AIChE Wiley InterScience DISCOVER SOMETHING GREAT

An Improved Discretized Tracer Mass Distribution of Hounslow et al.

M. Peglow, J. Kumar, G. Warnecke, S. Heinrich, E. Tsotsas, and L. Mörl
Institute for Processing Engineering, Institute for Analysis and Numerics, Institute of Process Equipment and Environmental Technology, Otto-von-Guericke-University Magdeburg, Germany

M. J. Hounslow
Particle Products Group, Dept. of Chemical and Process Engineering, The University of Sheffield, Sheffield, U.K.

DOI 10.1002/aic.10748
Published online December 29, 2005 in Wiley InterScience (www.interscience.wiley.com).

The discretized tracer population balance equations (DTPBE) for aggregation problems is discussed and modified. It is shown that the original version was not entirely consistent with the associated discretized population balance equation (DPBE). These inconsistencies are remedied in a new formulation that retains the advantages of the original DTPBE, such as conservation of total tracer mass, prediction of tracer-weighted mean particle volume, and so on. Furthermore, the DTBE has been extended to an adjustable discretization. Numerous comparisons are made of the validity of the extended and modified formulation. © 2005 American Institute of Chemical Engineers *AIChE J,* 52: 1326–1332, 2006
Keywords: aggregation, discretization, population balance

Introduction

In order to describe the temporal change of a particle-size distribution (PSD), one-dimensional (1-D) population balances for growth, agglomeration and breakage of particles are frequently used to describe processes, such as crystallization, granulation and agglomeration. Usually, the only significant property considered for the dispersed phase is particle size. In many applications, however, besides knowledge of the particle size, other properties of populations are of interest. In a fluidized-bed granulation, for example, in addition to the temporal change of granule size, the amount of water or binder within a granule, the enthalpy of a granule and number of primary particles within a granule are also important. From the numerical point of view, the complete two (or higher) dimensional formulation is quite difficult to solve. To decrease the computational efforts Hounslow et al.[1] reduced a 2-D population balance equation to two 1-D population balance equations. They proposed a discretized population balance for both 1-D equations, and calculated the temporal change of PSD and tracer-mass distribution (TMD) simultaneously.

Frequently, the state of a dispersed phase is described in terms of extensive properties. Extensive properties, such as mass, mole number or enthalpy, are defined as the properties which depend directly on the size of a thermodynamic system. If such a system is divided into several parts, the extensive property changes according to the size of these parts. It is often useful to give an extensive property in a form which is not directly proportional to the size of system, in other words, to convert an extensive property into an intensive property. For certain applications it might be necessary to determine intensive properties of the disperse phase, such as density, concentration, or temperature. For example, to model the mass transfer between disperse and gas phase, the temperature of the disperse phase is necessary to calculate the equilibrium state at the phase boundary. In general an intensive property is defined as the quotient of an extensive property with respect to another extensive property.

Correspondence concerning this article should be addressed to M. Peglow at mirko.peglow@vst.uni-magdeburg.de.

Problems with the existing formulation

One can use the DTPBE of Hounslow et al.[1] for the computation of various extensive properties (amount of water within the particle, enthalpy of particles, and so on) of aggregating systems. The equations predict the total amount of extensive properties exactly but it fails to predict intensive properties which are proportional to the ratio of extensive properties and mass of granules. For the purpose of illustration, let us consider an example where a particle system is described by the two properties: volume of particles and amount of water within the particles. Our interest is to calculate the particle moisture content as the ratio of water mass to solid particle mass — an intensive property. We assume that the solid density is constant and solid particle volume is equal to the solid particle mass. For simplicity, let us assume that initially the water mass inside a particle is equal to the solid particle mass. In other words, the ratio between water mass and solid particle mass or equivalently particle moisture content is constant over the particle size range and is equal to one. Since agglomeration is the only governing mechanism which changes the particle size, the particle moisture content should be constant throughout the process.

