Finite-Element Scheme for Solution of the Dynamic Population Balance Equation

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A first-order finite-element scheme is proposed for solving the 1-D dynamic population balance equation. The model includes the kinetic processes of nucleation, growth, aggregation, and breakage in a spatially uniform system with or without flow. The numerical scheme features improved stability by taking into account the convective nature of the growth term, and enhanced conservation of moments at nonuniform grids by identifying and mitigating the sources of conservation error in the evaluation of the aggregation source term. Exhaustive benchmarking is carried out against analytical solutions for size-dependent growth and nucleation, size-dependent aggregation with a variety of kernels, binary and multiple breakage with uniform and parabolic daughter particle distributions, and combinations of these, respectively. In all cases tested the method proves capable of producing accurate results at a reasonable CPU time, while being relatively simple and easy to implement.

Introduction

During the last century, the population-balance equation (PBE) has been established as a rigorous way of describing a variety of particulate systems such as cell populations, droplet and bubble swarms, granular solids, crystals, polymers, coagulating aerosols, and even astrophysical systems. In all of these situations, the aim is to predict the evolution of the distribution of one or more properties that characterize the individual particulate entities, and the dynamic PBE is in essence a number-balance equation describing this evolution.

The PBE originates in the work of Smoluchowski (1917), whose discrete equation for pure aggregation grew out of his study of Brownian motion. The equation was later cast in a continuous form and extended to include the process of breakage. Thus, in the 1960s Hulburt and Katz (1964) and Randolph (1964) formulated the PBE for nucleation and growth phenomena. Subsequent milestones, such as Drake's comprehensive survey (1972), the analysis of Ramkrishna and Borwanker (1973), and Ramkrishna's review (1985), did much to rationalize the PBE and establish it as a mainstream method for the analysis of particulate systems. In its most general form, the continuous PBE is a dynamic transport equation that describes the temporal evolution of population density as a result of four particulate mechanisms: nucleation, growth, aggregation, and breakage, as well as transport due to the flow field

$$\frac{\partial n(x_i,t)}{\partial t} = -\frac{\partial\left[u_i \cdot n(x_i,t)\right]}{\partial x_i} + D\frac{\partial^2 n(x_i,t)}{\partial x_i^2} + B_{\text{nuc}} + B_{\text{agg}} - D_{\text{agg}} + B_{br} - D_{br} \quad (1)$$

Written thus, the PBE refers to a state space comprised of time, physical space, and an additional number of dimensions that represent the characteristic properties of the particles whose distribution we wish to calculate (often referred to as internal coordinates). The gradient (index notation is used, so that repeated indices imply summation) term in Eq. 1, apart from input/output due to convective flow (differentiation in physical space), represents growth (differentiation in the particle size domain), or any other mechanism that operates on the population density in a convective way, as well as input/output due to convective flow. Similarly the diffusion term may represent both diffusion in physical space and effective diffusion of the population density, though the later has usually no physical interpretation.

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Unfortunately, the PBE yields analytical solutions for only a few special cases. Numerical solution of the PBE remains a considerable challenge, however, largely due to the integral form of the aggregation and breakage terms, and most approaches seem to have been tailored to specific applications (Rawlings et al., 1993). The aim of this work is to develop a numerical scheme that is capable of dealing effectively with an arbitrary particulate process, rather than being derived to match the needs of individual problems, while maintaining simplicity and speed. Particular emphasis will be put on stability and proper conservation of the moments. Solutions to the discrete Smoluchowski equation are beyond the scope of this article (see, for example, Drake, 1972); instead we shall focus on the continuous PBE. Monte Carlo methods will not be considered either; they are generally more efficient for multivariate problems, where the other approaches become too involved (Ramkrishna, 1985).

Review of previous work

One of the oldest and most widespread methods of solving the PBE is the method of moments (Hulburt and Katz, 1964; Randolph and Larson, 1988; McCoy and Madras, 2001), according to which the PBE is being transformed into a set of ordinary differential equations that provide the exact solution for the moments, but do not retrieve the entire distribution. Moreover, not all PBE formulations can be reduced to moment equations—in fact, this is usually the case with formulations that include aggregation and/or breakage. Thus, the use of moments is limited to special cases, and numerical methods are the only option if one wishes to retrieve the entire distribution from a continuous PBE that may include any combination of particulate mechanisms. Numerical methods for the PBE can be classified in two broad categories:

(1) Methods that retrieve the distribution by approximating the solution with a series of trial functions, whose coefficients are to be determined so that their sum will satisfy the PBE. When these functions are global, the scheme is a variation of the method of weighted residuals; in essence, the method of moments also belongs to this class (Ramkrishna, 1985). When the functions are local, we have a variation of the finite-element method.

(2) Methods that discretize the spectrum of the independent variable into a number of intervals and subsequently use the mean-value theorem to transform the continuous PBE into a series of equations in terms of either number of particles—termed the M-I approach—or average population density in each class—the M-II approach. These methods are often referred to as "discretized population balances" (DPBs).

Weighted residual methods with global functions were among the first to be tried, and one can consult Ramkrishna (1985) for a review of early attempts. It was soon realized, however, that global functions cannot capture the features of an arbitrarily-shaped distribution, especially if it exhibits sharp changes and discontinuities. If *a priori* knowledge of the shape of the resulting distribution is available, the trial functions can be tailored to accommodate it; in that case, the method converges and may even be computationally attractive. If generality and flexibility are required, however, the trial functions must clearly be local. Finite-element methods approximate the solution with piecewise low-order polynomials that are only locally nonzero, and are, thus, flexible and capable of capturing highly irregular solutions. The first notable attempt to solve the PBE with finite elements was carried out by Gelbard and Seinfeld (1978), who considered both orthogonal collocation and spline collocation for the solution of a dynamic PBE with nucleation, growth, and aggregation.

Meanwhile, an alternative approach emerged: the discretization of the domain of the independent variable. As pointed out by Gelbard and Seinfeld (1978), this approach, if straightforward, leads to severe errors unless a uniform grid is employed—unrealistic for anything but systems exhibiting a very small size range. For processes such as crystallization or coagulation of aerosols, where large particles can be created rapidly via aggregation, a nonuniform, preferably exponential grid must be employed. Bleck (1970) pointed out that the computational effort can be severely reduced if the grid assumes a geometric progression of (2^j)—this way, the number of combinations of particles that must be considered to evaluate the aggregation terms is substantially reduced. The next challenge for advocates of DPBs was to find a way of conserving the moments in such a grid. Batterham et al. (1981) modified this approach so that it conserves exactly the first moment with respect to volume (that is, the total mass) by dividing particles that were created at nonnodal points into fragments whose volume would lie at the nearby nodal points. This artificial way of conserving mass, however, introduced errors in all the other moments, as well as in the predicted population density. The most established method in this series, proposed by Hounslow et al. (1988), is capable of conserving exactly both total mass and number of particles, as well as accommodating simultaneous growth and aggregation. All these methods are, however, constrained with respect to the choice of grid, as they are based on exploiting the properties of the geometric progression. Recent works by Litster et al. (1995) and Kumar and Ramkrishna (1996a,b,c) have succeeded in extending the concept of DPBs to finer grids; the latter also offers the advantage of conserving any two chosen moments. Also notable are the "method of classes" due to Marchal et al. (1988), which treats aggregation as a chemical reaction, and the work of Hill and Ng (1995) on breakage processes, respectively.

