for a wide range of aggregation rate kernels as well as various nucleation and growth rate functions. 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 MC method typically employed 1000-10000 particles. For the case of pure size-independent particle aggregation, the bi-variate PBE was solved for a constant aggregation kernel and compared to the analytical solution (Gelbard and Seinfeld, 1978). It was found that the SGT displayed significant diffusion error in the tail region of the distribution just as in the uni-variate case. However, the peak of the bi-variate PSD was well predicted (i.e., in terms of position and height) and the PSD moments were

found to be accurate (i.e., errors < 0.5%). The most accurate distributions were obtained by the GFEM but required more computational time. Although, the MC method produced moderately accurate moments (to within 1% of analytical values) it could only resolve the peak region of the PSD. Note that for all pure-aggregation problems the bi-variate distribution converged to a uni-variate distribution (i.e., of constant concentration x/v) due to aggregation-driven mixing. The evolution of the PSD in a batch particulate process under the combined action of a size-independent particle aggregation kernel and a linear power law particle growth rate was also examined. The bi-variate distributions computed using the FEM, SGT and MV methods were compared to the available analytical solution (Gelbard and Seinfeld, 1978). For this problem the moving SGT was found to be the more accurate than the stationary-grid GFEM. Note that at long times the stationary non-adaptive grids typically used in the bi-variate SGT and FEM become inefficient and, thus, the MC method becomes competitive. Finally, the combined constant nucleation and constant aggregation case was examined. It was found that the GFEM was the most accurate method but required more computational time. It should be noted that the small-volume region of the bi-variate distribution was found to converge to a "steady-state" characterized by a constant total particle number just as in the uni-variate case.

References Alexopoulos, A.H., A.I. Roussos and C. Kiparissides, 2004, "Part I: Dynamic evolution of the particle size distribution in particulate processes undergoing combined particle growth and aggregation", Chem. Eng. Sci. 59, 5751-5769. Gelbard, F.M. and J.H. Seinfeld, 1978, "Coagulation and Growth of a Multicomponent Aerosol", J. Colloid Interf. Sci. 63, 472-479. Kumar, S. and Ramkrishna, D. 1996, "On the Solution of Population Balance Equations by Discretization-I. A Fixed Pivot Technique", Chem. Eng. Sci., 51(8), 1311-1332. Ramkrishna, D. 1985, "The Status of Population Balances", Rev. in Chem. Eng., 3(1), 49-95. Ramkrishna, D. and A.W. Mahoney, 2002, "Population balance modelling. Promise for the future", Chem. Eng. Sci. 57, 595. Roussos, A.I., A.H. Alexopoulos and C. Kiparissides, "Part III: Dynamic Evolution of the Particle Size Distribution in Continuous Flow and Batch Processes Using the Galerkin on Finite Element Method", Chem. Eng. Sci., 2005 (submitted).