Let us consider the following initial condition for the PSD with volume as the distributed property

$$n(v) = \frac{N_0}{v_0} \exp\left(-\frac{v}{v_0}\right) \tag{1}$$

In the DTPBE (Hounslow et al.[1]), the particle size domain is divided into discrete size ranges using a geometric discretization of the type

$$\frac{v_{i+1}}{v_i} = 2 \tag{2}$$

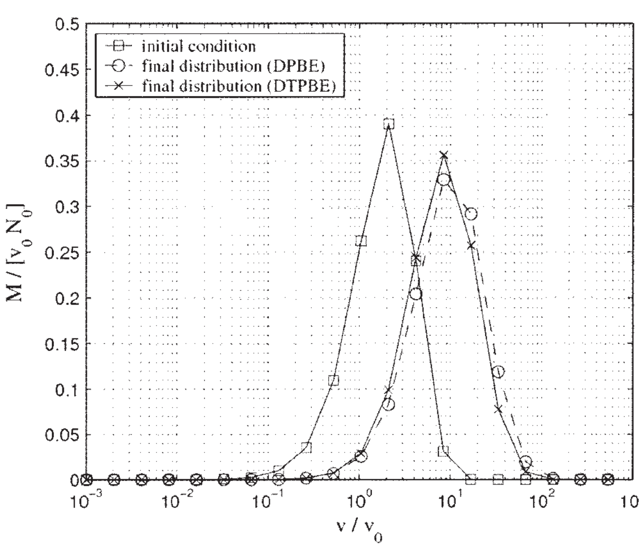

Figure 1. Initial and final distribution for size independent aggregation of a charge given by Eq. 1.

Results for particle mass calculated using the DBPE and for water mass calculated using the DTPBE of Hounslow et al. (2001): $I_{agg} = 4/5$.

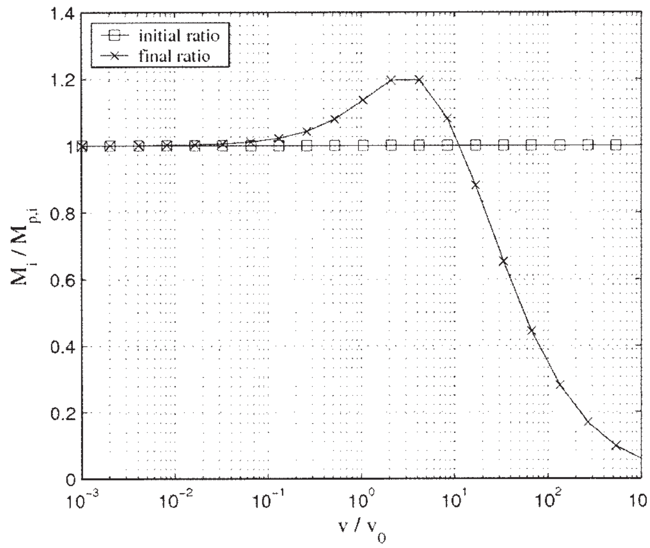

Figure 2. Initial and final ratio of particle mass and water mass distribution for the results of Figure 1.

Integration of Eq. 1 over an interval $[v_i, v_{i+1}]$ gives the total number of particles within the interval

$$N_i = \frac{N_0}{2}\left[\exp\left(-\frac{v_i}{v_0}\right) - \exp\left(-\frac{v_{i+1}}{v_0}\right)\right] \tag{3}$$

The mass of particles in an interval $[v_i, v_{i+1}]$ is approximated as

$$M_{P,i} = N_i\left(\frac{v_i + v_{i+1}}{2}\right) \tag{4}$$

Here we make an assumption of constant solid density, so that the solid mass could be replaced by solid volume. The initial condition for water mass distribution is chosen in such a way that the total mass of water $M_{w,i}$ within the particles in the interval $[v_i, v_{i+1}]$ is equal to the total solid mass of particles in this interval

$$M_{w,i} = M_{P,i} \tag{5}$$

The computation is made for a size independent kernel. We calculated the particle mass and the water content within the particles as a function of particle volume using Hounslow's discrete population balance equation (Hounslow et al.[2]) and Hounslow's DTPBE (Hounslow et al.[1]), respectively. The numerical results at $I_{agg} = 0.8$ together with initial condition have been plotted in Figure 1. Clearly the DTPBE fails to predict the water distribution within the particles correctly since both water distribution and mass distribution must be the same during the process. Furthermore, Figure 2 includes initial and final ratio of water mass and particle mass within the intervals. It can be seen from the figure that the final ratio is not constant. However, it should be pointed out that both discretized formulations conserve mass.