After Gelbard and Seinfeld's pioneering work (1978), finite-element and other function approximation methods seemed to attract less attention than DPBs; even these authors made their own contribution to the DPBs (Gelbard et al., 1980). The main reasons for this trend are assumed to be the excessive, for that time, computational demands raised by these methods, as well as the complexity of their implementation, especially when applied to aggregation and breakage problems. Steemson and White (1988) used spline collocation to solve steady-state problems of nucleation, growth, and size dispersion; Eyre et al. (1988) also employed spline collocation, complemented with an adaptive grid. The recent advances in computational power, however, have brought finite-element methods back to the forefront. Nicmanis and Hounslow (1998) solved the steady-state PBE with nucleation, growth, aggregation, and breakage using collocation and Galerkin finite elements with cubic Lagrangian trial functions. The approach of Liu and Cameron (2001) to the dynamic PBE is also a function approximation method that uses wavelets to represent the solution, and focuses on accu-

rate prediction of discontinuities. Recent reviews have been contributed by Rawlings et al. (1993), Kostoglou and Karabelas (1994), Vanni (2000), and Lee (2001).

A comparison of finite-element methods with DPBs reveals that the former:

- Are a general and very flexible class of numerical methods and as such, when applied to the PBE, they can easily cope with any possible formulation, for example, size-dependent growth and different aggregation kernels;
- Retrieve the entire distribution with detail;
- Are more difficult to implement and computationally demanding;
- Their prediction of any moment is subject to errors.

On the other hand, DPBs are:

- Custom-derived methods tailored to solve individual problems efficiently but lack generality;
- Valued for their ease of use and computational efficiency;
- Engineered to predict some moments (usually one or two) *exactly*, but other moments may exhibit severe errors, as may their reproduction of the distribution.

The new approach

In light of these conclusions, the aim of this work is to derive a finite-element scheme that exhibits the following features:

- *Generality*. The dynamic PBE with nucleation, growth, dispersion, aggregation, and breakage will be considered, the only assumptions being spatial uniformity and one-dimensional (1-D) domain of the particulate variable.
- *Stability*. Linear trial functions will be employed and collocated in a way that reflects the convective nature of the growth term, resulting in a scheme that is stable even in problems with shocks.
- *Enhanced Conservation of the Moments*. A good numerical method must be conservative, if inaccurate, even in a coarse grid, but with the PBE, conservation is not straightforward due to the presence of the integral terms. The possible sources of error will be identified, and ways of mitigating them will be proposed.
- *Ease of implementation.* Owing to the use of linear trial functions, the resulting scheme will be very simple and easy to incorporate in a computer code.
- *Good balance of accuracy and speed*. As will be demonstrated by the test cases, very accurate results can be achieved within reasonable CPU time, even in desktop computers. The lower accuracy of the linear trial functions will be compensated by the use of a finer grid.

Formulation of the PBE

In Eq. 1, the independent variables are the dimensions of physical space (x_i, $i = 1,2,3$) and the internal coordinates. The most common internal coordinate is a measure of particle size, and previous work has often favored the use of particle diameter (for example, Gelbard and Seinfeld, 1978; Hounslow et al., 1988), due to the fact that it is the variable most commonly measured by particle-sizing equipment. In this work, however, it was decided to formulate the PBE in terms of particle volume in order to emphasize conservation through correct discretization of aggregation and breakage terms. In these two processes volume is being conserved, and, therefore, it is the natural choice of an independent variable. Furthermore, if particle diameter is chosen, the aggregation terms must include shape factors that are system dependent.

A spatially distributed PBE implies coupling with fluid dynamics. Such a coupling raises extreme computational demands, and so far has only been accomplished through simplification of other aspects of the problem (such as solution with the method of moments). In this work, the PBE will be formulated for a spatially uniform system by integrating Eq.1 over a uniform finite domain. The resulting equation is

$$\frac{\partial n(v,t)}{\partial t} = \frac{n_{in}(v,t) - n(v,t)}{\tau} - \frac{\partial}{\partial v}\left(G(v)\cdot n(v,t)\right) + B_0\,\delta(v - v_0) + \frac{1}{2}\int_0^v \beta_a(v - v', v')\,n(v - v',t)\,n(v',t)\,dv' - n(v,t)\int_0^\infty \beta_a(v,v')\,n(v',t)\,dv' + \int_v^\infty \beta_b(v')\,N_p(v')\,b(v/v')\,n(v',t)\,dv' - \beta_b(v')\,n(v) \quad (2)$$

What follows is only a brief explanation of the symbolism used; one is referred to Ramkrishna (1985) or Drake (1972) for an in-depth discussion. $\beta_a(v - v', v')$ is the aggregation kernel, in general a function of the volumes of the aggregating particles ($v - v'$, v'). The breakage-source term has to account for three things: the rate at which breakage events occur, the number of daughter particles created in each event, and the size distribution of these particles. The first one is represented by the breakage kernel, $\beta_b(v - v', v')$, while the other two are accommodated by the functions $N_p(v')$ and $b(v/v')$, respectively.

Nucleation is a point source implemented via a Dirac function. Growth is a convective mechanism that has the same dimensions as velocity – [unit of independent variable]/[unit of time]. Occasionally an effective diffusion term in the size domain is incorporated to account for growth-rate variations among particles of the same size. This phenomenon, occurring mainly in crystallization, is attributed to the following reasons (Randolph and White, 1974): (1) spatial effects (such as nonuniform supersaturation field, size classification due to flow field), and (2) variations in particle structure. Diffusional transport, however, implies the transport of a property due to the presence of gradients; rather than this, it is clear that the fluctuations mentioned earlier occur due to another variable not included in the model. For this reason, diffusion is not included in this formulation.

Equation 2 is a first-order hyperbolic differential equation that requires two initial conditions. The initial condition for the time domain is straightforward

$$n(v,0) = n_0(v) \quad (3)$$

With respect to the size domain, the most physically realistic choice is to state that there are no particles of zero volume

$$n(0,t) = 0 \quad (4)$$

The volume domain must then start from zero (rather than nuclei volume). Randolph and Larson (1988) proposed instead the condition $n(\nu_0,t) = B_0/G$ for nucleation–growth problems, which implies that the number of nuclei is determined by a balance between nucleation and growth. This condition is not valid for aggregation problems, because in that case nuclei may also be reduced via the aggregation mechanism, and moreover it implies that nuclei are of zero size, which is not physically realistic (Rojkowski, 1990).

Preliminary considerations

Truncation of the domain

To carry out computations, the infinite domain of the independent variable must be truncated to a finite value. For a dynamic simulation, the best practice is to use the moments to monitor the solution and determine whether the truncation has any effect. Aggregation–breakage problems can be monitored in this way on the basis of the first moment. In growth-dominated problems, the values of the population density that lie within the computational domain are not affected by the excluded tail. Since the whole body of the distribution is moving toward larger sizes, however, once it reaches the truncation point, the solution is clearly of no value. Usually the size range to be covered can be estimated beforehand from the growth kinetics.

Grid generation

The choice of grid has a profound effect on both accuracy and conservation. In aggregation and volume-dependent growth problems very big particles may be rapidly created, meaning that a wide range of volume must be covered, while nucleation creates a large number of particles in the smaller size ranges, thus giving rise to steep gradients that require a locally refined grid. In this work, the cases including aggregation will be solved on an exponential grid generated with the following series

$$\nu_i = \nu_{\min} + \nu_{\min}\frac{1-\alpha^{i-1}}{1-\alpha} \tag{5}$$

where α is real. For best performance the grid must be tailored to the needs of the problem under consideration, while an adaptive grid is probably the only way to cope efficiently with different cases and long-time simulations. Remeshing algorithms, however, are beyond the scope of this work.

