New Numerical Method for Solving the Dynamic Population Balance Equations

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A new numerical scheme is proposed for solving general dynamic population balance equations (PBE). The PBE considered can simultaneously include the kinetic processes of nucleation, growth, aggregation and breakage. Using the features of population balance, this method converts the PBE into a succession of algebraic equations which can be solved easily and accurately. The new method is free from stability and dispersion problems of general numerical techniques. Some benchmark problems with analytic solutions were tested. In all cases tested this method gave accurate results with very few computational requirements. For nucleation and size-independent growth without aggregation and breakage, the numerical method gives exactly the same result as analytic solution. © 2005 American Institute of Chemical Engineers *AIChE J,* 51: 3000–3006, 2005

Introduction

The population balance equation (PBE) has been used to model a variety of particulate systems. Analytical solutions to the PBE can only be obtained for very few special cases. Numerical solution of the PBE remains a considerable challenge due to aggregation and breakage terms. Most of the existing numerical methods are tailored to handle specific applications and lack generality (Raphael et al., 1995).

The method of weighted residuals with global functions is one of the most popular methods to solve the PBE (Ramkrishna, 1985). In the method of weighted residuals, the solution is approximated by a linear combination of a series of chosen basis functions, whose unknown coefficients are determined by satisfying the PBE to define a residual. The idea of weighted residuals is to find the coefficients that force the residuals to be orthogonal to a chosen set of weighting functions. The method of moments is equivalent to the method of weighted residuals if the weighting functions are chosen to be polynomials. Since polynomials weights are often a poor choice for population balances on semi-infinite intervals, and not all PBE formulations can be reduced to moment equations, the method of moments is not recommended. A limitation of global functions is that they cannot always capture the features of the solution, especially when there are sharp changes and discontinuities in the solution. Discontinuities arise in crystallizer systems with product classification or fines destruction. Finite-element methods approximate the solutions with local functions which can be tailored to handle discontinuities and sharp changes.

Gelbard and Seinfeld (1978) considered orthogonal collocation and spline collocation on finite elements to solve population balance equations with nucleation, growth and aggregation. The semi-infinite particle size domain is truncated at some large value, and then the finite domain is divided into elements. Lower order polynomials are used to approximate the solution on each elements. Nicmanis and Hounslow (1998) solved the steady-state PBE with aggregation, breakage, nucleation and growth using collocation and Galerkin methods. The method is based on an error estimate of the second moment. Raphael et al. (1995) used orthogonal collocation to solve the problem of isoelectric precipitation of sunflower protein. Bennett and Ro-

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hani (2001) solved a PBE by combining Lax-Wendroff and Crank-Nicholson methods. Rigopoulos and Jones (2003) used linear elements to represent the solution of dynamic PBE. The method is expected to be faster than higher-order finite element collocation methods, but it is less accurate.

An alternative numerical technique for solving PBE is discretization method, which divides the particles into discrete but contiguous size ranges (Baterham et al., 1981; Hounslow et al., 1988; Kostoglou and Karabelas, 1994). This method requires a uniform particle distribution, which is unrealistic for processes such as crystallization or coagulation of aerosols where agglomeration exists. Kumar and Ramkrishna (1997) extended this method for solving PBE for breakage and aggregation of particles. The method combines the features of discretization technique with the method of characteristics. Detailed reviews of previous work on solving PBEs have been made by Kostoglou and Karabelas (1994), Vanni (2000), and Lee (2001).

Most of the earlier numerical methods are difficult to implement, computationally demanding, or lacking in accuracy. They usually are tailored to solve individual problems. Their predictions of moments are subject to errors. The objective of this paper is to present a new technique for solving dynamic PBE for nucleation, growth, breakage and aggregation, all processes occurring simultaneously. The approach makes use of the properties of population balance. The new method is free from problems due to stability and dispersion of the numerical solutions. The proposed approach has been tested on a number of benchmark problems. Comparison between the analytic solutions and the numerical solutions indicates the distributions can be accurately predicted by the proposed method. In addition, the proposed method is easy to implement with few computational requirements.