Modifications

Let us discuss this ratio problem in detail. Consider that N_jN_k is the birth contribution in the *i*th interval due to collision of particles between *j*th and *k*th intervals. In DTPBE this birth contribution is replaced by $N_jM_k + M_jN_k$ as a tracer mass-birth contribution, which is the actual amount of tracer coming from *j*th and *k*th intervals. The cause for the nonconstant ratio is that the assignment of the tracer mass is different from that of granule mass in the *i*th interval. Let us observe the actual amount of granule mass corresponding toN_jN_k birth in the *i*th interval. If $\bar{v}$ represents the mean volume (or mass) of an interval then $N_jN_k(\bar{v}_j + \bar{v}_k)$ is the total mass carried with N_jN_k particles from *j*th and *k*th intervals. On the other hand, corresponding to N_jN_k particles in *i*th interval we assign $N_jN_k\ \bar{v}_i$ mass to ith interval. Clearly

$$N_jN_k(\bar{v}_j + \bar{v}_k) \neq N_jN_k\bar{v}_i, \quad \text{for all } i, j, k \tag{6}$$

The authors did not assign the mass of tracer in the same ratio as granule mass. This inconsistency can be repaired by introducing some correction factors. For instance, a correction factor *K* in this case can be introduced in the following way

$$\frac{\text{Assigned tracer mass}}{\text{Actual tracer mass}} = \frac{\text{Assigned granule mass}}{\text{Actual granule mass}} = \frac{\bar{v}_i}{\bar{v}_j + \bar{v}_k} = K \tag{7}$$

By introducing correction factors in each term of the DTPBE, we obtain the following set of equations

$$\frac{dM_i}{dt} = \sum_{j=1}^{i-2} 2^{j-i+1}\beta_{i-1,j}(M_{i-1}N_j + N_{i-1}M_j)K_1 + N_i\sum_{j=1}^{i-1}(1-2^{j-i})\beta_{i,j}M_jK_2 + \beta_{i-1,i-1}N_{i-1}M_{i-1} - M_i\sum_{j=1}^{i-1}2^{j-i}\beta_{i,j}N_jK_3 - M_i\sum_{j=i}^{M}\beta_{i,j}N_j \tag{8}$$

with correction factors

$$K_1 = \frac{2}{2^{j-i+1}+1}, \quad K_2 = \frac{1}{2^{j-i}+1}, \quad K_3 = \frac{2}{2^{j-i}+1} \tag{9}$$

The correction factors corresponding to the third and the last terms of Eq. 8 are 1. It can be shown easily that the total tracer mass is still conserved, see Appendix A.

This modified formulation has been now applied to the same problem as before. As expected, Figure 3 shows the same prediction by both the DPBE and the modified DTPBE.

The DTPBE can easily be extended to adjustable discretization of the type $v_{i+1} = 2^{1/q}v_i$, see Litster et al.[3] and Wynn[4]

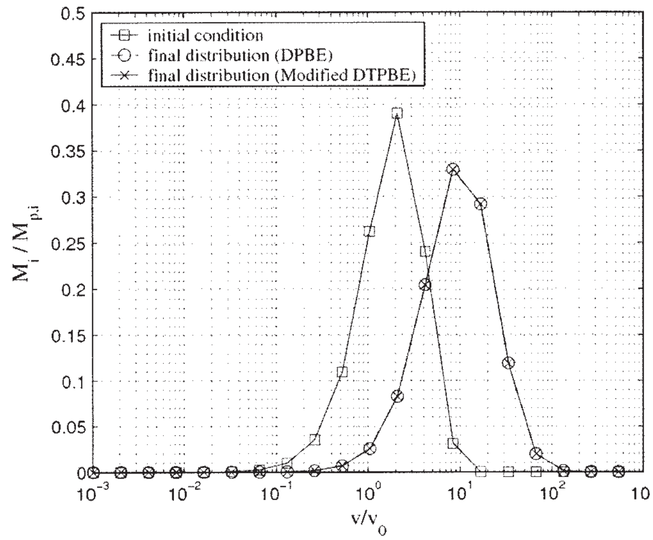

Figure 3. Initial and final distribution for size independent aggregation of a charge given by Eq. 1.