The Main Steps of the Method

The time-dependent PBE (Eq. 2) is a hyperbolic partial integrodifferential equation with respect to time and the particulate coordinate. The procedure for its solution can be summarized in the following steps:

(1) Approximation of the solution with trial functions.

(2) Evaluation of the integral terms. The aggregation birth, death, and breakage birth terms must be evaluated via numerical integration.

(3) Formulation of weighted residual expressions. The trial functions are substituted into the resulting terms and any derivatives are analytically elaborated. The resulting function, the residual, is multiplied by appropriate weighting functions, and subsequently integrated over each element or, in the case of collocation, evaluated at the nodal points.

(4) Integration of the resulting ODEs along time, which can be carried out with any initial-value ODE integration algorithm.

It is interesting to compare this procedure with the implementation of the finite-element method in solid and fluid mechanics. The common practice there is to derive a weak form of the original equation, a step omitted here because the PBE is only first order with respect to both time and volume and the boundary conditions are all of Dirichlet type. Moreover, the discretized equations are often derived in terms of local variables defined within each element by a coordinate transformation, not possible in the PBE because of the aggregation source term that involves values of the independent variable defined within separate elements. Finally, the application of the finite-element method to an integrodifferential equation requires an extra step that must precede all others, the evaluation of the integrals with the trial functions. In what follows, it will be shown that this step is a potential source of errors in the conservation of moments if carried out in a straightforward manner.

Approximation of the solution

Previous applications of finite elements to the PBE employed cubic splines or high-order Lagrangian polynomials (for example, Gelbard and Seinfeld, 1978; Nicmanis and Hounslow, 1998). The scheme presented here, however, is based on linear trial functions for two main reasons. Higher-order trial functions require more evaluations and interpolations for the same number of nodal points, while first-order functions can accommodate a much finer grid in the same amount of computational time—note that higher order does not necessarily imply higher accuracy, especially when the solution is irregular. Second, we suggest a way of collocating the linear functions that improves stability for growth problems. Furthermore, these functions simplify considerably the scheme and facilitate the analysis of conservation errors.

Two linear trial functions are required:

$$c_{1,2}(\nu) = \left(\frac{\nu-\nu_{i-1}}{\nu_i-\nu_{i-1}}, \frac{\nu_i-\nu}{\nu_i-\nu_{i-1}}\right) \tag{6}$$

The solution over an element is then approximated as

$$n(\nu)_{[i-1,i]} = n(\nu_{i-1})\cdot\frac{\nu_i-\nu}{\nu_i-\nu_{i-1}} + n(\nu_i)\cdot\frac{\nu-\nu_{i-1}}{\nu_i-\nu_{i-1}} \tag{7}$$

In what follows, index i shall be used to denote both a nodal point and an element bounded by $[i-1,\ i]$ (point 0 corresponds to the lower boundary).

Evaluation of the integral terms

The three integral terms of the PBE (aggregation birth, aggregation death, breakage birth) are the main potential sources of conservation error in the numerical solution. Note that conservation is not a synonym of accuracy, which can be improved mainly by refining the grid. The aim of a conserva-

tive numerical method is to conserve the moments of the distribution (the most important being the first, whose conservation is essentially the mass balance) even in a coarse grid. To attain this, the sources generated by the discretization must be balanced by the sinks over all elements. In the following, two potential sources of conservation error will be identified:

- Error due to grid nonuniformity;
- Error due to nonconservative quadrature.

These errors will be elaborated below, and ways of mitigating them will be proposed.

Aggregation Source Term. The aggregation source term links each element with all elements preceding it. To evaluate it at an element boundary, v_m, the integral may be divided into a sum of contributions (note that to speed up the calculations, the aggregation source term may be evaluated up to $v/2$ instead)

$$B_{\text{agg}}(v_m,t) = \sum_{i=1}^{m}\left[\frac{1}{2}\int_{v_{i-1}}^{v_i}\beta_a(v_m-v',v')n(v-v',t)n(v',t)\,dv'\right] \quad (8)$$

To calculate the contribution from each preceding element $[i-1,i]$, we need the products of the nodal values at its boundaries, (n_{i-1}, n_i) with the values at its complementary points, that is, the points $(v_m - v_{i-1}, v_m - v_i)$. Suppose that these lie in a distant element k. Substituting the trial functions into Eq. 8 we obtain:

$$B_{\text{agg}}(v_m,t) = \sum_{i=1}^{m}\frac{1}{2}\left\{\int_{v_{i,1}}^{v_{i,2}}\beta_a(v_m-v',v')n(v_{i-1},t)\cdot\left[\frac{v_i-v'}{v_i-v_{i-1}}+n(v_2,t)\frac{v'-v_{i-1}}{v_i-v_{i-1}}\right]_i\cdot\left[n(v_{k-1},t)\frac{v_k-(v-v')}{v_k-v_{k-1}}+n(v_k,t)\frac{(v-v')-v_{k-1}}{v_k-v_{k-1}}\right]_k dv\right\} \quad (9)$$

Further elaboration of Eq. 9 is not possible, however, because it involves values of the independent variable that lie in two distinct, possibly isolated elements. This is the reason why the common practice of transformation into local element coordinates cannot be applied here. Another important point is that in a nonuniform grid the complementary points are not, generally, element boundaries, and may not even lie within the same element. In that case, their nodal values must be interpolated from the boundaries of the elements where they belong. This is a potential source of conservation error, best illustrated via a geometric representation of the aggregation source term. Figure 1 shows the distribution in the element i and the two neighboring elements where the complementary points lie. By determining the nodal values by interpolation, the resulting integration corresponds to aggregation of the particles represented by the area of the following tetrahedra

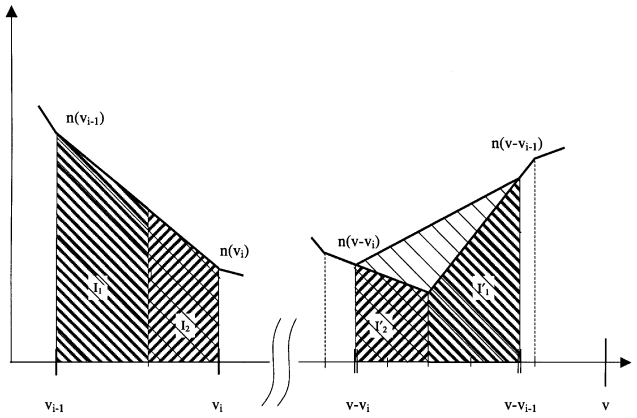

Figure 1. Geometrical representation of the conservation error arising in nonuniform grids.

$$\left[(v_i,0),(v_{i-1},0),(v_i,n(v_i)),(v_{i-1}n(v_{i-1}))\right] + \left[(v-v_i,0),(v-v_{i-1},0),(v-v_1,n(v-v_i))\,(v-v_{i-1},n(v-v_{i-1}))\right]$$

In the case of Figure 1, however, the second tetrahedron includes a triangle of nonexisting particles, which is the source of the conservation error. The correct aggregating particles are defined by the areas (I_1, I'_1) and (I_2, I'_2). To obtain a conservative algorithm, the interval defined by the mirror points will be divided into a number of subintervals, separated by the interelement boundaries that are located within that interval. If K-1 boundaries are contained, the integral over each element in Eq. 8 is split into a sum of K contributions from the resulting subintervals

$$B_{\text{agg}}(v_m,t) = \frac{1}{2}\sum_{i\text{-}1}^{m}\sum_{k=1}^{K}\int_{v_{i,k-1}}^{v_{i,k}}\beta_a(v',v_m-v')n(v_m-v',t)\,n(v',t)dv' \quad (10)$$

The integrand of Eq. 10 is nonlinear, meaning that numerical integration must be applied. This can be another source of conservation error, since any errors in it will result in an imbalance between the aggregation birth and death terms. To eliminate this error, the integration must be exact. Since the integrand comprises the product of the two trial functions, both first-order polynomials, and the aggregation kernel, if the latter is linear exact integration, it can be accomplished by adopting the following rule

$$\langle\text{Order of quadrature}\rangle = \langle\text{Order of kernel}\rangle + 2$$

Since the endpoints of the elements are readily available, a Newton–Cotes closed formula is preferable. The second-order formula, the familiar Simpson's rule, is also exact for third-order polynomials and needs only one interpolation.