Population Balances

In this work, we consider the general PBE of the form (Randolph and Larson, 1988)

$$\frac{\partial n(v,t)}{\partial t} + \frac{\partial (G(v)n(v,t))}{\partial v} = B_{nuc}(v) + B_{agg}(v) - D_{agg}(v) + B_{br}(v) - D_{br}(v) \quad (1)$$

where $n(v, t)$ is the population density of particles of volume v and time t, $G(v)$ is growth rate for particles of volume v, $B_{nuc}(v)$ is the nucleation rate of particles of volume v, $B_{agg}(v)$ and $D_{agg}(v)$ are the birth and death rates of particles of volume v due to aggregation, and B_{br} and D_{br} are the birth and death rates of particles of volume v due to breakage. $B_{agg}(v)$ and $D_{agg}(v)$ can be written as (Hulburt and Katz, 1964)

$$B_{agg}(v) = \frac{1}{2}\int_0^v \beta(v-v', v')n(v-v't)n(v', t)dv' \quad (2)$$

$$D_{agg}(v) = n(v,t)\int_0^\infty \beta(v, v')n(v', t)dv' \quad (3)$$

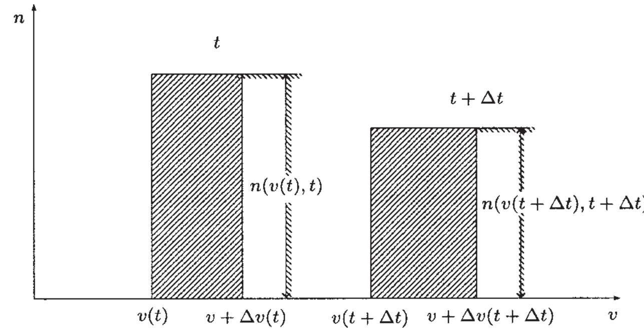

Figure 1. Population balance.

where $\beta(v, v')$ is the aggregation kernel. The birth and death rates of particles due to breakage can be written as (Prasher, 1987)

$$B_{br}(v) = \int_v^\infty \rho(v, v')S(v')n(v', t)dv' \quad (4)$$

$$D_{br}(v) = S(v)n(v, t) \quad (5)$$

where $\rho(v, v')$ is the breakage function and $S(v)$ is the rate of breakage for particles of volume v. Equation 1 is a first order hyperbolic differential equation for which very few analytical solutions have been found.

Formulation of the Algorithm

Approximation of solution

In the absence of aggregation and breakage, a representation of population balance is shown in Figure 1, where population balance distributions at time t and $t + \Delta t$ are demonstrated. The particles grow into the volume range $[v(t + \Delta t), v + \Delta v(t + \Delta t)]$ from volume range $[v(t), v + \Delta v(t)]$ over the time interval Δt. $n(v(t), t)$ and $n(v(t + \Delta t), t + \Delta t)$ represent the population density at time t and $t + \Delta t$, respectively. The population balance implies (Hu et al., 2004)

$$n(v(t), t)\Delta v(t) = n(v(t + \Delta t), t + \Delta t)\Delta v(t + \Delta t) \quad (6)$$

However, in the presence of nucleation, aggregation and breakage, the Eq. 6 is not satisfied anymore. In this case, we assume that

$$n(v(t), t)\Delta v(t) = n(v(t + \Delta t), t + \Delta t)\Delta v(t + \Delta t) - \alpha(v(t))\Delta v(t)\Delta t \quad (7)$$

The last term in Eq. 7 is included due to nucleation, aggregation and breakage, and $\alpha(v(t))$ is a parameter to be determined.