Results for particle mass calculated using the DBPE and for water mass calculated using the modified DTPBE: $I_{agg} = 4/5$.

$$\begin{aligned}\frac{dM_i}{dt} = {} & \sum_{j=1}^{i-S_1}\frac{2^{(j-i+1)/q}}{2^{1/q}-1}\beta_{i-1,j}(M_{i-1}N_j + N_{i-1}M_j)K_1 \\ & + \beta_{i-q,i-q}N_{i-q}M_{i-q} + \sum_{p=1}^{q-1}\sum_{j=i+1-S_p}^{i+1-S_{p+1}}\frac{2^{1/q} - 2^{(j-i)/q} - 2^{-p/q}}{2^{1/q}-1} \\ & \times \beta_{i-p,j}(M_{i-p}N_j + N_{i-p}M_j)K_2 + \sum_{p=2}^{q}\sum_{j=i-S_{p-1}}^{i-S_p} \\ & \times \frac{2^{(j-i+1)/q} - 1 + 2^{-(p-1)/q}}{2^{1/q}-1}\beta_{i-p,j}(M_{i-p}N_j + N_{i-p}M_j)K_3 \\ & + \sum_{j=1}^{i-S_1+1}\left(1 - \frac{2^{(j-i)/q}}{2^{1/q}-1}\right)\beta_{i,j}N_iM_jK_4 - \sum_{j=1}^{i-S_1+1}\frac{2^{(j-i)/q}}{2^{1/q}-1}\beta_{i,j}M_iN_jK_5 \\ & - \sum_{j=i-S_1+2}^{I}\beta_{i,j}M_iN_j\end{aligned} \tag{10}$$

where

$$S_p = INT\left(1 - \frac{q\ln(1-2^{-p/q})}{\ln 2}\right) \tag{11}$$

and

$$K_1 = \frac{2^{(i-j)/q}}{1 + 2^{(j-i-1)/q}} \tag{12}$$

$$K_2 = K_3 = \frac{2^{(i-j)/q}}{1 + 2^{(j-i-p)/q}} \tag{13}$$

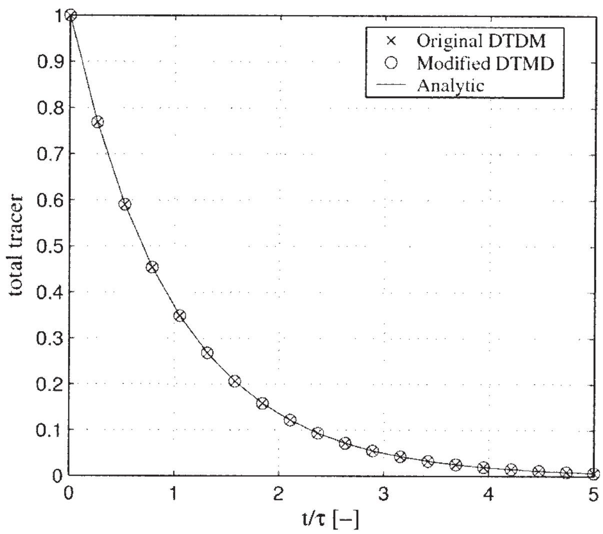

Figure 4. Progress of total tracer mass for size-independent aggregation in Ilievski and Hounslow's problem, I_{agg} = 1/4.

$$K_4 = \frac{1}{1 + 2^{(j-i)/q}} \tag{14}$$

$$K_5 = \frac{2^{1/q} - 1}{2^{(j-i)/q}} - \frac{-2^{i/q} + 2^{(2i-j)/q}(2^{1/q} - 1)}{2^{j/q} + 2^{i/q}} \tag{15}$$

The inclusion of the correction factors allows us to predict the change of the mass of tracer according to the change of mass of particles (see Eq. 7). In Hounslow's previous approach, Hounslow et al.[1], the change of number of particles within an interval was considered to determine the change of tracer. Setting all corrections factors K_1–K_5 to 1, the set of Eqs. 8 and 10 reduces exactly to that given by Hounslow et al.[1] and its

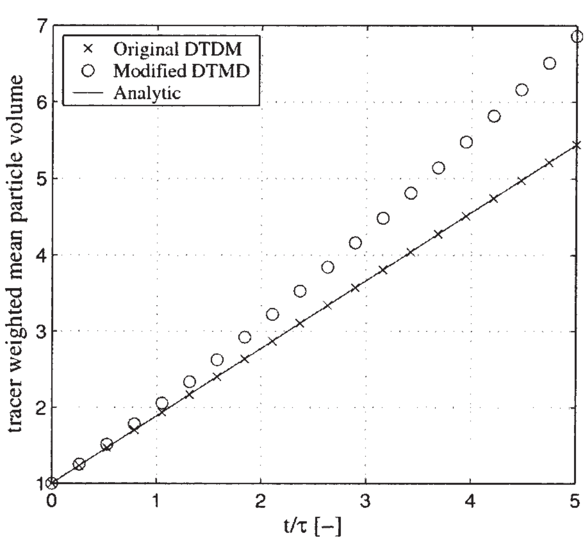

Figure 5. Progress of tracer-weighted particle volume for size-independent aggregation in Ilievski and Hounslow's problem, I_{agg} = 1/4.