The use of linear trial functions proves advantageous here, as higher-order functions would require more interpolations.

Finally, a note will be made about nonlinear kernels. In that case, exact integration can be attained via Gaussian quadrature, by using the kernel as a weight function. Press et al. (1992) describe a procedure for computing the coefficients of Gaussian quadrature with arbitrary weight functions. This process is rather complex, however, and for practical purposes a Newton–Cotes formula is usually acceptable, as the test cases will show.

Aggregation Sink Term. The aggregation sink term is easier to treat, since its integrand involves just the product of the population density and the aggregation kernel

$$D_{\text{agg}}(\nu,t) = n(\nu,t)\int_0^\infty \beta_a(\nu-\nu',\nu')n(\nu',t)\,d\nu' \quad (11)$$

As earlier, the evaluation should follow the rule

$$\langle\text{Order of quadrature}\rangle = \langle\text{Order of kernel}\rangle + 1$$

Breakage Source Term. This is the last integral term of the PBE, and may be written as

$$B_{br}(\nu,t) = \int_\nu^\infty \beta_b(\nu')b'(\nu/\nu')n(\nu',t)\,d\nu' \quad (12)$$

where $b'(\nu/\nu')$ is a function that incorporates both a degree of fragmentation and size distribution of fragments. To maintain conservation, integration should follow the same rules proposed earlier for aggregation: the quadrature should ideally be exact for the combination of trial functions, breakage kernel, and daughter particles distribution

$$\langle\text{Order of quadrature}\rangle = \langle\text{Order of kernels}\rangle + \langle\text{Order of daughter particle distribution}\rangle + 1$$

Formulation of weighted residual equations

So far, the integral terms of the PBE have been evaluated by way of numerical integration. The weighted residual expressions over each element are formulated by multiplying the PBE with an appropriate weight function and integrating. Common choices of weight functions are

$$\text{Collocation:} \quad w(\nu) = \delta(\nu-\nu_k) \quad (13)$$

$$\text{Subdomain:} \quad w(\nu) = 1 \quad (14)$$

$$\text{Galerkin:} \quad w(\nu) = c_i(\nu) = \left(\frac{\nu-\nu_{i-1}}{\nu_i-\nu_{i-1}}, \frac{\nu_i-\nu}{\nu_i-\nu_{i-1}}\right) \quad (15)$$

Collocation is an evaluation of the residual at the nodal points. The other schemes require integration of the residual over an entire interval, and are, thus, more expensive to compute. Moreover, integral schemes cannot be combined with the algorithm proposed earlier for a conservative evaluation of the aggregation source term, because they require the integrand to be continuous. For these reasons, we chose to employ the collocation scheme.

Growth. In orthogonal collocation on finite elements (Carey and Finlayson, 1975), the trial functions are higher than first order and the evaluation is performed at points interior to each element. As a result, information flows to the collocation point from both ends of the element. This strategy is not efficient for discretizing the growth term because growth is a convective mechanism, which implies that the population at one point is affected only by upstream points (since particles can only grow from smaller to bigger sizes). Without a diffusion term to dampen the convective transport, the use of schemes where information flows in both directions can result in instabilities and oscillations. The following "upwind" evaluation scheme is proposed

$$\int_{\nu_{i-1}}^{\nu_i}\left[w(\nu)\cdot\frac{\partial}{\partial\nu}(G(\nu)\cdot n(\nu,t))\right]d\nu = \frac{n_i(t)}{\nu_i-\nu_{i-1}} - \frac{n_{i-1}(t)}{\nu_i-\nu_{i-1}} + n_i(t)\cdot\frac{\partial}{\partial\nu}(G(\nu))\bigg|_i \quad (16)$$

Nucleation. Nucleation is implemented as a point source at the first nodal point. With finite elements it is impossible to have true point sources, however, as trial functions are valid over a whole interval, and the source results in a "spike" spreading over the first two intervals. Care must be taken, therefore, in order to make sure that the nucleation rate $B_0(t)$ reflects the total number of particles produced in the first two intervals that are affected, and has units of (time^{-1} volume^{-1})

$$\int_{\nu_{i-1}}^{\nu_i}\left[w(\nu)\cdot B_0(t)\,\delta(\nu-\nu_0)\right]d\nu = \frac{B_0}{\frac{1}{2}(\nu_1-\nu_0)} + \frac{B_0}{\frac{1}{2}(\nu_0-0)} \quad (17)$$

Integration of ODEs

Having performed the pointwise evaluations of the PBE terms at the nodal points, we have succeeded in transforming the original partial integrodifferential equation into a set of ordinary differential equations (ODEs), where the dependent variables are the nodal values. The integration of these ODEs along time is bound to introduce further errors in terms of both accuracy and conservation, which depend on the accuracy of the integration algorithm. The results presented here were obtained using the LSODE integration package (Hindmarsh, 1983), using an implementation of the Adams class of methods with adaptive time-stepping. The source code of LSODE is available in the public domain (such as via netlib.org). In most cases, the error in the first moment was maintained below 1%. The code was compiled in FORTRAN 90.

Results and Discussion

The method will now be tested against both time-dependent analytical solutions of batch systems and steady-state so-

Table 1. Aggregation

Case	I-1		I-2		I-3	
Kernel	k_a	k_a	$k_a(v+v')$	$k_a(v+v')$	Shear (Eq. 23)	Gravitational (Eq. 24)
k_a	1	1	1	1	1	1
N_0	1	1	1	1	1	1
v_0	1	1	1	1	1	1
t (s)	10	20	0.5	2	0.5	0.5
T	10	20	0.5	2	—	—
Grid type	Exp.	Exp.	Exp.	Exp.	Exp.	Exp.
α (grid exponent)	1.05	1.05	1.04	1.04	1.04	1.04
No. of elements	100	100	150	150	200	200
Size range	0.1–100	0.1–100	0.1–100	0.1–100	0.1–100	0.1–100
I_{agg}, %	82	90	39	86	77	63
Error in M_1, %	0.12	0.51	0.39	1.021	0.87	0.91
CPU (s)	0.5	1	0.8	1.5	2	20

lutions of systems with flow. The CPU times reported correspond to an average desktop PC (Pentium III, 933 MHz).

Aggregation

The continuous time-dependent PBE for aggregation only is

$$\frac{\partial n(v,t)}{\partial t} = \frac{1}{2}\int_0^v \beta_a(v-v',v')n(v-v',t)n(v',t)\,dv' - n(v,t)\int_0^\infty \beta_a(v,v')n(v',t)\,dv' \quad (18)$$

Most analytical solutions have been derived with the exponential initial distribution

$$n(v)|_{t=0} = \frac{N_0}{v_0}\cdot e^{(v/v_0)} \quad (19)$$

For the pure aggregation cases an exponential grid is preferable. The extent of aggregation in each case may be quantified by the aggregation index due to Hounslow (1990)

$$I_{agg}(t) = 1 - \frac{m_0(t)}{m_{0\text{initial}}} \quad (20)$$

Table 1 summarizes parameters and results for the aggregation cases.