On the basis of the definition of growth rate, we have

$$v(t + \Delta t) \approx v(t) + G(v(t))\Delta t \quad (8)$$

which implies

$$\Delta v(t+\Delta t) \approx \left(1 + \frac{\partial G(v)}{\partial v}\bigg|_{v=v(t)} \Delta t\right)\Delta v(t) \quad (9)$$

Using the Taylor series, we have

$$n(v(t+\Delta t), t+\Delta t) \approx n(v(t), t+\Delta t) + \frac{\partial n(v, t+\Delta t)}{\partial v}\bigg|_{v=v(t)} (v(t+\Delta t) - v(t)) \quad (10)$$

With Eq. 8 and Eq. 10 can be written as

$$n(v(t+\Delta t), t+\Delta t) \approx n(v(t), t+\Delta t) + \frac{\partial n(v, t+\Delta t)}{\partial v}\bigg|_{v=v(t)} G(v(t))\Delta t \quad (11)$$

Substituting Eq. 9 and Eq. 11 into Eq. 7 yields

$$n(v(t), t) \approx n(v(t), t+\Delta t) + \frac{\partial n(v, t+\Delta t)}{\partial v}\bigg|_{v=v(t)} G(v(t))\Delta t + n(v(t), t+\Delta t)\frac{\partial G(v)}{\partial v}\bigg|_{v=v(t)} \Delta t + \frac{\partial n(v(t), t+\Delta t)}{\partial v}\bigg|_{v=v(t)} \frac{\partial G(v)}{\partial v}\bigg|_{v=v(t)} G(v(t))(\Delta t)^2 - \alpha(v(t))\Delta t \quad (12)$$

which can be written as

$$\frac{n(v(t), t+\Delta t) - n(v(t), t)}{\Delta t} \approx -G(v(t))\frac{\partial n(v(t), t+\Delta t)}{\partial v}\bigg|_{v=v(t)} - n(v(t), t+\Delta t)\frac{\partial G(v)}{\partial v}\bigg|_{v=v(t)} + \alpha(v(t)) - \frac{\partial n(v, t+\Delta t)}{\partial v}\bigg|_{v=v(t)} \frac{\partial G(v)}{\partial v}\bigg|_{v=v(t)} G(v(t))\Delta t \quad (13)$$

If $\Delta t \to 0$, Eq. 13 becomes

$$\frac{\partial n(v, t)}{\partial t} + G(v)\frac{\partial n(v, t)}{\partial v} + n(v, t)\frac{\partial G(v)}{\partial v} = \alpha(v) \quad (14)$$

which is equivalent to Eq. 1 if

$$\alpha(v) = B_{nuc}(v) + B_{agg}(v) - D_{agg}(v) + B_{br}(v) - D_{br}(v) \quad (15)$$

therefore, we can use Eq. 7 to solve Eq. 1. Using Eq. 8 and Eq. 9, Eq. 7 can be changed to

$$n(v(t+\Delta t), t+\Delta t) \approx \frac{n(v(t), t) + \alpha(v(t))\Delta t}{1 + \frac{\partial G(v)}{\partial v}\Big|_{v=v(t)} \Delta t} \quad (16)$$

Thus, given the population density at time t, the population density at time $t + \Delta t$ can be solved by Eq. 16.

For numerically solving the PBE (Eq. 1), the infinite domain of the particle volume v must be truncated to a finite upper limit. The error in the ith moment of the solution due to domain truncation is

$$M_i^e = \int_{v_{\max}}^{\infty} v^i n(v)dv \quad (17)$$

where $v_{\max}$ is the upper limit of the finite domain. In practice, the population density $n(v)$ tends to zero at sufficiently large particle volumes, so $v_{\max}$ can be chosen to be sufficiently large such that M_i^e is negligibly small.