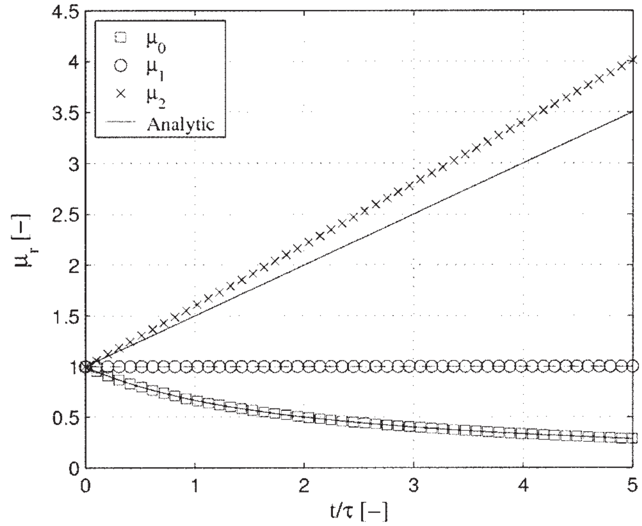

Figure 6. Progress of moments in a batch aggregation with size independent kernel, I_{agg} = 5/7.

extended version for geometric grid of the type, $v_{i+1} = 2^{1/q}v_i$, respectively.

In order to validate the extended and modified formulation we consider the same problem which was considered in Hounslow et al.[1] for a CST with size-independent aggregation. Ilievski and Hounslow[5] have also presented analytical solutions for sum and product kernels. They considered a well-mixed continuous process initially at steady state to which a spike of monodisperse tracer was added. Figure 4 compares the total tracer mass calculated using both formulations. The prediction of total tracer mass is exactly the same for both in each case. A comparison of tracer-weighted mean particle volumes, as drawn in Figure 5, illustrates that a good prediction is only possible by the original version. It should be pointed out, that

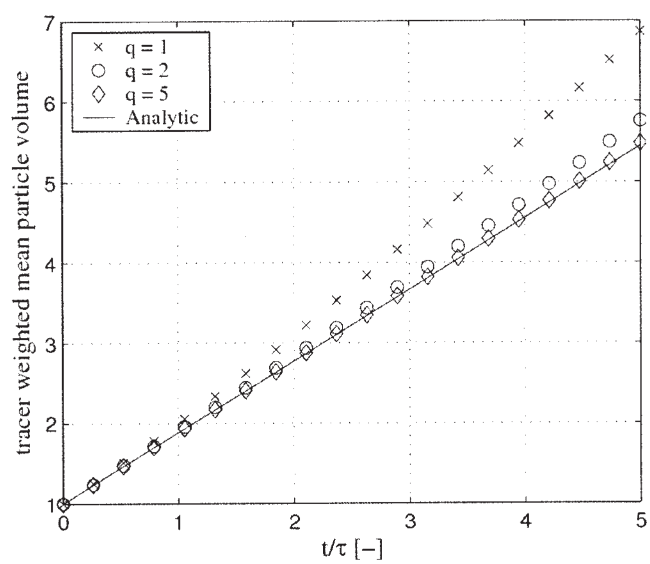

Figure 7. Progress of mean weighted tracer mass for size independent kernel and different geometric discretizations, for Ilievski and Hounslow's problem, I_{agg} = 1/4.