Case I-1: Size-Independent Kernel

$$\beta_a(v-v',v') = k_a \quad (21)$$

This kernel can be used as an approximation of Smoluchowski's equations for Brownian coagulation (perikinetic aggregation). An analytical solution for the exponential initial distribution has been derived by Scott (1968). Comparison with the numerical and analytical solutions for the population density (Figure 2) for two values of a dimensionless time ($T = b_a N_0 t$, where N_0 is the initial number of particles) shows excellent agreement. The conservation of the first moment is also good, as shown in Table 1, despite the use of a fairly coarse grid.

Case I-2: Sum (Golovin) Kernel

$$\beta_a(v-v',v') = k_a(v+v') \quad (22)$$

A solution for this kernel and an exponential initial distribution has also been provided by Scott (1968). Results for the population density are plotted in Figure 3, and as with the case of the constant kernel agreement, is very good.

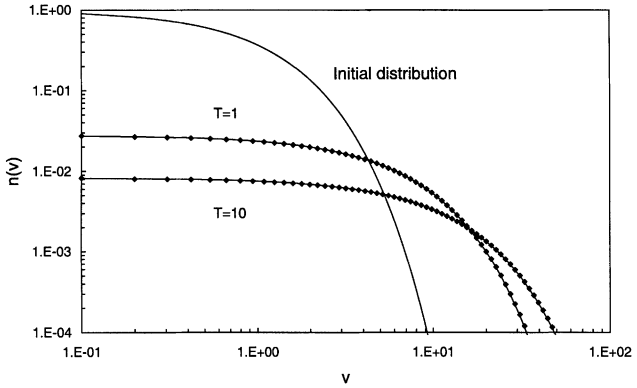

Figure 2. Aggregation, constant kernel, comparison of simulation with the analytical solution of Scott (1968).

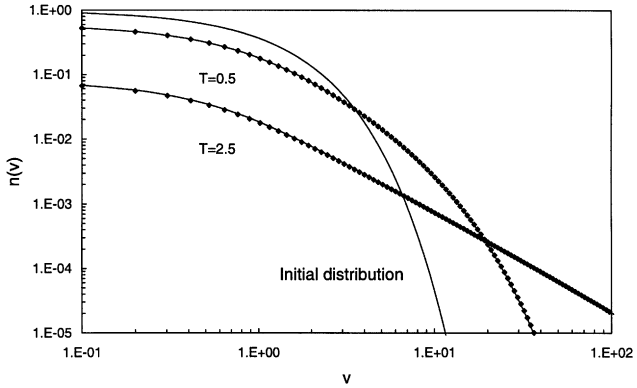

Figure 3. Aggregation, sum kernel, comparison of simulation with the analytical solution of Scott (1968).

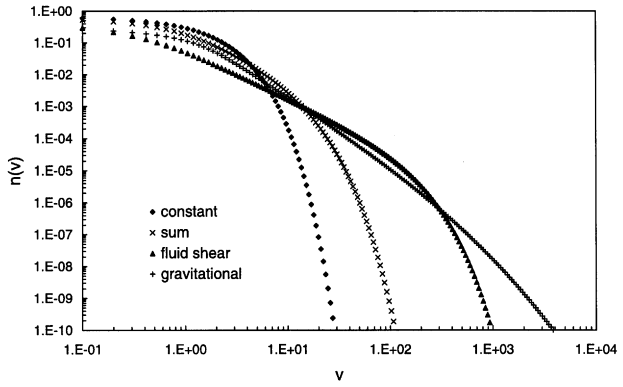

Figure 4. Aggregation, constant, sum, fluid shear, and gravitational kernels, simulation results.

Case I-3: Fluid Shear and Gravitational Kernels. Smoluchowski (1917) derived the following kernel to represent aggregation due to hydrodynamic effects such as laminar shear flow (orthokinetic aggregation)

$$\beta_a(v-v',v') = k_a\left[(v-v')^{1/3}+v'^{1/3}\right]^3 \qquad (23)$$

Another kernel derived on the basis of physical arguments is the following, used to describe gravitational coalescence (Drake, 1972)

$$\beta_a(v-v',v') = k_a\left[(v-v')^{1/3}+v'^{1/3}\right]^2\cdot\left|(v-v')^{2.3}-v'^{2/3}\right| \qquad (24)$$

In these kernels the parameter k_a is usually a function of hydrodynamic variables; such functions are not supplied here, since our interest in the kernels is only from the point of view of numerical analysis. Although there are no analytical solutions for these kernels that we are aware of, they will be considered according to their physical significance. The results shown in Table 1 have been obtained using Simpson's rule for integrating the kernel over the subintervals, and evidently the conservation error is negligible. In Figure 4, the distributions resulting from these kernels are compared with those of the sum and constant kernels.

Nucleation growth

Nucleation-growth problems have been studied extensively by, among others, Randolph and Larson (1988). The dynamic PBE for nucleation-growth with flow is

$$\frac{\partial n(v,t)}{\partial t} = \frac{n_{in}(v,t)+n(v,t)}{\tau} - \frac{\partial}{\partial v}\left(G(v)\cdot n(v,t)\right) + B_0\,\delta(v-v_0) \qquad (25)$$

Because this is a hyperbolic equation, the main challenge for a numerical method is to capture the steep moving fronts and shocks that may emerge. Growth problems are best studied on a uniform grid. Parameters and results are shown in Table 2.

Case II-1: Size-Independent Growth and Nucleation with Flow. Most analytical solutions refer to size-independent growth. Since our formulation employs volume as an independent variable, for comparison we must consider volume-independent growth. This is rather unrealistic, but in a formulation without aggregation the choice of an independent

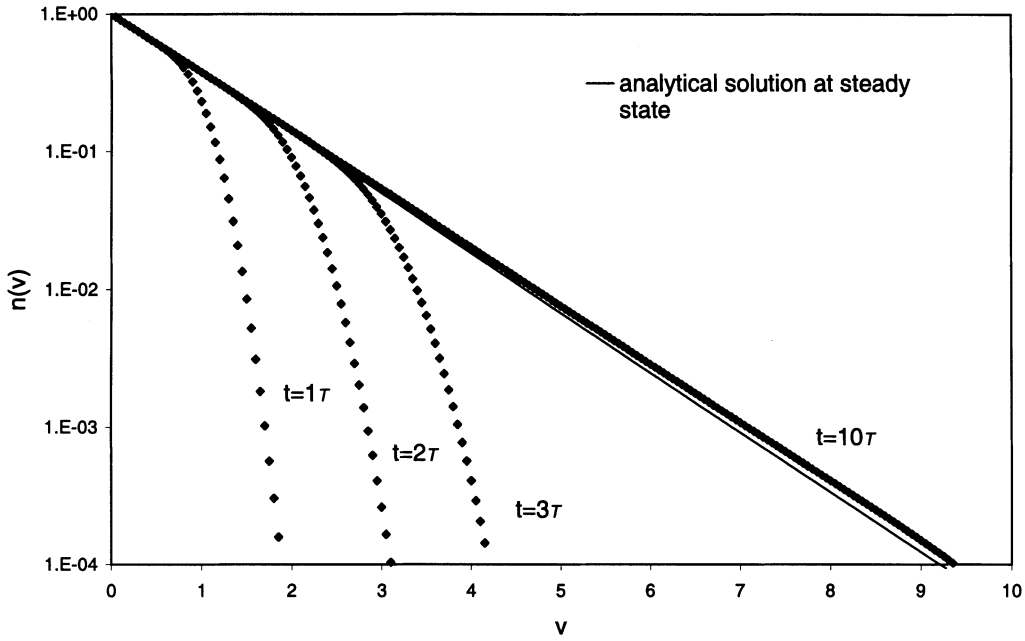

Figure 5. Steady-state size-independent nucleation growth, comparison of simulation with the analytical solution of Randolph and Larson (1988).