In this method, the truncated domain $v \in [0, v_{\max}]$ is partitioned into N discrete and contiguous elements, that is, we subdivide it into subintervals with common endpoints, called nodes. The time step is set to Δt. We use $(v_{j,i}, n_{j,i})$ to represent a node on the particle distribution profile on the vn-coordinate plane. The index j denotes the time $j\Delta t$, and the index $i = 0, 1, \ldots, N$ indicates the series number of the nodes on the particle distribution profile. Note that only the initial truncated domain (at $t = 0$) needs to be partitioned, and the nodes on the vn-coordinate plane at other time can be approximated successively using Eq. 8 and Eq. 16 which imply

$$v_{j+1,i} \approx v_{j,i} + G(v_{j,i})\Delta t \quad (18)$$

$$n_{j+1,i} \approx \frac{n_{j,i} + \alpha(v_{j,i})\Delta t}{1 + \frac{\partial G(v)}{\partial v}\Big|_{v=v_{j,i}} \Delta t} \quad (19)$$

for $i = 0, 1, \ldots, N$ and j is a nonnegative integer.

Approximation of birth and death terms

In Eq. 19, the term $\alpha(v_{j,i})$ still needs to be solved. We use cubic spline interpolation to find additional nodes we may need in this section, while $(n_{j,1} - n_{j,0})/(v_{j,1} - v_{j,0})$, $(n_{j,N} - n_{j,N-1})/(v_{j,N} - v_{j,N-1})$ are used as the end slopes for the cubic spline.

Gauss-Legendre quadrature is used to perform integrations over each element $[v_{j,i}, v_{j,i+1}]$ (Hildebrand, 1956). An approximation to the integral

$$\int_{-1}^{1} f(x)dx \approx \sum_{i=1}^{N} w_{N,i} f(z_{N,i}) \quad (20)$$

is obtained by sampling $f(x)$ at the N unequally spaced abscissas $z_{N,1}, z_{N,2}, \ldots, z_{N,N}$, where the corresponding weights are $w_{N,1}, w_{N,2}, \ldots, w_{N,N}$. The abscissas and weights for Gauss-Legendre quadrature can be computed analytically for small N. For $N = 5$

$$\int_{-1}^{1} f(x)dx \approx w_1 f(z_1) + w_2 f(z_2) + w_3 f(z_3) + w_4 f(z_4) + w_5 f(z_5) \quad (21)$$

where

$$z_1 = -\frac{1}{21}\sqrt{245 + 14\sqrt{70}}, \quad w_1 = \frac{1}{900}(322 - 13\sqrt{70})$$

$$z_2 = -\frac{1}{21}\sqrt{245 - 14\sqrt{70}}, \quad w_2 = \frac{1}{900}(322 + 13\sqrt{70})$$

$$z_3 = 0, \quad w_3 = \frac{128}{225}$$

$$z_4 = \frac{1}{21}\sqrt{245 - 14\sqrt{70}}, \quad w_4 = \frac{1}{900}(322 + 13\sqrt{70})$$

$$z_5 = \frac{1}{21}\sqrt{245 + 14\sqrt{70}}, \quad w_5 = \frac{1}{900}(322 - 13\sqrt{70})$$

For ease of reading the earlier list the notation z_i and w_i instead of $z_{N,i}$ and $w_{N,i}$, has been used respectively.

To apply the rule over the interval $[a, b]$, use the change of variable

$$t = \frac{a+b}{2} + \frac{b-a}{2}x \quad \text{and} \quad dt = \frac{b-a}{2}dx$$

then the relationship $\int_a^b f(t)dt = \int_{-1}^1 f((a+b)/2 + ((b-a)/2)x)((b-a)/2)dx$ is used to obtain the quadrature formula

$$\int_a^b f(t)dt \approx \frac{b-a}{2}\sum_{i=1}^{N} w_{N,i} f\left(\frac{a+b}{2} + \frac{b-a}{2}z_{N,i}\right)$$

If there is no aggregation and breakage, the solution Eq. 19 becomes

$$n_{j+1,i} \approx \frac{n_{j,i} + B_{nuc}(v_{j,i})\Delta t}{1 + \left.\frac{\partial G(v)}{\partial v}\right|_{v=v_{j,i}} \Delta t} \quad (22)$$

then the accuracy of the solution can be increased by simply selecting a small Δt. Furthermore, if the growth rate is volume-independent, that is, $\partial G/\partial v = 0$, (Eq. 22) can be simplified to

$$n_{j+1,i} = n_{j,i} + B_{nuc}(v_{j,i})\Delta t \quad (23)$$

which indicates that accurate solution can be obtained. This is a very useful property for some batch crystallization processes.