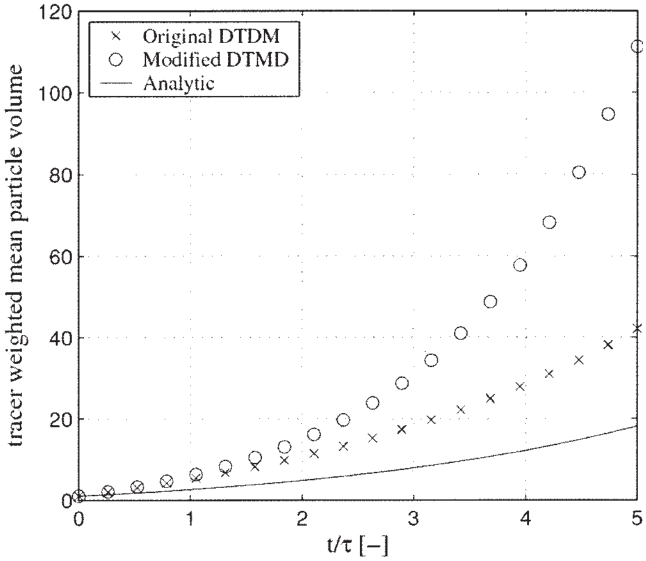

Figure 8. Progress of total tracer mass for sum kernel in Ilievski and Hounslow's problem, I_{agg} = 1/4, β = u+v.

this over prediction of tracer-weighted mean particle volume is a consequence of the assumption in Eq. 7. Since the tracer-weighted mean particle volume is similar to the second moment of PSD, and the DPBE gives an over prediction of the second moment, we have this over prediction in tracer-weighted mean particle volume. In Figure 6, the progress of the first three moments of the PSD

$$\mu_r = \int_0^\infty v^r n(v)dv \tag{16}$$

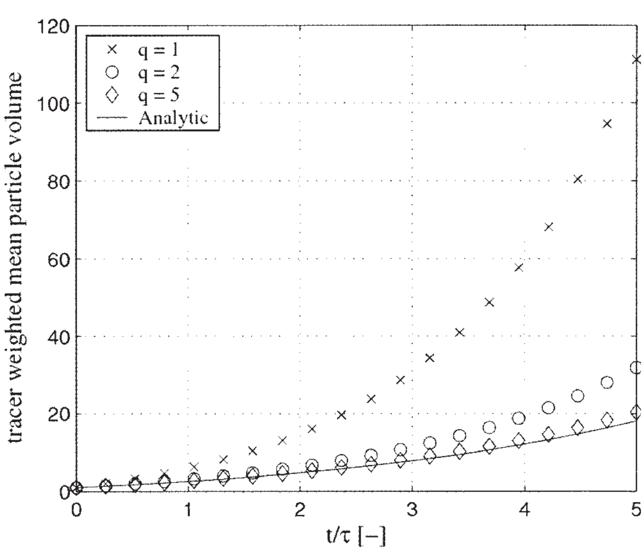

Figure 9. Progress of total tracer mass for constant kernel and different geometric discretisations for Ilievski and Hounslow's problem, I_{agg} = 1/4, β_0 = u+v.

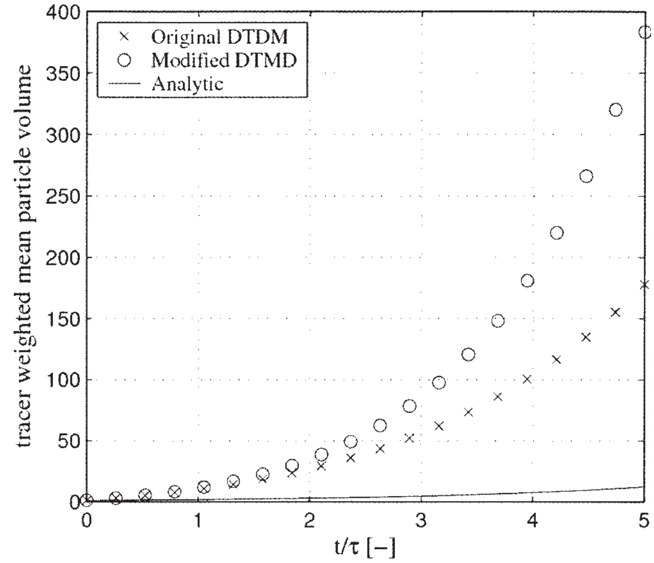

Figure 10. Progress of total tracer mass for product kernel for Ilievski and Hounslow's problem I_{agg} = 1/8, β = u v.

for a simple batch aggregation process with size independent kernel are presented. As one can see, the 2nd moment of particle number deviates from the analytical solution in the same manner as the tracer-weighted mean particle volume obtained from our new formulation of the DTPBE, Eqs. 8 and 10, respectively. For a better prediction of numerical results we have to use a finer grid. This is true for the DTPBE and for the DBPE as well. The numerical solutions obtained for different discretizations are plotted in Figure 7. However, by q = 5, the results are in good agreement with the analytical solutions.