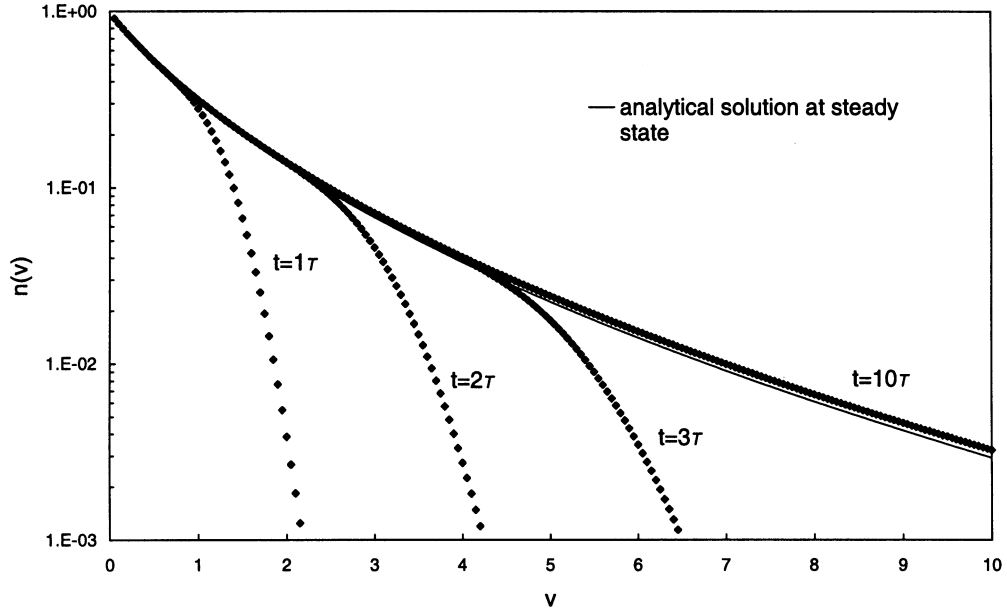

Figure 6. Steady-state-size-dependent nucleation-growth, comparison of simulation with the analytical solution of Randolph and Larson (1988).

Table 2. Nucleation Growth

Case	II-1	II-2	II-3	
Growth kernel	G_0	$G_0(1+\gamma v)^\beta$	G_0	G_0
B_0	1	1	1	1
G_0	1	1	1	1
γ	1	1	—	—
β	—	1	—	—
t (τ)	10	10	1	2
Grid type	Uniform	Uniform	Uniform	Uniform
Grid increment	0.5	0.5	0.002	0.002
No. of elements	200	200	1200	1200
Size range	0.1–10	0.1–10	0.002–2.4	0.002–2.4
CPU (s)	1	1.5	5	8

variable is irrelevant. At steady state, the solution is the well-known MSMPR equation (Randolph and Larson, 1988). Figure 5 shows the time-dependent numerical solution and the steady-state analytical solution. After several residence times, the two curves merge, and comparison is very good. Table 2 shows the numerical parameters and CPU time required for 10 residence times.

Case II-2: Size-Dependent Growth and Nucleation with Flow. A commonly used size-dependent growth function is (Randolph and Larson, 1988)

$$G = G_0(1+\gamma\nu)^\beta \tag{26}$$

This function involves the growth rate of nuclei, G_0, and two empirical parameters. These authors also provide a steady-state solution for this class of functions. Figure 6 shows the numerical solution at increasing values of the residence time. After 10 residence times, the simulation has approximated the analytical solution at steady state.

Case II-3: Batch Size-Independent Nucleation Growth. The analytical time-dependent solution to Eq. 25 for size-independent growth, no flow and no particles present at $t = 0$, can be found in Hounslow et al. (1988). This is a very ill-conditioned problem, as the solution is simply a discontinuity that moves with time. Such a discontinuity is very difficult to capture, and a potential source of numerical instability. The method of Hounslow et al. (1988), being of zero order, proved stable enough, but its prediction of the front was smeared by numerical dispersion. This effect can only be mitigated by the use of a finer grid, but in that method the grid is constrained to follow a geometrical progression. The finite-element method, on the other hand, may employ a uniform grid that is most suitable for this case. Figure 7 compares numerical and analytical results. The stability of the method is owing to the first-order trial functions and the upwind discretization of growth. Dispersion around the discontinuity is inevitable, since this is a first-order method, but it can be controlled by refining the grid.

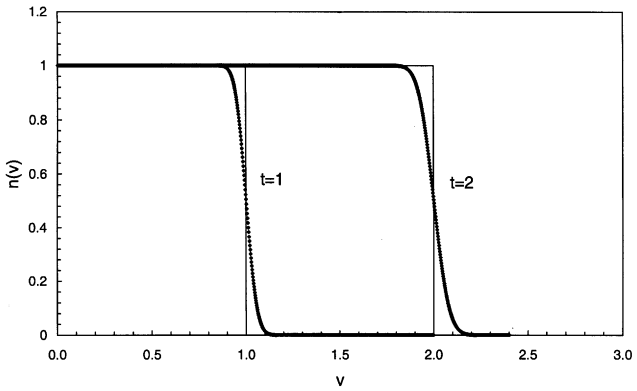

Figure 7. Batch size-independent nucleation-growth, comparison of simulation with the analytical solution of Hounslow et al. (1988).

Table 3. Breakage

Case	III-1			III-2			III-3	
Breakage kernel	$k_b v$	$k_b v$	$k_b v$	$k_b v$	$k_b v$	$k_b v$	$k_b v^\lambda$	$k_b v^\lambda$
Daughter particle distribution	$2/v$	$2/v$	$2/v$	Parabolic (Eq. 30)	Parabolic (Eq. 30)	Parabolic (Eq. 30)	Multiple (Eq. 31)	Multiple (Eq. 31)
k_b	1	1	1	1	1	1	1	1
N_0	1	1	1	1	1	1	1	1
v_0	1	1	1	1	1	1	1	1
t	1	10	20	10	10	10	0.1	1
Grid type	Exp.	Exp.	Exp.	Exp.	Exp.	Exp.	Uniform	Uniform
Grid exponent	1.02	1.02	1.02	1.02	1.02	1.02	0.1	0.1
No. of elements	200	200	200	200	200	200	200	200
Size range	0.01–25	0.01–25	0.01–25	0.01–10	0.01–10	0.01–10	0.01-10	0.01-10
Error in M_1, %	0.41	0.53	0.64	0.54	0.53	0.48	0.11	0.68
CPU (s)	14	20	23	20	20	20	10	14

Breakage

The equation for breakage without flow is

$$\frac{\partial n(v,t)}{\partial t} = \int_v^\infty \beta_b(v')b'(v/v')n(v',t)\,dv' - \beta_b(v)n(v,t) \tag{27}$$

Three representative cases will be considered here: uniform binary breakage, parabolic binary breakage, and fragmentation (multiple breakage). Results are shown in Table 3.