Case Studies

The method was tested for six cases of the PBE, where analytical solutions are available. The numerical results were compared with analytic solutions. The CPU time reported corresponds to a desktop computer with 1.6 GHz Intel Pentium 4 processor.

Aggregation and size-dependent growth

By letting the nucleation rate $B_{nuc}(v)$, breakage function $\rho(v, v')$, and the rate of breakage $S(v)$ to be zero, the PBE for aggregation and size-dependent growth is

$$\frac{\partial n(v,t)}{\partial t} + \frac{\partial (G(v)n(v,t))}{\partial v} \approx \frac{1}{2}\int_0^v \beta(v-v', v')n(v - v't)n(v', t)dv' - n(v,t)\int_0^\infty \beta(v, v')n(v', t)dv' \quad (24)$$

The initial distribution is

$$n(v, 0) = \frac{N_0}{v_0}\exp\left(-\frac{v}{v_0}\right) \quad (25)$$

and size-dependent growth rate is

$$G(v) = \sigma_1 v \quad (26)$$

Case A1. For the size-independent aggregation kernel

$$\beta(v, v') = \beta_0 \quad (27)$$

the analytical solution has been derived by Ramabhadran et al. (1976)

$$n(v,t) = \frac{M_0^2}{M_1}\exp\left(-\frac{M_0}{M_1}v\right) \quad (28)$$

where $M_0(t)$ and $M_1(t)$ are the first two moments

$$M_0(t) = \int_0^\infty n(v,t)dv = \frac{2N_0}{2 + \beta_0 N_0 t} \quad (29)$$

$$M_1(t) = \int_0^\infty v n(v,t)dv = N_0 v_0 \exp(\sigma_1 t) \quad (30)$$

The constant N_0, v_0, σ_1 and β_0 were set to 1. The truncation point (v_{max}) was chosen as 10, and the initial truncated domain was evenly partitioned into 20 discrete elements with common ends. The time step Δt was chosen to be 0.02.

The problem (Eq. 24) was solved using the proposed method. The particle density distributions for the initial time, $t = 0.5$, $t = 1$ and $t = 2$ are shown in Figure 2. The solid lines represent the analytical solution, while the symbols are the

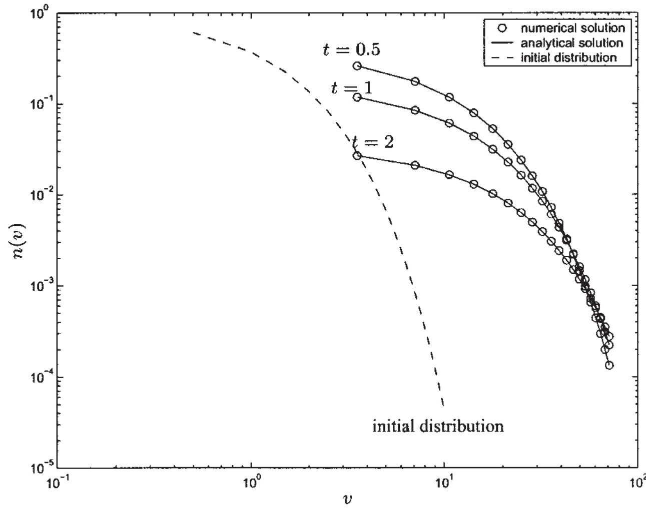

Figure 2. Aggregation with constant kernel $\beta(v, v') = 1$, and size-dependent growth rate $G(v) = v$.