In Figure 8 and Figure 9, a similar comparison of tracer-weighted mean particle volumes for the size-dependent sum

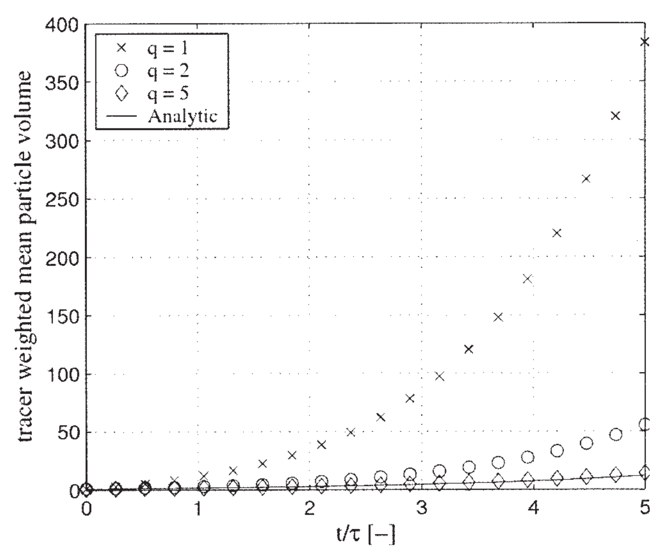

Figure 11. Progress of total tracer mass for the product kernel and different geometric discretizations in Ilievski and Hounslow's problem, I_{agg} = 1/8, β = u v.

kernel is made. The decay of total tracer mass is the same in each case, we refrain from plotting this comparison here. For this kernel both formulations deviate significantly from the analytical solutions, see Figure 8. The effectiveness of the grid for different values of q has been shown in Figure 9. Once again the numerical results are satisfactory by q = 5. A similar observation has been made for the product kernel shown in Figures 10 and 11.

Conclusions

In this work we presented a new formulation of the DTPBE of Hounslow et al.[1]. The new version retains all the advantages of the original version. It has been tested for a simple aggregation problem in a batch system. Contrary to the original version, the new version predicts a constant ratio of tracer mass and granule mass during aggregation in a batch system. It has been shown that both versions predict the same results for total tracer mass, while the original version is more accurate for tracer-weighted mean particle volume. Moreover, the DTPBE has been extended for the geometric grid of the type $v_{i+1} = 2^{1/q}v_i$, and validated by many problems where analytical solutions are available.

Acknowledgments

This work was supported by the Graduiertenkolleg-828, "Micro-Macro-Interactions in Structured Media and Particles Systems", Otto-von-Guericke-Universität Magdeburg. The authors gratefully acknowledge for funding through this PhD program.

Notation

I_{agg} = index of aggregation
K = correction factor
M = tracer mass, mass, kg/m^3
n = number density, 1/m^3.m^3
N = number of particles, 1/m^3
N_0 = initial number of particles, 1/m^3
q = adjustable parameter for grids
t = time, 1/s
v = volume, m^3
v_0 = initial average volume, m^3
$\bar{v}$ = mean volume of an interval, m^3

Greek symbols

β = agglomeration rate, 1/s
τ = dimensionless time, m/s
μ_r = r-th -moment of particle-size distribution, M^{3r}

Subscripts

i = interval
p = particle
w = water

Acronyms

$DPBE$ = A discretized population balance equation
$DTPBE$ = A discretized tracer population balance equation

Literature Cited

1. Hounslow MJ, Pearson JMK, Instone T. Tracer studies of high shear granulation: II. Population balance modeling. *AIChE J*. 2001;47:1984-1999.
2. Hounslow MJ, Ryall RL, Marshall VR. A discretized population balance for nucleation, growth and aggregation. *AIChE J*. 1988;34:1821-1831.
3. Litster JD, Smit DJ, Hounslow MJ. Adjustable discretized population balance for growth and aggregation. *AIChE J*. 1995;41:591-603.
4. Wynn EJW. Improved accuracy and convergence of discretized population balance of Lister et al. *AIChE J*. 1996;42:2084-2086.
5. Ilievski D, Hounslow MJ. Agglomeration during precipitations: II. Mechanism deduction from tracer data. *AIChE J*. 1995;41:525-535.