Case III-1: Uniform Binary Breakage. Uniform breakage implies that fragments of every size have the same probability to form. This situation can arise in polymer chains which, being one-dimensional, are equally likely to break at any point. If the breakage is also binary, it can be represented by the following overall function

$$b'(v/v') = \frac{2}{v'} \tag{28}$$

It can be easily confirmed that this function satisfies the above constraints just cited. Ziff and McGrady (1985) derived an analytical time-dependent solution for this case, with a size-dependent kernel and an exponential initial distribution. Figure 8 shows the initial distribution and the distributions resulting from breakage at different times. The numerical solution compares very well.

Case III-2: Parabolic Binary Breakage. Rigid particles are more likely to break into unequal pieces. This can be accounted for by using a parabolic "U-shape" function to describe the daughter particle-size distribution. An overall

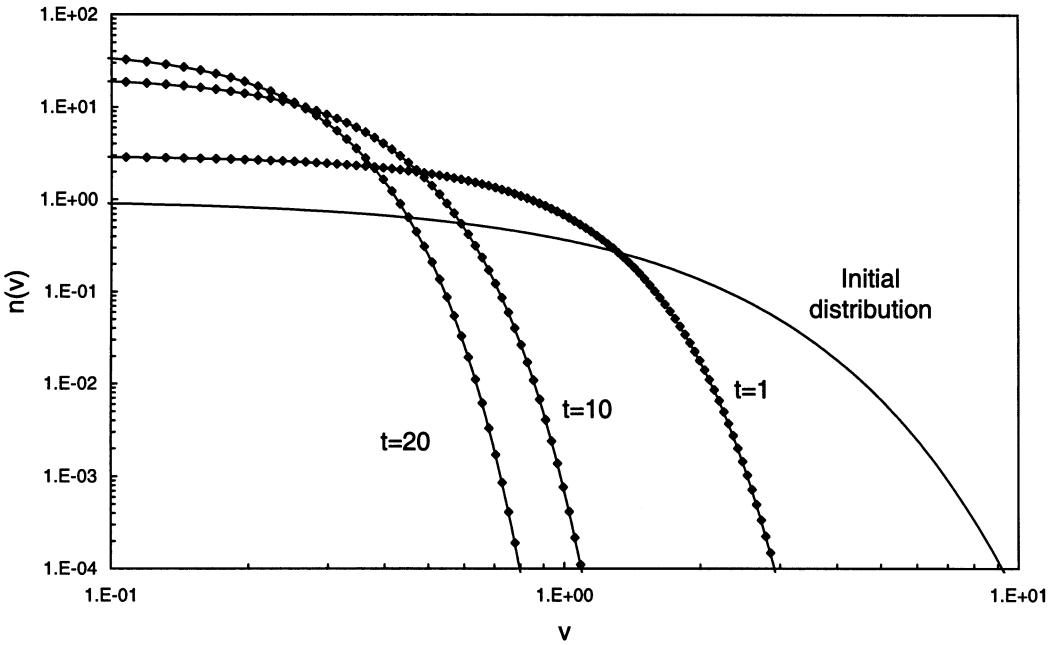

Figure 8. Batch uniform binary breakage, comparison of simulation with the analytical solution of Ziff and McGrady (1985).

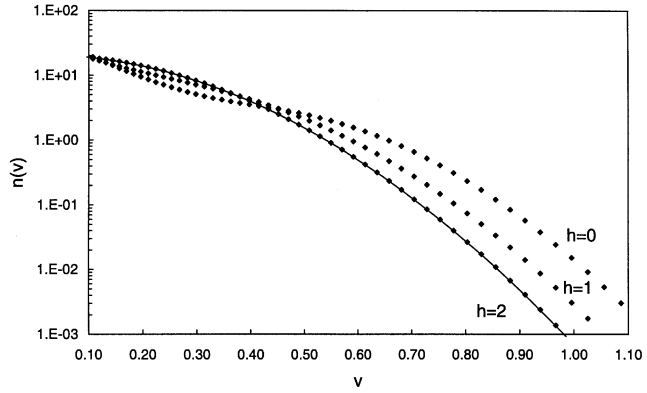

Figure 9. Batch parabolic breakage, simulation for $h = 0,1,2$ and comparison with analytical solution (Ziff and McGrady, 1985) for $h = 2$.

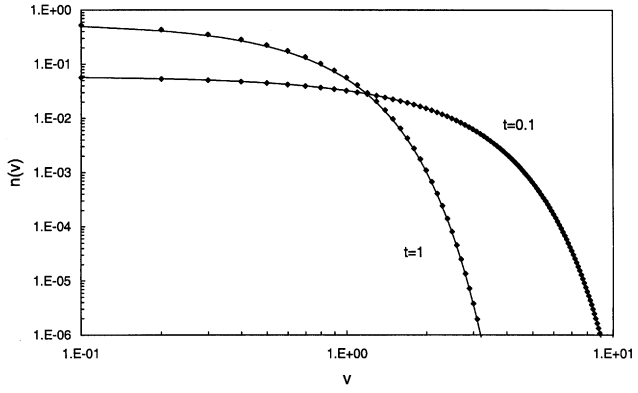

Figure 10. Batch ternary fragmentation, comparison of simulation with the analytical solution of Ziff (1991).

function describing parabolic binary breakage has been suggested by Hill and Ng (1995)

$$b'(v/v') = \frac{24\left(1-\frac{h}{2}\right)\left(v^2 - vv' + \frac{v'^2}{4}\right)}{v'^3} + \frac{h}{v'} \qquad (29)$$

For $0 < h < 2$, the function represents a concave parabolic profile with variable steepness. At $h = 2$ the function is equal to the uniform binary breakage, while for $2 < h < 3$, the function represents a convex parabola. Outside these bounds the function assumes unrealistic (negative) values. Figure 9 shows the simulation results for three values of h. To the authors' knowledge there is no general analytical solution except for $h = 2$, in which case the simulation approximates very well the solution for uniform binary breakage. The first moment is properly conserved in all three cases. As expected, parabolic breakage favors smaller and bigger particle sizes in the expense of the intermediate ones, resulting in a fairly bimodal distribution.

Case III-3: Multiple Breakage (Fragmentation). Ziff (1991) presented an overall distribution function that describes fragmentation

$$b'(v/v') = \frac{a\gamma}{v'}\left(\frac{v}{v'}\right)^{\gamma-2} + \frac{(1-a)\delta}{v'}\left(\frac{v}{v'}\right)^{\delta-2} \qquad (30)$$

where α, γ, δ are parameters. The average number of fragments produced can be determined by the following

$$N_p(v') = \int_0^{v'} b'(v/v')\, dv \qquad (31)$$

Ziff (1991) derived complete time-dependent solutions for a subset of this class, defined by

$$b'(v/v') = \frac{1}{v'}\frac{\delta\lambda}{\delta-\lambda}\left(\frac{\lambda}{v'}^{\lambda-2} - \frac{v}{v'}^{\delta-2}\right) \qquad (32)$$

$$\beta_b(v) = k_b \cdot v^{\lambda} \qquad (33)$$

for a monodisperse initial condition. Figure 10 shows the numerical and analytical solution to the case of ternary fragmentation ($\lambda = 2,\ \delta = 3$). A uniform grid was employed for this simulation. To implement the monodisperse initial condition, an initial distribution resembling a spike had to be employed, resulting in a rather ill-conditioned problem; nevertheless, solution was easily obtained.