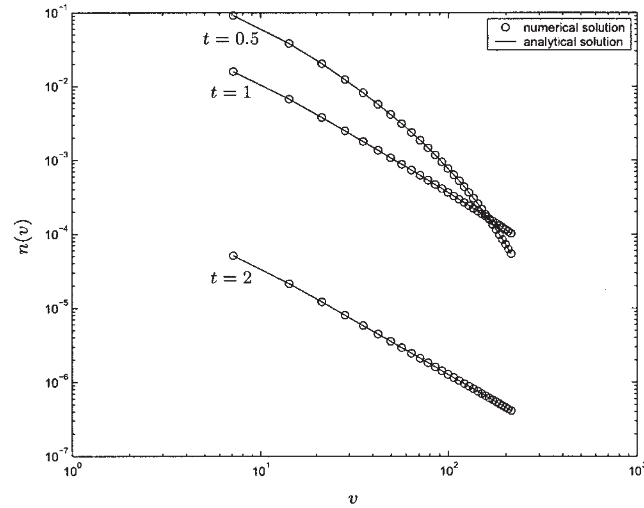

Figure 3. Aggregation with sum kernel $\beta(v, v') = v + v'$, and size-dependent growth rate $G(v) = v$.

nodal values obtained by the proposed method. The numerical and analytical solutions are in excellent agreement.

Case A2. For the size-dependent aggregation kernel

$$\beta(v, v') = \beta_1(v + v') \tag{31}$$

The analytical solution is (Ramabhadran et al., 1976)

$$n(v, t) = \frac{\dfrac{M_0^2}{M_1}\exp\left[-\dfrac{M_0}{M_1}\left(\dfrac{2N_0}{M_0} - 1\right)v\right]I_1\left(2\sqrt{1 - \dfrac{M_0}{N_0}}\dfrac{N_0 v}{M_1}\right)}{\dfrac{M_0 v}{M_1}\sqrt{1 - \dfrac{M_0}{N_0}}} \tag{32}$$

where $I_1(\cdot)$ is the modified Bessel function of the first kind of order one, and

$$M_0(t) = N_0\exp\left[\frac{\beta_1 N_0 v_0}{\sigma_1}(1 - \exp(\sigma_1 t))\right] \tag{33}$$

$$M_1(t) = N_0 v_0 \exp(\sigma_1 t) \tag{34}$$

The constant β_1 was set to 1. The truncation point $v_{\max}$ was selected to be 30. The domain $[0, v_{\max}]$ was evenly partitioned into 30 elements. The time interval was 0.02. Comparison of simulation with the analytical solution is shown in Figure 3. The solids lines represent the analytical solution, and the symbols are the numerical results. Again, the numerical solution overlaps the analytical solution excellently.

Case A3. If the initial distribution is

$$n(v, 0) = \frac{N_0 v}{v_0^2}\exp\left(-\frac{v}{v_0}\right) \tag{35}$$

and $\beta(v, v')$ and $G(v)$ are the same as in Case A1, then the analytical solution is (Ramabhadran et al., 1976)

$$n(v, t) = \frac{M_0^2}{M_1}\frac{1}{\sqrt{1 - M_0/N_0}}\exp\left(-\frac{N_0 v}{M_1}\right)\sinh\left(\sqrt{1 - \frac{M_0}{N_0}}\frac{N_0 v}{M_1}\right) \tag{36}$$

where M_0 and M_1 are defined in Eq. 29 and Eq. 30, respectively. All constants and truncation are the same as in Case A1. Figure 4 compares the numerical solution with the analytical solution. The solids lines represent the analytical solution, while the symbols are the nodal values. The agreement is quite good.

Errors in the first moment of the numerical solution and CPU time are recorded in Table 1. The accuracy of the numerical solutions is satisfactory, as shown in Table 1, despite the use of a fairly coarse grid.