Appendix A

Mass conservation in modified discretized tracer mass distribution

$$\sum_i \frac{dM_i}{dt} = \sum_i \sum_{j=1}^{i-2} \frac{2^{j-i+2}}{2^{j-i+1}+1} \beta_{i-1,j}(M_{i-1}N_j + N_{i-1}M_j) + \sum_i N_i \sum_{j=1}^{i-1} (1 - 2^{j-i})\beta_{i,j}M_j \frac{1}{2^{j-i}+1} + \sum_i \beta_{i-1,i-1}N_{i-1}M_{i-1} - \sum_i M_i \sum_{j=1}^{i-1} \frac{2^{j-i+1}}{2^{j-i}+1}\beta_{i,j}N_j - \sum_i M_i \sum_{j=i} \beta_{i,j}N_j$$

expending the terms, we get

$$\sum_i \frac{dM_i}{dt} = \sum_i M_{i-1} \sum_{j=1}^{i-2} \frac{2^{j-i+2}}{2^{j-i+1}+1} \beta_{i-1,j}N_j - \sum_i M_i \sum_{j=1}^{i-1} \frac{2^{j-i+1}}{2^{j-i}+1}\beta_{i,j}N_j + \sum_i N_{i-1} \sum_{j=1}^{i-2} \frac{2^{j-i+2}}{2^{j-i+1}+1}\beta_{i-1,j}M_j - \sum_i N_i \sum_{j=1}^{i-1} \frac{2^{j-i}}{2^{j-i}+1}\beta_{i,j}M_j + \sum_i \beta_{i-1,i-1}N_{i-1}M_{i-1} + \sum_i N_i \sum_{j=1}^{i-1} \beta_{i,j}M_j \frac{1}{2^{j-i}+1} - \sum_i M_i \sum_{j=i} \beta_{i,j}N_j$$

splitting the third term into two parts, we obtain

$$\sum_i \frac{dM_i}{dt} = \sum_i M_{i-1} \sum_{j=1}^{i-2} \frac{2^{j-i+2}}{2^{j-i+1}+1} \beta_{i-1,j}N_j - \sum_i M_i \sum_{j=1}^{i-1} \frac{2^{j-i+1}}{2^{j-i}+1}\beta_{i,j}N_j + \sum_i N_{i-1} \sum_{j=1}^{i-2} \frac{2^{j-i+1}}{2^{j-i+1}+1}\beta_{i-1,j}M_j + \sum_i N_{i-1} \sum_{j=1}^{i-2} \frac{2^{j-i+1}}{2^{j-i+1}+1}\beta_{i-1,j}M_j - \sum_i N_i \sum_{j=1}^{i-1} \frac{2^{j-i}}{2^{j-i}+1}\beta_{i,j}M_j + \sum_i \beta_{i-1,i-1}N_{i-1}M_{i-1} + \sum_i N_i \sum_{j=1}^{i-1} \beta_{i,j}M_j \frac{1}{2^{j-i}+1}$$

$$
\begin{aligned}
-\sum_i M_i \sum_{j=i} \beta_{i,j}N_j = \sum_i M_{i-1} \sum_{j=1}^{i-2} \frac{2^{j-i+2}}{2^{j-i+1}+1}\beta_{i-1,j}N_j - \sum_i M_i \sum_{j=1}^{i-1} \frac{2^{j-i+1}}{2^{j-i}+1}\beta_{i,j}N_j + \sum_i N_{i-1} \sum_{j=1}^{i-2} \frac{2^{j-i+1}}{2^{j-i+1}+1}\beta_{i-1,j}M_j \\
-\sum_i N_i \sum_{j=1}^{i-1} \frac{2^{j-i}}{2^{j-i}+1}\beta_{i,j}M_j + \sum_i \beta_{i-1,i-1}N_{i-1}M_{i-1} + \sum_i N_i \sum_{j=1}^{i-1}\beta_{i,j}M_j - \sum_i M_i \sum_{j=i}\beta_{i,j}N_j = 0.
\end{aligned}
$$

Manuscript received Apr. 17, 2005, and revision received Nov. 4, 2005.