Combined problems

Case IV-1: Nucleation–Aggregation with Flow. Hostomský (1987) derived a semianalytical solution for steady-state nucleation and size-independent aggregation with flow and no particles in the feed. Under these conditions, the PBE is written as

$$\frac{1}{2}k_a\int_0^{v} n(v-v')n(v')\, dv' - n(v)k_a\int_0^{\infty} n(v')\, dv' + B_0\delta(v-v_0) - \frac{n(v)}{\tau} = 0 \qquad (34)$$

Hostomský's solution is semianalytical because its derivation involves a Taylor series expansion, and is, therefore, subject to error. It holds for $v > 10v_0$ and is more accurate for higher volumes. Hostomský also provided an equation for the zeroth moment. Figure 11 compares this solution with the numerical simulation. A discrepancy in the first points is due to the semi-analytical solution, which is inaccurate at that range. At $v > 10v_0$ agreement is quite good, and the zeroth moment is also well predicted (Table 4).

Case IV-2: Batch Aggregation-Growth. This case was studied by Ramabhadran et al. (1976). They derived an analytical solution for a size-independent aggregation kernel, a first-order linear size-dependent growth term ($G = k_g v$), and the exponential initial distribution. Figure 12 shows the analytical and numerical results. According to the exponential initial distribution used to derive the analytical solution, $n(v)$ assumes the value N_0/v_0 at $v = 0$. This is unrealistic, however, and the present formulation has the boundary condition $n(0,t) = 0$, while the population density starts to assume the initial distribution at the size of the nuclei. Thus, the initial

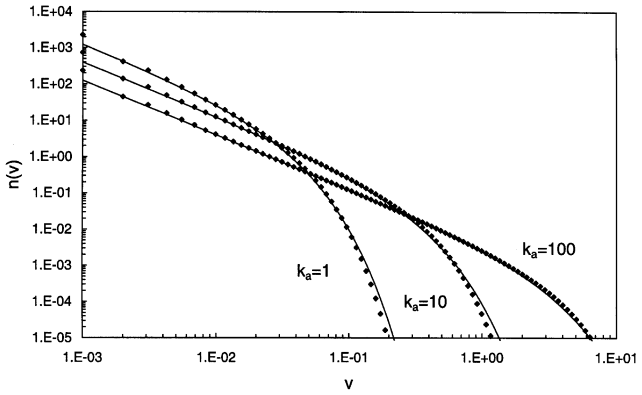

Figure 11. Steady-state aggregation (constant kernel) and nucleation, comparison of simulation with the semianalytical solution of Hostomsky (1987).

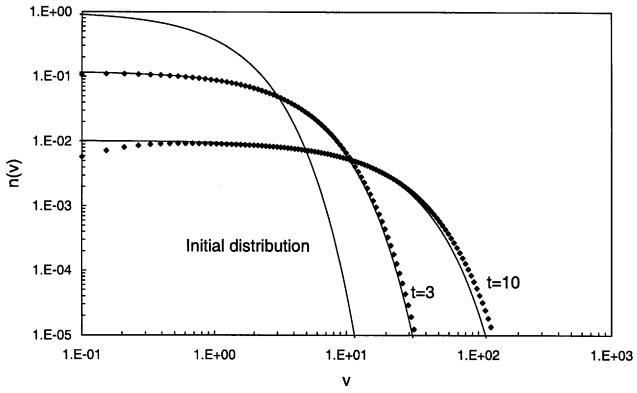

Figure 12. Batch aggregation (constant kernel) and growth (size-dependent), comparison of simulation with the analytical solution of Ramabhadran et al. (1976).

distribution used in the simulation inevitably deviates from the one used to derive the analytical solution, resulting in a slight disagreement of the results at the lowest end of the size range.

Conclusions

In this work, a finite-element scheme was proposed for solving dynamic population-balance problems. The PBE and boundary conditions were formulated to accommodate an arbitrary problem involving any combination of the four main particulate mechanisms: nucleation, growth, aggregation, and breakage. The numerical scheme consists of collocation on linear elements with an "upwind" propagation of first-order derivatives (that is, growth) that results in enhanced stability. Integral terms are evaluated with Newton–Cotes quadrature, and a three-point formula (Simpson's rule) requiring just one interpolation was found to be sufficient for most kernels. Three sources of error in the conservation of moments were identified: the nonuniform grid, the quadrature employed for the integral terms, and the temporal integration of the resulting ODEs. An accurate way of evaluating the aggregation source term, based on a geometric interpretation of it, and a rule for determining the order of quadrature were proposed to deal with the first two sources of error, respectively. Although the third source is inevitable, the results demonstrated that if the preceding rules are followed, the conservation error can be mitigated.

Table 4. Combined Problems

Case	IV-1		IV-2		
Agg. kernel	k_a	k_a	k_a	k_a	k_a
Growth kernel	$G_0 v$	$G_0 v$	1	1	100
B_0	—	—	10	10	10
k_a	1	1	1	1	1
N_0	1	1	—	—	—
v_0	1	1	—	—	—
G_0	0.1	0.1	—	—	—
β	1	1	—	—	—
t (sec)	3	10	—	—	—
τ	—	—	1	1	1
Grid type	Exp.	Exp.	Exp.	Exp.	Exp.
Grid exponent	1.05	1.05	1.08	1.08	1.08
No. of elements	120	120	100	100	100
Size range	0.005–300	0.005–300	0.001–25	0.001–25	0.001–25
Error in M_0, %	—	—	0.021	0.023	0.029
CPU (s)	5	7	2	2	2

Direct comparison with CPU times reported for other methods is not straightforward because they refer to different computers, but an evaluation can be attempted based on how each method works. The use of linear elements makes the number of computations and interpolations substantially fewer than those required for higher-order elements (for most kernels just one interpolation is enough). Thus, the method is expected to be much faster than higher-order finite element collocation methods, such as cubic spline or orthogonal collocation, while integral finite-element schemes such as the Galerkin require many more computations. Higher-order methods may be more accurate, but the CPU time saved due to the first-order elements can be spent on refining the grid. DPBs, especially those based on a geometric series (such as the Hounslow method) are likely to be even faster, because their way of evaluating the integral terms requires fewer computations. However, the important conclusion is that the CPU demands raised by this method are affordable even with a desktop PC.

Another notable feature is that the ease and speed with which the dynamic method converges to steady-state solutions with ease and speed. Nicmanis and Hounslow (1998), who studied the steady-state PBE with finite elements, reported numerical problems, such as matrix ill-conditioning in growth problems, that could only be resolved by resorting to Galerkin methods, as well as difficulties in inverting the dense matrices resulting from the discretization. On the other hand, the dynamic method is guided by the solution at the previous time step and is, thus, much easier to converge. This is a commonly encountered situation in numerical analysis: a steady-state problem is easier to solve by considering the dynamic case.

When comparing any finite-element scheme with DPBs, it must be borne in mind that the mechanisms of aggregation and breakage are essentially discrete, and, therefore, DPBs are a more natural choice for them. Moreover, being zero

order, DPBs can accommodate more easily certain situations that are ill-conditioned for higher-order methods (such as monodisperse initial conditions). The value of the finite-element method is better realized when considering combined problems that involve both discrete and continuous mechanisms, in which case the generality of the method allows it to tackle a variety of different problems without being tailored to the needs of individual ones. The scheme proposed here features this generality while retaining some of the advantages of DPBs (stability and ability to deal with ill-conditioned problems) due to its low order.

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