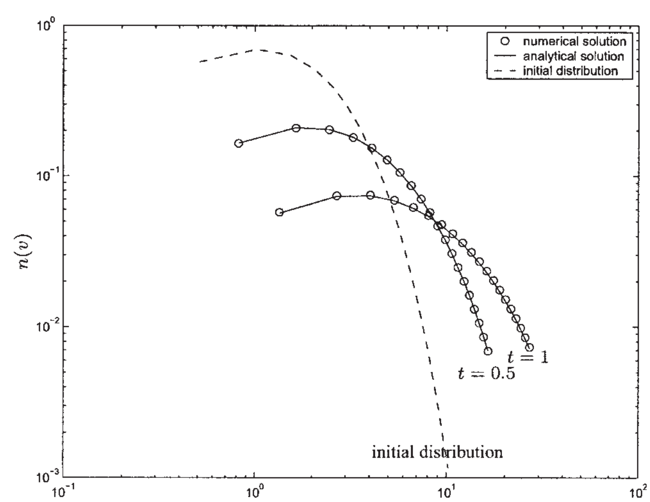

Figure 4. Aggregation with constant kernel $\beta(v, v') = 1$, size-dependent growth rate $G(v) = v$, and initial distribution (Eq. 35).

Table 1. Aggregation and Size-Dependent Growth Rate

Case	Time	Δt	No. of Elements	Error in M_1%	CPU (s)
A1	1	0.02	20	0.15	34
A2	1	0.02	30	0.46	76
A3	1	0.02	20	0.28	34

Breakage

The PBE for pure breakage is obtained by setting the growth rate $G(v)$, the nucleation rate $B_{nuc}(v)$, and the aggregation kernel $\beta(v, v')$ to zero, that is

$$\frac{\partial n(v, t)}{\partial t} = -S(v)n(v, t) + \int_v^\infty \rho(v, v')S(v')n(v', t)dv' \quad (37)$$

Case B1. If the probability that a particle breaks up into two pieces is independent of both the size of the object and of the pieces, then

$$\rho(v, v') = \frac{2}{v'} \quad (38)$$

$$S(v) = v \quad (39)$$

For the initial distribution

$$n(v, 0) = \exp(-v) \quad (40)$$

the analytical solution is (Ziff and McGrady, 1985)

$$n(v, t) = (1 + t)^2\exp[-v(1 + t)] \quad (41)$$

Figure 5 shows the initial distribution and the distributions at $t = 0.5$, and $t = 1$ for this case. The solid line represents the analytical solutions, and the symbols represent the numerical solutions. The numerical solutions match the analytical solutions very well.

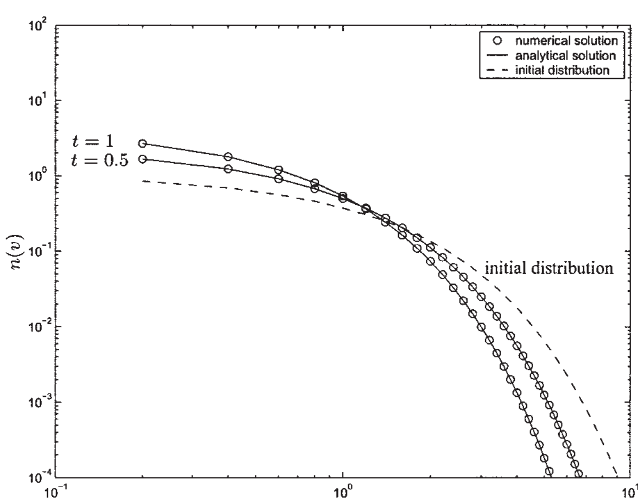

Figure 5. Breakage with a binary breakage function $\beta(v, v') = 2/v'$, and size-dependent breakage rate $S(v) = v$.

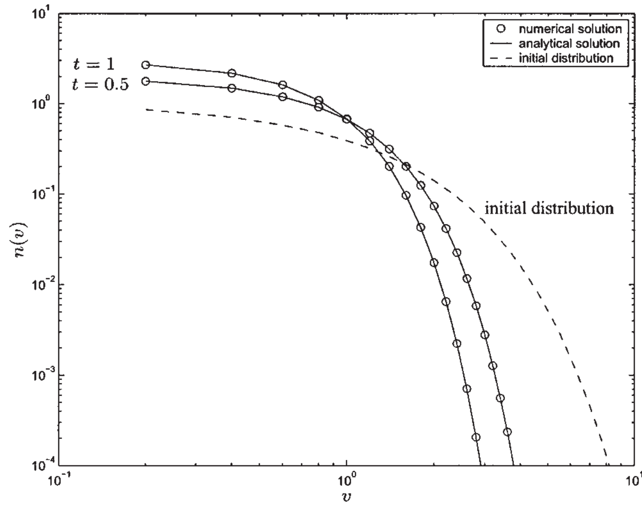

Figure 6. Breakage with breakage rate proportional to the particle volume $S(v) = v^2$.

Case B2. In this case, the rate of breakage is proportional to the size, and

$$\rho(v, v') = \frac{2}{v'} \quad (42)$$

$$S(v) = v^2 \quad (43)$$

For the exponential initial distribution (Eq. 40), the analytical solution is (Ziff and McGrady, 1985)

$$n(v, t) = \exp(-tv^2 - v)[1 + 2t(1 + v)] \quad (44)$$

Distributions at different moments for this case are shown in Figure 6. The numerical solutions are in good agreement with the analytical solutions.

Parameters used for numerical solution, errors in the first moment and CPU time were summarized in Table 2.

Nucleation and growth

For simultaneous nucleation and size independent growth the PBE is

$$\frac{\partial n(v, t)}{\partial t} + G\frac{\partial n(v, t)}{\partial v} = B_0\delta(v) \quad (45)$$

Table 2. Breakage

Case	Time	Δt	No. of Elements	Error in M_1%	CPU (s)
B1	1	0.02	50	0.50	80
B2	1	0.02	50	0.55	80

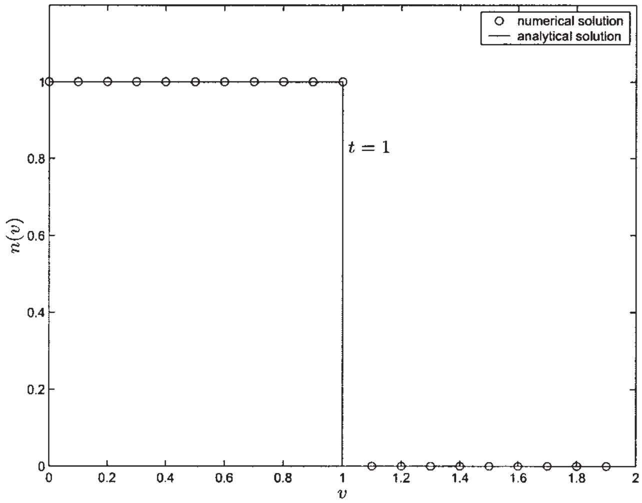

Figure 7. Nucleation and growth.

where B_0 and G are constant. The analytical solution for this model is (Hounslow et al., 1988)

$$n(v, t) = \frac{B_0}{G} u\left(t - \frac{v}{G}\right) \tag{46}$$

where u is the unit step function. With $G = 1$ and $B_0 = 1$, comparison between the numerical solution with the analytical solution is shown in Figure 7, where the solid line represents the analytical solution, and the symbols are for the numerical solution. It should be noted that the proposed method provides absolutely accurate solution in this case and is better than any other existing numerical methods. The CPU time was only 0.02 s.

Conclusions

In this work, a new numerical method for solving dynamic PBE was proposed. The new method is able to accurately predict the solution of PBE involving any combination of particulate system subprocess, that is, nucleation, growth, aggregation and breakage, with reasonable computational requirement. The proposed approach is free from stability and dispersion problems of other numerical methods such as finite-element method. The numerical scheme requires a partition of the particle-size domain, but there are no restrictions on the location of nodes when using this method. An evenly spaced partition was used in this method, although other partition methods can also be applied. Accuracy of the numerical solution can be simply improved by smaller time step and more partition elements of particle size domain without any stability problems, which makes this method particularly useful for developing hierarchical solution for particulate processes. A compromise between computational time and accuracy can be easily achieved by choosing the node resolution.

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